# ELECTROCHEMICAL REMOVAL OF HEAVY METAL IONS FROM SIMULATED INDUSTRIAL EFFLUENTS

**A** Thesis

Submitted to the College of Engineering of Nahrain University in Partial Fulfillment of the Requirements for the Degree of Doctor of Philosophy in Chemical Engineering

by

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#### Abstract

This investigation represents an experimental study which was conducted on a flow-by fixed bed electrochemical reactor in which electrolyte is perpendicular to the current flow direction.

The experiments were performed in two types of cells (1) Non-flow system cell (2) Flow system cell, using a porous bed of highly conductive mesh brass of 50 mesh no. (50x50 pore in inch<sup>2</sup>), in order to design and construct a once through (single compartment) electrochemical reactor to investigate the removal and recovery of copper (Cu<sup>+2</sup>), nickel (Ni<sup>+2</sup>), and cadmium (Cd<sup>+2</sup>) metal ions, concentration profile with deposition time, and also to find and calculate the current efficiency for removal of metal ions.

The electrochemical reduction (deposition) of  $Cu^{+2}$  ion only to its metallic form on single, double, and triple meshes was conducted in the non-flow system cell at different total applied currents, and deposition time (0.5, 1.0, and 1.5 h) in 0.5 M sulfuric acid as supporting electrolyte at 40  $^{\circ}C$  to study the effect of these parameters on removal of metal ions and current efficiency.

The electrochemical reduction (deposition) of  $Cu^{+2}$ ,  $Ni^{+2}$ , and  $Cd^{+2}$  metal ions to its metallic form on single, double, and triple meshes was conducted in the flow system at different total applied currents, flow rates of (100, 200, and 300 l/h), concentration ratio of Cu:Ni, Cu:Cd, and Cu:Ni:Cd for co-deposition, and withdrawn sampling time of (0.5, 1.0, 1.5, and 2.0 h) for single mesh (0.5, 1.0, and 2.0 h) for double and triple meshes in 0.5 M sulfuric acid as supporting electrolyte at 40  $^{0}C$ .

Results are presented on the electrodeposition of  $Cu^{+2}$ ,  $Ni^{+2}$ , and  $Cd^{+2}$  metal ions to their metallic form in terms of current efficiency. The application of more positive total applied current to the electrolytic cell resulted in the decrease in current efficiency values but for  $Cu^{+2}$  ions there is an increase in the current efficiency at potentials before hydrogen evolution. Also the increase in flow rate leads to increase in removal of metal, hence increase in current efficiency. It was found that as long as the deposition time gives an increase in removal and current efficiency.

The presence of  $Cu^{+2}$  ion with  $Ni^{+2}$  and  $Cd^{+2}$  as a co-deposition resulted an increase in current efficiency for metal ions removal. Also the increase in  $Cu^{+2}$  ion concentrations of (100, 200, and 300 ppm) resulted in a relative increase in current efficiency of metal ions removal. In addition, it was found that the addition of  $Ni^{+2}$  ion caused a slight increase in current efficiency of  $Cd^{+2}$  ion.

Three number of meshes were used in this work to investigate the best effective bed thickness. The results showed that the current efficiency increases with increasing the number of meshes just as for two meshes and the current efficiency decreases when a third one was used due to a bad electric field penetration and also the dissolution of metal ions is greater than the deposition of metal.

The Atomic Absorption Spectrophotometer technique was used to measure the remaining concentration of metal ions through different deposition sampling times in order to assess the remaining concentrations of the different metal ions in solution, and also the current efficiencies of each metal ion under all the above experimental conditions.

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# Notations

## Symbols

# Symbols Notations

Α	The electrode area
$A_e$	Cross sectional area of an electrolyte
$A_e$	The electrochemically active surface area per unit volume of the three dimensional material in eq. (3.24)
a	Specific surface area
à	Tafel constant ={ $(RT/\alpha zF)\ln i_o$ }
b	Tafel constant = $(-2.303RT/\alpha zF)$
b	Constant in eq. (2.19) = $(-RT/\alpha zF)$
С	Concentration
$C_o$	Inlet concentration
$C_s$	Concentration at the electrode surface
$C_b$	Bulk electrolyte concentration
$C_{Ob}$	Concentration of oxidize species at the bulk
$C_{Rb}$	Concentration of reduced species at the bulk
$C_{Os}$	Concentration of oxidize species at the surface
$C_{Rs}$	Concentration of reduced species at the surface
$C_{Ej}$	Current efficiency of species j
D	The diffusion coefficient of reacting ions
E	Electrode potential
$E^{o}$	Equilibrium electrode potential at standard condition
E <sub>eq.</sub>	Equilibrium electrode potential
F	Faraday's constant (=96487)
Ι	Total applied current
i	Current density

<i>i</i> <sub>a</sub>	Anodic current density
$i_c$	Cathodic current density
$i_l$	Limiting current density
$i_o$	Exchange current density
$K_m$	Mass transfer coefficient
$k_a$	Electrochemical rate constant for anodic reaction
$k_c$	Electrochemical rate constant for cathodic reaction
$k_a^o$	Electrochemical standard rate constant for anodic reaction
$k_c^o$	Electrochemical standard rate constant for cathodic reaction
L <sub>e</sub>	Resistance path
$M_{wt}$	Molecular Weight
М	Metal
$M^{+n}$	Metal ion
m	The mass electro-processed during the time interval $dt$
Ν	Molar flux
n	Positive exponent lower than one in eq. (2.18)
$n_i$	Number of moles of species i
0	Molecule of an oxidized species
Q	The amount of electricity passed through the cell
R	Molecule of reduced species in eq. (2.1)
R	Gas constant (=8.314)
$R_o$	Resistance of the electrolyte solution
Т	Temperature
t	Deposition time
V	The volume of affluent to be treated
	The volume of enfluent to be treated

W	Weight of metal deposited
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- $W_{th}$  Theoretical weight of metal deposited
- *z* Number of electrons transferred in the reaction

#### **Greek letters**

### Symbols Meaning

α	Charge transfer coefficient
$\delta_d$	Thickness of diffusion layer
З	Porosity
η	Overpotential
μ	Electrolyte viscosity
ρ	Electrolyte density
σ	Conductivity of the electrolyte solution

## Subscripts

<u>Symbols</u>	<u>Meaning</u>
a	Anode
b	Bulk
С	Cathode
d	Diffusion
E	Efficiency
i,j	Species
т	Mass transfer
0	Oxidized
0	Standard
R	Reduced
S	Surface or solution

### Abbreviations

## Symbols Meaning

AAS	Atomic Absorption Spectrophotometer
Co.	Company
Conc.	Concentration
Cont.	Continued
Exp.	Experiment
exp	Exponential
Μ	Molarity
No.	Number
SCE	Standard calomel electrode
UPD	Under potential deposition

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# Chapter One Introduction

#### **1.1 Definition**

Water pollution is one of the largest environmental problems facing society today. Each year, millions of tons of toxic pollutants are discharged into rivers, lakes, and oceans by industry. The major source for this discharge is the wastewater or process waters that are emitted in mass quantities as a result of industrial processes. After entering the environment, these pollutants adversely affect the quality of life, not only for flora and fauna that live next to these bodies of water but for human as well. Because of this, the treatment of wastewater is of utmost importance.

Wastewater containing heavy metal ions is generated in large quantities from the mining, ore processing, microelectronics, metal finishing and photographic industries. These water contain toxic Ag, Au, Cd, Co, Cr, Cu, Ni, Pb, and Zn ions in the range of 50 to 1000 parts per million (ppm). They must be adequately treated before discharged into waste streams or be recycled within a processing plant. The specific discharge concentration limits vary according to the size of the industrial operations and the type of the pollutants. They range from 0.1-0.3 ppm for Ag, Pb and Cd ions to 0.8-1.1 ppm for As and Au ions [1].

Over the past thirty years or so there has been a considerable resurgence of interest in the use of electrochemical routes in the chemical and process industries. However, electrochemical processes remain an active quite apart from their technical experience. Any device in which chemical reaction occurs directly due to the input of electrical energy can be defined as an electrochemical reactor. The term "electrochemical reactor" describes a device, which is more familiarly known as an "electrochemical activities has brought about a considerable gap between electrochemistry and chemical engineering. However, an electrochemical reactor

involves kinetics, heat and mass transfer and fluid flow, all of which are basic chemical engineering topics, but in electrochemical reactor there is an ionic transfer. The transfer of ions in an electrolyte solution arises from migration in the potential field and from mass transfer due to concentration difference in the solution [2].

Electrochemical systems find widespread technical applications. Industrial electrolytic processes include electroplating and refining, electropolishing and machining, and the electrochemical production of aluminum, chlorine, caustic soda, and other products, energy conversion in fuel cells and in primary and secondary batteries. Electrochemical corrosion should not be neglected. And some systems for desalting water involve electrochemical process. Electrochemical methods are used for qualitative and quantitative analysis. Idealized electrochemical systems are also of interest for studies of mass transfer processes and the mechanisms of electrode reactions and for the determination of basic data on transport properties [3].

The need to protect the environment from further contamination by transition and heavy metal ions is well recognized and universally reinforced by legislation which sets limits on the levels in effluents which may be discharged into sewers and local waters. All chemical plants, factories and other facilities employing solutions of such metals should therefore be treating their wastewater before discharge. Electrochemical methods compete with a number of other technologies including evaporation, precipitation, ion exchange and solvent extraction to offer solutions to the needs of the many industries involved. Electrochemical methods, however, are uniquely capable of recovering pure metal for recycle with a high purity approaching 99.99% of metal that is recovered [4].

Although electrochemical technology for metal ion removal has been available for some time, it continues to develop to meet the challenges of lower consent levels and more complex effluent compositions. Moreover the technology now on the market is based on diverse electrochemical concepts [5].

Due to the ecological and economic problems they cause, recovery of heavy metals (Cu, Pb, Zn, Ni, Cd and Cr) from dilute aqueous solutions is necessary.

2

These metals are produced as industrial wastes by electroplating industries and by metal extraction.

From an ecological standpoint, effluents with even low concentrations of heavy metal ions are extremely toxic and can cause several types of pollution, such as mud formation, exterminate aquatic life and create hazards to human health.

The methods normally used for metal ion recovery are precipitation as hydroxides, sulphides and oxalates, ion exchange by chemical or electrochemical means, reverse osmosis, chemical or physical adsorption, chemical reduction and biochemical remediation [6]. As an alternative to conventional methods, an electrochemical process was developed. This process can be described as a clean technology, as permanent residues are absent. From an economic point of view, the electrochemical method may offer some advantages, such as labour reduction, partial or total elimination of stock areas and reintegration of metals into the main process (due to the high degree of purity of the metal obtained).

However, for metal electrodeposition from dilute solutions, the plain electrode was inadequate because of poor mass transfer rates. Therefore, the electrolytic removal and recovery of metal ions from dilute solutions requires the use of electrodes with large surface areas, like the three-dimensional particulate electrode. Although the fixed bed reactor has been reported to be the most efficient for metal electrodeposition [8], it has the disadvantage that its porous matrix becomes clogged with metallic deposit, which renders continuous operation of the process impossible. However, with dilute solutions the operational time becomes so long that use of the porous matrix as a refill may be justified.

#### **1.2 Types of Electrochemical Reactors**

There are several types of electrochemical reactors, which can be considered for specific processes [9] as shown in Fig. 1-1. Among them are the fixed bed and fluidized bed reactor Fig.1-1, e and f.



Figure 1-1 Types of electrochemical reactors.

A good deal of attention has been given in recent years to the use of particulate electrodes in electrochemical reactors. These consist of beds of conducting particles through which electrolyte solutions flow [2].

The porous beds can be used as electrodes in monopole, bipole, and divided or undivided reactors. The "bed" can consist of particles, screens, felts, fabrics or reticulate materials. Electrolyte is pumped through the bed parallel or perpendicular to the current. Fixed bed reactors are well suited to processing multiphase electrolyte and give good gas-liquid, liquid-liquid, and liquid-solid mass transport.

Electrodes which are porous to the electrolyte solution are presently of great technical importance. They provide a very large electrode bed area in proportion to their size and this is several times greater in magnitude than that for non-porous structure [9].

Flows through porous electrodes seem to have several potential applications for example in electro-recuperation of metals from dilute solutions. Fixed bed electrodes have been proposed as concentrating systems for such solutions and fluidized electrodes could be used for continuous electrowining and electrorefining operations. There could be advantages in carrying out electrochemical reactions without phase change and requiring low current densities in fixed flow through porous electrodes.

The high specific interfacial area of such electrodes allows them to act as three dimensional electrodes and in comparison with the classical planer electrodes reduces the investment cost. The maximum productivity would be obtained for operating conditions at limiting current. Consequently it seems interesting to understand the behavior of flow through fixed electrodes for such operating conditions particularly their efficiency and the local metal potential distribution. The latter has to be known in order to avoid the discharge of undesirable ions inside an electrode [10].

#### **1.3 The Scope of Present Work**

The present work deals with studying experimentally the electrochemical reduction (electrodeposition) of (Cu, Ni, and Cd) ions from their salts: copper sulfate (CuSO<sub>4</sub>.5H<sub>2</sub>O, M.wt.=249.68), nickel sulfate (NiSO<sub>4</sub>.6H<sub>2</sub>O, M.wt.=262.85), and cadmium sulfate (3CdSO<sub>4</sub>.8H<sub>2</sub>O, M.wt.=769.5) respectively in acidic media (0.5 M H<sub>2</sub>SO<sub>4</sub>) to their metals. The real objective of this work is merely to meet the legal limit on discharge or also to recover and recycle the metal and also to form such alloy from above ions that is needed in industry. Experimental work was divided into two parts:

a) Non-flow electrodeposition system for copper deposition only.

b) Flow electrodeposition system which was conducted in fixed bed of mesh cylindrical electrode (working electrode) constructed of highly conducting material (Brass) as cathode and cylindrical graphite (surrounded the cathode) as anode. The electrochemical reactor of a concentric flow-by electrode was used to deposit  $Cu^{+2}$  ion to Cu metal, Ni<sup>+2</sup> ion to Ni metal, and Cd<sup>+2</sup> ion to Cd Metal, first individually, double co-deposit ( $Cu^{+2}$ -Ni<sup>+2</sup>,  $Cu^{+2}$ - Cd<sup>+2</sup>) and finally triple co-deposit ( $Cu^{+2}$ -Ni<sup>+2</sup>-Cd<sup>+2</sup>) on cathode by using single, double, and triple meshes as cathode, with 200 ppm initial  $Cu^{+2}$ , Ni<sup>+2</sup>, and Cd<sup>+2</sup> ion concentration for single metal deposition and (100, 200, and 300 ppm) initial  $Cu^{+2}$  ion concentration, in the presence of sulfuric acid (0.5 M) as supporting electrolyte, all experiments were conducted at constant temperature of 40 °C.

The objective of present work can be described as follows:

- 1.To design and construct an electrochemical reactor of flow-by mesh electrode by using available materials compatible with original researches.
- 2.To determine the best bed electrode thickness (no. of meshes) to get best current efficiency, applied total current, initial concentration of metal ions, and time.
- 3.To study the effect of different flow rates, applied total current, sampling time, and initial concentration on current efficiency of removal of heavy metals.
- 4. To determine the current efficiency and concentration profile with time.

# Chapter Two General Aspects of Electrochemical Reactor

#### **2.1 Scope of Electrochemical Industries**

Electrochemistry has been classically defined as the science of the chemical changes caused by the electric current and the production of electricity from the energy of chemical reactions [11].

Industrially, however, the chemical and physical changes produced by the electric current are far more important.

Electrochemical engineering is a branch of chemical engineering upon which parts and aspects of electrical engineering and metallurgy have been grafted. Electrochemical engineering concerns itself, not only with all the topics generally considered as electrochemical, but also with entire furnishing of power to the industries, utilization of that power; the design, construction and operation of the equipment machinery and plants; the economic considerations in the competition of chemical and electrochemical methods for the same or similar products, as well as their sale, distribution and consumption [12].

Electrochemical industries may be roughly divided into:

- (1) Those of an electrolytic nature, further subdivided into those of
  - a) The electroseparation type, represented by alkali and chlorine.
  - b) The electrowinning type, as represented by the copper or zinc industries in which ore is leached.
  - c) The electorefining group, of which electrolytic copper or nickel are examples.
  - d) The electrodeposition type, as electroplating and electroforming, and
  - e) A section which, as the result of oxidation or reduction, concerns chlorites, peroxides, metallic oxides, as well as organic materials.
- (2) The fused electrolyte group represented by aluminum, magnesium and the alkali metals.
- (3) The electrothermal group, examples of which are calcium carbide, cyanamid, graphite and the synthetic abrasives.
- (4) The electro furnace ferroalloy and special alloys.
- (5) Those industries in which the primary interest is concerned with the electric current effecting reactions in the gaseous phase.
- (6) The batteries group, of both the primary and secondary type, as well as corrosion and the stability of structures and systems. Table 2-1 gives a list of the major products of the electric furnace and electrolytic cell [11].

Increasing use of electrochemical processes is expected as electrochemical techniques become better and more widely understood and as the economics of electrochemical processing improve through use of larger and more efficient cells, continuation of the decrease in cost of electrical energy over the long term relative to other forms of energy or to chemical oxidizing and reducing agents would add further impetus to use electrochemical processes [2].

Product	Raw material	Elec- tric furnace	Fused electro- lyte	Electro- lytic cell	Application
Alumina, fused	Bauxite (natural alu-	x		• • •	Abrasives and refrac-
Aluminum, pure	minum oxide) Aluminum metal		x	•••	tories Corrosion-resistant
Aluminum metal	Bauxite	••••	x		coatings Electric-power cable;
					alloys, deoxidizing agent for steel; mate-
Ammonium persulfate	Ammonium bisulfate	•••		x	rial of construction Oxidizing and bleach
Antimony	solution Antimony ores			x	ing agent, batteries
Barium	Fused barium		x	· · · ·	Alloys, electron emis-
Beryllium alloys	Beryllia		x		sion
Bismuth	Lead refining slimes	• • • •		x	Alloys
Boron carbide	Anhydrous boric acid and coke	x		• • •	Abrasive
Cadmium	Zinc electrowinning slimes		• • •	x	Alloys, plating
Calcium	Calcium chloride	•••	x ,	• • •	Deoxidizers, CaH2,
				40 40	radio tubes, lamps. allovs
Galeium carbide	Lime and coke	x	•••		Acetylene for welding
			×		acetone, acetic acid,
Colsium announde	Calaium carbida	x	De la constante de		synthetic resins
Calcium cyanamide	(nitrogen of the air)				Fertilizer, ammonia,
Calcium cyanide	Cyanamide and salt	x			Cyanidation of ores,
Carbon bisulfide	Coke and sulfur	x			Solvent, insecticide,
					artificial silk (viscose)
Caustic	Water, salt	•••		x	Soap, paper industry
Cerium metals	Rare earth chlorides	* * *	x		Pyrophoric alloys, auto-
			414.5		bullets and shells
Chlorine gas	Water, salt	• • •		x	Bleaching. organic
					chemicals. metal
· ·					chlorides, water puri-
• •					minum chloride for
				H	oil refining
Chromic acid	Oxidation of chro- mium sulfate solu-	•••		x	Chemicals, oxidizing
	tions				agent
Chromium	Chromic acid and sulfuric acid solu-		• • •	x	Plating coatings
Chromium metal	tion Complex chromium	• • •		x	Alloying agent
Cobalt	ores Complex cobalt ores			x	
Copper, pure	Copper ore	• • •		x	Alloying agent
	Crude copper	•••		x	industries
Cuprous oxide	Copper	•••		x	Paint pigment

 Table 2-1 Products of electric furnace and electrolytic cell [11]

Electro-Elec-Fused Raw material lytic Application Product tric electrofurnace cell lyte Deuterium..... Water . . . . . . x Chemical for scientific use Ferroalloys..... Ores, carbon and x Alloys, addition agents silicon to steel, stainless steels Fluorine..... Fused potassium x . . . Chemical acid fluoride or anhydrous HF Gold..... Copper refining . . . . . . x Jewelry, coinage, indusslimes trial alloys Graphite..... Coal and coke x . . . Electrodes, lubricants, and paints Hydrogen..... Water, NaOH . . . ... x Ballooning, hydrogenated fats Hydrogen peroxide ..... Sulfate solutions ... X Chemicals, antiseptic, bleaching, oxidizing agent Hypochlorite..... Water, salt ..... x Disinfectants, bleaches Indium..... Indium oxide in sul-. . . 3 Nontarnishable silver furic acid solution alloy, jewelry, television Iron, electrolytic..... Ferrous.ammonium ... .... X Electromagnetic sulfate solution purposes Iron, pig..... Iron ore, ilmenite x Steel industry Iron powder..... Iron ores or ferrous . . . X Powder metallurgy sulfate Lead, refined..... Crude lead Σ Alloys, fittings, acid chambers Lithium metal..... Lithium salts T Light alloys, reagent Magnesium metal..... Magnesium chloride, X Castings, sheet, cathodsea water ic protection anodes, lightweight alloys. fiares Manganese, electrolytic. Manganese ore ... Σ Ferrous and nonferrous alloys, stainless steels Manganese dioxide..... Manganese ore ... X Depolarizer for dry batteries Mercuric oxide..... Mercury x Catalyst and chemical Nickel, refined ..... Crude nickel X Alloys, plating industry, dairy equipment, utensils Nickel powder..... Nickel x Powder metallurgy Oxygen..... Water, NaOH . . . X Oxywelding. oxycutting Ozone..... Air x Sterilization of water, sanitation Palladium..... Nickel refining slimes x Industrial allovs Perborates..... Borax x Bleaching agents for textiles Perchloric acid..... Hydrochloric acid x Salts of perchloric acid Phosphoric acid..... Phosphate rock, x Acid phosphates. coke, and sand cleaners, food products Phosphorus..... Phosphate rock, х Matches, phosphorus coke, and sand compounds, phosphor bronze, smoke screens

Table 2-1 Cont.

Table 2-1 Cont.

Platinum	Product	Raw material	Elec- tric furnace	Fused electro- lyte	Electro- lytic cell	Application
Potassium chlorate       Sillies       dustrial alloys         Potassium hydroxide.       Potassium chloride        x         Potassium perchlorate.       Sodium chlorate       soan, chemicals, e         Potassium perchlorate.       Sodium chlorate       solution and con-         verted to potassium       salt       x       Oxidizing agent, bk         Quartz, fused       Quartz rock       x        x         Rhodium       Nickel refining slimes        x       Industrial alloys         Rubber       Latex containing the        x       Industrial alloys         Silicon carbide       Sand and coke       x        x       Industrial alloys.         Silicon       Latex containing the        x       Household and ir       trial rubber goods         Silicon       Sand and coke       x        x       Silicon steel, hydr         Silicon carbide       Sand and coke       x        x       Silicon steel, hydr         Sodium chlorate	Platinum	Nickel refining			x	Catalyst, jewelry, in-
Potassium hydroxide.       Potassium chloride        x       Soap, chemicals, e         Potassium perchlorate.       Sodium chlorate       sodium chlorate        x       Soap, chemicals, e         Potassium perchlorate.       Sodium chlorate        x       Oxidizing agent, e       sives, medicine         Potassium persulfate.       Potassium sulfate        x       Oxidizing agent, bls       sives, medicine         Quartz, fused.       Quartz rock       x        x       Oxidizing agent, bls         Rubber.       Latex containing the usual components for compounding rubber       x        x       Industrial alloys, trial alloys, trial silicides, si trial alloys for halloons, resist units, silicides, si terrachloride         Silicon carbide.       Sand, and coke       x        x       Silicon steel, hydr for halloons, resist units, silicides, si terrachloride         Sodium chlorate.       Sodium chloride        x        x       Silicon steel, nydr for halloons, resist units, silicides, si terrachloride         Sodium chlorate.       Sodium chloride         x       Silicon steel, nydr for halloons, resist units, silicides, si terrachloride         Sodium chlorate.       Sodium chloride         x       <	Potassium chlorate	Potassium chloride			x	dustrial alloys Primers, matches, dye-
Potassium perchlorate.       Sodium chlorate solution and converted to potassium salt       x       Oxidizing agent. e sives, medicine         Potassium persulfate.       Potassium sulfate solution       x        x       Oxidizing agent. bit ing         Quartz fused.       Quartz rock       x        x       Silica tubes, heat-resisting material optical uses, lens, ling         Rubber.       Latex containing the usual components for compounding rubber       x        x       Household and in trial rubber goods         Silicon.       Sand and coke       x        x       Bileicon steel, hydr for halloons, resist units, silicides, si tetrachloride coke         Silicon carbide.       Sand, sawdust, and coke       x        x       Jewerly, coinage, in trial alloys         Sodium chlorate.       Sodium chloride solutions       x        x       Jewerly, coinage, in trial alloys         Sodium chlorate.       Sodium salts.        x        x       Jewerly, coinage, in trial alloys         Sodium perchlorate.       Sodium salts.        x        x       Jewerly, coinage, in trial alloys.         Sodium chlorate       Sodium salts.        x        x       Jewerly, coinage, in trial alloys.	Potassium hydroxide.	Potassium chloride	147 ANI		x	ing Soap, chemicals, elec-
solution and con- verted to potassium salt       sives, medicine         Potassium persulfate       Potassium sulfate solution        x         Quartz, fused       Quartz rock       x        x         Rhodium       Nickel refining slimes       x        x       Industrial alloys, ing         Rubber       Latex containing the usual components for compounding rubber       x        x       Household and ir trial rubber goods         Silicon carbide       Sand, sawdust, and coke       x        x       Silicon steel, hydr for halloons, resist units, silicides, si tetrachloride         Silver       Sodium chloride solutions       x	Potassium perchlorate.	Sodium chlorate	No. And and a		x	troplating Oxidizing agent, explo-
saltxOxidizing agent, bbPotassium persulfateguartz, fusedyOxidizing agent, bbQuartz, fusedQuartz rockxSilica tubes, heat-resisting material optical uses, least- resisting material alloys solitonsSilicon carbideSand and cokexXSilicon carbideSand, sawdust, and cokexXSilicon carbideSand, sawdust, and cokexXSolium chlorateSodium chloride solutionsxyySodium metalSaltxYySodium perchlorateSodium, salts, NaClO3xMetal joiningTantalumForassium tantalum fuoride Impure tin, tin	pirteit a l	solution and con- verted to potassium	internation	1.1.1		sives, medicine
Solution Quartz, fused.Solution Quartz rockingQuartz, fused.Quartz rockxQuartz, fused.Nickel refining slimesxRubber.Latex containing the usual components for compounding 	Potassium persulfate	salt Potassium sulfate			x	Oxidizing agent, bleach-
Rhodium.       Nickel refining slimes       x       resisting material optical uses, lens, industrial alloys, ing         Rubber.       Latex containing the usual components for compounding rubber       x       Household and ir trial rubber goods         Silicon.       Sand and coke       x       X       Silicon steel, hydr for halloons, resist units, silicides, si tetrachloride         Silicon carbide.       Sand, sawdust, and coke       x       X       Silicon steel, hydr for halloons, resist units, silicides, si tetrachloride         Siliver.       Copper refining slimes       x       X       Jewelry, coinage, in trial alloys         Sodium chlorate.       Sodium chloride solutions       x       Yeed killer, explos matches, oxidizing agent agent alloys, tetraethyl organic synthesis         Sodium perchlorate.       Sodium, salts, NaClO, Import in-lead alloys       x       Metal joining alloys         Tantalum.       Potassium tantalum fluoride thoride thoride solutions       x       Acid-resistant lining chemical equipme tin, tin dross       x       Tin, refined.	Quartz, fused	solution Quartz rock	x			ing Silica tubes, heat-
Rhodium	naster. Naster: Constant	· · · · · ·				resisting materials.
RubberLatex containing the usual components for compounding rubberxHousehold and ir trial rubber goodsSiliconSand and cokex	Rhodium	Nickel refining slimes	• • • •		x	Industrial alloys, plat-
for compounding rubber       for compounding rubber       that Pubber goods         Silicon       Sand and coke       x       x       Silicon steel, hydr for balloons, resist units, silicides, si tetrachloride         Silicon carbide       Sand, sawdust, and coke       x       x       Abrasives and refu- tories         Silver       Copper refining slimes       x       y       Jewelry, coinage, in trial alloys         Sodium chlorate       Sodium chloride solutions       x       x       Weed killer, explos matches, oxidizing agent elloys, tetrachly il organic synthesis         Sodium perchlorate       Sodium salts, NaClO;       x       X       Metal joining         Tantalum       Potassium tantalum fuoride        x       Metal joining         Thorium       Fused potassium thorium fuoride       x        x       Acid-resistant linin, chemical equipme         Tin, refined       Impure tin, tin dross        x       Tin-plate industry,	Rubber	Latex containing the usual components			x	Household and indus-
SiliconSand and cokexSilicon steel, hydr for halloons, resist units, silicides, si tetrachlorideSilicon carbideSand, sawdust, and cokexSilicon steel, hydr for halloons, resist units, silicides, si tetrachlorideSilicon carbideSand, sawdust, and cokexSilicon steel, hydr for halloons, resist units, silicides, si tetrachlorideSilicon carbideSand, sawdust, and cokexSilicon steel, hydr for halloons, resist units, silicides, si tetrachlorideSilicon carbideSand, sawdust, and cokexXSilicon carbideSand, sawdust, and cokexXSilicon carbideSodium chloridexXSodium chlorateSodium chlorideXWeed killer, explos matches, oxidizing agentSodium metalSaltxYSodium perchlorateSodium salts, NaClO1XYSolderImpure tin-lead alloysXXTantalumPotassium tantalum fuorideXXThoriumFused potassium thorium fluorideXXTin, refinedImpure tin, tin drossXXTin, peinedImpure tin, tin drossXX	Contra	for compounding	6 A 31-00	1.1.1.1		trial rubber goods
Silicon carbideSand, sawdust, and cokexfor balloons, resist units, silicides, si tetrachlorideSilver.Copper refining slimesxSodium chlorateSodium chloride solutionsxJewelry, coinage, in trial alloysSodium metalSaltxWeed killer, explos matches, oxidizing agentSodium perchlorateSodium salts, NaClO1xPeroxides, cyanide: alloys, tetraethyl i organic synthesisSolderImpure tin-lead alloysxMetal joiningTantalumPotassium tantalum fuoridexAcid-resistant lining chemical equipme Electron emission, X-ray targetsTin, refinedImpure tin, tin drossxTin-plate industry,	Silicon	Sand and coke	x		subjects	Silicon steel, hydrogen
Silicon carbide.Sand, sawdust, and cokextetrachlorideSilver.Copper refining slimesxJawelry, coinage, in toriesSodium chlorate.Sodium chloride solutionsxYeed killer, explos matches, oxidizing agentSodium metal.SaltxYeed killer, explos matches, oxidizing agentSodium perchlorate.Sodium salts. NaClO;xxPeroxides, cyanides alloys, tetracthyl i organic synthesisSolder.Impure tin-lead alloysxMetal joiningTantalum.Potassium tantalum fuoridexAcid-resistant lining chemical equipme thorium fluorideTin, refined.Impure tin. tin drossxTin-plate industry,	1. Juni 19	a stituent	and pro-	Sec. 1	2 <sup>16</sup>	for balloons, resistance units, silicides, silicon
Silver.Cove slimestoriesSodium chlorate.Sodium chloride solutionsxJewelry, coinage, ir trial alloysSodium chlorate.Sodium chloride solutionsxWeed killer, explos 	Silicon carbide	Sand, sawdust, and	x			Abrasives and refrac-
Sodium chlorateSodium chloride solutionstrial alloysSodium metalSaltxWeed killer, explos matches, oxidizing agentSodium metalSaltxPeroxides, cyanides alloys, tetraethyl i organic synthesisSodium perchlorateSodium salts. NaClO2xxYSolderImpure tin-lead alloysxMetal joiningTantalumPotassium tantalum fluoridexAcid-resistant lining chemical equipme Electron emission, X-ray targetsTin, refinedImpure tin, tin drossxXTin-plate industry,	Silver	Copper refining			x	tories Jewelry, coinage, indus-
Sodium metal.Saltxagent agent alloys, tetraethyll organic synthesis FireworksSodium perchlorateSodium salts, 	Sodium chlorate	Sodium chloride solutions	in the second		x	trial alloys Weed killer, explosives, matches, oxidizing
Sodium perchlorateSodium salts, NaClO2xrerokides, cyanides alloys, tetraethyl organic synthesisSolderImpure tin-lead alloysxMetal joiningTantalumPotassium tantalum fluoridexMetal joiningThoriumFused potassium thorium fluoridexAcid-resistant lining chemical equipme Electron emission, X-ray targetsTin, refinedImpure tin, tin drossxxTin-plate industry,	Sodium metal	Salt	1.111 2.24	x		agent Baravides exercides
Sodium perchlorate       Sodium salts. NaClO3       x       Fireworks         Solder       Impure tin-lead alloys       x       Metal joining         Tantalum       Potassium tantalum fluoride       x       Acid-resistant lining chemical equipme         Tin, refined       Impure tin. tin dross       x       x	availa 1-	Stars.	in ran a			alloys, tetraethyl lead,
SolderImpure tin-lead alloysxMetal joiningTantalumPotassium tantalum fluoridexAcid-resistant linin, chemical equipmeThoriumFused potassium thorium fluoridexElectron emission, X-ray targetsTin, refinedImpure tin, tin drossxx	Sodium perchlorate	Sodium salts. NaClO		inter "	<b>X</b>	Fireworks
TantalumPotassium tantalum fluoridexAcid-resistant linin, chemical equipme Electron emission, X-ray targetsTin, refinedImpure tin, tin drossxx	Solder	Impure tin-lead alloys			x	Metal joining
Thorium     Fused potassium     x     Chemical equipme       Tin, refined     Impure tin, tin dross     x     X	Tantalum	Potassium tantalum fluoride	• • •		x	Acid-resistant lining for
Tin, refined     Impure tin, tin dross     X     X-ray targets       X     Tin-plate industry,     X	Thorium	Fused potassium		x		Electron emission,
	Tin, refined	Impure tin, tin dross			x	X-ray targets Tin-plate industry,
Titanium slag Ilmenite x Raw material for	Titanium slag	Ilmenite	x			bronzes Raw material for pig-
Tungsten Sodium tungstate x Alloys, plating, ele-	Tungsten	Sodium tungstate			x	ments and metal Alloys, plating, electric
Uranium	Uranium	Fused potassium		. x	And a second	bulbs Alloys
Zinc, pure	Zinc, pure	Zinc ore			x	Brass industry,
Zinc powder x galvanizing Powder metallurgy	Zinc powder	Zinc	Add to a		x	galvanizing Powder metallurgy

# 2.2 The Components and Operation of an Electrochemical Reactor

Any device in which chemical reactions occur directly due to the input of electrical energy can be defined as an electrochemical reactor. Consequently the term "Electrochemical Reactor" describes a device which is more familiarly known as an "Electrolyser", 'Electrolytic Cell" or "Electrochemical Cell".

With three apparently adequate names already available, it might seem strange to bring forward another one. The reason for doing so is because the word "Reactor" has definite chemical engineering implications [2].



A typical electrochemical reactor is shown schematically in Fig.2-1.

Figure 2-1 Schematic diagram of electrochemical reactor [2].

It consists essentially of two electronic conductors called electrodes immersed in a bath of electrically conducting liquid called the electrolyte or electrolytic solution. The electrodes are connected outside the bath to the terminals of a DC power supply. When an emf of a sufficient magnitude is applied electron transfer occurs between each electrode and the liquid, resulting in a flow of electricity in the external circuit and chemical reactions at each electrode. This phenomenon is referred to "Electrolysis".

The electric conduction in liquid phase is due to the motion of charged species within it called ions. These ions are made available from a chemical compound (an electrolyte) either by melting it or by dissolving it in an ionizing solvent; this solvent is most frequently water.

The potential difference between the two electrodes causes a movement of the negatively charged ions, the anions, towards the positive electrode or anode. Simultaneously the positively charged ions known as cations move towards the negative electrode or cathode [2].

In general, electrochemical process may be considered as being either cathodic process, or anodic process depending on the electrode at which the economically important reaction occurs. Most cathodic processes involve either electrodeposed of metal or electrolytic reduction of a constituent of the electrolyte in the presence of hydrogen formed at the cathode. Most anodic processes involve either the discharge of anion from solution at an essentially stable electrode (anode) or the dissolution of the anode itself [13].

# 2.3 Description and Classification of Electrochemical Reactors

Industrial electrochemical cells deal with a wide variety of liquid and gaseous reactants and products at temperatures from below ambient to 1000 °C or above. Electrolytes may be aqueous, organic and molten solid salt. The electrolyte may be very corrosive in combination with the reactants, products, or intermediates. The state of technology and material available changes with time. It is therefore not surprising to see a large variety of shapes and sizes of electrochemical cells [12,13].

The electrochemical reactor can be classified according to the type of electrode into: planar, rod, tube, mesh or screen and porous electrode. A classification depending on the electrode types is given in Table 2-2 [2].

Also electrochemical reactors can be classified according to the type of cell into: one-compartment cell, two-compartment and multi compartment cell. A classification [12] depending on the cell types is given in Table 2-3. In two compartment type, the cell consists of two chambers separated by diaphragm or membrane. Separation may be necessary because different compositions or pH are required for each electrode or in order to avoid transport of impurities or the product from one electrode to the other. A diaphragm may allow bulk flow or convection of solution from one compartment to the other as well as migration of ions and diffusion of various species.

An ion-exchange membrane allows transport of predominantly one ion species or charge. Typical diaphragms and membranes [13] are shown in Table 2-4.

Another classification of electrochemical reactors according to the method of electrical connection to the power supply is as shown in Fig.2-2 [2].

Туре	Example
l Planar	
A. Inert	Graphite anode in chlor—alkali cell Platinized titanium or ruthenium—oxide coated titanium anodes
	Platinum anodes in perchlorate cell iron cathodes in chlorate and perchlorate cells
	Device Transformers The Second
B Active consumable	Carbon anode in Hall-Heroult cell Copper anodes in refining cell
C Base for deposition	Mercury cathode in chlor—alkalı cell Aluminum cathode in Hall—Heroult cell Stainless steel cathodes in manganese metal cell Copper cathodes in refining cell
n en les re	Electroplated objects Carbon base for deposition of MnO <sub>2</sub>
	sada area of electro cheu
D. Phase shapper on	Anodizing aluminum
surface	Thin-tilm battery Solid-state coulometer
11 Mesh or screen	Steel cathode in diaphragm type Chlor—alkali cell Metal anodes in chlor—alkali cells

 Table 2-2 Types of electrodes by physical forms [2]



Table 2-2 Cont.

# Table 2-3 Types of cells [12]

Туре	Type Applications		Disadvantages or limitations	
I. One-Compartment A. Inert vessel, immersed electrodes	Electrorefining of Cu, Zn Electroplating, Electroforming, Anodizing, Batteries — lead—acid,	Simple Inexpensive Electrodes readily changed	Gases produced must be capable of dispersal in atmosphere Electrolyte, electrodes, reactants and products must not be affected by atmosphere	
B. Vessel is one electrode	Sodium chlorate Sodium perchlorate Leclanche cell (consumable case)	Simple Inexpensive Electrodes readily changed (except for Leclanche cell)		
C. Horizontal liquid-metal cathode 1. Mercury cell Cl <sub>2</sub> Brine Hg Amalgam to decomposer	Chlorine—caustic	Produces concentrated, high—purity sodium hydroxide	Large floor area Higher energy consumption per unit of product than diaphragm cell Must avoid mercury loss to environment	
C. Horizontal liquid-metal catnode (Cont'd) 2. Aluminum reduction cell	Aluminum reduction (A three-layer variation used in electrorefining aluminum)	The only cell design for producing aluminum that has reached commercial feasibility	Large floor area High capital cost	
D. Tube cell	Tetraaikyllead Plating wires	Good heat transfer Good current distribution		
II. Two compartment A. Open-diaphragm cell Convection parallel to current through diaphragm	Electrolytic manganese Electrorefining Ag, Au, etc. Electroplating (bagged anooes)	Controlled concentration and pH in each compartment Each electrode is exposed to appropriate solution Impurities from one electrode are prevented from reaching other	increased cost Plugging of diaphragms	

Table 2-3 Cont.

Туре	Applications	Advantages	Disadvantages or limitations
B. Closed-diaphragm cell Convection parallel to current Gases	Chlorine—caustic—hydrogen Hydrogen—oxygen	Same as A Separate collection of gasses	
C. Membrane cell Convection perpendicular to current + + + Flow	Oxidation and reduction Acid—base generator Electrodialysis Electrodimerization for adiponitrile	Electrolytes may be different and have only one common ion transported by membrane; complete separation of neutral reactants or products	
D. Cells for producing liquid metals—convection not desired Liquid metal Liquid metal		Separation of metal and chlorine products	High voltage drop Fouling of separators
II. Multicompartment Filter-press construction A. Frames Membranes or diaphragms Flow channels	Hydrogen—oxygen generation Hydrogen—oxygen fuel cell Otner fuel cell systems Electrodialysis Adiponitrile HCI electrolysis	Compactness Precision assembly Can operate under pressure Flow channels can be built in Adaptable to a wide variety of configurations with solid and porous electrodes, membranes, diaphragms and flow channels	Limited to inert or long-lived electrodes Long-time operation between disassembly required

Cloth Glass Polypropylene Polyvinyl chloride Fluorocarbon Felt Asbestos Plastic fiber Porous plastic Polyethylene Polyvinyl chloride Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Nonpermslective
Glass Polypropylene Polyvinyl chloride Fluorocarbon Felt Asbestos Plastic fiber Porous plastic Polyethylene Polyvinyl chloride Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Cloth
Polypropylene Polyvinyl chloride Fluorocarbon Felt Asbestos Plastic fiber Porous plastic Polyethylene Polyvinyl chloride Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Glass
Polyvinyl chloride Fluorocarbon Felt Asbestos Plastic fiber Porous plastic Polyethylene Polyvinyl chloride Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Polypropylene
Fluorocarbon Felt Asbestos Plastic fiber Porous plastic Polyethylene Polyvinyl chloride Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Polyvinyl chloride
Felt Asbestos Plastic fiber Porous plastic Polyethylene Polyvinyl chloride Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Fluorocarbon
Asbestos Plastic fiber Porous plastic Polyethylene Polyvinyl chloride Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Felt
Plastic fiber Porous plastic Polyethylene Polyvinyl chloride Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Asbestos
Porous plastic Polyethylene Polyvinyl chloride Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Plastic fiber
Polyethylene Polyvinyl chloride Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Porous plastic
Polyvinyl chloride Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Polyethylene
Fluorocarbon Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Polyvinyl chloride
Rubber Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Fluorocarbon
Porous ceramic Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Rubber
Aluminum oxide Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Porous ceramic
Metal mesh Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Aluminum oxide
Permselective Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Metal mesh
Homogeneous (sulfonated or aminated) Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Permselective
Styrene-divinyl,/benzene copolymer Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Homogeneous (sulfonated or aminated)
Polyethylene Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Styrene-divinyl,/benzene copolymer
Fluorocarbon Reinforced homogeneous Heterogeneous (particles in binder)	Polyethylene
Reinforced homogeneous Heterogeneous (particles in binder)	Fluorocarbon
Heterogeneous (particles in binder)	Reinforced homogeneous
Heterogeneous (particles in binder)	
· ·	Heterogeneous (particles in binder)

Table 2-4 Cell separator, diaphragm, and membranes [13]





(a) Monopolar connection and (b) Bipolar connection [2].

In Fig. 2-2 a, alternate electrodes are connected to the opposite power supply terminals giving a number of individual reactor units electrically in parallel with one another. This configuration is referred to as monopolar connection and in this system each unit operates at the same voltage, the total current being the sum of the individual unit currents. In practice a number of monopolar stacks will often be connected electrically in parallel so as to be compatible with the main electricity supply voltage.

The alternative configuration of Fig. 2-2 b is known as bipolar connection. Here only the end electrodes in the stack are connected to the power supply, electrical continuity being due to both intermediate electrolytic and electronic conduction. Each electrode except for those at each end thus functions as an anode on one face and as a cathode on the other. Thus every two adjacent electrodes and the intervening solution are a single unit as with monopolar connection. Each reactor unit is electrically in series with the others and the same current flows through every unit apart from leakage current, which occurs sometimes between every other electrode. The total voltage drop across a bipolar stack is equal to the sum of the individual unit voltages and connection of stacks in parallel to the power supply is frequently adapted.

Both monopolar and bipolar methods of connection are extensively used. The bipolar system is preferred because fewer electrical connections are required and less power dissipation occurs in the external circuit

Electrochemical reactors can be classified according to their operation mode into batch, plug-flow and continuous stirred tank electrochemical reactors.

The essential features of a batch process are well known. In a batch cycle the reactor is charged, electrolysis takes place and the reactor is emptied. This intermittence in operation makes the batch reactor unsuitable for high production rates, also during electrolysis process; conditions (i.e. concentrations and temperature) change with time and involve frequent adjustment of the voltage to maximize current efficiency.

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A plug flow electrochemical reactor is one in which there is no element of fluid flowing through mixers with any other element either in front of it or behind it. All fluid elements thus have equal residence times in the reactor.

Plug flow operation is most widely used particularly for high product rates. Usually plug flow is associated with a continuous process but it can occur in batch recirculating systems to overcome some inherent operational difficulties in batch systems. The continuous stirred tank electrochemical reactor (CSTER) is operated in a manner such that complete mixing of electrolyte solution is approximately obtained. This is achieved by mechanical agitation or sometimes by the stirring action of evolved gases.

In general CSTERs have a less compact form than their plug-flow equivalents and this can result in higher manufacturing costs and operating charges [2].

#### **2.4 Electrode Processes**

#### **2.4.1 The Electrical Double Layer**

The simplest electrical process possible at the interface between an electrode and an electrolyte solution is indicated in Fig. 2-3 a. A charge due to an excess or deficit of electrons at the metal surface electrostatically attracts ions of opposite charge sign on the solution side. These two charged layers are referred to as the electrical double layer.

The study of electrical double layers is an important area of electrochemistry because the charge distribution in a given system influences the electron transfer and the course of the electrochemical reaction [2].



Figure 2-3 The electrical double layer(a) Simplest model (b) Modern layer [2]

The modern idea of the double layer structure [14] is shown diagramatically in Fig. 2-3. It is composed of an adsorbed layer of oriented water molecules together with adsorbed anions referred to as specifically adsorbed ions held in a single layer with the metal atoms. The plane passing through the electrical centers of the specifically adsorbed ions is called the inner Helmholtz plane. Other ions, predominantly hydrated cations, are then held electrostatically in an outer layer called the outer Helmholtz layer. The plane through the centers of these nonspecifically adsorbed ions is referred to as the outer Helmholtz plane and the region bounded by the outer Helmholtz plane and the electrode is known as the compact double layer. The region between compact double layer and the bulk of the solution, where there is complete equality of opposite charge (electro-neutrality) is called the diffuse double layer.

#### **2.4.2 Electrode Potential**

The presence of electrical double layer leads to form an electric potential difference at the boundary of two phases, this potential difference is called the electrode potential [15]. It is difficult to obtain the absolute potential differences; therefore, the electrode potential can be related to that of reference electrode. This reference electrode must be capable of being brought rapidly to equilibrium and exhibits partial reversibility [2]. For simplicity let us consider an electrochemical process in which only one reaction occurs at each electrode. The electrode reaction can be expressed by:

$$O + ze \rightleftharpoons R \qquad \dots (2.1)$$

where: O represents a molecule of an oxidized species and R a molecule of reduced species. At equilibrium the forward and reverse reactions have identical rates so that no net current flows.

In this case, electrode potential is called the equilibrium potential. At standard conditions, the electrode potential is measured with reference to standard hydrogen electrode and tabulated as standard redox potentials at 25 °C, some of them are given in Table 2-5. The equilibrium potential depends on the pressure and temperature and the activities of compounds participating in the cell reaction and is given by the Nernst equation [15].

$$E_{eq.} = E^o - \frac{RT}{zF} \ln \frac{[R]}{[O]}$$
 ... (2.2)

The equilibrium of the reaction (2.1) is disturbed by altering the electrode potential, this phenomenon is called polarization, and the difference between the electrode potential when polarized and the equilibrium potential is called the overvoltage or overpotential [2,15].

$$\eta = E - E_{eq.} \qquad \dots (2.3)$$

Na/Na <sup>+</sup>	-2.713	$\operatorname{Sn}^{4+}$ + 2e $\rightarrow$ $\operatorname{Sn}^{2+}$	+0.154
Zn/Zn <sup>2+</sup>	-0.763	$Cu^{2+} + e \rightarrow Cu^{+}$	+0.167
Fe/Fe <sup>2+</sup>	-0.44	$Fe(CN)_{6}^{3-} + e \rightarrow Fe(CN_{6})^{4-}$	+0.356
ca/ca <sup>2+</sup>	-0.402	$I_3 + 2e \rightarrow 3I$	+0.535
Co/Co <sup>2+</sup>	-0.27	$Fe^{3+} + e \rightarrow Fe^{2+}$	+0.771
Ni/Ni <sup>2+</sup>	-0.23	$2Hg^{2+} + 2e \rightarrow Hg_{2}^{2+}$	+0.905
Sn/Sn <sup>2+</sup>	-0.141	$Tl^{3+} + 2e \rightarrow Tl^{+2}$	+1.25
Pb/Pb <sup>2+</sup>	-0.126	$Cr_{2}O_{7}^{2-} + 14H^{+} + 6e$	
Cu/Cu <sup>2+</sup>	+0.34	$\rightarrow 2Cr^{3+} + 7H_2O$	+1.36
Hg/Hg2+	+0.798	$HOBr + H^+ + 2e^2 \rightarrow Br^- + H_2O$	+1.33
Ag/Ag+	+0.799	$MnO_{2} + 4H^{+} + 2e \rightarrow Mn^{2+} + 2H_{2}O$	+1.236
		$Clo_{A}^{2} + 8H^{+} + 8e \rightarrow Cl^{-} + 4H_{2}O$	+1.35
Pt OH /02	+0.401	$PbO_{2}^{4} + 4H^{+} + 2e \rightarrow Pb^{2+} + 2H_{2}O$	+1.456
Pt I /I2	+0.536	$Clo_{2}^{2} + 6H^{+} + 6e \rightarrow Cl^{-} + 3H_{2}^{2}O$	+1.45
Pt Br /Br2	+1.066	HOC1 + $H^+$ + 2e $\rightarrow$ C1 <sup>-</sup> + $H_2O$	+1.50
Pt Cl <sup>-</sup> /Cl <sub>2</sub>	+1.358	$Mn^{3+} + e \rightarrow Mn^{2+}$	+1.51
-		$Ce^{4+} + e \rightarrow Ce^{3+}$	+1.610
$Cr^{3+} + e \rightarrow Cr^{2+}$	-0.41	$H_2O_2 + 2H^+ + 2e \rightarrow 2H_2O$	+1.77
$Ti^{3+} + e \rightarrow Ti^{2+}$	-0.37	$Co^{3+} + e \rightarrow Co^{2+}$	+1.842

Table 2-5 Standard redox potentials at 25 °C [2].

There are three types of polarization [2]:

#### a) Activation Polarization:

Activation polarization which arises due to phenomena associated with an electrode reaction. The essential feature of any electrode reaction is electron transfer across the electrode/solution interface but this process is only one in a sequence of reaction steps. The actual sequence is unique for any reaction but, typically, could include interface adsorption and desorption of reactants, products and intermediates together with surface diffusion and surface chemical reactions. The rate of the reaction, however, will be determined by the slowest step, known as the "rate-determining step". The rate-determining step represents the state of maximum energy for a system and the difference in energy between this state and the original state is known as the activation energy.

Activation polarization is characterized by slow electrochemical reaction and by low exchange current density  $i_o$ . The surface concentration does not differ much from bulk concentration.

The current which flows when the electrode is polarized cathodically represents the difference between the rates of the forward (cathodic) and reverse (anodic) reactions. The current density (current per unit area), i, which will be considered a positive quantity, which in this case is given by:

$$i = i_c - i_a \qquad \dots (2.4)$$

where  $(i_c)$  is the partial current density for the cathodic reaction and  $(i_a)$  that for the anodic reaction.

By analogy with chemical kinetics the rate of the forward reaction can be written as

$$\frac{i_c}{zF} = k_c C_{0_s} \qquad ...(2.5)$$

where  $k_c$  is the electrochemical rate constant and  $Co_s$ , is the concentration of (*O*) at that point close to the electrode surface where (*O*) is discharged. In an analogous way the rate of the reverse reaction is:

$$\frac{i_a}{zF} = k_a C_{R_s} \qquad \dots (2.6)$$

where  $k_a$  and  $C_{R_s}$  have corresponding meanings to those above.

Consequently we can substitute Eqs. (2.5 and 2.6) into Eq. (2.4) and obtain

$$i = zFk_c C_{O_s} - zFk_a C_{R_s} \qquad \dots (2.7)$$

By using an Arrhenius type of rate constant/activation energy relationship [1], the rate constant  $k_c$  and  $k_a$  can be expressed in terms of the electrode potential, which is a measure of the free energy requirements by the formulae:

$$k_c = k_c^o \exp\left\{-\frac{\alpha z F E}{RT}\right\} \qquad \dots (2.8)$$

$$k_a = k_a^o \exp\left\{-\frac{(1-\alpha)zFE}{RT}\right\} \qquad \dots (2.9)$$

where  $k_c^o$  and  $k_a^o$  are standard rate constants referenced to some particular electrode potential. Consequently we can substitute Eqs. (2.8 and 2.9) into Eq. (2.7) and obtain

$$i = zFk_c^o \exp\left\{-\frac{\alpha zFE}{RT}\right\}C_{O_s} - zFk_a^o \exp\left\{\frac{(1-\alpha)zFE}{RT}\right\}C_{R_s}\dots(2.10)$$

at equilibrium  $E = E_{eq.}$ , the bulk concentrations of *O* and *R* denoted by  $C_{O_b}$  and  $C_{R_b}$  respectively are uniform throughout and i = 0 so that

$$zFk_c^o exp\left\{\frac{-\alpha zFE_{eq.}}{RT}\right\}C_{O_b} = zF k_a^o exp\left\{\frac{(1-\alpha)zFE_{eq.}}{RT}\right\}C_{R_b} = i_o \dots (2.11)$$

 $i_o$  is called the exchange current density and represents the rates of the forward and reverse reactions at equilibrium. Using  $i_o$  to eliminate the rate constants in Eq. (2.10) and recalling that the overpotential,  $\eta$ , is defined by:

$$\eta = E - E_{eq.} \qquad \dots (2.3)$$

we can easily manipulate Eq. (2.10) into the general form

$$i = i_o \left\{ \frac{C_{O_s}}{C_{O_b}} \exp\left(\frac{-\alpha z F \eta}{RT}\right) - \frac{C_{R_s}}{C_{R_b}} \exp\left(\frac{(1-\alpha) z F \eta}{RT}\right) \right\} \qquad \dots (2.12)$$

Eq. (2.12) serves as the general expression for the rate of an electrode reaction in terms of the electrode overpotential.

At very low overpotential surface concentrations of (O and R) do not differ much from their values in the bulk so that Eq. (2.12) can be very closely approximated by

$$i = i_o \left\{ exp\left(\frac{-\alpha z F\eta}{RT}\right) - exp\left(\frac{(1-\alpha)z F\eta}{RT}\right) \right\} \qquad \dots (2.13)$$

Eq. (2.13) is known as Butler-Volmer equation. It is expressed for the anodic reaction only in terms of overpotential and when  $\eta < 100$  mV, the second on the right hand side of Eq. (2.13) becomes negligible as compared with first term.

A further simplification can be achieved by approximating the individual exponential terms in Eq. (2.13) by the first two terms of their respective power series so that:

$$i = i_o \left\{ 1 + \left( \frac{-\alpha z F \eta}{RT} \right) - \left( 1 + \frac{(1 - \alpha) z F \eta}{RT} \right) \right\} \qquad \dots (2.14)$$

And

$$i = -i_o \frac{zF\eta}{RT} \qquad \dots (2.15)$$

Thus around  $\eta = 0$ , the current density is a linear function of the overpotential with slope numerically equal to  $(i_o zF/RT)$ . The negative sign in Eq. (2.15) reflects the fact that a cathodic overpotential is negative.

For appreciable cathodic polarization the reverse reaction is almost entirely suppressed and the second exponential term is very nearly zero. Consequently

$$i = i_o \exp\left(\frac{-\alpha z F \eta}{RT}\right) \qquad \dots (2.16)$$

It has been found experimentally that, for a great many reactions taking place with an appreciable current density and overpotential of more than 100 mV in absolute magnitude, the experimental curve  $(i = f(\eta))$  can be expressed by the following equations:

$$\eta = \dot{a} + b \log i \qquad \dots (2.17)$$

$$i = i_o \frac{zF}{RT} \alpha^{1-n} \eta^n$$
 ... (2.18)

Eq. (2.17) is called the Tafel equation,  $\dot{a}$  and b being known as "Tafel constants", where  $\dot{a} = \{(RT/\alpha_z F) \ln i_o\}$  and  $b = \{(-2.303RT/\alpha_z F)\}$  [2].

Eq. (2.18) is called power function of  $\eta$ , in which (*n*) would be a positive exponent lower than one and  $\alpha$  a dimensional coefficient [15].

Thus, the rate of an electrode reaction in term of the electrode overpotential is taking an exponential or power function form with overpotentials more than 100 mV in absolute magnitude.

#### b) Concentration Polarization:

Concentration polarization is caused by changes in the concentration of species participating in an electrode reaction. The early ideas about mass

transfer in electrochemical system were proposed by Nernst and Marriam [2] using an analogy with the dissolution of a solid. They suggested that mass transfer occurs solely by molecular diffusion through a thin layer of solution adjacent to an electrode. This layer has a linear concentration gradient across it and the outer edge is assumed to be maintained at the constant bulk concentration by migration and convection. Within this layer it was postulated that diffusion alone controlled the transfer of substances to the electrode. This layer has become known as "Nernst diffusion layer ( $\delta_d$ ) as shown in Fig. 2-4. The diffusion layer thickness  $\delta_d$  is influenced by the shape of the electrode, the geometry of the system, and by the velocity of the solution or agitation [16]. Concentration polarization refers to fast electrochemical reactions that are limited by mass transfer of species to and from electrode. In other words, concentration polarization occurs when one of the reactants is consumed on at an electrode faster than it can be supplied from the bulk of the solution and the rate of the reaction is limited by diffusion from the solution to the electrode surface [17]. As the current density is increased concentration of (O) decreases until it becomes so small that a substantially constant current is reached. This constant current density is called the limiting current density [13].



Figure 2-4 The Nernst diffusion layer [2].

At higher overpotentials the surface concentrations of (O and R) .differ much from their values in the bulk, therefore, the relation between rate of an electrode reaction and overpotential will be [13].

$$\eta = \dot{a} + \dot{b} \ln i + \dot{b} \ln \frac{C_{O_b}}{C_{O_s}} \qquad ... (2.19)$$

where  $\vec{b} = -RT/\alpha zF$ 

where the surface concentration (O) becomes so small that a substantially constant current density is reached which is called the limiting current density. In this case, the rate of an electrode reaction will depend on the mass transfer coefficient according to the following relation [2]:

$$\frac{i_l}{zF} = k_m C_{O_b} \qquad ... (2.20)$$

where  $i_l$  = the limiting current density, which represents the maximum rate of possible reaction for a given system, and

 $k_m$  = the mass transfer coefficient which is equal to  $(D/\delta_d)$ . also Eq.(2.20) can be written in another form:

$$i_l = \frac{DzFC_{0_b}}{\delta_d} \qquad \dots (2.21)$$

where D = the diffusion coefficient of reacting ions, and

 $\delta_d$  = the thickness of diffusion layer (Nernst diffusion layer).

The value of the concentration polarization depends on the limiting current; the latter depends on the concentration, temperature, and diffusion boundary layer thickness. For a particular electrode in any system, the diffusion layer thickness is dependent on the velocity of the solution past the electrode surface. As the velocity increases, the thickness of this layer decreases and the limiting current increases.

#### c) Resistance Polarization:

Resistance polarization refers to electrochemical reactions that are working under high reaction rate or current densities, or reaction using electrolyte with poor conductivity or high resistance. These conditions would result in a potential drop through a portion of the electrolyte surrounding the electrode, or through reaction product, i.e. films, on the surface of the electrode. This usually is called the ohmic potential drop, contribution to the polarization as [2].

Ohmic Potential Drop (IR Drop) = 
$$i \cdot R_o$$
 ...(2.22)

where i = reaction rate expressed as current density.

 $R_o$  = resistance of the electrolyte solution.

$$R_o = \frac{1}{\sigma} \frac{L_e}{A_e} \qquad \dots (2.23)$$

where  $\sigma$  = conductivity of the electrolyte solution.

 $L_e$  = resistance path (i.e., separation distance between electrode).

 $A_e$  = cross sectional area of an electrolyte.

The value of the ohmic potential drop is influenced by the conductivity of the electrolyte; the latter is usually a strong function of temperature and composition. This term of polarization is usually neglected in highly conductive solutions.

#### 2.4.3 The Rate of an Electrochemical Reaction

The two laws which express the relationships between the amounts of products formed during electrolysis and Faraday discovered quantities of electricity passed. These can be conveniently combined in the following statement: the passage of 96487 coulombs through an electrochemical reactor produces in total one gram equivalent of products at an electrode [2]. This quantity of electricity is called the Faraday constant and is designated by F.

In the case of a single reaction occurring at an electrode with  $n_j$  moles of j are produced by a current  $I_j$  flowing for t seconds.

$$n_j = \frac{l_j t}{z_j F} \qquad \dots (2.24)$$

where  $z_j$  = the number of electrons associated with the production of a molecule of *j*. For a number of reactions at an electrode we can write:

$$\sum \mathbf{n}_j = \frac{t}{F} \sum \frac{l_j}{z_j} \qquad \dots (2.25)$$

where  $\sum \mathbf{n}_j$  = the total number of moles produced.  $I_j$  now refers to the partial current associated with the production of a given species. The total current  $I_t$  is evidently given by:

$$I_t = \sum I_j \qquad \dots (2.26)$$

The current efficiency,  $C_{E_j}$ , for the production of a given species *j* when other reactions occur is defined by:

$$C_{E_j} = \frac{I_j}{I_t} \qquad \dots (2.27)$$

and is the ratio of the current producing *j* to the total current.

Practical current efficiencies are always quoted as percentage ratios, 100  $C_{Ej}$ , but Eq. (2.27) will be used as more convenient relationship.

Since  $I_j$  can not be measured directly, except for the obvious case when  $C_{E_j} = 1$ , a more useful expression for the current efficiency is obtained by eliminating  $I_j$  between Eqs. (2.24 and 2.27) to give

$$C_{E_j} = \frac{n_j}{I_t t/z_j F}$$
 ... (2.28)

 $(I_t t/z_j F)$  being the theoretical amount of *j* produced by the current  $I_t$  assuming no other reaction occurs. In principle  $C_{E_j}$  can be measured by chemical analysis [2].

#### 2.4.4 Space Time Yield

One of the most important quantitative criteria related to the performance of electrochemical reactors is the space time yield defined by [2].

$$Y_{st} = \frac{Amount \ of \ Product \ Obtained}{Unit \ Time \ \times \ Unit \ Cell \ Volum} \qquad \dots (2.29)$$

On basis of Faraday's law the space time yield of an electrolytic cell is given by the following expression:

$$Y_{st} = a \, \frac{i \, C_{E_j} \, M_j}{zF} \qquad ... (2.30)$$

In this equation the specific electrode area term (*a*) is only determined by cell construction, whereas the term ( $i C_{E_j} M_j / zF$ ) only depends on the properties of the reaction system.

With respect to the critical importance of capital costs for the realization of an industrial process, the space time yield should be as large as possible. If the magnitude of reaction term in Eq. (2.30) is limited by kinetic control or small concentration it follows that the specific electrode surface area must be increased to reach an acceptable space time yield.

According to such considerations, three dimensional electrode, (packed bed or fluidized bed electrode) must be used [18].

## **2.5 Description of Ionic Transfer Reactions**

The transfer of ions in an electrolyte solution arises from migration in the potential field and from mass transfer due to concentration differences in the solution

For slow electrochemical reactions, or for fast reaction occurring at very 10w current densities, differences in concentration of species between the bulks of the solution close to an electrode are small. Consequently mass transfer is not a dominant process in these instances [2].

A fast electrochemical reaction proceeding at a high current density, however, can be limited by the rate of mass transfer to the electrode surface.

Let us consider a fast metal-deposition reaction occurring at a limiting current density. Figure 2-5a shows an electrode with stationary electrolyte solution and a plot of the concentration distribution on the basis of Nernst's theory. The concentration gradient is assumed to give rise to diffusion, a process which occurs at a molecular level.

The depletion of metal ions at the electrode surface produces a density difference between the solution at the surface and that in the bulk. This density difference causes a bulk flow of concentrated solution towards the electrode, (for the corresponding anodic process, i.e. metal dissolution, the flow will be away from the electrode), and will increase the limiting current density.

The associated concentration distribution is shown in Fig. 2-5b and it will be seen that the concentration gradient is confined to a smaller region than in the case of diffusion alone. Of course, a diffusional process ultimately occurs very close to the electrode surface because there any motion ceases due to frictional force. However, natural convection gives a more efficient electrode process at a given current density because the overpotential is lowered.



Figure 2-5 Concentration profiles adjacent to an electrode with a fast reaction [2].

Natural convection is not associated with every fast electrode reaction because density differences do not necessarily occur, but in every case the most efficient supply of fresh concentrated solution is provided by enhancing the bulk motion. This is achieved by stirring the solution or by flowing it past the electrode. A typical concentration gradient is shown in Fig. 2-5c indicating a higher limiting current density compared with the previous cases. Once again, the ultimate process at the electrode surface is diffusional. However, a quantitative description of forced convection effects in any system requires the application of fluid mechanics [2].

# Chapter Three Technologies of Electrochemical Removal and Literature Survey

## **3.1 Introduction**

The electroplating industry generates a significant amount of heavy metalcontaining discharge water in its daily operation. Heavy metals are toxic to living organisms and tend to accumulate in the environment over a period of time [19].

The largest volume of waste water is generated in the rinsing operation which is applied to every workpiece to remove residual plating bath solution. Because of the large volume of rinsewater used in comparison to the residual solution, the metal ions are low in concentration. Effluent standards for heavy metal-containing discharge waters have been proposed by the Environmental Protection Agency for the metal finishing industry and are given in Table 3-1. Along with the metal cations, the associated anions, possibly cyanide, chloride, or sulfate, are also present [20].

Species	Concentration mg/l
CN <sup>-</sup> , total	1.9
Cu	4.5
Ni	4.1
Cr	7.0
Zn	4.2
Pb	0.6
Cd	1.2

**Table 3-1** Proposed maximum effluent standards for metal-finishingplants with effluent of 10,000 gpd or more [20].

Various standard technologies exist to remove these pollutants. Among others, chemical oxidation and reduction, ion exchange, electrodialysis, electrolytic activated-carbon adsorption, and reverse osmosis have been utilized for heavy metal and cyanide removal. Probably the most used for heavy metal removal is a precipitation reaction which generates metallic hydroxides or sulfides. A solid sludge results which in turn must be dealt with. Precipitation requires ancillary equipments such as mixing tanks, clarifiers, filters and an inventory of chemicals (e.g., caustic). Since the precipitates are usually colloidal, coagulating agents must be added in order to assure effective solids removal **[20]**.

#### **3.2 Electrochemical Reactors for Metal Recovery**

The electrochemical recovery of metals has been practiced in the form of electrometallurgy since long time ago. The earliest reported application of electrochemical phenomena in chemical subjects was supposed to be carried out in protecting iron with lead electroplating. The first recorded example of electrometallurgy was in mid-17th century in Europe. It involved the recovery of copper from cupriferous mine water electrochemically. During the past two and half centuries, electrochemical technologies have grown into such areas as energy storage, chemical synthesis, metal production, surface treatment, etc. [21].

The electrochemical mechanism for metal recovery is very simple. It basically is the cathodic deposition as

$$M^{+n} + ne \to M \tag{3.1}$$

#### **3.2.1 Typical Reactors Applied**

There are quite a few types of reactors found applications in metal recovery, from very basic reactors such as tank cells, plate and frame cells, rotating cells, to complicated three-dimensional reactor systems like fluidized bed, packed bed cell, or porous carbon packing cells, mesh or screen bed cells. Tank cells, Fig. 3-1, are one of the simplest and hence the most popular designs. It can be easily scaled up or

down depending on the load of a process. The electrode can be arranged in monopolar or bi-polar mode. The main application of this type of reactor system is the recovery of metals from high concentration process streams such as effluents from the electroplating baths, ethants, and eluates of an ion-exchange unit [22]. The number of electrodes in a stack may vary from 10 to 100. The water flow is usually induced by gravity.



Figure 3-1 Tank cell reactor [22].

The plate and frame cell or sometimes called filter press, as shown in Fig. 3-2, is one of the most popular electrochemical reactor designs. It conveniently houses units with an anode, a cathode, and a membrane (if necessary) in one module. This module system makes the design, operation and maintenance of the reactor relatively simple [23].



Figure 3-2 Filter press reactor [23].

In order to enhance mass transfer from the bulk to the electrode surface and also to remove the deposited metal powders from the cathode, the rotating cathode cell was designed and employed, as illustrated in Fig. 3-3. It was found that this system can reduce copper content from 50 to 1.6 ppm by using the systems in a cascade version [24].



Figure 3-3 Rotating cylinder electrode reactor [24].

Since the metal deposition happens at the surface of the cathode, it is necessary to increase the specific surface area in order to improve the space–time yield. Fluidized bed electrode was therefore designed as shown in Fig. 3-4. The cathode is made of conductive particles in contact with a porous feeder electrode. The electrode can give a specific area of  $200 \text{ m}^2/\text{m}^3$ . Because of the fluidization of the particles by the water flow, the electrical contact is not always maintained thus the current distribution is not always uniform and the ohmic drop within the cell is high. In order to improve the contact between the electrode feeder and cathode particles, a large number of additional rod feeders are used [25].



Figure 3-4 Fluidized bed reactor [25].

The packed bed cell overcomes sometimes the non-contacting problem met in fluidized bed, as shown in Fig. 3-5. Carbon granules were packed in a cell. The anode was separated by a diaphragm. This three-dimensional porous, carbon cathode provides 500 times more plating area than conventional two-dimensional cells. In order for dilute metal pollutants to deposit properly on the cathode, it is suggested to seed metal powders by having concentrated metal solution at the beginning of the recovery process. Control of pH in the feed tank of a recirculating electrolyte is important to avoid precipitation of the metal [26,27].



Figure 3-5 Fixed bed reactor [27].

#### 3.2.2 Application Area

The electrochemical recovery of metals can be used in the metal surface finishing industry. One has to bear in mind that it is unable to provide a complete solution to the industry's waste management problems because it cannot treat all the metals either technically or economically. The electrolytic recovery of metals here involves two steps: collection of heavy metals and stripping of the collected metals. The collection step involves plating and the stripping can be accomplished chemically or electrochemically. Nowadays, metal powders can be formed on the surface of carbon cathodes. Therefore, physical separation is sufficient. The metals recovered can be of quite high purity.

Another application is in the printed circuit board manufacturing industry. Because of the well-defined process, the treatment can be accomplished relatively easily for this industry. For dilute effluent, an ion-exchange unit can be used to concentrate the metal concentration. For high concentration streams, they can be treated directly using a recovery system as in metal surface finishing industry. Application of metal recovery should be very much useful in metal winning in mining industry especially in the production of precious metals such as gold [22]

# **3.3 Methods of Electrochemical Metal Recovery**

#### **3.3.1 Electrocoagulation (EC)**

Electrocoagulation involves the generation of coagulants in situ by dissolving electrically either aluminum or iron ions from respectively aluminum or iron electrodes. The metal ions generation takes place at the anode; hydrogen gas is released from the cathode. The hydrogen gas would also help to float the flocculated particles out of the water. This process sometimes is called electrofloculation. It is schematically shown in Fig. 3-6. The electrodes can be arranged in a mono-polar or bi-polar mode. The materials can be aluminum or iron in plate form or packed form of scraps such as steel turnings, millings, etc.

The chemical reactions taking place at the anode are given as follows.

For aluminum anode:

$$Al - 3e \rightarrow Al^{+3}$$
 ...(3.2)

at alkaline conditions

$$Al^{+3} + 30H^{-} \rightarrow Al(0H)_{3} \qquad \dots (3.3)$$

at acidic conditions

$$Al^{+3} + 3H_20 \rightarrow Al(OH)_3 + 3H^+$$
 ...(3.4)

For iron anode:

$$Fe - 2e \rightarrow Fe^{+2}$$
 ...(3.5)

at alkaline condition

$$Fe^{+2} + 30H^- \rightarrow Fe(0H)_3 \qquad \dots (3.6)$$

at acidic condition

$$4Fe^{+2} + O_2 + 2H_2O \rightarrow 4Fe^{+3} + 4OH^- \qquad \dots (3.7)$$

In addition, there is oxygen evolution reaction

$$2H_2O - 4e \rightarrow O_2 + 4H^+$$
 ...(3.8)

The reaction at the cathode is

$$2H_2O + 2e \rightarrow H_2 + 2OH^- \qquad \dots (3.9)$$

The nascent  $Al^{+3}$  or  $Fe^{+2}$  ions are very efficient coagulants for particulates flocculating. The hydrolyzed aluminum ions can form large networks of Al-O-Al-OH that can chemically adsorb pollutants such as  $F^-$  [28]. Aluminum is usually used for water treatment and iron for wastewater treatment. The advantages of electrocoagulation include high particulate removal efficiency, compact treatment facility, relatively low cost and possibility of complete automation.

One of the factors affecting electrocoagulation is the current density or charge loading. The supply of current to the electrocoagulation system determines the amount of  $Al^{+3}$  or  $Fe^{+2}$  ions released from the respective electrodes. A large current means a small electrocoagulation unit. However, when too large current is used, there is a high chance of wasting electrical energy in heating up the water. More importantly, a too large current density would result in a significant decrease in current efficiency. The current efficiency for aluminum electrode can be 120–140% while that for iron is around 100%. The over 100% current efficiency for

aluminum is attributed to the pitting corrosion effect especially when there are chlorine ions present. The current efficiency depends on the current density as well as the types of the anions [29].

Electrocoagulation is efficient in removing suspended solids as well as oil and greases. It has been proven to be effective in water treatment such as drinking water supply for small or medium sized community, for marine operation and even for boiler water supply for industrial processes where a large water treatment plant is not economical or necessary. It is very effective in coagulating the colloidal found in natural water so as to reduce the turbidity and color. It is also used in the removal or destruction of algeas or microorganisms. It can be used to remove irons, silicates, humus, dissolved oxygen, etc. [30].



Figure 3-6 Electrocoagulation unit [29].

#### **3.3.2 Electroflotation (EF)**

Electroflotation is a simple process that floats pollutants to the surface of a water body by tiny bubbles of hydrogen and oxygen gases generated from water electrolysis as shown in Fig. 3-7. Therefore, the electrochemical reactions at the
cathode and anode are hydrogen evolution and oxygen evolution reactions, respectively [31].



Figure 3-7 A typical electroflotation unit design [31].

The performance of an electroflotation system is reflected by the pollutant removal efficiency and the power and/or chemical consumptions. The pollutant removal efficiency is largely dependent on the size of the bubbles formed. For the power consumption, it relates to the cell design, electrode materials as well as the operating conditions such as current density, water conductivity, etc [32].

The size variation of the bubbles depends on water pH as well as the electrode material as shown in Table 3-2 [33]. The hydrogen bubbles are smallest at neutral pH. For oxygen bubbles, their sizes increase with pH. It should be noted, however, the cathode materials affect the size of the hydrogen bubbles, and so do the anode materials [33].

pН	Hydrogen (µm)			Oxygen (µm)	
	Pt	Fe	С	Pt	
2	45-90	20-80	18-60	15-30	
7	5-30	5-45	5-80	17-50	
12	17-45	17-60	17-60	30-70	

Table 3-2 The range of gas bubbles at different pH and electrode materials [33].

The gas bubbles depend also on the current density. The surface condition affects the particle size, too. The polished mirror surface of the stainless steel plate gives the finest bubbles. Besides size of bubble, the bubble flux, defined as the number of gas bubbles available per second per unit cross-section area of the flotation cell, also plays a role in mineral flotation, recovery of different sized particles. A decrease in gas bubble sizes was found with the increase in current density as shown in Table 3-3 [34].

Electrode	Current density (A/m <sup>2</sup> )					
	125	200	250	300	375	
Hydrogen gas l	bubbles d	iameter (µ	.m)			
SS plate	34	32	29	26	22	
200 mesh	39	35	32	31	28	
100 mesh	45	40	38	30	32	
60 mesh	49	45	42	40	37	
Oxygen gas bu	bbles dia	neter (µm	ı)			
Pt plate	48		46		42	
200 mesh	50		45		38	

**Table 3-3** The mean gas bubble size at different current densities and electrodematerials, pH 9 [34].

Also it was found that such a decrease in bubble size with increase in current density is true only at the low end of current densities. When the current density is higher than 200 A/m<sup>2</sup>, no clear trend can be observed with gas bubbles ranging from 20 to 38  $\mu$ m [**35**].

In the arrangement of the electrodes in electroflotation unit, an anode usually is installed at the bottom, while a stainless steel screen cathode is fixed at 10–50 mm above the anode. Such an electrode arrangement cannot ensure quick dispersion of the oxygen bubbles generated at the bottom anode into wastewater flow, affecting flotation efficiency. Moreover, if the conductivity of wastewater is low, energy consumption will be unacceptably high due to the large inter-electrode spacing required for preventing the short-circuit between the upper flexible screen cathode and the bottom anode [**36**].

For a conventional electrode system, only the upper screen cathode faces the wastewater flow, while the bottom anode does not interact with the flow directly. Therefore, the oxygen bubbles generated at the bottom anode cannot be dispersed immediately into the wastewater being treated. Consequently, some oxygen bubbles may coalesce to form useless large bubbles. This not only decreases the availability of the effective small bubbles, but also increases the possibility of breaking the flocs formed previously, affecting the flotation efficiency. When the anode and the cathode are leveled, such an open configuration allows both the cathode and the anode to contact the wastewater flow directly. Therefore, the bubbles generated at both electrodes can be dispersed into wastewater rapidly and attach onto the flocs effectively, ensuring high flotation efficiency [**37**].

## **3.3.3 Electrooxidation (EO)**

The study on electrooxidation for wastewater treatment goes back to the 19th century, when electrochemical decomposition of cyanide was investigated. Extensive investigation of this technology commenced since the late 1970s. During the last two decades, research works have been focused on the efficiency in oxidizing various pollutants on different electrodes, improvement of the electrocatalytic activity and electrochemical stability of electrode materials, investigation of factors affecting the process performance, and exploration of the mechanisms and kinetics of pollutant degradation [**38**].

Electrooxidation is divided into two processes

## 1. Indirect electrooxidation processes

Electrooxidation of pollutants can be fulfilled in different ways. Use of the chlorine and hypochlorite generated anodically to destroy pollutants is well known. This technique can effectively oxidize many inorganic and organic pollutants at high chloride concentration, typically larger than 3 g/l. The possible formation of chlorinated organic compounds intermediates or final products hinders the wide application of this technique. Moreover, if the chloride content in the raw wastewater is low, a large amount of salt must be added to increase the process efficiency. The disadvantage of this method, however, is that where water is recycled one has to accept a level of chloride ion concentration of several percent.

In the situation where chlorine or hypochlorite is formed by addition of sodium chloride or sea water, the anode reaction is simply [38].

$$2Cl^{-} \rightarrow Cl_{2} + 2e \qquad \dots (3.10)$$

This is done in a cell in which anode and cathode products are not separated, the product of the reaction is sodium hypochlorite, NaClO.

Thus, in such an undivided cell the anode reaction is Eq. (3.10) and the cathode reaction is

$$2H_2O + 2e \rightarrow 2OH^- + H_2$$
 ...(3.9)

which on mixing gives

$$20H^{-} + Cl_{2} \rightarrow Cl^{-} + Cl0^{-} + H_{2}0$$
 ...(3.11)

**Farmer et al. in 1992 [39]** proposed another kind of electrooxidation, mediated electrooxidation, in treating mixed and hazardous wastes. In this process, metal ions, usually called mediators, are oxidized on an anode from a stable low valence state to a reactive high valence state, which in turn attacks organic

pollutants directly, and may also produce hydroxyl free radicals that promote destruction of the organic pollutants. Subsequently, the mediators are regenerated on the anode, forming a closed cycle. The typical mediators include  $Ag^{+2}$ ,  $Co^{+3}$ ,  $Fe^{+3}$ ,  $Ce^{+4}$  and  $Ni^{+2}$ . Mediated electrooxidation usually needs to operate in highly acidic media. In addition, there exists the secondary pollution from the heavy metals added. These disadvantages limit its application.

## 2. Direct anodic oxidation

Electrooxidation of pollutants can also occur directly on anodes by generating physically adsorbed "active oxygen" (adsorbed hydroxyl radicals, OH) or chemisorbed "active oxygen" (oxygen in the oxide lattice,  $MO_{x+1}$ ) [40]. This process is usually called anodic oxidation or direct oxidation. The physically adsorbed "active oxygen" causes the complete combustion of organic compounds (*R*), and the chemisorbed "active oxygen" ( $MO_{x+1}$ ) participates in the formation of selective oxidation products

$$R + MO_x (\bullet OH)_z \rightarrow CO_2 + zH^+ + ze + MO_x \qquad \dots (3.12)$$

$$R + MO_{x+1} \longrightarrow RO + MO_x \qquad \dots (3.13)$$

In general, OH is more effective for pollutant oxidation than O in  $MO_{x+1}$ . Because oxygen evolution, can also take place at the anode, high overpotentials for  $O_2$  evolution is required in order for reactions above Eqs. (3.13) and (3.14) to proceed with high current efficiency. Otherwise, most of the current supplied will be wasted to split water.

The anodic oxidation does not need to add a large amount of chemicals to wastewater or to feed  $O_2$  to cathodes, with no tendency of producing secondary pollution and fewer accessories required. These advantages make anodic oxidation more attractive than other electrooxidation processes [40].

## **3.3.4 Electrodeposition (ED)**

Electrodeposition is often also called "electroplating", and the two terms are used interchangeably. As a matter of fact, "electroplating" can be considered to occur by the process of electrodeposition. Electrodeposition is the process of producing a coating, usually metallic, on a surface by the action of electric current. The deposition of a metallic coating onto an object is achieved by putting a negative charge on the object to be coated and immersing it into a solution which contains a salt of the metal to be deposited (in other words, the object to be plated is made the cathode of an electrolytic cell). The metallic ions of the salt carry a positive charge and are thus attracted to the object. When they reach the negatively charged object (that is to be electroplated or electrodeposited), it provides electrons to reduce the positively charged ions to metallic form. Figure 3-8 is a schematic presentation of an electrolytic cell for electrodeposition of a metal "M" from an aqueous (water) solution of metal salt "MA" [41].



Figure 3-8 Schematics of an electrolytic cell for plating metal "M" from a solution of the metal salt "MA" [41].

## **3.3.4.1** Mechanism of Electrodeposition

To further illustrate the foregoing, let us assume that one has an object made of one of the common metals, like copper, and that it has been properly pre-cleaned. We should want to deposit it with, say, nickel. A wire will have to be attached to the object while the other end of the wire should be attached to the negative pole of a battery (or a power supply). To the positive pole of the battery (or power supply) we connect another wire with its other end connected to a rod made of nickel. Next we fill the cell with a solution of the metal salt to be plated. It is possible to use a molten salt and in some not so common cases. In most, more common, cases through the salt is simply dissolved in water. In our present example the nickel chloride salt dissociates in water to positively charged nickel cations and negatively charged chloride anions. As the object to be plated is negatively charged it attracts the positively charged nickel cations, and electrons flow from the object to the cations to neutralize them (to reduce them) to metallic form. Meanwhile the negatively charged chloride anions are attracted to the positively charged nickel rod (known as the anode of the electrolytic cell). At the anode electrons are removed from the nickel metal, oxidizing it to the nickel cations. Thus we see that the nickel dissolves as ions into the solution. That is how replacement nickel is supplied to the solution for that which has been deposited out and one retains a solution of nickel chloride in the cell.

By now it should be evident that electrodeposition or electroplating should be defined as the process in which the deposit of a (usually) thin layer (of metal) is formed electrolyticly upon a substrate (that is often, but not always, also a metal). The purpose of such process may be to enhance or change the substrate's appearance and/or attributes (such as corrosion resistance). Examples are the deposition of gold or silver on jewelry and utensils, and the deposition of chrome on automobile parts. Electrodeposition is performed in a liquid solution called an electrolyte, otherwise referred to as the "deposition bath". The bath is a specially designed chemical solution that contains the desired metal (such as gold, copper, or nickel) dissolved in a form of submicroscopic metallic particles (positively charged ions). In addition, various substances (additives) may be introduced in the bath to obtain smooth and bright deposits. The object that is to be deposited is submerged into the electrolyte (deposition bath). Placed usually at the center of the bath, the object that is to be deposited acts as a negatively charged cathode. The positively charged anode completes the electric circuit; those may be at opposite edges of the deposition tank, thus causing film deposit on both sides of the cathode. A power source in the form of a battery (which converts AC electricity to regulated low voltage DC current) is providing the necessary current. This type of circuit arrangement directs electrons (negative charge carriers) into a path from the power supply to the cathode (the object to be deposited). Now, in the bath the electric current is carried largely by the positively charged ions from the anode toward the negatively charged cathode. This movement makes the metal ions in the bath to migrate toward extra electrons that are located at or near the cathode's surface outer layer. Finally, by way of electrolysis the metal ions are removed from the solution and are deposited on the surface of the object as a thin layer. It is this process to which we refer to as "electrodeposition" [41].

From the above it would appear that the thickness of the electrodeposition layer on the substrate is determined by the time duration of the deposition. In other words, the longer time the object remains in the operating deposition bath the thicker the resulting electrodeposited layer will be. Typically, layer thicknesses may vary from 0.1 to 30 microns (micron = one millionth of a meter), though nothing prevents the deposition of thicker or thinner layers, as desired. The geometric shape and contour of an object to be plated affects the thickness of the deposited layer. In general, objects with sharp corners and features will tend to have thicker deposits on the outside corners and thinner ones in the recessed areas. The cause of this difference in the resulting layer thicknesses is that DC current flows more densely to sharp edges than to the less accessible recessed areas, in other words, the current distribution is not uniform. (Another, more accurate, explanation of this phenomenon involves the geometry of the electric field lines that exist between cathode and anode in the solution). In practice, an item such as, say, a watch or similar item with sharp faceted corners is difficult (almost impossible, actually) to deposit uniformly. A deposition method known as "electroless deposition", assures uniformity of deposited thickness even on highly irregular shaped objects. In the case of electrodeposition, judicial placement of the anode as well as modifications of the current density is required to overcome the thickness irregularity effects. Electrodeposition processes will not, as a rule, conceal preexisting surface imperfections such as scratches, dents or pits. Actually, the deposition process will more often than not, make most surface blemishes even more pronounced. Thus it is important to remove any undesirable surface marks prior to the deposition action.

The process of metal deposition generally consists of several partial processes:

- 1. Transport of ions to be deposited from the bulk of the solution to the cathode surface through migration and diffusion.
- 2. Charge transfer reaction: the simple metallic ions are then discharged at the electric double layer.
- 3. Electrocrystallization process: the discharged metal atoms are then changed into crystalline state.

In the case of materials such as gold, the anode is not sacrificial (gold does not dissolve easily), but it is made out of material that does not dissolve in the electrolyte, such as titanium. The deposited gold comes out of the solution. Plating is an oxidation-reduction reaction, where one material gives up electrons (gets oxidized) and the other material gains electrons (gets reduced). The anode is the electrode at which oxidation occurs, and the cathode is the electrode at which reduction occurs.

The electrodepositing process depends on a great number of factors affecting the nature and properties of deposits, among these factors are current density, conductivity of the solution, presence of addition agents, agitation, temperature and nature of the base metal (substrate) [42].

Electrodeposition is a "clean" process for treating wastewater containing heavy metal ions. The method is capable of freeing toxic metal ion by

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electrodepositing them in metallic form at the cathode of an electrolytic cell. The deposited metals can be recycled in a metal or alloy processing plants. The wastewater free of the pollutants may be then discharged to waste stream. One inherent problem associated with the electrolytic method is that the cathode current efficiency for the metal recovery decreases with metal ion concentrations in wastewater. The current efficiency is nearly 100% when the metal ion concentrations are greater than 50-100 ppm. However, the current efficiency decreases to less than 1% when the metal ion concentration becomes less than 1 ppm, leading to a high-energy consumption for treating wastewater containing dilute metal ions [43].

## **3.3.4.2 Current Efficiency**

The current efficiency,  $C_{E_j}$ , of an electrode deposition process is the ratio of the current flowing that is utilized in the electrodeposition reaction to the total current passing, if all the electric current has been used for depositing in the electrolytic process, expressed as a percentage. The current efficiency is given by [44]:

Current efficiency 
$$(C_{E_j}) \% = \frac{W}{W_{th}} \times 100$$
 ... (3.14)

where W = weight of metal deposited, and

 $W_{th}$  = theoretical weight of metal deposited.

The theoretical weight of the individual metal deposited can be calculated as follows:

$$W_{th} = \frac{Q}{z_j F} M_{wt_j} \qquad \dots (3.15)$$

where Q = the amount of electricity passed through the cell ( $Q = I \times t$ ).

So we can expressed Eq. 3.15 in another form

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$$C_{E_j} = \frac{z_j F}{M_{wt_j} I} \frac{dm}{dt} \qquad \dots (3.16)$$

where m = the mass electro-processed during the time interval dt.

Equation 3.17 was proposed by [45] during the study of the electroreduction of copper ions. As explained by these researchers, the overall *CE* given by Eq. 3.17 represents the combined effects of overpotential and current density profiles and also the effect of the decrease in porosity, which are difficult to measure individually in practice, especially in the case of porous electrodes.

The partial current efficiencies of the parent metals in the alloy  $C_{E_1}$ % and  $C_{E_2}$ % are determined from the relations:

$$C_{E_1} \% = \frac{W_1}{W_{th}} \times 100$$
 ...(3.17)

$$C_{E_2} \% = \frac{W_2}{W_{th}} \times 100$$
 ...(3.18)

The total efficiency of the alloy  $C_{E_t}$  % is equal to the sum of the partial current efficiencies  $C_{E_1}$  % and  $C_{E_2}$  %.

$$C_{E_t} \% = C_{E_1} \% + C_{E_2} \% \qquad \dots (3.19)$$

## 3.3.4.3 Deposition of Complex Ion

If a metal ion in solution is associated with chemical groups, or ligands (other than water), thus forming a complex ion, its activity is very much lower than when it is merely hydrated ("free" or "simple" ion) and, accordingly, its standard potential is shifted to a more negative value. The changes may be as great as 0.6 to 0.8 V, and alter the relative position of metal in the so-called electromotive or activity series [46].

In any electrodeposition process there is usually more than one cathodic reaction occurring. For example, metal deposition will often occur with concomitant hydrogen evolution. The position of the individual cathodic polarization curves will determine the magnitude of the partial current densities corresponding to the individual cathodic reaction as shown in Fig. 3-9.

If a metal A has a more noble polarization curve than some other metal B in Fig. 3-9a, then only deposition of metal A will occur and the deposition potential of B is not attained. At cathode potential  $E_1$ , current density  $i_1$  is found which corresponds only to the deposition of metal A. If, however, the current density is increased then deposition of metal B will also occur as the cathode potential became more active (that is, more negative). When the polarization curves or the deposition potential  $E_1$  the current density  $i_1$  corresponds to deposition of both metals A and B although even at low current density the proportion of A deposited will be greater than that of metal B. The most favorable conditions for co-deposition for both metals A and B occur when the polarization curves of the two metals are identical.

This situation, however, is rarely found in practice. More commonly polarization curves intersect in Fig. 3-9c, under these conditions there will be simultaneous and equal deposition of metal A and B at the intersection potential  $E_1$ , at potential more noble than  $E_1$  the proportion of B deposition will exceed that of metal A, whilst at potentials active or negative to  $E_1$ , a greater proportion of metal A will be deposited [47].

It is common to find some intersection of the partial polarization curves. Typically the deposition of more noble metals can occur at less noble cathode potentials than that observed for discharge of the noble metal on its own.

Conversely, the less noble metal can be deposited at nobler cathode potentials than when deposited alone. For a given electrolyte composition and operating conditions, the position of the cathode polarization curves for alloy relative to the curves of the separated metals will depend on the composition and electrochemical properties of the alloy to be deposited. It can be found, for example that the less noble metal will deposit preferentially (that is it functions as a more noble metal) during simultaneous deposition of the two metals. This can be found with nickel-zinc alloys. The general rule for co-deposition of alloys is that the metal which is, nobler under the electrolysis conditions and which is favored during discharge is preferentially deposited to a greater degree the lower the polarization existing during deposition. All factors that increase the polarization will shift the deposition ratio in favor of the less noble metal such that its proportion in the deposit is increased. Similarly, an increase in the ratio of the less noble metal deposit is produced by a decrease in the solution temperature and in static solution. Conversely, a decrease in polarization favors discharge of the more noble metal. Thus a high solution temperature, a low current density and solution agitation will lead to the deposit having greater proportion of the more noble metal.

There are, however, exceptions as for example, in gold-copper deposition where the gold is always preferentially deposited although an increase in the temperature will lead to the greater proportion of copper deposited. This is due to a shift in the equilibrium to the left so that the concentration polarization for copper decreases.

Many alloys are commercially electrodeposited and these depositions generally involve complex solution such as cyanide or pyrophosphates, simple solutions are, however, also known. For example, cadmium-nickel alloys may be deposited from sulfate solution. The important factor in alloy deposition is the deposition potential which is determined by the polarization processes occurring at the cathode rather than the standard potentials of the two metals as determined by the e.m.f. series [47].

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Figure 3-9 Schematic polarization curves for two metal A and B [47].

## **3.3.4.4 Underpotential Deposition (UDP)**

The phenomenon of underpotential deposition (UPD) has been discovered and studied for decades. Due to the stronger bond between the foreign metal adatoms with the metal substrate as compared to the same type substrate, the (UPD) foreign metals would deposit at a potential more positive to the Nernst potential, where bulk deposition takes place, forming one monolayer or sub-monolayers onto the substrate surface [48].

Underpotential deposition (UPD) is the deposition of a metal ion at a voltage anodic to the equilibrium potential predicted by the Nernst equation [49]. Underpotential deposition provides one possible means for catalysis. This phenomenon occurs when a metal adsorbate has a greater attraction to a different metal substrate than its own.

To understand (UPD) behavior, the energetic and entropic effects between the adsorbate and substrate (surface) must be considered. Differences in electronegativity between atoms cause a strong attraction. A transfer of charge which occurs from the less electronegative atom to the more electronegative atom which is similar to ionic bonding may explain this attraction. The more electronegative atom has a greater attraction for the shared electrons in a bond. A more convenient measure of an atom's attraction for electrons is its electronic work function. The electronic work function is defined as the work required to remove a single electron from a solid surface to a vacuum [49].

**Trasatti S. in 1971 [50]** has shown a direct correlation between an atom's electronic work function and its electronegativity by showing that a different correlation exists for the transition metals. Values of the electronic work function and effective electronegativities for several metal atoms of interest are given in Table 3-4. The effective electronegativities reported below, which are made based on relationships observed between the electronic work function, the potential of zero charge, and the electronegativity of different groups of atom

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Metal	Work Function (eV)	Effective Electronegativity
Au	4.78	2.10
Cd	4.12	1.77
Cr	4.40	1.65
Cu	4.70	2.06
Ni	4.73	1.82
Pb	4.18	1.80
Pt	5.03	2.22
Zn	4.30	1.60

Table 3-4 Selected work functions and effective electronegativities [50].

Difference in electronegativities between atoms can cause a transfer of a partial charge to occur. This results in a bond formed between the two atoms.

A large attraction by the metal at adsorbate for a foreign substrate occurs when the work function of the metallic surface is greater than that of the metal adsorbate [51]. An UPD phenomenon has not been observed experimentally for systems where the electronic work function of the metal adsorbate was greater than that of the metal substrate.

#### **3.3.5** The Electrochemical Precipitation of Metal Ion

Several technologies are based on the precipitation of the metal ions as the hydroxide. The hydroxide ion results from the cathodic reduction of water:

$$2H_2O + 2e \rightarrow H_2 + 2OH^- \qquad \dots (3.9)$$

which then reacts with metal ions in the medium. In such a process for (e.g. Ionsep process) [4], the contaminated solution is fed to the anode compartment of a cell separated by a cation exchange membrane. During water electrolysis within the cell, the metal ions migrate through the cation permeable membrane into the catholyte where they are precipitated by the hydroxide formed by reaction, as mentioned above, at the cathode. The process has been used for the removal of ions such as  $Cu^{+2}$ ,  $Cd^{+2}$ ,  $Fe^{+2}$ ,  $Ni^{+2}$ ,  $AI^{+3}$  and  $Cr^{+3}$ .

Another process (e.g. Andco process) [4] employs an undivided cell with an iron anode. During operation, the cell produces  $Fe^{+2}$  at the anode and hydroxide at the cathode; these combine to precipitate iron hydroxide and it is found that other ions in solution co-precipitate with the iron hydroxide. The co-precipitation occurs by a combination of mechanisms which include simple precipitation of the metal hydroxide/oxide as well as surface complexation adsorption and electrostatic interaction with the surface of the iron hydroxide. The addition of polymers assists the co-precipitation and filtration and large amounts of ion removal are possible for relatively low charges through the cell. For example, the arsenic content of an effluent from a *GaAs* semiconductor plant could be reduced from 5.2 ppm to 45 ppb with 50 ppm of anodically generated iron hydroxide.

From the viewpoints of being able to handle readily very low levels of metal ions and also meeting consent limits for many ions, this method is very attractive. Moreover, even the lowest metal ion levels are effectively treated in simple and cheap cells. On the other hand, it is difficult to introduce any specificity and the metals exit from the system in their least valuable form, solids with a complex composition. Certainly, in most applications it is not possible to recycle the metals and the solids, after filtration, are sent to landfill [4].

## 3.3.6 The Cathodic Deposition of Metal Ion

This method has the attraction that the metals can be recovered from the cell in a valuable form, either high purity metal or a concentrated and pure metal salt solution. In this technology, the effluent will be fed to the cathode in a divided or undivided cell where the reaction

$$M^{+n} + ne \to M \qquad \dots (3.1)$$

is carried out.

Clearly, the method is limited to those ions which reduce to the metal at potential less negative than water reduction (Eq. 3-9) which is now an unwanted,

competing reaction leading to loss of current efficiency. Even so, many heavy and transition metals including Ag, Au, Pt, Pd, Ir, Rh, Cu, Ni, Hg, Cd, Pb, Bi, Zn, Co, As, Sb, and Te can be removed. The presence of oxygen or other reducible species also reduces the current efficiency and increases the process power consumption. Even so, the power consumption is normally low for effluent treatment (where the metal ion concentration is typically < 100 ppm) and by controlling the electrolysis conditions; it may be possible to introduce selectivity for single metal ions.

No electrode reaction can occur more rapidly than the rate at which reactant reaches the electrode surface. To obtain the highest rate of removal, the reduction of  $M^{n+}$  should be mass transport controlled and under these conditions the rate of removal of metal ion from an effluent by cathodic reduction may be written as [52].

$$-V \frac{dc}{dt} = -\frac{I_l}{nF} = k_m A c \qquad ... (3.20)$$

where V = the volume of effluent to be treated.

c = the concentration of metal ion  $M^{n+}$ .

t =the time.

 $I_l$  = the mass transport limited current.

F = the Faraday constant.

 $k_m$  = the mass transfer coefficient. and

A = the electrode area.

Integration of Eq. 3-20 with respect to time gives an expression for the fraction of metal ion removed, i.e.

$$\frac{c(t)}{c(o)} = exp - \frac{k_m A}{V} t \qquad ...(3.21)$$

where c(o) = the initial concentration of metal ion.

c(t) = the concentration after electrolysis for time t.

Hence, it can be seen the rate of removal depends on two factors:

- (i) the electrode area, and
- (ii) the mass transport regime determined by the flow conditions (electrolyte flow rate or electrode movement) and the presence of turbulence promoters.

Note that although the current density (I/A) is proportional to the concentration of  $M^{n+}$  and a higher current density leads to more rapid removal, the time taken to achieve a defined fractional removal is not dependent on the concentration. For the technology of metal ion removal by cathodic reduction, the conclusions are clear. Cells must be designed to give very high mass transport coefficients and/or to have a very high cathode surface area.

When plated in the cells under mass transfer control (i.e. by using flow conditions such as electrolyte flow rate or electrode movement), the metal is deposited as a rough layer and rotation of this rough layer leads to greatly enhanced values of  $k_m$ . Increases in both  $k_m$  and A enhance the performance of cells with three dimensional electrodes. In such cells,

$$A = A_e V_e \qquad \dots (3.22)$$

where  $V_e$  = the volume of the three dimensional cathode, and

 $A_e$  = the electrochemically active surface area per unit volume of the three dimensional material.

In employing three dimensional electrodes, there is always a concern about the potential distribution through the electrode and hence whether the entire electrode is active for deposition.

Fortunately the treatment of effluents fits well with three dimensional electrodes because of the low metal ion concentration, hence low current density and the minimum possibility of unwanted voltage drop.

Several companies offer technology for the removal of metal ions by cathodic deposition, and again it should be stressed that the systems are designed to achieve different goals, which is that any cell must be intended to recover large quantities of metal from relatively concentrated solution and a major requirement is that the pure metal is easily recovered from the cell. Hence, the cathodes are designed to be lifted from the cell.

## **3.4 Literature Survey**

This section will be mainly concerned with previous works about the removal of heavy metals for different methods, configuration, and type of metals.

## **3.4.1 Electrolytic Method**

**Hickman K. et al. in 1993 [53]** used a silver tower electrolyzer to recover silver from spent photographic fixing solutions. **Zhou C. D., and Chin D. T. in 1994 [54]** described an electrolytic process for simultaneously recovering heavy metal ions from wastewater at the cathode and destruction of cyanide ions at the anode. **Khristoskova S., and Lazavou D. in 1984 [55]** studied an electrolytic process to remove hexavalent chromium from wastewater. **Shifrin S. M. et al. in 1979 [56]** discussed the advantage of adding RuO<sub>2</sub> to a TiO<sub>2</sub> anode to treat wastewater. **Saito K. in 1979 [57]** described a process for the recovery of bromide from a solution containing carboxylic or phenolic compounds. **O'Keefee T. J., and Ettel V. A. in 1997 [58]** discussed the environmental and economic factors of the electrolytic metal recovery process.

To treat large volume of wastewater and to reduce metal concentration to a low level, many different cell designs have been reported in the literature. **Hertwig K. et al. in 1992 [59] and Tison R. P. in 1981 [60]** used a rotary drum electrode to recover copper. **Fleishmann M. et al. in 1971a [61]** used a fluidized bed as the cathode to plate copper onto metal particles. **Bennion D. N. and Newman J. in 1972 [62]** used a flow-through porous-electrode to remove copper ions from dilute solutions. **Robertson P. M. and Dossenbach O. in 1981 [63]** developed a gas-sparging cell to improve mass transfer in an electrolytic cell for wastewater treatment.

Holland F. S. in 1978[64] described an ECO-cell developed by Ecological Engineering Ltd, in the United Kingdom. The cell consisted of an inner rotating

cylinder cathode with a diaphragm and an outer stationary cylinder as the anode. To obtain a high conversion for metal recovery, the cathode compartment was divided into 6 to 12 sections by internal baffles. Electrolyte flowed through a small annular gap between the baffles and the rotating cylinder to effectively provide a high mass transfer rate.

**Robertson P. M. et al. in 1983 [65]** developed a Swiss-roll cell. Two foil and two thin separators were placed on one another and were rolled around an axis and pressed into a cylinder container. The electrolyte was pumped through the gap provided by the cell separator. The cell was suited for wastewater treatment due to its large surface area.

**Lopez-Cacicedo C. L. in 1975** [66] developed a Chemelec cell, this cell consisted of a vertical container in which two horizontal parallel metal meshes were used as the electrodes and glass beads were employed to generate fluidization between the electrodes to obtain a high mass transfer rate.

Fleishmann M. et. al. in 1971b [67] developed a bipolar trickle tower cell, in this cell, the anode and cathode were located on the top and bottom of the cell, and layers of electrical conductive material separated by insulating meshes were sandwich between the anode and cathode. When a terminal voltage was applied to the tower, bipolarity was induced in each conductive layer. The trickle tower possessed a large electrode surface area.

Fleishmann et al. in 1976 [68] developed an electrochemical flow cell using a rotating disk as an electrode. In this cell, the gap between the rotating disk electrode and the counter electrode was small, yielding a low IR drop in the solution.

**Entwistle J. E. in 1976 [69]** compared various CN<sup>-</sup> removal schemes for concentrated plating bath solutions. He studied incineration biological oxidation, combustion of HCN liberated from acidic solutions, chlorination at high pH and electrooxidation. He concluded that electrooxidation is the most cost effective for concentrated waste stream.

Bennion D. N. and Newman J. in 1972 [70] removed  $Cu^{+2}$  in porous flowthrough electrode of graphite particles. They found the procedure to be very effective in removing the  $Cu^{+2}$  to levels near 0.02 mg/l but only at very low flowrates. The cell's performance was limited by the evolution of hydrogen gas at the most cathodic potentials.

Van Zee J. and Newman J. in 1977 [71] examined the electrochemical recovery of  $Ag^+$  from (thiosulfate-photographic) fixing bath solutions. They also used a porous graphite electrode. Even though the silver cation is a highly electropositive ion, they observed large hydrogen evolution rates with resulting 10w current efficiencies for the metal deposition. This result is explained by the complexation of  $Ag^+$  ions with thiosulfate which makes it more stable in the solution and hence less electropositive. Adams G. B. et al in 1976 [72] have shown that a porous graphite electrode can be used to reduce the concentration of ferric ions found in mine waste drainage by reduction to the less noxious ferrous species.

Jansson R. E. W. and co-workers in 1982 [73, 74, 75] demonstrated that a (trickle-tower) electrode can simultaneously remove  $Pb^{+2}$ ,  $Cd^{+2}$ , or  $Cu^{+2}$  and the associated  $CN^{-}$  anion from an aqueous stream. The trickle tower consists of a series of conductive particles with each layer of particles separated by an insulating mesh. The fluid trickles down from one particle layer to the next below and the two current collectors are placed at the top and bottom of such a stack. This arrangement unfortunately, results in large power requirements and does not seem to be industrially promising. The electrodes were made of graphite and were uncatalyzed.

**Yaniv D. and Ariel M. in 1981 [76]** have demonstrated  $Cu^{+2} Pb^{+2} Cd^{+2}$  may be simultaneously removed from  $Cl^{-}$  solution on an uncatalyzed graphite cloth electrode. Not surprisingly, they found that the more electropositive metal was preferentially removed.

**Giannopoulou I. and Panias D. in 2007** [77] generated an acidic polymetallic wastewaters during the pyrometallurgical treatment of chalcopyrite for the production of primary copper. They found that copper can be recovered electrolyticly followed by bismuth and the two metalloids arsenic and antimony that exhibits almost the same electronegativity as copper. The other high electropositive metals Ni, Pb, Zn, and Fe remain, as it was expected, in the solution from which

nickel can be recovered with neutralization, contaminated with Cu, Fe, Zn and traces of bismuth, arsenic and antimony.

**Roventi G., Bellezze T., and Fratesi R. in 2005** [78] investigated the effect of the underpotential deposition of zinc on Zn–Co alloy electrodeposition from chloride baths without complexing agents by cyclic voltammetry (CV) and anodic linear sweep voltammetry (ALSV), to identify the deposited phases at the various potentials and to investigate the cause of the inhibition observed at low polarizations.

**Gulnaziya I. et. al. in 2005** [44] obtained activated carbon from palm shells has good electrochemical properties and may be used as a working electrode material to remove ions of heavy metals from industrial wastewaters. Results are presented on the electrodeposition of copper and lead ions onto palm shell activated carbon electrodes in terms of current efficiency (%). They showed that the current efficiency increases with an increase in the flow rate. The application of more negative current to the electrolytic cell results in the decrease in current efficiency values and they found that the presence of malonic acid results in a relative increase in the current efficiency compared to the single metal system for both pH 3 and 5 and also found that the presence of boric acid also results in a similar overall increase in the current efficiency.

Albert B. et al in 1996 [79] demonstrated the removal of  $Cu^{+2}$ ,  $Pb^{+2}$ ,  $Cd^{+2}$ , and  $Ni^{+2}$  from aqueous samples using a small-scale electrolytic cell. They developed various novel carbon materials with a large ratio of surface area to volume for the electrochemical removal of heavy metal ions from aqueous wastes. Removal efficiencies above 90% have been observed. They demonstrated the continuous removal of lead over a 72 h period at these levels, they also observed an effluent concentration below 10 ppm for an inlet solution feed of 100 ppm. They found that the use of electrolytic remediation on waste streams has proved successful in the selective removal and recovery of metals. They also found that the removal of nickel ions from solution directly onto the bare carbon material is inefficient.

**Kaminari N. M. S. et al in 2006 [80]** studied an electrochemical reactor using metallic particles as cathode for  $Pb^{+2}$ ,  $Cu^{+2}$ , and  $Ni^{+2}$  ions recovery from acidified aqueous solutions for different process parameters as metallic ion concentration, current density and bed expansion. They evaluated the reactor performance considering the system efficiency and energy consumption. They obtained the highest current efficiencies were 75.8%, 89.9%, and 30.3% for  $Pb^{+2}$ ,  $Cu^{+2}$ , and  $Ni^{+2}$ , respectively.

#### **3.4.2 Electrodialytic Method**

Membrane process has proved its reliability in a large number of applications. Electrodialysis has been practically applied to purification by means of desalination in medical, food, chemical and metallurgical industries The electrodialysis technique was also developed mainly for desalination and concentration of seawater **[81, 82]**, but later it was applied to the recovery of metals from the metal finishing wastewater and in the metallurgical industries **[83, 84]**.

Recently, electrodialysis has been applied to the removal and treatment of industrial effluents from wastewater [85, 86]. In this process, ion migration through an ion exchange membrane takes place when a potential gradient is applied across the membrane; electrostatic interaction within the membrane plays a key role in the transfer of ions. Electrodialysis has the advantage of linking energy expenditure to the quantity of electrolytes to be extracted and not to the volume of water to be treated. This process enables low ion concentrations to be brought up to higher levels with low energy requirement.

**Xue Z. et al. in 1992 [87]** used a continuous flow electrodialysis cell for the purification of industrial wastewater consisting of an acidic process water stream and a spent alkaline process stream. **Saracco G. et al. in 1993 [88]** described an electrodialytic process for the separation of NaCl and Na<sub>2</sub>CO<sub>3</sub> salts in an industrial wastewater of a leaching operation using ion-permselective membranes.

Cherif A. T. et al. in 1997 [89] described an electrodialysis method for the recovery of nitric acid and sodium hydroxide from an industrial wastewater

containing NaNO<sub>3</sub>, using a three compartment electrodialysis cell. In another study electrodialysis has been successfully applied to the removal of copper from wastewater. The pH of the treated water was 6.5-7.5 whereas pH of the wastewater was 9.0. This eliminates the need for adjusting pH of the wastewater before discharging into the waste streams. In addition, the operation cost of this method was reduced because of the recovery of copper in the cathode compartment.

**Rodrigues M. A. S. et al. in 1999 [90]** studied the electrodialysis of an industrial wastewater (or rinse water from chromating bath) containing 4600 ppm of Cr(VI) besides other metallic contaminants using ion exchange membranes (Selemiom AMT and CMT). The results show that 99.9% of Cr(VI) was recovered in a treated water with a chromium concentration of 4 mg/l. These results show that electrodialysis can be used to treat these effluents, and this water could be reused as rinse water. The solution in the anodic compartment reached 7200 mg/l of Cr(VI), which could be reused on the chromating bath itself.

Shim J. B. et al. in 1999 [91] discussed the removal of vanadium(III) and Iron(II) ions from a simulated decontamination waste solution by electrodialysis technique through the (Nafion 117 cation exchange membrane). Zhou K. et al in 1999 [92] studied the treatment of industrial wastewater from an alumina plant to produce deionized water which was carried out by membranes technology by the combination of the electrodialysis and reverse osmosis techniques. The research shows that the method is effective and on the basis of experimental results a process for the wastewater treatment with a capacity of 120 tons.

**Ribeiro A. B. et al. in 2000 [93]** studied the removal of Cu, Cr, and As from chromated copper arsenate timber waste using electrodialysis cell. The highest recovery rates obtained were 93% of Cu, 95% of Cr, and 99% of As.

Wisniewski J. A. et al. in 1999 [94] investigated water and acid recovery from the effluent after metal etching rinsing using electrodialysis systems. The results showed that water of good quality, with no acid and metal salts, was obtained. The hydrochloric acid from the electrodialysis concentrate was recovered by

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monoselective electrodialysis and the acid solution was concentrated by 74 times compared to raw wastewater.

Hunsom M. et. al. in 2005 [95] tested an electrochemical technique in a laboratory scale to treat heavy metals ( $Cu^{+2}$ ,  $Cr^{+6}$  and  $Ni^{+2}$ ) from plating industrial effluent. The experiments were performed in a membrane reactor. They found that a membrane reactor with plane electrode was capable of treating plating wastewater with low energy consumption, and low operating cost. More than 99% of metal reduction was achieved and the final concentrations of  $Cu^{+2}$ ,  $Cr^{+6}$ , and  $Ni^{+2}$  in treated water were 0.10–0.13, 0.19–0.20, and 0.05–0.13 ppm, respectively.

# Chapter Four Experimental Work

# **4.1 Introduction**

This chapter describes the experiment setup and the construction of the main equipment, which is the integrated cell. In addition, this chapter describes the experimental procedure that was followed to achieve the desired objectives and the range of control variables changed while experiments were done.

# 4.2 Experimental Set-up

The work reported presently describes an electrochemical fix bed reactor used to remove heavy metal ions  $(Cu^{+2}, Ni^{+2}, and Cd^{+2})$  by cathodic deposition on a brass cylindrical mesh electrode (50 mesh no. that means there is 50x50 pore in 1 in<sup>2</sup>). Brass has been chosen as the electrode material during this work because of the following reasons:

- 1. It's a good corrosion resistance.
- 2. Its low in cost.
- 3. Its good in electrical conductivity.

The experimental work was divided into the following parts:

- 1. Preliminary set of experiments were performed in a non flow system to eliminate Cu<sup>+2</sup> ions only to Cu metal on a (single, double, and triple) brass meshes electrode (working electrode) by cathodic deposition in order to get a best current density which would be used in a major set.
- 2. Major set of experiments were performed in a flow system as follows:
  - i) Experiments were performed in one compartment electrochemical fixed bed reactor to eliminate individually (Cu<sup>+2</sup>, Ni<sup>+2</sup>, and Cd<sup>+2</sup>) ions to their metals on a (single, double, and triple) brass meshes (working electrode) by cathodic deposition in sulfuric acid to measure degradation of concentration with time.

- ii) Experiments were performed in a once through electrochemical fixed bed reactor to eliminate a couple of (Cu<sup>+2</sup>-Ni<sup>+2</sup> and Cu<sup>+2</sup>-Cd<sup>+2</sup>) ions to their metals on a (single, double, and triple) brass meshes (working electrode) by cathodic co-deposition in sulfuric acid to measure degradation of concentration with time.
- iii) Experiments were performed in a once through electrochemical fixed bed reactor to eliminate a triple (Cu<sup>+2</sup>-Ni<sup>+2</sup>-Cd<sup>+2</sup>) ions to their metals on a (single, double, and triple) brass meshes (working electrode) by cathodic co-deposition in sulfuric acid to measure degradation of concentration with time.

For the preliminary sets of experiments, five different total currents for single mesh electrode (15, 20, 25, 70, and 90 mA), five different total currents for double meshes electrodes (30, 40, 50, 140, and 180 mA), and five different total currents for triple meshes electrodes (45, 60, 75, 210, and 270 mA) were considered for deposition of  $Cu^{+2}$  ion only to Cu metal at 40  $^{0}C$  and three deposition times (0.5, 1.0, and 1.5 h).

For a major set of experiments:

- i) Five different total currents for single mesh electrode (30, 40, 50, 145, and 190 mA), three different total currents for double meshes electrodes (60, 100, and 380 mA), and one total current for triple meshes electrodes (90 mA) were considered for deposition of Cu<sup>+2</sup> ion to Cu metal, with 200 ppm initial concentration of Cu<sup>+2</sup> ion for individual and co-deposition experiments.
- ii) Five different total currents for single mesh electrode (145, 160, 175, 190, and 205 mA), three different total currents for double meshes electrodes (290, 350, and 410 mA) were considered for deposition of Ni<sup>+2</sup> ion to Ni metal and co-deposition of Cu<sup>+2</sup>-Ni<sup>+2</sup> ion to their metals respectively, and one total current for triple meshes electrodes (435 mA) were considered for deposition of Ni<sup>+2</sup> ion and co-deposition of Cu<sup>+2</sup>-Ni<sup>+2</sup> ion to their metals, with 200 ppm initial concentration of Cu<sup>+2</sup> and Ni<sup>+2</sup> ion for

individual deposition and (100, 200, and 300 ppm) of  $Cu^{+2}$  ion for codeposition experiments.

iii) Five different total currents for single mesh electrode (175, 190, 205, 220, and 235 mA), and three total currents for double meshes electrodes (350, 410, and 470 mA) were considered for deposition of Cd<sup>+2</sup> ion to Cd metal, double metal co-deposition of Cu<sup>+2</sup>-Cd<sup>+2</sup> ions, and triple metal co-deposition of Cu<sup>+2</sup>-Ni<sup>+2</sup>-Cd<sup>+2</sup> ions to their metals respectively, and one total current for triple meshes electrodes (525 mA) were considered for deposition of Cd<sup>+2</sup> and co-deposition of (Cu<sup>+2</sup>-Cd<sup>+2</sup> and Cu<sup>+2</sup>-Ni<sup>+2</sup>-Cd<sup>+2</sup>) ion to their metals, with 200 ppm initial concentration of Cu<sup>+2</sup>, Ni<sup>+2</sup>, and Cd<sup>+2</sup> ions for individual deposition and (100, 200, and 300 ppm) of Cu<sup>+2</sup> ion for co-deposition experiments.

The major sets of experiments were considered with three volumetric flow rates (100, 200, and 300 l/h), four sampling times (0.5, 1.0, 1.5, and 2.0 h) for single mesh electrode, three sampling times (0.5, 1.0, and 2.0 h) for double and triple meshes electrode which were taken to examine the remaining concentration for  $Cu^{+2}$ , Ni<sup>+2</sup>, and Cd<sup>+2</sup> using Atomic Absorption Spectrophotometer (AAS) at 40  $^{0}C$ .

# 4.3 Electrochemical Reactor Design (Electrolytic Cell)

## **4.3.1 Preliminary Experiments**

An integrated electrochemical reactor (electrolytic cell) was fabricated in a beaker of 2 L, for more details see Fig 4-1. The electrolytic cell consisted of the following parts for a non flow system (static condition):

## **4.3.1.1** Working Electrode (Cathode)

The cathode was a rectangular brass mesh (50 mesh no.) with dimension (80x80x0.5 mm) and having a surface area equal to  $(171.9 \text{ cm}^2)$ . The cathode was placed vertically and held above the bottom of the solution container.

Brass alloy consisted of the following compositional elements:

[Copper (Cu) 87.8%, Zinc (Zn) 9.5%, Tin (Sn) 2.62%, Iron (Fe) 0.08% by weight]. This analysis was achieved by Geological Survey and Mining Company.

## 4.3.1.2 Counter Electrode (Anode)

The anode was a rectangular pure graphite flat plate with dimensions (80x80x1 mm), located vertically opposite to the working electrode in the solution container.

## 4.3.1.3 Water Bath

A water bath was used for heating electrolyte solution to the required experiment temperature within  $\pm 1$  <sup>0</sup>C with heater range 0-100 <sup>0</sup>C of type Memmert.



Figure 4-1 Schematic diagram of experimental electrochemical cell (non-flow system).

#### **4.3.2 Major Experiments**

An integrated electrochemical reactor (electrolytic cell) was fabricated in a cylindrical concentric electrochemical reactor of inside cathode electrode and surrounded by outside anode electrode, for more details see Figs. (4-2 and 4-3). The electrolytic cell consisted of the following parts:

#### 4.3.2.1 Calming Section

Calming section was made of teflon tube at the inlet and outlet of the electrochemical reactor of 5 cm internal diameter and 5 cm long which was held with a flange. The calming section was used to reduce the entrance effect of the entering electrolyte to the cathode section. Two taps were located before and after electrochemical cell at the end of calming section and extended through flexible plastic tube to the U-tube manometer to measure if there was any high pressure drop resulting from block up of mesh pore.

#### **4.3.2.2** Working Electrode (Cathode)

The cathode was made of brass mesh (50 mesh no.) alloy of cylindrical shape of 15 cm long and 2.5 cm in diameter with 0.05 cm thick (for experiments of a single mesh electrode) or 0.2 cm thick (for experiments of a double meshes electrode) or 0.4 cm thick (for experiments of a triple meshes electrode).

Two Teflon plates were placed at the top and the bottom of the edge of the counter electrode (anode), each plate had a neck to hold the mesh electrode at the center of the electrochemical reactor and make it stable when the electrolyte was flowing through the cell. The plate at the bottom was penetrated at the center from its neck with a 2 cm in diameter as the inlet of the electrolyte solution. The plate at the top was penetrated with small holes except its neck. So, the flow entered from the center of the bottom plate and then passed from the mesh pore along the cathode to the anode towards the top plate.

At the center of working electrode two glass capillary tubes were fixed, one for the experiments of single mesh electrode and the other for the double and triple mesh electrode which set for the outer mesh electrode through a small hole at the center of anode and extended through flexible plastic tube to the saturated calomel electrode (SCE) as indication to find out when the potential drop be same as the inner mesh. The capillary tips were positioned 1.0-2.0 mm from the surface of cathode in order to avoid interference with the fluid flow.

# 4.3.2.3 Counter Electrode (Anode)

The anode was of pure graphite tube of 5 cm internal diameter and 15 cm long. The anode electrode was held with a flange at the top and the bottom and connected with a flange of calming section.



Figure 4-2 Photo of experimental electrochemical reactor.



Figure 4-3 Schematic diagram of experimental electrochemical reactor (flow system).

Screws were used to join these parts, in this way the construction and reproducibility of the arrangement of the electrodes were made easy to handle, clean, and refreshing the bed.

# 4.4 The Flow System

The flow system was made of the following items. For more details see Figs. (4-4 and 4-5).

## 4.4.1 Electrolyte Solution Reservoir

The reservoir was made of a Perspex rectangular container having a capacity of 10.0 liter. The reservoir has one hole from the bottom for the outgoing electrolyte controlled by a PVC valve and three holes or connecting points, two holes at the top and the other one at side bottom used as follows:

- 1) Electrolyte circulation through bypass PVC valve.
- 2) Electrolyte return from electrochemical reactor.
- 3) Samplings withdraw of electrolyte solution through Pyrex valve.

## 4.4.2 Electrolyte Pump

The electrolyte flows in a closed circuit with provision for circulation by means of magnetic centrifugal pump made of PVDF having a capacity of (0.9 m<sup>3</sup>/hr and 0.25 KW) and maximum head of 4 m supplied by Clechorn Waring Pumps UK (type).

## 4.4.3 Flowmeter (Rotameter)

A PVC rotameter with a range of 0-2000 liter  $H_2O/hr$  at 20  $^{0}C$  having a Teflon float was used to measure electrolyte flow rates. The flow meter was calibrated by measuring the volume of electrolyte leaving the cell during a given interval of time at a test temperature.

## 4.4.4 Connecting Pipes and Valves

All connecting pipes (hoses) were made of reinforced transparent polyethylene and all valves were made of PVC. Thus all the components in the system were fabricated from non-metallic materials except the cathode.

# 4.4.5 Thermometer and Heater

They were made of glass to measure temperature up to 40  $^{0}$ C and heating the electrolyte solution respectively. The heater has a controller to set the temperature of the electrolyte to the required temperature.


Figure 4-4 Photo of experimental rig (flow system)



Figure 4-5 Schematic diagram of flow system.

## 4.5 Description of the Electrical Circuit

The electrical circuit consisted of the following items, for more details see Fig. 4-6.

## 4.5 1 D.C. Power Supply

A filtered DC power supply which is often equipped with current and voltage limiters offers better stability and control and can be used for applying potensotatic technique (constant voltage) or galvanostatic technique (constant current) boundary condition. It was supplied by Metcix Company, France (type AX 503), with range of 0-30 Volts and 0-6 Amperes.

#### 4.5 2 Resistance Box

A variable resistance (resistance box) made by (Donbriolge company). It provides an electrical resistance up to 0.1 M $\Omega$  with accuracy of 0.1  $\Omega$ .

## 4.5 3 Multirange Ammeter

The total current passed to the electrochemical reactor from the D.C. power supply during the electrolysis experiments was measured using digital multirange ammeter supplied by Hewlett-Packard Company, USA (type 3466A), with range of 0-4 Amperes.

#### 4.5 4 Multirange Voltmeter

The potential difference between the working electrode (cathode) and the reference electrode (Saturated Calomel Electrode) during the practical experiments was measured using digital multirange voltmeter supplied by Metcix Company, France (type AX 553), with range 0-450 Volts.

## 4.5 5 Reference Electrode

A saturated calomel reference electrode type (Inglod Electrode 303-57) was used in this work having the following reversible reaction.

$$Hg_2Cl_2 + 2e^- \longrightarrow 2Hg^+ + 2Cl^- \qquad \dots (4.1)$$

A saturated solution of potassium chloride is very convenient to use and the potential of saturated calomel electrode (SCE) is then 0.242 V with respect to the standard hydrogen electrode (SHE) at 25  $^{\circ}$ C with a temperature coefficient of  $-0.76 \text{ mV}/ {^{\circ}}$ C [2].

The reference electrode was mounted in a volumetric flask filled with the same electrolyte solution. A luggin capillary bridge led to the reference electrode by a transport plastic tube mounted near the working electrode [96] to within  $\approx$ 1-2 mm [97] perpendicular to the working electrode to minimize the ohmic resistance of the solution. For this purpose, a capillary about 1 mm in diameter was introduced between the working electrode and reference electrode. It was always made sure that no bubbles remained in the capillary and plastic tubes.



Figure 4-6 Schematic diagram of electric circuit.

## **4.6 Experimental Program**

#### **4.6.1 Electrolyte Preparation**

The electrolytic solutions used in this work were as follows:

a) Preliminary experiments:

Sulfuric acid solution (0.5 M) prepared from Analar grade acid supplied by s d fine-chem limited Mumbai (purity > 98 wt%, sp.gr. = 1.838) + Analar copper sulfate (CuSO<sub>4</sub>.5H<sub>2</sub>O with M.wt. = 249.68, supplied by B D H chemicals Ltd. Poole England) to deposit 200 ppm of Cu<sup>+2</sup> to Cu metal.

b) Major experiments

Deposition of individual ion

- 1) Sulfuric acid (0.5 M) + Analar copper sulfate (CuSO<sub>4</sub>.5H<sub>2</sub>O) with initial concentration of 200 ppm of Cu<sup>+2</sup>.
- 2) Sulfuric acid (0.5 M) + Analar nickel sulfate (NiSO<sub>4</sub>.6H<sub>2</sub>O with M.wt. = 262.85, supplied by B D H chemicals Ltd. Poole England) with initial concentration of 200 ppm of Ni<sup>+2</sup>.
- Sulfuric acid (0.5 M) + Analar cadmium sulfate (3CdSO<sub>4</sub>.8H<sub>2</sub>O with M.wt. = 769.5, supplied by B D H chemicals Ltd. Poole England) with initial concentration of 200 ppm of Cd<sup>+2</sup>.

Co-deposition of ions

- 1) Sulfuric acid (0.5 M) + Analar copper sulfate (CuSO<sub>4</sub>.5H<sub>2</sub>O) with different initial concentrations of (100, 200, and 300 ppm) of Cu<sup>+2</sup> ion + Analar nickel sulfate (NiSO<sub>4</sub>.6H<sub>2</sub>O) with 200 ppm initial concentration.
- Sulfuric acid (0.5 M) + Analar copper sulfate (CuSO<sub>4</sub>.5H<sub>2</sub>O) with different initial concentrations of (100, 200, and 300 ppm) of Cu<sup>+2</sup> ion + Analar cadmium sulfate (3CdSO<sub>4</sub>.8H<sub>2</sub>O), with 200 ppm initial concentration.
- 3) Sulfuric acid (0.5 M) + Analar copper sulfate ( $CuSO_4.5H_2O$ ) with different initial concentrations of (100, 200, and 300 ppm) of  $Cu^{+2}$ ion + Analar nickel sulfate (NiSO<sub>4</sub>.6H<sub>2</sub>O) with 200 ppm initial

concentration + Analar cadmium sulfate  $(3CdSO_4.8H_2O)$  with 200 ppm initial concentration.

The physical properties of the electrolytic solutions, i.e. density and viscosity were measured experimentally at experiments temperature (40  $^{0}$ C). The density was measured using pycnometer, while the viscosity was measured by a rotational viscometer. The average physical properties of the electrolytic solutions are presented in Table 4-1

Property	Temperature 40 <sup>0</sup> C
Density $\rho$ (kg/m <sup>3</sup> )	1034
Viscosity µ (kg/m.s)	$7.5^{*}10^{-5}$

**Table 4-1** Physical properties of the electrolytic solutions.

#### **4.6.2 Electrode Surface Preparation**

Before and after each experimental run, the metal specimen (cathode brass mesh electrode) was prepared by immersing in 0.1 M  $H_2SO_4$  for two minutes in order to be free from any deposition or oxides, then washed with distilled water, dried with clean paper tissue followed by immersing in Analar methanol for two minutes, then dried with clean paper tissue, and then immersed in Analar acetone for two minutes, and finally left to dry for 0.5 h in the desiccater over silica gel, then weighing the cathode mesh were carried out using digital balance to 0.1 g accuracy.

The working cathode mesh electrode was fixed at the center of the reactor surrounded by the counter anode electrode making a concentric cylinder. The two electrodes (cathode and anode) were bolted together, and then the electrolytic cell was connected to the flow system and the electrical circuit.

#### **4.6.3 Experimental Procedure**

## A. Preliminary Experiments

The experiments were used to measure the cathodic deposition of  $Cu^{+2}$  ion to Cu metal on a single, double, and triple brass meshes electrode.

Before starting each run, 1.5711 g of copper sulfate (CuSO<sub>4</sub>.5H<sub>2</sub>O) which corresponded to 200 ppm (0.2 g/l) of Cu<sup>+2</sup> ion and 54.4 ml which corresponded to 0.5 M H<sub>2</sub>SO<sub>4</sub> were prepared and added to the beaker and diluted by using distilled water to 2 liter to obtain the required electrolyte solution. The electrolyte was stirred using a glass rod in order to obtain a homogenous solution, then the heater controller in bath was set to the required temperature 40  $^{0}$ C to achieve thermal equilibrium before starting the experimental run.

Cathode and anode were immersed in the 2 liter beaker to a distance above the bottom of it, and the two electrode were held by a glass tube. The space between cathode and anode was kept constant throughout the experiments in order to obtain a uniform result by making IR drop constant.

The electrical circuit was switched on as soon as the electrode was covered by the electrolyte. According to the galvanostatic technique, the voltage of the DC power supply remained constant during the experiments, and the resistance of the circuit was changed until obtaining a total current of 15 mA. The experimental run lasted 0.5 h. The above procedure was repeated for single, double, and triple meshes and total current (15, 20, 25, 70, and 90 mA for single mesh), (30, 40, 50, 140, and 180 mA for double meshes), and (45, 60, 75, 210, and 270 mA for triple meshes).

#### **B.** Major Experiments

The experiments were used to measure the individual cathodic deposition of  $Cu^{+2}$  ion to Cu metal,  $Ni^{+2}$  ion to Ni metal, and  $Cd^{+2}$  ion to Cd metal, and co-deposition of  $Ni^{+2}$  ion and  $Cd^{+2}$  ion with  $Cu^{+2}$  ion.

Before starting each run, 4.7133 g of copper sulfate (CuSO<sub>4</sub>.5H<sub>2</sub>O) which corresponded to 200 ppm (0.2 g/l) of Cu<sup>+2</sup> ion [2.3566 g for 100 ppm (0.1 g/l) of Cu<sup>+2</sup> ion and 7.0699 g for 300 ppm (0.3 g/l) Cu<sup>+2</sup> ion] and 163 ml which corresponded to 0.5 M H<sub>2</sub>SO<sub>4</sub> were prepared and added to the reservoir. The heater controller in the reservoir was set to the required temperature 40  $^{0}$ C, during the heating period until the electrolyte temperature reached the required temperature level; the electrical circuit was connected to the cell and to the reference electrode.

After checking all the electrical connections, valve 1 and 3 were opened and the pump was switched on and the electrolyte started to flow in the system through the bypass section without letting the electrolyte to flow through the cell in order to obtain a homogenous solution before getting in the cell. When the electrolyte reached the required temperature; the bypass was closed and the cell valve (valve 2) was opened to allow the electrolyte to flow through the electrochemical reactor and to the reference electrode through the connecting bridge.

The electrical circuit was switched on as soon as the electrode was covered by the electrolyte. According to the galvanostatic technique, the voltage of the DC power supply remained constant during the experiments, and the resistance of the circuit was changed until obtaining a total current of 30 mA. The experimental run lasted 2 h with sampling withdraw time each 0.5 h.

At the end of each run the power supply was switched off, the drain valve (valve 4) was opened in order to drain the used electrolytes. The system was then washed with distilled water several times in circulation to make sure that there was no electrolyte left in the system.

The above procedure was repeated for single, double, and triple mesh, individual deposition  $Cu^{+2}$ ,  $Ni^{+2}$  [5.3739 g of nickel sulfate (NiSO<sub>4</sub>.6H<sub>2</sub>O) which corresponded to 200 ppm (0.2 g/l) of Ni<sup>+2</sup> ion], and Cd<sup>+2</sup> [8.2136 g of cadmium sulfate (3CdSO<sub>4</sub>.8H<sub>2</sub>O) which corresponded to 200 ppm (0.2 g/l) of Cd<sup>+2</sup> ion, co-deposition (Cu-Ni ,Cu-Cd, and Cu-Ni-Cd) in different concentration ratios (100:200, 200:200, and 300:200 for Cu-Ni and Cu-Cd) and (100:200:200, 200:200, and 300:200 for Cu-Ni-Cd), different flow rates of (100, 200, and 300 l/h), different sampling times (0.5, 1.0, 1.5, and 2.0 h) for single mesh and (0.5, 1.0, and 2.0 h) for double and triple meshes which were taken to examine the remaining concentration for  $Cu^{+2}$ , Ni<sup>+2</sup>, and Cd<sup>+2</sup> by using Atomic Absorption Spectrophotometer (AAS), and finally different total current as mentioned above. Experiments were repeated to insure the accuracy when reproducibility was in doubt.

# Chapter Five Results and Interpretations

# **5.1 Introduction**

The main purpose of the present experiments, which were carried out in 0.5 M  $H_2SO_4$  as supporting electrolyte, is to investigate the removal of  $Cu^{+2}$  ion in non-flow system and the removal of  $Cu^{+2}$ , Ni<sup>+2</sup>, and Cd<sup>+2</sup> ions in flow system (individual deposit and co-deposit as described previously) and also study the behavior of a fixed bed flow-by electrochemical reactor in term of current efficiency of ions removal which is affected by hydrogen evolution, and also study the optimum bed thickness (no. of meshes) in different conditions as described previously.

The electrode potential is presented with reference to the saturated calomel electrode (SCE).

Tables of experimental runs are classified as groups, including the experimental conditions are presently reported. Concentration remaining curves versus time for selected conditions are presented, while the data for all the experiments are tabulated in Appendix A (for non-flow system) and Appendix B (for flow system).

# **5.2 Experimental Conditions Studied**

In the present work two sets of experiments have been studied:

## 1. Preliminary Experiments (Non-Flow System)

In this part of experiments five different total currents (15, 20, 25, 70, and 90 mA) were used for single mesh, duplicated for double meshes, and triplicated for triple meshes in 0.5  $H_2SO_4$  as supporting electrolyte with 200 ppm initial Cu<sup>+2</sup> ion concentration.

#### 2. Major Experiments (Flow System)

In this part of experiments the influences of five parameters were studied as follows:

#### A. Fixed Bed Thickness (No. of Meshes)

Single (0.05 cm), double (0.2 cm), and triple (0.4 cm) bed thicknesses were used to determine optimum bed thickness.

#### **B. Electrolyte Flow Rate**

Three different electrolyte flow rates (100, 200, and 300 l/h) were used to determine the effect of flow rate on the removal of metal ions.

#### **C. Total Current**

Five different total currents for single mesh electrode, three different total currents for double meshes electrode, and one total current for triple meshes electrode as mentioned previously, were used to determine the effect of total current on the removal of metal ions.

#### **D.** Copper Concentration

Three different copper concentrations (100, 200, and 300 ppm) were used to determine the influence of changing copper concentration on removal of metal ions  $(Ni^{+2} \text{ and } Cd^{+2})$  for co-deposition.

#### **E.** Sampling Time (Deposition Time)

Four different sampling withdraw times (0.5, 1.0, 1.5, and 2.0 h) for single mesh and three sampling withdraw times (0.5, 1.0, and 2.0 h) for double and triple meshes, to determine the concentration gradient with time.

# **5.3 Sets of Experiments**

## Part A Single Mesh (Individual Deposition)

In this part of experiments 200 ppm initial concentration of metal ions ( $Cu^{+2}$ ,  $Ni^{+2}$ , and  $Cd^{+2}$ ) were used for different flow rates, total currents, and deposition times. Conditions of these experiments were as follows:

Fyn	$Cu^{+2}$ ion	Total Current	Flow rate	Sam	nling
No	Conc (nnm)	$(m\Delta)$	(1/h)	Tim	ping e (h)
110.	Conc. (ppm)		(1/11)	0.5	
1	200	30	100	1.5	2.0
2	200	20	200	0.5	1.0
2	200	30	200	1.5	2.0
3	200	30	300	0.5	1.0
	200	50	500	1.5	2.0
1	200	40	100	0.5	1.0
-	200	40	100	1.5	2.0
5	200	40	200	0.5	1.0
5	200	40	200	1.5	2.0
6	200	40	200	0.5	1.0
0	200	40	300	1.5	2.0
7	200	50	100	0.5	1.0
/	200	50	100	1.5	2.0
Q	200	50	200	0.5	1.0
0	200	50	200	1.5	2.0
0	200	50	300	0.5	1.0
9	200	50	500	1.5	2.0
10	200	145	100	0.5	1.0
10	200	145	100	1.5	2.0
11	200	145	200	0.5	1.0
11	200	145	200	1.5	2.0
12	200	145	200	0.5	1.0
12	200	145	300	1.5	2.0
12	200	100	100	0.5	1.0
15	200	190	100	1.5	2.0
14	200	100	200	0.5	1.0
14	200	190	200	1.5	2.0
15	200	100	200	0.5	1.0
15	200	190	300	1.5	2.0

**Table 5-1** Conditions of experiments for  $Cu^{+2}$  deposition.

Exp.	Ni <sup>+2</sup> ion	Total Current	Flow rate	Sam	pling
No.	Conc. (ppm)	(mA)	(l/h)	Tim	e (h)
16	200	145	100	0.5	1.0
10	200	145	100	1.5	2.0
17	200	145	200	0.5	1.0
17	200	145	200	1.5	2.0
18	200	145	300	0.5	1.0
10	200	145	500	1.5	2.0
19	200	160	100	0.5	1.0
17	200	100	100	1.5	2.0
20	200	160	200	0.5	1.0
20	200	100	200	1.5	2.0
21	200	160	300	0.5	1.0
21	200	100	500	1.5	2.0
22	200	175	100	0.5	1.0
22	200	175	100	1.5	2.0
23	200	175	200	0.5	1.0
23	200	175	200	1.5	2.0
24	200	175	300	0.5	1.0
24	200	175	500	1.5	2.0
25	200	100	100	0.5	1.0
23	200	190	100	1.5	2.0
26	200	100	200	0.5	1.0
20	200	190	200	1.5	2.0
27	200	100	300	0.5	1.0
21	200	190	300	1.5	2.0
28	200	205	100	0.5	1.0
20	200	203	100	1.5	2.0
20	200	205	200	0.5	1.0
29	200	203	200	1.5	2.0
30	200	205	300	0.5	1.0
50	200	205	500	1.5	2.0

 Table 5-2 Conditions of experiments for Ni<sup>+2</sup> deposition.

Exp.	$Cd^{+2}$ ion	Total Current	Flow rate	Sam	pling
No.	Conc. (ppm)	(mA)	(l/h)	Tim	e (h)
31	200	175	100	0.5	1.0
51	200	175	100	1.5	2.0
32	200	175	200	0.5	1.0
52	200	175	200	1.5	2.0
33	200	175	300	0.5	1.0
55	200	175	500	1.5	2.0
34	200	190	100	0.5	1.0
51	200	150	100	1.5	2.0
35	200	190	200	0.5	1.0
55	200	170	200	1.5	2.0
36	200	190	300	0.5	1.0
50	200	170	500	1.5	2.0
37	200	205	100	0.5	1.0
57	200	203	100	1.5	2.0
38	200	205	200	0.5	1.0
50	200	203	200	1.5	2.0
39	200	205	300	0.5	1.0
57	200	203	500	1.5	2.0
40	200	220	100	0.5	1.0
10	200	220	100	1.5	2.0
41	200	220	200	0.5	1.0
11	200	220	200	1.5	2.0
42	200	220	300	0.5	1.0
12	200	220	500	1.5	2.0
43	200	235	100	0.5	1.0
15	200	233	100	1.5	2.0
44	200	235	200	0.5	1.0
	200	200	200	1.5	2.0
45	200	235	300	0.5	1.0
45	200	233	300	1.5	2.0

**Table 5-3** Conditions of experiments for Cd<sup>+2</sup> deposition.

## Part B Single Mesh (Two Metal Co-deposition)

In this part of experiments different concentrations ratio of Cu-Ni and Cu-Cd (100:200, 200:200, and 300:200) were used for different flow rates, total currents, and deposition times. Conditions of these experiments were as follows:

Exp.	Cu <sup>+2</sup> -Ni <sup>+2</sup> ion	Total Current	Flow rate	Sam	pling
No.	Conc. ratio (ppm)	(mA)	(l/h)	Tim	e (h)
16	100,200	145	100	0.5	1.0
40	100.200	145	100	1.5	2.0
17	100.200	145	200	0.5	1.0
47	100.200	145	200	1.5	2.0
19	100.200	145	200	0.5	1.0
40	100.200	143	300	1.5	2.0
40	100.200	160	100	0.5	1.0
49	100.200	100	100	1.5	2.0
50	100.200	160	200	0.5	1.0
50	100.200	100	200	1.5	2.0
51	100.200	160	200	0.5	1.0
51	100.200	100	300	1.5	2.0
50	100.200	175	100	0.5	1.0
32	100.200	175	100	1.5	2.0
53	100.200	175	200	0.5	1.0
55	100.200	175	200	1.5	2.0
54	100.200	175	300	0.5	1.0
54	100.200	175	500	1.5	2.0
55	100.200	190	100	0.5	1.0
55	100.200	170	100	1.5	2.0
56	100.200	190	200	0.5	1.0
50	100.200	170	200	1.5	2.0
57	100.200	190	300	0.5	1.0
57	100.200	170	500	1.5	2.0
58	100.200	205	100	0.5	1.0
50	100.200	203	100	1.5	2.0
59	100.200	205	200	0.5	1.0
	100.200	203	200	1.5	2.0
60	100.200	205	300	0.5	1.0
00	100.200	205	500	1.5	2.0

**Table 5-4** Conditions of experiments for  $Cu^{+2}$ -Ni<sup>+2</sup> co-deposition.

**Table 5-5** Conditions of experiments for  $Cu^{+2}$ -Ni<sup>+2</sup> co-deposition.

Exp.	Cu <sup>+2</sup> -Ni <sup>+2</sup> ion	Total Current	Flow rate	Sam	pling
No.	Conc. ratio (ppm)	(mA)	(l/h)	Tim	e (h)
61	200.200	145	100 -	0.5	1.0
01	200.200	145		1.5	2.0
62	200.200	145	200	0.5	1.0
02	200:200	145		1.5	2.0
63	200:200	145	200	0.5	1.0
			300	1.5	2.0

64	200:200	160	100	0.5	1.0
				1.5	2.0
65	200.200	160	200	0.5	1.0
03	200:200	100	200	1.5	2.0
	200.200	1.00	200	0.5	1.0
66	200:200	160	300	1.5	2.0
<b>67</b>	200.200	175	100	0.5	1.0
6/	200:200	1/5	100	1.5	2.0
69	200.200	175	200	0.5	1.0
08	200:200	175	200	1.5	2.0
(0)	200.200	175	200	0.5	1.0
69	200:200	1/5	300	1.5	2.0
70	200.200	100	100	0.5	1.0
70	200:200	190	100	1.5	2.0
71	200.200	100	200	0.5	1.0
/1	200.200	190	200	1.5	2.0
72	200.200	100	300	0.5	1.0
12	200.200	190	300	1.5	2.0
72	200.200	205	100	0.5	1.0
15	200.200	203	100	1.5	2.0
74	200.200	205	200	0.5	1.0
/+	200.200	205	200	1.5	2.0
75	200.200	205	200	0.5	1.0
15	200.200	205	300	1.5	2.0

Table 5-5 Cont.

**Table 5-6** Conditions of experiments for  $Cu^{+2}$ -Ni<sup>+2</sup> co-deposition.

Exp.	Cu <sup>+2</sup> -Ni <sup>+2</sup> ion	Total Current	Flow rate	Sam	pling
No.	Conc. ratio (ppm)	(mA)	(l/h)	Tim	e (h)
76	300.200	145	100	0.5	1.0
70	300.200	145	100	1.5	2.0
77	300.200	145	200	0.5	1.0
//	300.200	145	200	1.5	2.0
79	300.200	145	300	0.5	1.0
78	300.200	145	300	1.5	2.0
70	200.200	160	100	0.5	1.0
19	500:200	100	100	1.5	2.0
80	300.200	160	200	0.5	1.0
80	300.200	100	200	1.5	2.0
<b>Q</b> 1	300.200	160	300	0.5	1.0
01	300.200	100	500	1.5	2.0
87	300.200	175	100	0.5	1.0
02	300.200	175	100	1.5	2.0
02	200.200	175	200	0.5	1.0
65	300.200	175	200	1.5	2.0

84	300.200	175	300	0.5	1.0
04	500.200	175	500	1.5	2.0
85	300.200	100	100	0.5	1.0
65	300.200	190	100	1.5	2.0
96	200.200	100	200	0.5	1.0
80	500.200	190	200	1.5	2.0
07	200.200	190 300	200	0.5	1.0
0/	500.200		500	1.5	2.0
00	200.200	205	100	0.5	1.0
00	500.200	203	100	1.5	2.0
80	200.200	205	200	0.5	1.0
89	300:200		200	1.5	2.0
90	200.200	205	300	0.5	1.0
	300:200			1.5	2.0

Table 5-6 Cont.

**Table 5-7** Conditions of experiments for  $Cu^{+2}$ - $Cd^{+2}$  co-deposition.

Exp.	$Cu^{+2}$ - $Cd^{+2}$ ion	Total Current	Flow rate	Sam	pling
No.	Conc. ratio (ppm)	(mA)	(l/h)	Tim	e (h)
01	100,200	175	100	0.5	1.0
91	100.200	175	100	1.5	2.0
02	100.200	175	200	0.5	1.0
92	100.200	175	200	1.5	2.0
02	100.200	175	300	0.5	1.0
93	100.200	175	300	1.5	2.0
04	100.200	100	100	0.5	1.0
94	100.200	190	100	1.5	2.0
05	100.200	100	200	0.5	1.0
95	100.200	190	200	1.5	2.0
06	100.200	100	200	0.5	1.0
90	100.200	190	300	1.5	2.0
07	100.200	205	100	0.5	1.0
97	100.200	203	100	1.5	2.0
08	100.200	205	200	0.5	1.0
90	100.200	203	200	1.5	2.0
00	100.200	205	200	0.5	1.0
99	100.200	203	300	1.5	2.0
100	100.200	220	100	0.5	1.0
100	100.200	220	100	1.5	2.0
101	100.200	220	200	0.5	1.0
101	100.200	220	200	1.5	2.0
102	100.200	220	300	0.5	1.0
102	100.200	220	300	1.5	2.0
103	100.200	235	100	0.5	1.0
105	100.200	233	100	1.5	2.0

Table 5-7 Cont

104	100:200	235	200	0.5	1.0
				1.5	2.0
105	100:200	235	300	0.5	1.0
				1.5	2.0

**Table 5-8** Conditions of experiments for  $Cu^{+2}$ - $Cd^{+2}$  co-deposition.

Exp.	$Cu^{+2}$ - $Cd^{+2}$ ion	Total Current	Flow rate	Sam	pling
No.	Conc. ratio (ppm)	(mA)	(l/h)	Tim	e (h)
106	200.200	175	100	0.5	1.0
100	200.200	175	100	1.5	2.0
107	200.200	175	200	0.5	1.0
107	200.200	175	200	1.5	2.0
108	200.200	175	300	0.5	1.0
100	200.200	175	500	1.5	2.0
109	200.200	190	100	0.5	1.0
107	200.200	170	100	1.5	2.0
110	200.200	190	200	0.5	1.0
110	200.200	190	200	1.5	2.0
111	200.200	100	300	0.5	1.0
111	200.200	190	300	1.5	2.0
112	200.200	205	100	0.5	1.0
112	200.200	203	100	1.5	2.0
112	200.200	205	200	0.5	1.0
115	200.200	203	200	1.5	2.0
114	200.200	205	300	0.5	1.0
114	200.200	203	300	1.5	2.0
115	200.200	220	100	0.5	1.0
115	200.200	220	100	1.5	2.0
116	200.200	220	200	0.5	1.0
110	200.200	220	200	1.5	2.0
117	200.200	220	300	0.5	1.0
11/	200.200	220	300	1.5	2.0
110	200.200	225	100	0.5	1.0
110	200.200	233	100	1.5	2.0
110	200.200	225	200	0.5	1.0
119	200:200	233	200	1.5	2.0
120	200.200	225	200	0.5	1.0
120	200:200	233	300	1.5	2.0

Exp.	$Cu^{+2}$ - $Cd^{+2}$ ion	Total Current	Flow rate	Sam	pling
No.	Conc. ratio (ppm)	(mA)	(l/h)	Tim	e (h)
121	300.200	175	100	0.5	1.0
121	300.200	175	100	1.5	2.0
122	300.200	175	200	0.5	1.0
122	500.200	175	200	1.5	2.0
123	300.200	175	300	0.5	1.0
123	500.200	175	500	1.5	2.0
124	300.200	190	100	0.5	1.0
127	500.200	170	100	1.5	2.0
125	300.200	190	200	0.5	1.0
123	500.200	170	200	1.5	2.0
126	300.200	190	300	0.5	1.0
120	500.200	170	500	1.5	2.0
127	300.200	205	100	0.5	1.0
127	500.200	205	100	1.5	2.0
128	300.200	205	200	0.5	1.0
120	500.200	203	200	1.5	2.0
129	300.200	205	300	0.5	1.0
12)	500.200	205	500	1.5	2.0
130	300.200	220	100	0.5	1.0
150	500.200	220	100	1.5	2.0
131	300.200	220	200	0.5	1.0
151	500.200	220	200	1.5	2.0
132	300.200	220	300	0.5	1.0
152	500.200	220	500	1.5	2.0
133	300.200	235	100	0.5	1.0
155	500.200	233	100	1.5	2.0
134	300.200	235	200	0.5	1.0
134	500.200	233	200	1.5	2.0
135	300.200	235	300	0.5	1.0
155	300.200	233	500	1.5	2.0

**Table 5-9** Conditions of experiments for  $Cu^{+2}$ - $Cd^{+2}$  co-deposition.

# Part C Single Mesh (Three Metal Co-deposition)

In this part of experiments different concentrations ratio of Cu-Ni-Cd (100:200:200, 200:200, and 300:200:200) were used for different flow rates, total currents, and deposition times. Conditions of these experiments were as follows:

Exp. No.	Cu <sup>+2</sup> -Ni <sup>+2</sup> -Cd <sup>+2</sup> ion Conc. ratio (ppm))	Total Current (mA)	Flow rate (1/h)	Samj Tim	pling e (h)
136	100:200:200	175	100	0.5	1.0
				1.5	2.0
137	100:200:200	175	200	0.5	1.0
				0.5	1.0
138	100:200:200	175	300	1.5	2.0
139	100.200.200	190	100	0.5	1.0
157	100.200.200	170	100	1.5	2.0
140	100.200.200	100	200	0.5	1.0
140	100.200.200	190	200	1.5	2.0
141	100.200.200	100	200	0.5	1.0
141	100.200.200	190	300	1.5	2.0
142	100.200.200	205	100	0.5	1.0
142	100:200:200	203		1.5	2.0
143	100:200:200	205	200	0.5	1.0
				1.5	2.0
144	100:200:200	205	300	0.5	1.0
				1.5	2.0
145	100:200:200	220	100	0.5	1.0
				1.5	2.0
146	100:200:200	220	200	0.5	1.0
				1.5	2.0
147	100.200.200	220	300	0.5	1.0
117	100.200.200		200	1.5	2.0
148	100.200.200	235	100	0.5	1.0
110	100.200.200	233	100	1.5	2.0
149	100.200.200	235	200	0.5	1.0
177	100.200.200	233	200	1.5	2.0
150	100.200.200	225	300	0.5	1.0
150	100.200.200	233	300	1.5	2.0

**Table 5-10** Conditions of experiments for  $Cu^{+2}$ -Ni<sup>+2</sup>-Cd<sup>+2</sup> co-deposition.

**Table 5-11** Conditions of experiments for  $Cu^{+2}$ -Ni<sup>+2</sup>-Cd<sup>+2</sup> co-deposition.

Exp. No.	Cu <sup>+2</sup> -Ni <sup>+2</sup> -Cd <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (l/h)	Samj Tim	pling e (h)
151	200:200:200	175	100	0.5	1.0 2.0
152	200:200:200	175	200	0.5	1.0 2.0

153	200:200:200	175	300	0.5	1.0 2.0
15/	200.200.200	190	100	0.5	1.0
134	200.200.200	170	100	1.5	2.0
155	200.200.200	100	200	0.5	1.0
155	200:200:200	190	200	1.5	2.0
156	200.200.200	100	200	0.5	1.0
150	200:200:200	190	300	1.5	2.0
157	200.200.200	205	100	0.5	1.0
157	200:200:200	205	100	1.5	2.0
170	200 200 200	205	200	0.5	1.0
158	200:200:200	205	200	1.5	2.0
1.50	200 200 200	205	200	0.5	1.0
159	200:200:200	205 300	1.5	2.0	
1.00	200 200 200	220	100	0.5	1.0
160	200:200:200	220	100	1.5	2.0
1.61	200 200 200	220	200	0.5	1.0
161	200:200:200	220	200	1.5	2.0
1.00	200 200 200	220	200	0.5	1.0
162	200:200:200	220	300	1.5	2.0
1.02	200.200.200	225	100	0.5	1.0
163	200:200:200	255	100	1.5	2.0
1.6.4	200 200 200	225	200	0.5	1.0
164	200:200:200	235	200	1.5	2.0
1.65	200 200 200	225	200	0.5	1.0
165	200:200:200	235	300	1.5	2.0
				1	

 Table 5-11 Cont.

**Table 5-12** Conditions of experiments for  $Cu^{+2}$ -Ni<sup>+2</sup>-Cd<sup>+2</sup> co-deposition.

Exp. No.	Cu <sup>+2</sup> -Ni <sup>+2</sup> -Cd <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (l/h)	Samj Tim	pling e (h)
166	300:200:200	175	100	0.5	1.0 2.0
167	300:200:200	175	200	0.5 1.5	1.0 2.0
168	300:200:200	175	300	0.5	1.0 2.0
169	300:200:200	190	100	0.5	1.0 2.0
170	300:200:200	190	200	0.5 1.5	1.0 2.0

$\begin{array}{c ccccccccccccccccccccccccccccccccccc$						
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	171	300.200.200	100	300	0.5	1.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	1/1	500.200.200	190	500	1.5	2.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	172	300.200.200	205	100	0.5	1.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	172	300.200.200	203	100	1.5	2.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	172	300.200.200	205	200	0.5	1.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	175	300.200.200	203	200	1.5	2.0
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	174	200.200.200	205	200	0.5	1.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	1/4	500.200.200	203	300	1.5	2.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	175	200.200.200	220	100	0.5	1.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	175	500.200.200	220	100	1.5	2.0
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	176	200.200.200	220	200	0.5	1.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	170	500.200.200	220	200	1.5	2.0
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	177	200.200.200	220	200	0.5	1.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	1//	500.200.200	220	300	1.5	2.0
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	179	300.200.200	225	100	0.5	1.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	170	300.200.200	233	100	1.5	2.0
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	170	200.200.200	225	200	0.5	1.0
180         300:200:200         235         300         0.5         1.0           1.5         2.0	179	500:200:200	255	200	1.5	2.0
180 300.200.200 235 300 1.5 2.0	190	200.200.200	225	200	0.5	1.0
	160	500:200:200	233	300	1.5	2.0

Table 5-12 Cont.

## Part D Double Mesh (Individual deposition)

In this part of experiments 200 ppm initial concentration of metal ions ( $Cu^{+2}$ ,  $Ni^{+2}$ , and  $Cd^{+2}$ ) were used for different flow rates, total currents, and deposition times. Conditions of these experiments were as follows:

Exp. No.	Cu <sup>+2</sup> ion Conc. (ppm)	Total Current (mA)	Flow rate (l/h)	Sa T	amplii ime (l	ng h)
181	200	60	100	0.5	1.0	2.0
182	200	60	200	0.5	1.0	2.0
183	200	60	300	0.5	1.0	2.0
184	200	100	100	0.5	1.0	2.0
185	200	100	200	0.5	1.0	2.0
186	200	100	300	0.5	1.0	2.0
187	200	380	100	0.5	1.0	2.0

**Table 5-13** Conditions of experiments for Cu<sup>+2</sup> deposition.

Table 5-13 Cont.

188	200	380	200	0.5	1.0	2.0
189	200	380	300	0.5	1.0	2.0

 Table 5-14 Conditions of experiments for Ni<sup>+2</sup> deposition.

Exp. No	Ni <sup>+2</sup> ion Conc. (ppm)	Total Current (mA)	Flow rate	Sa T	amplin ime (l	ng h)
190	200	290	100	0.5	1.0	2.0
191	200	290	200	0.5	1.0	2.0
192	200	290	300	0.5	1.0	2.0
193	200	350	100	0.5	1.0	2.0
194	200	350	200	0.5	1.0	2.0
195	200	350	300	0.5	1.0	2.0
196	200	410	100	0.5	1.0	2.0
197	200	410	200	0.5	1.0	2.0
198	200	410	300	0.5	1.0	2.0

**Table 5-15** Conditions of experiments for Cd<sup>+2</sup> deposition.

Exp. No.	Cd <sup>+2</sup> ion Conc. (ppm)	Total Current (mA)	Flow rate (l/h)	Sa T	amplii ime (l	ng h)
199	200	350	100	0.5	1.0	2.0
200	200	350	200	0.5	1.0	2.0
201	200	350	300	0.5	1.0	2.0
202	200	410	100	0.5	1.0	2.0
203	200	410	200	0.5	1.0	2.0
204	200	410	300	0.5	1.0	2.0
205	200	470	100	0.5	1.0	2.0
206	200	470	200	0.5	1.0	2.0
207	200	470	300	0.5	1.0	2.0

## Part E Double Meshes (Two Metal Co-deposition)

In this part of experiments different concentrations ratio of Cu-Ni and Cu-Cd (100:200, 200:200, and 300:200) were used for different flow rates, total currents, and deposition times. Conditions of these experiments were as follows:

Exp. No.	Cu <sup>+2</sup> -Ni <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (1/h)	Sa T	amplii ime (l	ng 1)
208	100:200	290	100	0.5	1.0	2.0
209	100:200	290	200	0.5	1.0	2.0
210	100:200	290	300	0.5	1.0	2.0
211	100:200	350	100	0.5	1.0	2.0
212	100:200	350	200	0.5	1.0	2.0
213	100:200	350	300	0.5	1.0	2.0
214	100:200	410	100	0.5	1.0	2.0
215	100:200	410	200	0.5	1.0	2.0
216	100:200	410	300	0.5	1.0	2.0

**Table 5-16** Conditions of experiments for Cu<sup>+2</sup>-Ni<sup>+2</sup> co-deposition.

**Table 5-17** Conditions of experiments for Cu<sup>+2</sup>-Ni<sup>+2</sup> co-deposition.

Exp.	$Cu^{+2}$ -Ni <sup>+2</sup> ion	Total Current $(mA)$	Flow rate	Sa	amplii ime (l	ng
217	200:200	290	100	0.5	1.0	2.0
218	200:200	290	200	0.5	1.0	2.0
219	200:200	290	300	0.5	1.0	2.0
220	200:200	350	100	0.5	1.0	2.0
221	200:200	350	200	0.5	1.0	2.0
222	200:200	350	300	0.5	1.0	2.0
223	200:200	410	100	0.5	1.0	2.0
223	200:200	410	200	0.5	1.0	2.0
224	200:200	410	200	0.5	1.0	2.0
223	200:200	410	300	0.5	1.0	2.0

Exp. No.	Cu <sup>+2</sup> -Ni <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (1/h)	Sa T	amplin ime (l	ng h)
226	300:200	290	100	0.5	1.0	2.0
227	300:200	290	200	0.5	1.0	2.0
228	300:200	290	300	0.5	1.0	2.0
229	300:200	350	100	0.5	1.0	2.0
230	300:200	350	200	0.5	1.0	2.0
231	300:200	350	300	0.5	1.0	2.0
232	300:200	410	100	0.5	1.0	2.0
233	300:200	410	200	0.5	1.0	2.0
234	300:200	410	300	0.5	1.0	2.0

 Table 5-18 Conditions of experiments for Cu<sup>+2</sup>-Ni<sup>+2</sup> co-deposition.

**Table 5-19** Conditions of experiments for Cu<sup>+2</sup>-Cd<sup>+2</sup> co-deposition.

Exp. No.	Cu <sup>+2</sup> -Cd <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (1/h)	Sa T	amplin ime (l	ng h)
235	100:200	350	100	0.5	1.0	2.0
236	100:200	350	200	0.5	1.0	2.0
237	100:200	350	300	0.5	1.0	2.0
238	100:200	410	100	0.5	1.0	2.0
239	100:200	410	200	0.5	1.0	2.0
240	100:200	410	300	0.5	1.0	2.0
241	100:200	470	100	0.5	1.0	2.0
242	100:200	470	200	0.5	1.0	2.0
243	100:200	470	300	0.5	1.0	2.0

Exp.	$Cu^{+2}$ - $Cd^{+2}$ ion	Total Current	Flow rate	Sampling		
No.	Conc. ratio (ppm)	(mA)	(l/h)	Т	ime (l	h)
244	200:200	350	100	0.5	1.0	2.0
245	200:200	350	200	0.5	1.0	2.0
246	200:200	350	300	0.5	1.0	2.0
247	200:200	410	100	0.5	1.0	2.0
248	200:200	410	200	0.5	1.0	2.0
249	200:200	410	300	0.5	1.0	2.0
250	200:200	470	100	0.5	1.0	2.0
251	200:200	470	200	0.5	1.0	2.0
252	200:200	470	300	0.5	1.0	2.0

**Table 5-20** Conditions of experiments for Cu<sup>+2</sup>-Cd<sup>+2</sup> co-deposition.

**Table 5-21** Conditions of experiments for Cu<sup>+2</sup>-Cd<sup>+2</sup> co-deposition.

Exp. No.	Cu <sup>+2</sup> -Cd <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (l/h)	Sa T	amplii ime (l	ng h)
253	300:200	350	100	0.5	1.0	2.0
254	300:200	350	200	0.5	1.0	2.0
255	300:200	350	300	0.5	1.0	2.0
256	300:200	410	100	0.5	1.0	2.0
257	300:200	410	200	0.5	1.0	2.0
258	300:200	410	300	0.5	1.0	2.0
259	300:200	470	100	0.5	1.0	2.0
260	300:200	470	200	0.5	1.0	2.0
261	300:200	470	300	0.5	1.0	2.0

## Part F Double Mesh (Triple Metal Co-deposition)

In this part of experiments different ratio concentration of Cu-Ni-Cd (100:200:200, 200:200, and 300:200:200) were used for different flow rates, total currents, and deposition times. Conditions of these experiments were as follows:

Exp. No.	Cu <sup>+2</sup> -Ni <sup>+2</sup> -Cd <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (l/h)	Sa T	amplii ime (l	ng h)
262	100:200:200	350	100	0.5	1.0	2.0
263	100:200:200	350	200	0.5	1.0	2.0
264	100:200:200	350	300	0.5	1.0	2.0
265	100:200:200	410	100	0.5	1.0	2.0
266	100:200:200	410	200	0.5	1.0	2.0
267	100:200:200	410	300	0.5	1.0	2.0
268	100:200:200	470	100	0.5	1.0	2.0
269	100:200:200	470	200	0.5	1.0	2.0
270	100:200:200	470	300	0.5	1.0	2.0

**Table 5-22** Conditions of experiments for Cu<sup>+2</sup>-Ni<sup>+2</sup>-Cd<sup>+2</sup> co-deposition.

**Table 5-23** Conditions of experiments for  $Cu^{+2}$ -Ni<sup>+2</sup>-Cd<sup>+2</sup> co-deposition.

Exp. No.	Cu <sup>+2</sup> -Ni <sup>+2</sup> -Cd <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (l/h)	Sa T	Sampling Time (h)	
271	200:200:200	350	100	0.5	1.0	2.0
272	200:200:200	350	200	0.5	1.0	2.0
273	200:200:200	350	300	0.5	1.0	2.0
274	200:200:200	410	100	0.5	1.0	2.0
275	200:200:200	410	200	0.5	1.0	2.0
276	200:200:200	410	300	0.5	1.0	2.0

Table 5-23 Cont.

277	200:200:200	470	100	0.5	1.0	2.0
278	200:200:200	470	200	0.5	1.0	2.0
279	200:200:200	470	300	0.5	1.0	2.0

**Table 5-24** Conditions of experiments for  $Cu^{+2}$ -Ni<sup>+2</sup>-Cd<sup>+2</sup> co-deposition.

Exp. No.	Cu <sup>+2</sup> -Ni <sup>+2</sup> -Cd <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (l/h)	Sa T	amplin ime (1	ng h)
280	300:200:200	350	100	0.5	1.0	2.0
281	300:200:200	350	200	0.5	1.0	2.0
282	300:200:200	350	300	0.5	1.0	2.0
283	300:200:200	410	100	0.5	1.0	2.0
284	300:200:200	410	200	0.5	1.0	2.0
285	300:200:200	410	300	0.5	1.0	2.0
286	300:200:200	470	100	0.5	1.0	2.0
287	300:200:200	470	200	0.5	1.0	2.0
288	300:200:200	470	300	0.5	1.0	2.0

## Part G Triple Mesh

In this part of experiments one total current for single metal deposition of  $Cu^{+2}$ ,  $Ni^{+2}$  and  $Cd^{+2}$  ions, one total current for double metal co-deposition  $Cu^{+2}$ - $Ni^{+2}$  and  $Cu^{+2}$ - $Cd^{+2}$  ions with one concentration ratio 200:200, and one total current for triple metal co-deposition  $Cu^{+2}$ - $Ni^{+2}$ - $Cd^{+2}$  ions with one concentration ratio 200:200:200 were used for different flow rates and deposition times. Conditions of these experiments were as follows:

Exp. No.	Cu <sup>+2</sup> ion Conc. (ppm)	Total Current (mA)	Flow rate (l/h)	Sampling Time (h)		ng h)
289	200	90	100	0.5	1.0	2.0
290	200	90	200	0.5	1.0	2.0
291	200	90	300	0.5	1.0	2.0

**Table 5-25** Conditions of experiments for Cu<sup>+2</sup> deposition.

 Table 5-26 Conditions of experiments for Ni<sup>+2</sup> deposition

Exp. No.	Ni <sup>+2</sup> ion Conc. (ppm)	Total Current (mA)	Flow rate (l/h)	Sampling Time (h)		
292	200	435	100	0.5	1.0	2.0
293	200	435	200	0.5	1.0	2.0
294	200	435	300	0.5	1.0	2.0

 Table 5-27 Conditions of experiments for Cd<sup>+2</sup> deposition

Exp. No.	Cd <sup>+2</sup> ion Conc. (ppm)	Total Current (mA)	Flow rate (l/h)	Sampling Time (h)		ng h)
295	200	525	100	0.5	1.0	2.0
296	200	525	200	0.5	1.0	2.0
297	200	525	300	0.5	1.0	2.0

**Table 5-28** Conditions of experiments for Cu<sup>+2</sup>-Ni<sup>+2</sup> co-deposition.

Exp. No.	Cu <sup>+2</sup> -Ni <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (l/h)	Sampling Time (h)		ng h)
298	200:200	435	100	0.5	1.0	2.0
299	200:200	435	200	0.5	1.0	2.0
300	200:200	435	300	0.5	1.0	2.0

Exp. No.	Cu <sup>+2</sup> -Cd <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (l/h)	Sampling Time (h)		
301	200:200	525	100	0.5	1.0	2.0
302	200:200	525	200	0.5	1.0	2.0
303	200:200	525	300	0.5	1.0	2.0

**Table 5-29** Conditions of experiments for Cu<sup>+2</sup>-Cd<sup>+2</sup> co-deposition

**Table 5-30** Conditions of experiments for  $Cu^{+2}$ -Ni<sup>+2</sup>-Cd<sup>+2</sup> co-deposition.

Exp. No.	Cu <sup>+2</sup> -Ni <sup>+2</sup> -Cd <sup>+2</sup> ion Conc. ratio (ppm)	Total Current (mA)	Flow rate (l/h)	Sampling Time (h)		
304	200:200:200	525	100	0.5	1.0	2.0
305	200:200:200	525	200	0.5	1.0	2.0
306	200:200:200	525	300	0.5	1.0	2.0

# **5.4 Electrochemical Reactions**

The electrochemical reactions which occurred in the cell are as follows:

At the cathode

$$Ni^{+2} + 2e^{-} \rightarrow Ni \qquad \dots (5.2)$$

$$Cd^{+2} + 2e^{-} \rightarrow Cd \qquad \dots (5.3)$$

$$2H^{+2} + 2e^- \longrightarrow H_2(g) \uparrow \qquad \dots (5.4)$$

At the anode

$$2H_2 0 \rightarrow O_2(g) \uparrow + 4H^+ + 4e^- \qquad \dots (5.5)$$

# **5.5 Experimental Results**

# A. Single Mesh Electrode with Single Metal Deposition:

The cathodic deposition of  $Cu^{+2}$ ,  $Ni^{+2}$ , and  $Cd^{+2}$  ion metal under different conditions (Exps. No. 1 to 45) are presented in Figs. (5-1 to 5-11). These experiments were carried out to investigate the effect of changing the experimental parameters on removal of metals.

From Figures below it can be noticed that the removal of  $Cu^{+2}$ ,  $Ni^{+2}$ , and  $Cd^{+2}$  ion metal increasing with increasing total applied current, flow rate, and deposition time.



deposition time at I=50 mA



deposition time at I=175 mA



deposition time at I=205 mA



**Figure 5-14** Cadmium concentration vs. deposition time at I=220 mA



**Figure 5-15** Cadmium concentration vs. deposition time at I=235 mA

## **B. Single Mesh Electrode with Double Metal Co-Deposition:**

The cathodic co-deposition of  $Cu^{+2}$ -Ni<sup>+2</sup>, and  $Cu^{+2}$ -Cd<sup>+2</sup> ion metal under different conditions (Exps. No. 46 to 135) are presented in Figs. (5-16 to 5-45). These experiments were carried out to investigate the effect of changing the experimental parameters on removal of metals.

From figures below it can be noticed that the removal of  $Ni^{+2}$  and  $Cd^{+2}$  metal ion increasing when amount of  $Cu^{+2}$  ion was added in different concentration due to UPD phenomena and also when the concentration of  $Cu^{+2}$  ion increased. In the addition it was found that the more positive metal ( $Cu^{+2}$  ion) effect on more negative metal ( $Ni^{+2}$  and  $Cd^{+2}$  ion) not vice versa.



deposition time at I=175 mA


deposition time at I=175 mA



deposition time at I=175 mA



deposition time at I=205 mA



**Figure 5-34** Cadmium concentration vs. deposition time at I=220 mA



**Figure 5-35** Cadmium concentration vs. deposition time at I=235 mA



deposition time at I=205 mA



**Figure 5-39** Cadmium concentration vs. deposition time at I=220 mA



Figure 5-40 Cadmium concentration vs. deposition time at I=235 mA



deposition time at I=205 mA



Figure 5-44 Cadmium concentration vs. deposition time at I=220 mA



Figure 5-45 Cadmium concentration vs. deposition time at I=235 mA

# **C. Single Mesh Electrode with Triple Metal Co-Deposition:**

The cathodic co-deposition of  $Cu^{+2}$ -Ni<sup>+2</sup>-Cd<sup>+2</sup> ion metal under different conditions (Exps. No. 136 to 180) are presented in Figs. (5-46 to 5-60). These experiments were carried out to investigate the effect of changing the experimental parameters on removal of metals.

From figures below it can be noticed that the addition amount of  $Ni^{+2}$  ion led to increase in removal of  $Cd^{+2}$  ion due to UPD phenomena also. Again it was found that the more positive metal effect on more negative metal not vice versa as mentioned above.



concentration vs. deposition time at I=175



**Figure 5-47** Nickel, cadmium concentration vs. deposition time at I=190



concentration vs. deposition time at I=205 mA



concentration vs. deposition time at I=220



Figure 5-50 Nickel, cadmium concentration vs. deposition time at I=235 mA



concentration vs. deposition time at I=175



**Figure 5-52** Nickel, cadmium concentration vs. deposition time at I=190



concentration vs. deposition time at I=205 mA



concentration vs. deposition time at I=220



Figure 5-55 Nickel, cadmium concentration vs. deposition time at I=235 mA



concentration vs. deposition time at I=175



**Figure 5-57** Nickel, cadmium concentration vs. deposition time at I=190



concentration vs. deposition time at I=205 mA



concentration vs. deposition time at I=220



Figure 5-60 Nickel, cadmium concentration vs. deposition time at I=235 mA

## **D. Double Mesh Electrode with Single Metal Deposition:**

The cathodic deposition of  $Cu^{+2}$ ,  $Ni^{+2}$ , and  $Cd^{+2}$  ion metal under different conditions (Exps. No. 181 to 207) are presented in Figs. (5-61 to 5-69). These experiments were carried out to investigate the effect of changing the experimental parameters on removal of metals.

From figures below it can be noticed that the removal of  $Cu^{+2}$ ,  $Ni^{+2}$ , and  $Cd^{+2}$  ion metal increases with increasing no. of meshes to two meshes.





deposition time at I=410 mA

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## **E. Double Mesh Electrode with Double Metal Co-Deposition:**

The cathodic co-deposition of  $Cu^{+2}$ -Ni<sup>+2</sup>, and  $Cu^{+2}$ -Cd<sup>+2</sup> ion metal under different conditions (Exps. No. 208 to 261) are presented in Figs. (5-70 to 5-87). These experiments were carried out to investigate the effect of changing the experimental parameters on removal of metals.







deposition time at I=410 mA



Figure 5-73 Nickel concentration vs. deposition time at I=290 mA



Figure 5-74 Nickel concentration vs. deposition time at I=350 mA



Figure 5-75 Nickel concentration vs. deposition time at I=410 mA



Figure 5-76 Nickel concentration vs. deposition time at I=290 mA



Figure 5-77 Nickel concentration vs. deposition time at I=350 mA



Figure 5-78 Nickel concentration vs. deposition time at I=410 mA



Figure 5-79 Cadmium concentration vs. deposition time at I=350 mA



Figure 5-80 Cadmium concentration vs. deposition time at I=410 mA



Figure 5-81 Cadmium concentration vs. deposition time at I=470 mA



Figure 5-82 Cadmium concentration vs. deposition time at I=350 mA



Figure 5-83 Cadmium concentration vs. deposition time at I=410 mA



Figure 5-84 Cadmium concentration vs. deposition time at I=470 mA



Figure 5-85 Cadmium concentration vs. deposition time at I=350 mA



Figure 5-86 Cadmium concentration vs. deposition time at I=410 mA



Figure 5-87 Cadmium concentration vs. deposition time at I=470 mA

## **F. Double Mesh Electrode with Triple Metal Co-Deposition:**

The cathodic co-deposition of  $Cu^{+2}$ -Ni<sup>+2</sup>-Cd<sup>+2</sup> ion metal under different conditions (Exps. No. 262 to 288) are presented in Figs. (5-88 to 5-96). These experiments were carried out to investigate the effect of changing the experimental parameters on removal of metals.





concentration vs. deposition time at I=470 mA





Figure 5-91 Nickel, cadmium concentration vs. deposition time at I=350 mA



Figure 5-92 Nickel, cadmium concentration vs. deposition time at I=410 mA



Figure 5-93 Nickel, cadmium concentration vs. deposition time at I=470 mA



Figure 5-94 Nickel, cadmium concentration vs. deposition time at I=350 mA



Figure 5-95 Nickel, cadmium concentration vs. deposition time at I=410 mA



Figure 5-96 Nickel, cadmium concentration vs. deposition time at I=470 mA

#### **<u>G. Triple Mesh Electrode:</u>**

The cathodic deposition of  $Cu^{+2}$ ,  $Ni^{+2}$ ,  $Cd^{+2}$  ions, double co-deposition of  $Cu^{+2}$ - $Ni^{+2}$  and  $Cu^{+2}$ - $Cd^{+2}$  ions, and triple co-deposition of  $Cu^{+2}$ - $Ni^{+2}$ - $Cd^{+2}$  ions under different conditions (Exps. No. 289 to 297) are presented in Figs. (5-97 to 5-99). These experiments were carried out to investigate the effect of changing the experimental parameters on removal of metals.

From figures below it can be noticed that a third meshes was used, led to almost the same removal of metals ion as in the double meshes cause of bad current field penetration.



deposition time at I=435 mA



Figure 5-99 Cadmium concentration vs. deposition time at I=525 mA



Figure 5-100 Nickel concentration vs. deposition time at I=435 mA



Figure 5-101 Cadmium concentration vs. deposition time at I=525 mA



Figure 5-102 Nickel, cadmium concentration vs. deposition time at I=525 mA

# Chapter Six Discussion

# 6.1 Introduction

In this chapter, which introduces abundant experimental results, the influence of the effective parameters on the deposition current efficiency in a concentric flow-by fixed bed reactor is explained and discussed briefly.

## **6.2 Effect of Parameters**

## 6.2 1 Effect of Applied Total Current

Figures (6-1 and 6-2) represent the relation between the current efficiency of cathodic deposition of  $Cu^{+2}$  ions on a brass electrode for single and double meshes, different applied total currents, different flow rates, and all at the same deposition time (2.0 h).

From figures below it can be seen that for the applied total current of (30, 40, and 50 mA at different flow rate) to single mesh and (60 and 100 mA at different flow rates) to double meshes, current efficiency increases with increasing applied total current because the deposition potential of  $Cu^{+2}$  ions is more positive than the potential of hydrogen (hydrogen evolution), thus the current efficiency goes up.

When the overpotential of  $Cu^{+2}$  ions limit is exceeded below the potential of hydrogen at applied total current (145 and 190 mA) to single mesh and (380 mA) to double meshes, hydrogen gas begins to evolve whose overpotential, according the conditions of this work, is typically equal to -260.4 mV vs. (SCE) and that is enough such that hydrogen evolution is very evident as the strongly reducing current efficiency, i.e., it competes with deposition of  $Cu^{+2}$  ions leading to the decrease in current efficiency. The same demonstration can be applied to Figs. (6-3 to 6-6) which represent the current efficiency of Ni<sup>+2</sup> and Cd<sup>+2</sup> ions deposition under different conditions.

Also it is noticed that for the applied total current (50 mA at flow rate =200 and 300 l/h) for single mesh and (100 mA at the different flow rates) for the double meshes the current efficiency is greater than 100% due to the fact that  $Cu^{+2}$  ions are adsorpted by graphite or some other non electrochemical form of metal removal, so there is a combination of adsorption phenomena [98] with electroreduction process especially at high pH. The above observation is demonstrated by different researchers who have proved that combination phenomena [99, 100].



**Figure 6-1** Current eff. vs. total app. current for  $Cu^{+2}$  ion deposition at t=2.0 h



**Figure 6-2** Current eff. vs. total app. current for  $Cu^{+2}$  ion deposition at t=2.0 h



**Figure 6-3** Current eff. vs. total app. current for  $Ni^{+2}$  ion deposition at t=2.0 h



**Figure 6-4** Current eff. vs. total app. current for  $Ni^{+2}$  ion deposition at t=2.0 h



Figure 6-5 Current eff. vs. total app. current for  $Cd^{+2}$  ion deposition at t=2.0 h



**Figure 6-6** Current eff. vs. total app. current for  $Cd^{+2}$  ion deposition at t=2.0 h

## 6.2.2 Effect of Co-deposition of Metal Ions

As metal contaminated wastewaters mostly include a variety of different metals, the influence of other metals was studied on the removal of  $Ni^{+2}$  and  $Cd^{+2}$  metal ions.

Figures (6-7 to 6-10) represent the relation between the current efficiency of cathodic co-deposition of  $Cu^{+2}$ -Ni<sup>+2</sup> and  $Cu^{+2}$ -Cd<sup>+2</sup> ions on a brass electrode for single and double meshes, at different applied total currents, all at the same flow rate=100 l/h and deposition time =2.0 h.

From figures below it can be noticed that the current efficiency of  $Ni^{+2}$  and  $Cd^{+2}$  ions deposition increase in comparison with no  $Cu^{+2}$  ion co-deposited due to the fact of UPD phenomena, as mentioned in chapter three, for deposition of  $Ni^{+2}$  and  $Cd^{+2}$  ions.

It is common practice in the electroplating industry to precoat a workpiece with a copper before plating the desired metal [101]. Many metal deposition reactions occur at an enhanced rate on a copper surface. This phenomenon has been exploited in this work. Copper should provide a catalytic effect for  $Ni^{+2}$  and  $Cd^{+2}$  ions deposition.

The presence of copper adsorbed on a platinum surface inhibits hydrogen adsorption which is desirable for inhibiting hydrogen evolution was showed by [49], and this also has been exploited in this work.



Figure 6-7 Current efficiency vs. total applied current for Ni<sup>+2</sup> ion co-deposition at t=2.0 h



Figure 6-8 Current efficiency vs. total applied current for Ni<sup>+2</sup> ion co-deposition at t=2.0 h



Figure 6-9 Current efficiency vs. total applied current for Cd<sup>+2</sup> ion co-deposition at t=2.0 h



Figure 6-10 Current efficiency vs. total applied current for  $Cd^{+2}$  ion co-deposition at t=2.0 h

From figs (6-11 and 6-12) it can be noticed that the current efficiency of  $Cd^{+2}$  ion is increased by addition of Ni<sup>+2</sup> ion for triple co-deposition, also due to the fact of UPD phenomenon.



Figure 6-11 Current efficiency vs. total applied current for  $Cu^{+2}$ - $Cd^{+2}$  ions co-deposition at t=2.0 h



Figure 6-12 Current efficiency vs. total applied current for  $Cu^{+2}$ - $Cd^{+2}$  ions co-deposition at t=2.0 h

#### 6.2.3 Effect of Flow Rate and Time

Figures (6-13 to 6-18) represent the current efficiency of cathodic deposition of  $Cu^{+2}$ ,  $Ni^{+2}$ , and  $Cd^{+2}$  ions on a brass electrode for single and double meshes, different flow rates, different deposition times, and at constant applied total currents.

From the figures below it can be noticed that the current efficiency increases with increasing flow rate because the reduction of  $Cu^{+2}$  to Cu,  $Ni^{+2}$  to Ni, and  $Cd^{+2}$  to Cd systems are mass transfer controlled.

This behavior is consistent with the fact that the velocity increases the reaction rate of deposition of metal ions if the process is under concentration polarization. This behavior is anticipated as the increase in velocity past the electrode surface tends to decrease the thickness of diffusion of boundary layer  $\delta_d$  (viscous layer adjacent to metal surface), and thus increases the diffusion and reaction rate as in equation below

$$i_{dep.} = \frac{DzF(C_b - C_s)}{\delta_d} \qquad \dots (6.1)$$

where  $i_{dep}$  represents the current density in which the metal ions are deposited on the metal surface (reaction rate). From above equation the reaction rate is inversely proportional to the diffusion layer thickness. This means that increasing the velocity facilities the passage of charged ions from the bulk of the solution through the diffusion boundary layer to electrode surface, thus increasing the reaction rate.

Figures below also show that current efficiency increases with the increasing of the period of deposition time. This indicates that the longer time results in better or higher efficiency because further deposition is allowed to take place when time is increased.

The same effect was observed for the rest of experiments under different conditions that would apply.



**Figure 6-13** Current efficiency vs. deposition time for Cu<sup>+2</sup> ion deposition at total applied current=30 mA



**Figure 6-14** Current efficiency vs. deposition time for Cu<sup>+2</sup> ion deposition at total applied current=60 mA



**Figure 6-15** Current efficiency vs. deposition time for Ni<sup>+2</sup> ion deposition at total applied current=145 mA



**Figure 6-16** Current efficiency vs. deposition time for Ni<sup>+2</sup> ion deposition at total applied current=290 mA



**Figure 6-17** Current efficiency vs. deposition time for Cd<sup>+2</sup> ion deposition at total applied current=175 mA



**Figure 6-18** Current efficiency vs. deposition time for Cd<sup>+2</sup> ion deposition at total applied current=350 mA

#### **6.2.4 Effect of Concentration Ratio**

Figures (6-19 to 6-22) represent the current efficiency of cathodic co-deposition of  $Cu^{+2}$ -Ni<sup>+2</sup>, and  $Cu^{+2}$ -Cd<sup>+2</sup> ions on a brass electrode for single and double meshes, different Cu concentration ratios, different deposition times, at flow rate=100 l/h, and at constant applied total currents.

From figures below it can be obviously noticed that the current efficiency increases with increasing the Cu concentration ratio. This result is due to the increase in the deposition rate. Also this behavior can be demonstrated by looking into Eq. 6-1:

$$i_{dep.} = \frac{DzF(C_b - C_s)}{\delta_d} \qquad \dots (6.1)$$

which shows that any increase in bulk concentration leads to increase in reaction rate  $(i_{dep.})$  because of the proportionality between bulk concentration (Cu concentration) and reaction rate which leads to increase deposition of metal ions, and hence increase current efficiency.



**Figure 6-19** Current efficiency vs. deposition time for co-deposition Cu<sup>+2</sup>-Ni<sup>+2</sup> ions at applied total current=145 mA



**Figure 6-20** Current efficiency vs. deposition time for co-deposition Cu<sup>+2</sup>-Ni<sup>+2</sup> ions at applied total current=290 mA



**Figure 6-21** Current efficiency vs. deposition time for co-deposition Cu<sup>+2</sup>-Cd<sup>+2</sup> ions at applied total current=175 mA


**Figure 6-22** Current efficiency vs. deposition time for co-deposition Cu<sup>+2</sup>-Cd<sup>+2</sup> ions at applied total current=350 mA

#### 6.2.5 Effect of No. of Meshes (Bed Thickness)

Figures (6-23 and 6-24) represent the current efficiency of cathodic co-deposition of  $Cu^{+2}$  ion and  $Cu^{+2}$ -Ni<sup>+2</sup>ions on a brass electrode for single, double, triple meshes, different deposition times, at flow rate=100 l/h, and at constant applied total currents.

From figures below it can be noticed current efficiency increases with increasing no. of mesh but for two meshes. When the no. of meshes increases over two meshes (three meshes are used as maximum no. of mesh in this work), it was shown that there is a sharp decrease in current efficiency. That means a two meshes (near the counter electrode) is more effective than a third one.

This phenomena can be explained as in three-dimensional electrodes, such as packed, fluidized, circulating and spouted beds, the electric field will not be uniform because of IR drop along the bed (electric conductivity of solution). This non-uniformity results in an overpotential profile [102] and a current density profile [103].

The intensity of the electric field is high near the counter electrode, where high potentials and high electric currents are found, providing higher reaction rates, as experimentally shown by the present work experiments.

The main reasons for this tendency might be (1) simultaneous reduction and dissolution of metal ions at the electrode vicinity, and (2) electrode bed thickness, which sets conditions for the electric field penetration at a given thickness [45].



**Figure 6.23** Current efficiency vs. deposition time for Cu<sup>+2</sup> ion deposition at constant total applied current.



**Figure 6.24** Current efficiency vs. deposition time for Cu<sup>+2</sup>-Ni<sup>+2</sup> ions co-deposition at constant total applied current.

# Chapter Seven Conclusions and Recommendations

#### 7.1 Introduction

The present work has been mainly aimed to investigate the removal and current efficiency of metal ions under different conditions and also improve the current efficiency by co-deposition of metal ions in  $0.5 \text{ M H}_2\text{SO}_4$  as supporting electrolyte solution in a concentric flow-by electrochemical reactor.

### 7.2 Conclusions

The following points can be concluded from the present work:

- Current efficiency of Cu<sup>+2</sup> ion increases with increasing total applied current till the hydrogen gas bubbles begin to evolve, then a dramatic decrease in current efficiency was shown because of the competition of hydrogen evolution reaction. The current efficiency of Ni<sup>+2</sup> and Cd<sup>+2</sup> ions decrease with increasing total applied current because they were deposited more negatively than hydrogen potential.
- Current efficiency of Cu<sup>+2</sup> ion exceeds 100% due to the fact that Cu<sup>+2</sup> ions are adsorpted by graphite or some other non electrochemical form of metal removal.
- 3. Current efficiency of Ni<sup>+2</sup> and Cd<sup>+2</sup> ions increases to some extent when codeposited with different Cu concentration ratios due to the application of UPD phenomenon for deposition of metal ions. And also a slight effect of Ni<sup>+2</sup> ion deposition is seen on increasing the current efficiency of Cd<sup>+2</sup> ion deposition in triple co-deposition.
- 4. Current efficiency of Ni<sup>+2</sup> and Cd<sup>+2</sup> ions increases with increasing the initial Cu concentration due to the highly conductive electrolyte.

- 5. Current efficiency of  $Cu^{+2}$ ,  $Ni^{+2}$ , and  $Cd^{+2}$  ions increases with increasing flow rate because the systems of  $Cu^{+2}$  to Cu metal,  $Ni^{+2}$  to Ni metal , and  $Cd^{+2}$  to Cd metal are mass transfer controlled.
- 6. Current efficiency of Cu<sup>+2</sup>, Ni<sup>+2</sup>, and Cd<sup>+2</sup> ions increases with increasing deposition time. This indicates that the longer time results in better or higher efficiency because further deposition is allowed to take place when time is increased.
- 7. The elctro-active bed thickness was found to be two no. of meshes of 0.4 cm thick in the direction of current flow, although the IR drop is decreased might be because of the dissolution of metal ions is greater than deposition of metal ions.

#### 7.3 Recommendations

It is recommended that the future work should study:

- 1. The present deposition and co-deposition by using flow-through mesh electrode in order to compare the result with flow-by mesh electrode results.
- 2. The effect of using different mesh no. especially small mesh no. in order to investigate the pressure drop when the pore is blocking and also different types of material should be used.
- 3. The effect of using different operational conditions such as low flow rate of electrolyte solution, and long deposition time.
- 4. The effect of different electrolyte temperature on current efficiency of metal ions.
- 5. Developing a large scale fixed bed (mesh electrode) electrochemical reactor, if possible.

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### **Appendix A**

#### **Experimental Data and Current Efficiency**

#### for Non-Flow System

**Table A-1** Experimental data and current efficiency (Single mesh, I=15 mA, 0.5 M H<sub>2</sub>SO<sub>4</sub>,  $Cu^{+2}$  conc.=200 ppm, Temp.=40  $^{0}C$ )

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0049	0.0089	55.06
1.0	0.0108	0.0178	60.67
1.5	0.0177	0.0267	66.39

**Table A-2** Experimental data and current efficiency (Single mesh, I=20 mA, 0.5 M H<sub>2</sub>SO<sub>4</sub>, Cu<sup>+2</sup> conc.=200 ppm, Temp.=40 <sup>0</sup>C)

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0075	0.0119	63.03
1.0	0.0164	0.0237	69.20
1.5	0.0267	0.0356	75.00

**Table A-3** Experimental data and current efficiency (Single mesh, I=25 mA, 0.5 M H<sub>2</sub>SO<sub>4</sub>,  $Cu^{+2}$  conc.=200 ppm, Temp.=40  $^{0}C$ )

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0106	0.0148	71.62
1.0	0.0232	0.0297	78.12
1.5	0.0372	0.0445	83.60

**Table A-4** Experimental data and current efficiency (Single mesh, I=70 mA, 0.5 M H<sub>2</sub>SO<sub>4</sub>,  $Cu^{+2}$  conc.=200 ppm, Temp.=40  $^{0}C$ )

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0176	0.0415	42.41
1.0	0.0404	0.0830	48.68
1.5	0.0679	0.1245	54.54

**Table A-5** Experimental data and current efficiency (Single mesh, I=90 mA, 0.5 M  $H_2SO_4$ ,<br/>Cu<sup>+2</sup> conc.=200 ppm, Temp.=40  $^{0}C$ )

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0182	0.0534	34.08
1.0	0.0426	0.1067	39.93
1.5	0.0738	0.1601	46.10

Deposition Time (h)	∆Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0106	0.0178	59.55
1.0	0.0236	0.0356	66.29
1.5	0.0392	0.0534	73.41

**Table A-6** Experimental data and current efficiency (Double meshes, I=30 mA, 0.5 M H<sub>2</sub>SO<sub>4</sub>, Cu<sup>+2</sup> conc.=200 ppm, Temp.=40 <sup>0</sup>C)

**Table A-7** Experimental data and current efficiency (Double meshes, I=40 mA, 0.5 M H<sub>2</sub>SO<sub>4</sub>, Cu<sup>+2</sup> conc.=200 ppm, Temp.=40 <sup>0</sup>C)

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0161	0.0237	67.93
1.0	0.0356	0.0474	75.11
1.5	0.0587	0.0712	82.44

**Table A-8** Experimental data and current efficiency (Double meshes, I=50 mA, 0.5 M H<sub>2</sub>SO<sub>4</sub>, Cu<sup>+2</sup> conc.=200 ppm, Temp.=40 <sup>0</sup>C)

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0228	0.0297	76.77
1.0	0.0499	0.0593	84.15
1.5	0.0815	0.0889	91.68

**Table A-9** Experimental data and current efficiency (Double meshes, I=140 mA, 0.5 M<br/>H<sub>2</sub>SO<sub>4</sub>, Cu<sup>+2</sup> conc.=200 ppm, Temp.=40 °C)

Deposition Time (h)	$\Delta \mathbf{Wt.} \mathbf{Exp.} (\mathbf{g})$	Wt. Theo. (g)	Cur. Eff.
0.5	0.0393	0.0830	47.35
1.0	0.0893	0.1660	53.80
1.5	0.1494	0.2490	60.00

**Table A-10** Experimental data and current efficiency (Double meshes, I=180 mA, 0.5 MH2SO4, Cu<sup>+2</sup> conc.=200 ppm, Temp.=40 °C)

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0421	0.1067	39.46
1.0	0.0986	0.2135	46.18
1.5	0.1688	0.3202	52.72

Deposition Time (h)	∆Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0168	0.0267	62.92
1.0	0.0371	0.0534	69.48
1.5	0.0611	0.0801	76.28

**Table A-11** Experimental data and current efficiency (Triple meshes, I=45 mA, 0.5 M H<sub>2</sub>SO<sub>4</sub>, Cu<sup>+2</sup> conc.=200 ppm, Temp.=40 <sup>0</sup>C)

**Table A-12** Experimental data and current efficiency (Triple meshes, I=60 mA, 0.5 M H<sub>2</sub>SO<sub>4</sub>, Cu<sup>+2</sup> conc.=200 ppm, Temp.=40 <sup>0</sup>C)

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0255	0.0356	71.63
1.0	0.0553	0.0712	77.67
1.5	0.0905	0.1067	84.82

**Table A-13** Experimental data and current efficiency (Triple meshes, I=75 mA, 0.5 M H<sub>2</sub>SO<sub>4</sub>, Cu<sup>+2</sup> conc.=200 ppm, Temp.=40 <sup>0</sup>C)

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0353	0.0445	79.33
1.0	0.0769	0.0889	86.50
1.5	0.1251	0.1334	93.78

**Table A-14** Experimental data and current efficiency (Triple meshes, I=210 mA, 0.5 M<br/>H<sub>2</sub>SO<sub>4</sub>, Cu<sup>+2</sup> conc.=200 ppm, Temp.=40 <sup>0</sup>C)

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0619	0.1245	49.72
1.0	0.1397	0.2490	56.10
1.5	0.2328	0.3736	62.31

**Table A-15** Experimental data and current efficiency (Triple meshes, I=270 mA, 0.5 M H<sub>2</sub>SO<sub>4</sub>, Cu<sup>+2</sup> conc.=200 ppm, Temp.=40 <sup>0</sup>C)

Deposition Time (h)	$\Delta$ Wt. Exp. (g)	Wt. Theo. (g)	Cur. Eff.
0.5	0.0669	0.1601	41.79
1.0	0.1563	0.3202	48.81
1.5	0.2651	0.4803	55.20

## **Appendix B**

### **Experimental Data and Current Efficiency**

### for Flow System

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	198.22	0.01068	0.0178	60.04
1.0	196.10	0.0234	0.0356	65.77
1.5	193.67	0.03798	0.0534	71.17
2.0	190.64	0.05616	0.0712	78.93

Table B-1 Experimental data and current efficiency for Ex. No. 1

Table B-2 Experimental data and current efficiency for Ex. No. 2

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	198.15	0.0111	0.0178	62.40
1.0	196.02	0.0239	0.0356	67.12
1.5	193.44	0.0394	0.0534	73.75
2.0	190.39	0.0577	0.0712	81.03

Table B-3 Experimental data and current efficiency for Ex. No. 3

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	198.09	0.0115	0.0178	64.42
1.0	195.89	0.0247	0.0356	69.31
1.5	193.21	0.0407	0.0534	76.34
2.0	190.13	0.0592	0.0712	83.23

Table B-4 Experimental data and current efficiency for Ex. No. 4

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	197.16	0.0170	0.0237	71.84
1.0	193.71	0.0377	0.0474	79.56
1.5	189.78	0.0613	0.0712	86.18
2.0	185.22	0.0887	0.0949	93.47

Table B-5 Experimental data and current efficiency for Ex. No. 5

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	197.07	0.0176	0.0237	74.12
1.0	193.52	0.0389	0.0474	81.96
1.5	189.56	0.0626	0.0712	88.03
2.0	184.96	0.0902	0.0949	95.12

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	196.98	0.0181	0.0237	76.40
1.0	193.36	0.0398	0.0474	83.99
1.5	189.31	0.0641	0.0712	90.14
2.0	184.66	0.0920	0.0949	97.01

**Table B-6** Experimental data and current efficiency for Ex. No. 6

**Table B-7** Experimental data and current efficiency for Ex. No. 7

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	196.11	0.0233	0.0296	78.72
1.0	191.60	0.0504	0.0593	85.00
1.5	186.53	0.0808	0.0889	90.87
2.0	180.59	0.1165	0.1186	98.20

Table B-8 Experimental data and current efficiency for Ex. No. 8

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	196.02	0.0239	0.0296	80.54
1.0	191.41	0.0515	0.0593	86.92
1.5	186.24	0.0826	0.0889	92.82
2.0	180.11	0.1193	0.1186	100.63

Table B-9 Experimental data and current efficiency for Ex. No. 9

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.91	0.0245	0.0296	82.77
1.0	191.22	0.0527	0.0593	88.84
1.5	185.92	0.0845	0.0889	94.98
2.0	179.62	0.1223	0.1186	103.11

Table B-10 Experimental data and current efficiency for Ex. No. 10

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.79	0.0433	0.0860	50.31
1.0	183.78	0.0973	0.1720	56.60
1.5	172.66	0.1640	0.2579	63.60
2.0	159.82	0.2411	0.3439	70.10

 Table B-11 Experimental data and current efficiency for Ex. No. 11

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	192.53	0.0448	0.0860	52.13
1.0	183.15	0.1011	0.1720	58.79
1.5	171.67	0.1700	0.2579	65.90
2.0	158.79	0.2473	0.3439	71.90

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	192.23	0.0466	0.0860	54.22
1.0	182.52	0.1049	0.1720	60.99
1.5	170.89	0.1747	0.2579	67.71
2.0	157.59	0.2545	0.3439	73.99

 Table B-12 Experimental data and current efficiency for Ex. No. 12

 Table B-13 Experimental data and current efficiency for Ex. No. 13

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.25	0.0465	0.1127	41.27
1.0	182.16	0.1070	0.2253	47.50
1.5	169.41	0.1835	0.3380	54.30
2.0	154.18	0.2749	0.4507	61.01

 Table B-14 Experimental data and current efficiency for Ex. No. 14

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.91	0.0485	0.1127	43.08
1.0	181.30	0.1122	0.2253	49.79
1.5	168.34	0.1900	0.3380	56.20
2.0	152.30	0.2862	0.4507	63.51

**Table B-15** Experimental data and current efficiency for Ex. No. 15

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.48	0.0511	0.1127	45.37
1.0	180.62	0.1163	0.2253	51.61
1.5	167.10	0.1974	0.3380	58.40
2.0	150.72	0.2957	0.4507	65.61

Table B-16 Experimental data and current efficiency for Ex. No. 16

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.96	0.0242	0.0794	30.54
1.0	190.34	0.0580	0.1588	36.51
1.5	183.05	0.1017	0.2381	42.71
2.0	174.49	0.1531	0.3175	48.21

 Table B-17 Experimental data and current efficiency for Ex. No. 17

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.77	0.0254	0.0794	31.97
1.0	189.79	0.0613	0.1588	38.59
1.5	182.30	0.1062	0.2381	44.60
2.0	173.70	0.1578	0.3175	49.70

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	195.5	0.0270	0.0794	34.01
1.0	189.31	0.0641	0.1588	40.40
1.5	181.70	0.1098	0.2381	46.11
2.0	172.85	0.1629	0.3175	51.30

**Table B-18** Experimental data and current efficiency for Ex. No. 18

 Table B-19 Experimental data and current efficiency for Ex. No. 19

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.90	0.0246	0.0876	28.09
1.0	190.31	0.0581	0.1752	33.19
1.5	182.74	0.1036	0.2628	39.41
2.0	173.49	0.1591	0.3504	45.40

Table B-20 Experimental data and current efficiency for Ex. No. 20

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.72	0.0257	0.0876	29.32
1.0	189.75	0.0615	0.1752	35.11
1.5	181.91	0.1085	0.2628	41.31
2.0	172.44	0.1654	0.3504	47.20

**Table B-21** Experimental data and current efficiency for Ex. No. 21

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.47	0.0272	0.0876	31.03
1.0	189.17	0.0650	0.1752	37.09
1.5	181.08	0.1135	0.2628	43.20
2.0	171.45	0.1713	0.3504	48.89

Table B-22 Experimental data and current efficiency for Ex. No. 22

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.93	0.0244	0.0958	25.49
1.0	190.07	0.0596	0.1916	31.10
1.5	182.23	0.1066	0.2874	37.10
2.0	172.47	0.1652	0.3832	43.10

 Table B-23 Experimental data and current efficiency for Ex. No. 23

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.58	0.0265	0.0958	27.68
1.0	189.46	0.0632	0.1916	33.01
1.5	181.32	0.1121	0.2874	39.00
2.0	171.45	0.1713	0.3832	44.70

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	195.39	0.0277	0.0958	28.87
1.0	188.92	0.0665	0.1916	34.70
1.5	180.50	0.1170	0.2874	40.71
2.0	170.37	0.1778	0.3832	46.39

 Table B-24 Experimental data and current efficiency for Ex. No. 24

 Table B-25 Experimental data and current efficiency for Ex. No. 25

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.89	0.0247	0.1040	23.71
1.0	189.77	0.0614	0.2080	29.51
1.5	181.64	0.1102	0.3120	35.30
2.0	171.64	0.1702	0.4161	40.90

Table B-26 Experimental data and current efficiency for Ex. No. 26

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.56	0.0266	0.1040	25.61
1.0	189.15	0.0651	0.2080	31.29
1.5	180.76	0.1154	0.3120	37.00
2.0	170.46	0.1772	0.4161	42.60

**Table B-27** Experimental data and current efficiency for Ex. No. 27

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.27	0.0284	0.1040	27.28
1.0	188.56	0.0686	0.2080	33.00
1.5	179.87	0.1208	0.3120	38.71
2.0	169.21	0.1847	0.4161	44.40

Table B-28 Experimental data and current efficiency for Ex. No. 28

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.88	0.0247	0.1122	22.03
1.0	189.56	0.0626	0.2245	27.91
1.5	181.03	0.1138	0.3367	33.81
2.0	170.60	0.1764	0.4489	39.30

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Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.53	0.0268	0.1122	23.90
1.0	188.90	0.0666	0.2245	29.67
1.5	180.02	0.1199	0.3367	35.61
2.0	169.33	0.1840	0.4489	40.99

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	195.20	0.0288	0.1122	25.66
1.0	188.25	0.0705	0.2245	31.41
1.5	179.10	0.1254	0.3367	37.25
2.0	167.98	0.1921	0.4489	42.80

**Table B-30** Experimental data and current efficiency for Ex. No. 30

 Table B-31 Experimental data and current efficiency for Ex. No. 31

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	193.73	0.0376	0.1835	20.50
1.0	184.46	0.0932	0.3670	25.41
1.5	172.84	0.1630	0.5505	29.60
2.0	158.16	0.2510	0.7340	34.20

**Table B-32** Experimental data and current efficiency for Ex. No. 32

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	193.24	0.0406	0.1835	22.10
1.0	183.61	0.0983	0.3670	26.80
1.5	171.56	0.1706	0.5505	31.00
2.0	156.57	0.2606	0.7340	35.50

**Table B-33** Experimental data and current efficiency for Ex. No. 33

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.70	0.0438	0.1835	23.87
1.0	182.75	0.1035	0.3670	28.20
1.5	170.10	0.1794	0.5505	32.59
2.0	154.86	0.2708	0.7340	36.90

Table B-34 Experimental data and current efficiency for Ex. No. 34

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	193.69	0.0379	0.1992	19.00
1.0	184.13	0.0952	0.3984	23.90
1.5	171.61	0.1703	0.5977	28.50
2.0	155.41	0.2675	0.7969	33.57

Table B-35 Experimental	data and	current efficienc	y for E	Ex. No.	35

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	193.16	0.0410	0.1992	20.60
1.0	183.20	0.1008	0.3984	25.30
1.5	169.92	0.1805	0.5977	30.20
2.0	153.40	0.2796	0.7969	35.09

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.60	0.0444	0.1992	22.29
1.0	182.20	0.1068	0.3984	26.80
1.5	168.03	0.1918	0.5977	32.10
2.0	151.92	0.2885	0.7969	36.20

**Table B-36** Experimental data and current efficiency for Ex. No. 36

 Table B-37 Experimental data and current efficiency for Ex. No. 37

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	193.59	0.0385	0.2149	17.89
1.0	183.59	0.0985	0.4299	22.90
1.5	170.66	0.1760	0.6448	27.30
2.0	153.28	0.2803	0.8598	32.60

 Table B-38 Experimental data and current efficiency for Ex. No. 38

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.98	0.0421	0.2149	19.60
1.0	182.59	0.1045	0.4299	24.30
1.5	168.94	0.1864	0.6448	28.90
2.0	151.28	0.2923	0.8598	34.00

**Table B-39** Experimental data and current efficiency for Ex. No. 39

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.37	0.0458	0.2149	21.30
1.0	181.59	0.1105	0.4299	25.69
1.5	166.90	0.1986	0.6448	30.80
2.0	149.56	0.3026	0.8598	35.20

Table B-40 Experimental data and current efficiency for Ex. No. 40

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	193.54	0.0388	0.2307	16.80
1.0	183.16	0.1010	0.4614	21.90
1.5	169.67	0.1820	0.6920	26.30
2.0	151.71	0.2897	0.9227	31.40

Table B-41 Experimental	l data and	current efficiency	for Ex. No. 4	1

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.89	0.0427	0.2307	18.49
1.0	182.01	0.1079	0.4614	23.40
1.5	167.71	0.1937	0.6920	28.00
2.0	149.56	0.3026	0.9227	32.80

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.20	0.0468	0.2307	20.29
1.0	180.93	0.1144	0.4614	24.80
1.5	165.74	0.2056	0.6920	29.70
2.0	147.56	0.3146	0.9227	34.10

**Table B-42** Experimental data and current efficiency for Ex. No. 42

 Table B-43 Experimental data and current efficiency for Ex. No. 43

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	193.51	0.0389	0.2464	15.80
1.0	182.83	0.1030	0.4928	20.90
1.5	168.58	0.1885	0.7392	25.50
2.0	150.23	0.2986	0.9856	30.30

**Table B-44** Experimental data and current efficiency for Ex. No. 44

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.77	0.0434	0.2464	17.61
1.0	181.52	0.1109	0.4928	22.50
1.5	166.61	0.2003	0.7392	27.10
2.0	147.76	0.3134	0.9856	31.80

**Table B-45** Experimental data and current efficiency for Ex. No. 45

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.03	0.0478	0.2464	19.41
1.0	180.37	0.1178	0.4928	23.90
1.5	164.64	0.2122	0.7392	28.70
2.0	145.46	0.3272	0.9856	33.20

Table B-46 Experimental data and current efficiency for Ex. No. 46

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	93.48	0.0391	0.0860	45.50
0.5	195.86	0.0248	0.0794	31.29
1.0	85.24	0.0886	0.1720	51.50
1.0	189.97	0.0602	0.1588	37.91
1.5	75.24	0.1486	0.2579	57.60
	182.18	0.1069	0.2381	44.90
2.0	63.49	0.2191	0.3439	63.70
2.0	173.01	0.1619	0.3175	51.00

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	93.24	0.0406	0.0860	47.17
0.5	195.66	0.0260	0.0794	32.80
1.0	84.72	0.0917	0.1720	53.32
1.0	189.55	0.0627	0.1588	39.49
1.5	74.46	0.1532	0.2579	59.41
	181.47	0.1112	0.2381	46.69
2.0	62.46	0.2252	0.3439	65.49
2.0	172.06	0.1676	0.3175	52.80

Table B-47 Experimental data and current efficiency for Ex. No. 47

**Table B-48** Experimental data and current efficiency for Ex. No. 48

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	92.98	0.0421	0.0860	48.99
0.5	195.42	0.0275	0.0794	34.62
1.0	84.18	0.0949	0.1720	55.20
	189.07	0.0656	0.1588	41.31
1.5	73.69	0.1579	0.2579	61.20
	180.75	0.1155	0.2381	48.50
2.0	61.48	0.2311	0.3439	67.20
	171.05	0.1737	0.3175	54.71

**Table B-49** Experimental data and current efficiency for Ex. No. 49

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	93.33	0.0400	0.0949	42.18
0.5	195.75	0.0255	0.0876	29.11
1.0	84.76	0.0914	0.1897	48.19
1.0	189.72	0.0617	0.1752	35.21
1.5	74.38	0.1537	0.2846	54.01
	181.61	0.1103	0.2628	41.99
2.0	62.11	0.2273	0.3795	59.91
	171.62	0.1703	0.3504	48.60

 Table B-50 Experimental data and current efficiency for Ex. No. 50

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	93.04	0.0418	0.0949	44.02
0.3	195.50	0.0270	0.0876	30.83
1.0	84.19	0.0949	0.1897	49.99
1.0	189.23	0.0646	0.1752	36.89
1.5	73.48	0.1591	0.2846	55.91
	180.82	0.1151	0.2628	43.79
2.0	61.04	0.2338	0.3795	61.60
	170.63	0.1762	0.3504	50.30

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	92.90	0.0426	0.0949	44.90
0.5	195.24	0.0286	0.0876	32.61
1.0	83.59	0.0985	0.1897	51.89
1.0	188.70	0.0678	0.1752	38.70
1.5	72.63	0.1642	0.2846	57.70
	179.99	0.1201	0.2628	45.69
2.0	59.96	0.2402	0.3795	63.31
2.0	169.52	0.1829	0.3504	52.20

 Table B-51 Experimental data and current efficiency for Ex. No. 51

 Table B-52 Experimental data and current efficiency for Ex. No. 52

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	93.13	0.0412	0.1038	39.72
0.5	195.61	0.0263	0.0958	27.49
1.0	84.37	0.0938	0.2075	45.19
1.0	189.24	0.0646	0.1916	33.69
1.5	73.07	0.1616	0.3113	51.90
	180.98	0.1141	0.2874	39.71
2.0	60.08	0.2395	0.4151	57.71
	170.43	0.1774	0.3832	46.30

**Table B-53** Experimental data and current efficiency for Ex. No. 53

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	92.84	0.0430	0.1038	41.40
0.5	195.32	0.0281	0.0958	29.31
1.0	83.78	0.0973	0.2075	46.89
1.0	188.66	0.0680	0.1916	35.51
1.5	72.14	0.1672	0.3113	53.70
	180.07	0.1196	0.2874	41.61
2.0	58.91	0.2465	0.4151	59.40
	169.22	0.1847	0.3832	48.19

 Table B-54 Experimental data and current efficiency for Ex. No. 54

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	92.51	0.0449	0.1038	43.31
0.5	195.02	0.0299	0.0958	31.19
1.0	83.12	0.1013	0.2075	48.80
1.0	188.06	0.0716	0.1916	37.39
1.5	71.21	0.1727	0.3113	55.49
	179.21	0.1247	0.2874	43.40
2.0	57.59	0.2545	0.4151	61.30
	168.07	0.1916	0.3832	49.99

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	93.11	0.0413	0.1127	36.69
0.5	195.51	0.0269	0.1040	25.90
1.0	83.85	0.0969	0.2253	43.00
	188.84	0.0670	0.2080	32.19
1.5	72.34	0.1660	0.3380	49.10
	179.93	0.1204	0.3120	38.59
20	58.84	0.2470	0.4507	54.80
2.0	168.86	0.1868	0.4161	44.91

 Table B-55 Experimental data and current efficiency for Ex. No. 55

 Table B-56 Experimental data and current efficiency for Ex. No. 56

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	92.77	0.0434	0.1127	38.50
0.5	195.18	0.0289	0.1040	27.80
1.0	83.18	0.1009	0.2253	44.79
	188.25	0.0705	0.2080	33.89
1.5	71.27	0.1724	0.3380	51.00
	178.94	0.1264	0.3120	40.49
2.0	57.56	0.2546	0.4507	56.51
	167.68	0.1939	0.4161	46.61

**Table B-57** Experimental data and current efficiency for Ex. No. 57

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	92.41	0.0455	0.1127	40.42
0.5	194.87	0.0308	0.1040	29.59
1.0	82.50	0.1050	0.2253	46.60
	187.59	0.0745	0.2080	35.79
1.5	70.31	0.1781	0.3380	52.71
	178.05	0.1317	0.3120	42.21
2.0	56.13	0.2632	0.4507	58.41
	166.37	0.2018	0.4161	48.50

 Table B-58 Experimental data and current efficiency for Ex. No. 58

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	93.11	0.0413	0.1216	34.01
0.5	195.47	0.0272	0.1122	24.22
1.0	83.71	0.0977	0.2431	40.20
1.0	188.52	0.0689	0.2245	30.69
1.5	71.74	0.1696	0.3647	46.50
	179.13	0.1252	0.3367	37.19
2.0	57.13	0.2572	0.4862	52.90
	167.53	0.1948	0.4489	43.40

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	92.73	0.0436	0.1216	35.88
0.5	195.14	0.0292	0.1122	25.98
1.0	83.02	0.1019	0.2431	41.91
1.0	187.81	0.0731	0.2245	32.59
1.5	70.64	0.1762	0.3647	48.31
	178.17	0.1310	0.3367	38.90
20	55.75	0.2655	0.4862	54.60
2.0	166.26	0.2024	0.4489	45.10

 Table B-59 Experimental data and current efficiency for Ex. No. 59

 Table B-60 Experimental data and current efficiency for Ex. No. 60

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	92.34	0.0460	0.1216	37.81
0.5	194.78	0.0313	0.1122	27.91
1.0	82.21	0.1067	0.2431	43.91
	187.17	0.0770	0.2245	34.30
1.5	69.49	0.1831	0.3647	50.20
	177.11	0.1373	0.3367	40.79
2.0	54.21	0.2747	0.4862	56.50
	164.91	0.2105	0.4489	46.90

 Table B-61 Experimental data and current efficiency for Ex. No. 61

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.69	0.0439	0.0860	51.01
0.5	195.77	0.0254	0.0794	31.97
1.0	183.69	0.0979	0.1720	56.91
1.0	189.65	0.0621	0.1588	39.12
1.5	172.96	0.1622	0.2579	62.90
	181.58	0.1105	0.2381	46.41
2.0	160.39	0.2377	0.3439	69.10
	171.95	0.1683	0.3175	53.01

 Table B-62 Experimental data and current efficiency for Ex. No. 62

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	192.47	0.0452	0.0860	52.55
0.5	195.57	0.0266	0.0794	33.48
1.0	183.23	0.1006	0.1720	58.51
1.0	189.20	0.0648	0.1588	40.82
1.5	172.19	0.1669	0.2579	64.69
	180.87	0.1148	0.2381	48.20
2.0	159.42	0.2435	0.3439	70.80
	171.05	0.1737	0.3175	54.71

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	192.20	0.0468	0.0860	54.43
0.5	195.34	0.0280	0.0794	35.22
1.0	182.69	0.1039	0.1720	60.40
	188.70	0.0678	0.1588	42.71
1.5	171.13	0.1732	0.2579	67.16
	180.20	0.1188	0.2381	49.89
2.0	158.39	0.2497	0.3439	72.59
2.0	170.10	0.1794	0.3175	56.50

 Table B-63 Experimental data and current efficiency for Ex. No. 63

**Table B-64** Experimental data and current efficiency for Ex. No. 64

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.44	0.0454	0.0949	47.81
0.5	195.59	0.0265	0.0876	30.21
1.0	183.08	0.1015	0.1897	53.50
	189.26	0.0644	0.1752	36.78
1.5	171.92	0.1685	0.2846	59.19
	180.69	0.1159	0.2628	44.09
2.0	158.70	0.2478	0.3795	65.30
	170.10	0.1794	0.3504	51.20

 Table B-65 Experimental data and current efficiency for Ex. No. 65

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.19	0.0469	0.0949	49.39
0.5	195.36	0.0278	0.0876	31.78
1.0	182.54	0.1048	0.1897	55.21
1.0	188.79	0.0673	0.1752	38.39
1.5	171.02	0.1739	0.2846	61.09
	179.94	0.1204	0.2628	45.80
2.0	157.56	0.2546	0.3795	67.10
	169.05	0.1857	0.3504	53.00

 Table B-66 Experimental data and current efficiency for Ex. No. 66

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	191.90	0.0486	0.0949	51.23
0.5	195.11	0.0293	0.0876	33.50
1.0	181.97	0.1082	0.1897	57.01
1.0	188.23	0.0706	0.1752	40.31
1.5	170.16	0.1790	0.2846	62.90
	179.20	0.1248	0.2628	47.49
2.0	156.36	0.2618	0.3795	69.00
	167.94	0.1924	0.3504	54.90

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	192.23	0.0466	0.1038	44.93
0.5	195.31	0.0281	0.0958	29.37
1.0	182.43	0.1054	0.2075	50.80
1.0	188.47	0.0692	0.1916	36.11
1.5	170.43	0.1774	0.3113	56.99
	179.31	0.1241	0.2874	43.19
2.0	156.48	0.2611	0.4151	62.91
2.0	167.87	0.1928	0.3832	50.31

 Table B-67 Experimental data and current efficiency for Ex. No. 67

 Table B-68 Experimental data and current efficiency for Ex. No. 68

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.94	0.0484	0.1038	46.60
0.5	195.03	0.0298	0.0958	31.13
1.0	181.81	0.1091	0.2075	52.59
1.0	188.93	0.0664	0.1916	34.67
1.5	169.54	0.1828	0.3113	58.71
	178.44	0.1294	0.2874	45.01
2.0	155.24	0.2686	0.4151	64.70
	166.73	0.1996	0.3832	52.09

**Table B-69** Experimental data and current efficiency for Ex. No. 69

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.63	0.0502	0.1038	48.40
0.5	194.75	0.0315	0.0958	32.88
1.0	181.15	0.1131	0.2075	54.50
	187.35	0.0759	0.1916	39.61
1.5	168.56	0.1886	0.3113	60.60
	177.53	0.1348	0.2874	46.91
2.0	153.99	0.2761	0.4151	66.51
	165.58	0.2065	0.3832	53.89

 Table B-70 Experimental data and current efficiency for Ex. No. 70

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.08	0.0475	0.1127	42.18
0.5	195.20	0.0288	0.1040	27.69
1.0	181.86	0.1088	0.2253	48.30
1.0	188.00	0.0720	0.2080	34.61
1.5	169.36	0.1838	0.3380	54.39
	178.42	0.1295	0.3120	41.49
2.0	154.55	0.2727	0.4507	60.51
	166.16	0.2030	0.4161	48.80

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	191.74	0.0496	0.1127	43.99
0.5	194.87	0.0308	0.1040	29.59
1.0	181.18	0.1129	0.2253	50.11
	187.41	0.0755	0.2080	36.31
1.5	168.40	0.1896	0.3380	56.10
	177.48	0.1351	0.3120	43.30
2.0	153.13	0.2812	0.4507	62.40
	165.39	0.2077	0.4161	49.91

Table B-71 Experimental data and current efficiency for Ex. No. 71

 Table B-72
 Experimental data and current efficiency for Ex. No. 72

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.42	0.0515	0.1127	45.69
0.5	194.54	0.0328	0.1040	31.50
1.0	180.47	0.1172	0.2253	52.00
1.0	186.76	0.0794	0.2080	38.19
1.5	167.33	0.1960	0.3380	58.00
	176.55	0.1407	0.3120	45.09
2.0	151.70	0.2898	0.4507	64.31
	164.08	0.2155	0.4161	51.80

**Table B-73** Experimental data and current efficiency for Ex. No. 73

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.04	0.0478	0.1216	39.29
0.5	195.14	0.0292	0.1122	25.98
1.0	181.65	0.1101	0.2431	45.29
	187.66	0.0740	0.2245	32.99
1.5	168.76	0.1874	0.3647	51.40
	177.55	0.1347	0.3367	40.01
2.0	153.48	0.2791	0.4862	57.41
	164.14	0.2152	0.4489	47.93

 Table B-74 Experimental data and current efficiency for Ex. No. 74

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.65	0.0501	0.1216	41.22
0.3	194.8	0.0312	0.1122	27.80
1.0	180.92	0.1145	0.2431	47.09
1.0	186.98	0.0781	0.2245	34.80
1.5	167.67	0.1940	0.3647	53.19
	176.6	0.1404	0.3367	41.70
2.0	152.03	0.2878	0.4862	59.19
	163.79	0.2173	0.4489	48.40

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	191.29	0.0523	0.1216	42.99
0.5	194.48	0.0331	0.1122	29.51
1.0	180.15	0.1191	0.2431	48.99
1.0	186.27	0.0824	0.2245	36.70
1.5	166.51	0.2009	0.3647	55.10
	175.59	0.1465	0.3367	43.50
2.0	150.57	0.2966	0.4862	61.00
2.0	162.52	0.2249	0.4489	50.10

 Table B-75 Experimental data and current efficiency for Ex. No. 75

 Table B-76 Experimental data and current efficiency for Ex. No. 76

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	292.08	0.0475	0.0860	55.27
0.5	195.70	0.0258	0.0794	32.50
1.0	282.43	0.1054	0.1720	61.31
	189.42	0.0635	0.1588	39.99
1.5	271.20	0.1728	0.2579	66.99
	181.19	0.1129	0.2381	47.39
2.0	258.04	0.2518	0.3439	73.20
	171.09	0.1735	0.3175	54.63

 Table B-77 Experimental data and current efficiency for Ex. No. 77

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	291.82	0.0491	0.0860	57.08
0.5	195.46	0.0272	0.0794	34.32
1.0	281.94	0.1084	0.1720	63.02
	189.02	0.0659	0.1588	41.50
1.5	270.85	0.1749	0.2579	67.81
	180.67	0.1160	0.2381	48.70
2.0	257.01	0.2579	0.3439	75.00
	170.79	0.1753	0.3175	55.20

 Table B-78 Experimental data and current efficiency for Ex. No. 78

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	291.53	0.0508	0.0860	59.11
0.3	195.21	0.0287	0.0794	36.21
1.0	281.40	0.1116	0.1720	64.90
1.0	188.57	0.0686	0.1588	43.20
1.5	270.12	0.1793	0.2579	69.51
	179.88	0.1207	0.2381	50.69
2.0	255.92	0.2645	0.3439	76.90
	169.84	0.1810	0.3175	56.99

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	291.78	0.0493	0.0949	51.98
0.5	195.52	0.0269	0.0876	30.69
1.0	281.72	0.1097	0.1897	57.80
1.0	188.99	0.0661	0.1752	37.71
1.5	270.02	0.1799	0.2846	63.20
	180.21	0.1187	0.2628	45.19
2.0	256.04	0.2638	0.3795	69.50
2.0	169.17	0.1850	0.3504	52.80

 Table B-79 Experimental data and current efficiency for Ex. No. 79

 Table B-80 Experimental data and current efficiency for Ex. No. 80

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	291.48	0.0511	0.0949	53.88
0.5	195.27	0.0284	0.0876	32.40
1.0	281.12	0.1133	0.1897	59.70
	188.44	0.0694	0.1752	39.59
1.5	269.17	0.1850	0.2846	64.99
	179.42	0.1235	0.2628	46.99
2.0	254.84	0.2710	0.3795	71.40
	168.18	0.1909	0.3504	54.49

 Table B-81 Experimental data and current efficiency for Ex. No. 81

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	291.18	0.0529	0.0949	55.78
0.5	194.99	0.0301	0.0876	34.32
1.0	280.52	0.1169	0.1897	61.60
	187.91	0.0725	0.1752	41.41
1.5	268.74	0.1876	0.2846	65.90
	179.07	0.1256	0.2628	47.79
2.0	253.70	0.2778	0.3795	73.20
	167.07	0.1976	0.3504	56.39

 Table B-82 Experimental data and current efficiency for Ex. No. 82

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	291.49	0.0511	0.1038	49.21
0.5	195.29	0.0283	0.0958	29.50
1.0	280.91	0.1145	0.2075	55.19
1.0	188.22	0.0707	0.1916	36.89
1.5	268.20	0.1908	0.3113	61.29
	178.59	0.1285	0.2874	44.70
2.0	253.44	0.2794	0.4151	67.30
	166.92	0.1985	0.3832	51.79

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	291.18	0.0529	0.1038	51.00
0.5	195.02	0.0299	0.0958	31.19
1.0	280.28	0.1183	0.2075	57.01
	187.67	0.0740	0.1916	38.61
1.5	267.26	0.1964	0.3113	63.10
	177.68	0.1339	0.2874	46.60
20	252.26	0.2864	0.4151	69.01
2.0	165.77	0.2054	0.3832	53.59

 Table B-83 Experimental data and current efficiency for Ex. No. 83

**Table B-84** Experimental data and current efficiency for Ex. No. 84

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	291.04	0.0538	0.1038	51.81
0.5	194.72	0.0317	0.0958	33.07
1.0	279.63	0.1222	0.2075	58.89
	187.04	0.0778	0.1916	40.58
1.5	266.33	0.2020	0.3113	64.89
	176.77	0.1394	0.2874	48.50
2.0	250.95	0.2943	0.4151	70.90
	164.62	0.2123	0.3832	55.40

**Table B-85** Experimental data and current efficiency for Ex. No. 85

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	291.27	0.0524	0.1127	46.49
0.5	195.16	0.0290	0.1040	27.92
1.0	280.32	0.1181	0.2253	52.40
	187.73	0.0736	0.2080	35.39
1.5	266.82	0.1991	0.3380	58.90
	177.69	0.1339	0.3120	42.90
2.0	251.32	0.2921	0.4507	64.81
	165.67	0.2060	0.4161	49.51

 Table B-86 Experimental data and current efficiency for Ex. No. 86

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	290.95	0.0543	0.1127	48.20
0.5	194.83	0.0310	0.1040	29.82
1.0	279.65	0.1221	0.2253	54.19
1.0	187.14	0.0772	0.2080	37.09
1.5	265.81	0.2051	0.3380	60.69
	176.81	0.1391	0.3120	44.59
2.0	250.05	0.2997	0.4507	66.50
	164.42	0.2135	0.4161	51.31

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	290.59	0.0565	0.1127	50.11
0.5	194.52	0.0329	0.1040	31.61
1.0	278.89	0.1267	0.2253	56.21
	186.48	0.0811	0.2080	38.99
1.5	264.74	0.2116	0.3380	62.59
	175.87	0.1448	0.3120	46.40
2.0	248.62	0.3083	0.4507	68.41
2.0	163.11	0.2213	0.4161	53.20

 Table B-87 Experimental data and current efficiency for Ex. No. 87

**Table B-88** Experimental data and current efficiency for Ex. No. 88

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	291.13	0.0532	0.1216	43.78
0.5	195.10	0.0294	0.1122	26.20
1.0	279.90	0.1206	0.2431	49.61
	187.36	0.0758	0.2245	33.79
1.5	266.15	0.2031	0.3647	55.69
	176.77	0.1394	0.3367	41.40
2.0	249.84	0.3010	0.4862	61.90
	163.94	0.2164	0.4489	48.20

**Table B-89** Experimental data and current efficiency for Ex. No. 89

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	290.78	0.0553	0.1216	45.51
0.5	194.78	0.0313	0.1122	27.91
1.0	279.17	0.1250	0.2431	51.41
	186.65	0.0801	0.2245	35.69
1.5	265.05	0.2097	0.3647	57.50
	175.82	0.1451	0.3367	43.09
2.0	248.38	0.3097	0.4862	63.70
	162.59	0.2245	0.4489	50.00

 Table B-90 Experimental data and current efficiency for Ex. No. 90

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	290.44	0.0574	0.1216	47.19
0.5	194.43	0.0334	0.1122	29.78
1.0	278.40	0.1296	0.2431	53.31
1.0	185.93	0.0844	0.2245	37.61
1.5	263.96	0.2162	0.3647	59.30
	174.75	0.1515	0.3367	45.00
2.0	246.76	0.3194	0.4862	65.70
	161.17	0.2330	0.4489	51.90
Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
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0.5	93.08	0.0415	0.1038	40.01
0.5	193.49	0.0391	0.1835	21.29
1.0	84.23	0.0946	0.2075	45.59
	183.61	0.0983	0.3670	26.80
1.5	73.28	0.1603	0.3113	51.50
	171.01	0.1739	0.5505	31.60
20	60.15	0.2391	0.4151	57.60
2.0	154.98	0.2701	0.7340	36.80

 Table B-91 Experimental data and current efficiency for Ex. No. 91

 Table B-92 Experimental data and current efficiency for Ex. No. 92

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	92.77	0.0434	0.1038	41.80
0.5	192.91	0.0425	0.1835	23.18
1.0	83.60	0.0984	0.2075	47.41
	182.45	0.1053	0.3670	28.69
1.5	72.40	0.1656	0.3113	53.20
	169.36	0.1838	0.5505	33.40
2.0	58.84	0.2470	0.4151	59.50
	152.78	0.2833	0.7340	38.60

**Table B-93** Experimental data and current efficiency for Ex. No. 93

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	92.46	0.0452	0.1038	43.60
0.5	192.32	0.0461	0.1835	25.11
1.0	82.98	0.1021	0.2075	49.21
	181.22	0.1127	0.3670	30.70
1.5	71.41	0.1715	0.3113	55.10
	167.52	0.1949	0.5505	35.40
2.0	57.52	0.2549	0.4151	61.41
	150.34	0.2980	0.7340	40.60

 Table B-94 Experimental data and current efficiency for Ex. No. 94

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	93.07	0.0416	0.1127	36.91
0.5	193.23	0.0406	0.1992	20.39
1.0	83.89	0.0967	0.2253	42.90
1.0	182.80	0.1032	0.3984	25.90
1.5	72.17	0.1670	0.3380	49.40
	169.02	0.1859	0.5977	31.10
20	58.46	0.2492	0.4507	55.31
2.0	150.86	0.2948	0.7969	37.00

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	92.73	0.0436	0.1127	38.72
0.5	192.63	0.0442	0.1992	22.20
1.0	83.18	0.1009	0.2253	44.79
	181.61	0.1103	0.3984	27.69
1.5	71.22	0.1727	0.3380	51.09
	167.13	0.1972	0.5977	33.00
20	57.11	0.2573	0.4507	57.10
2.0	148.60	0.3084	0.7969	38.70

**Table B-95** Experimental data and current efficiency for Ex. No. 95

Table B-96 Experimental data and current efficiency for Ex. No. 96

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	92.36	0.0458	0.1127	40.69
0.5	191.99	0.0481	0.1992	24.12
1.0	82.39	0.1057	0.2253	46.89
	180.41	0.1175	0.3984	29.50
1.5	70.14	0.1792	0.3380	53.01
	165.24	0.2086	0.5977	34.90
20	55.76	0.2654	0.4507	58.90
2.0	146.21	0.3227	0.7969	40.50

 Table B-97 Experimental data and current efficiency for Ex. No. 97

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	93.03	0.0418	0.1216	34.40
0.5	192.87	0.0428	0.2149	19.90
1.0	83.55	0.0987	0.2431	40.60
	181.73	0.1096	0.4299	25.50
1.5	71.62	0.1703	0.3647	46.69
	167.22	0.1967	0.6448	30.50
2.0	57.13	0.2572	0.4862	52.90
	147.84	0.3130	0.8598	36.40

 Table B-98 Experimental data and current efficiency for Ex. No. 98

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	92.67	0.0440	0.1216	36.18
0.5	192.19	0.0469	0.2149	21.80
1.0	82.82	0.1031	0.2431	42.40
1.0	180.37	0.1178	0.4299	27.40
1.5	70.58	0.1765	0.3647	48.41
	165.29	0.2083	0.6448	32.30
20	55.59	0.2665	0.4862	54.80
2.0	145.26	0.3284	0.8598	38.20

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	92.30	0.0462	0.1216	38.01
0.5	191.55	0.0507	0.2149	23.59
1.0	82.09	0.1075	0.2431	44.20
1.0	179.01	0.1259	0.4299	29.30
1.5	69.43	0.1834	0.3647	50.30
	163.24	0.2206	0.6448	34.20
20	53.97	0.2762	0.4862	56.80
2.0	142.25	0.3465	0.8598	40.30

**Table B-99** Experimental data and current efficiency for Ex. No. 99

 Table B-100 Experimental data and current efficiency for Ex. No. 100

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	93.02	0.0419	0.1305	32.10
0.5	192.70	0.0438	0.2307	18.99
1.0	83.35	0.0999	0.2609	38.29
1.0	181.08	0.1135	0.4614	24.61
1.5	70.98	0.1741	0.3914	44.49
	165.86	0.2048	0.6920	29.60
2.0	55.73	0.2656	0.5218	50.90
	145.56	0.3266	0.9227	35.40

 Table B-101 Experimental data and current efficiency for Ex. No. 101

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	92.63	0.0442	0.1305	33.90
0.5	192.04	0.0478	0.2307	20.70
1.0	82.56	0.1046	0.2609	40.11
	179.62	0.1223	0.4614	26.50
1.5	69.74	0.1816	0.3914	46.39
	163.90	0.2166	0.6920	31.30
2.0	54.17	0.2750	0.5218	52.70
	142.95	0.3423	0.9227	37.10

 Table B-102 Experimental data and current efficiency for Ex. No. 102

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	92.20	0.0468	0.1305	35.88
0.3	191.31	0.0521	0.2307	22.60
1.0	81.78	0.1093	0.2609	41.90
1.0	178.24	0.1306	0.4614	28.30
1.5	68.43	0.1894	0.3914	48.40
	161.71	0.2297	0.6920	33.20
20	52.52	0.2849	0.5218	54.60
2.0	140.02	0.3599	0.9227	39.00

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	92.94	0.0424	0.1393	30.40
0.5	192.61	0.0443	0.2464	17.99
1.0	83.05	0.1017	0.2787	36.49
	180.53	0.1168	0.4928	23.71
1.5	70.32	0.1781	0.4180	42.60
	164.40	0.2136	0.7392	28.90
2.0	54.57	0.2726	0.5574	48.90
2.0	143.49	0.3391	0.9856	34.40

 Table B-103 Experimental data and current efficiency for Ex. No. 103

 Table B-104 Experimental data and current efficiency for Ex. No. 104

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	92.52	0.0449	0.1393	32.21
0.5	191.91	0.0485	0.2464	19.70
1.0	82.16	0.1070	0.2787	38.41
	179.06	0.1256	0.4928	25.49
1.5	68.99	0.1861	0.4180	44.51
	162.18	0.2269	0.7392	30.70
2.0	52.99	0.2821	0.5574	50.60
	140.45	0.3573	0.9856	36.25

**Table B-105** Experimental data and current efficiency for Ex. No. 105

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	92.08	0.0475	0.1393	34.10
0.5	191.13	0.0532	0.2464	21.60
1.0	81.28	0.1123	0.2787	40.30
	177.41	0.1355	0.4928	27.50
1.5	67.53	0.1948	0.4180	46.60
	159.96	0.2402	0.7392	32.50
2.0	51.23	0.2926	0.5574	52.50
	137.58	0.3745	0.9856	38.00

 Table B-106 Experimental data and current efficiency for Ex. No. 106

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	192.20	0.0468	0.1038	45.10
0.3	193.27	0.0404	0.1835	22.01
1.0	182.32	0.1061	0.2075	51.11
	182.87	0.1028	0.3670	28.01
1.5	170.38	0.1777	0.3113	57.09
	169.45	0.1833	0.5505	33.30
2.0	156.35	0.2619	0.4151	63.10
2.0	152.29	0.2863	0.7340	39.00

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	191.89	0.0487	0.1038	46.89
0.5	192.72	0.0437	0.1835	23.80
1.0	181.74	0.1096	0.2075	52.79
	181.83	0.1090	0.3670	29.71
1.5	169.44	0.1834	0.3113	58.90
	167.71	0.1937	0.5505	35.20
2.0	155.10	0.2694	0.4151	64.90
2.0	151.31	0.2921	0.7340	39.80

 Table B-107 Experimental data and current efficiency for Ex. No. 107

 Table B-108 Experimental data and current efficiency for Ex. No. 108

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.58	0.0505	0.1038	48.69
0.3	192.17	0.0470	0.1835	25.60
1.0	181.08	0.1135	0.2075	54.70
	180.67	0.1160	0.3670	31.60
1.5	168.45	0.1893	0.3113	60.81
	165.87	0.2048	0.5505	37.20
2.0	153.79	0.2773	0.4151	66.80
	148.99	0.3061	0.7340	41.70

**Table B-109** Experimental data and current efficiency for Ex. No. 109

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	192.02	0.0479	0.1127	42.50
0.5	192.86	0.0428	0.1992	21.50
1.0	181.67	0.1100	0.2253	48.81
	181.74	0.1096	0.3984	27.50
1.5	169.13	0.1852	0.3380	54.80
	166.83	0.1990	0.5977	33.30
2.0	154.33	0.2740	0.4507	60.81
	149.91	0.3005	0.7969	37.71

 Table B-110 Experimental data and current efficiency for Ex. No. 110

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	191.66	0.0500	0.1127	44.42
0.3	192.23	0.0466	0.1992	23.40
1.0	181.04	0.1138	0.2253	50.49
1.0	180.48	0.1171	0.3984	29.39
1.5	168.12	0.1913	0.3380	56.59
	165.14	0.2092	0.5977	35.00
2.0	153.05	0.2817	0.4507	62.51
2.0	144.88	0.3307	0.7969	41.50

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	191.31	0.0521	0.1127	46.28
0.5	191.53	0.0508	0.1992	25.51
1.0	180.32	0.1181	0.2253	52.40
	179.08	0.1255	0.3984	31.50
1.5	167.05	0.1977	0.3380	58.49
	163.24	0.2206	0.5977	36.90
2.0	151.55	0.2907	0.4507	64.51
2.0	142.36	0.3458	0.7969	43.40

 Table B-111 Experimental data and current efficiency for Ex. No. 111

 Table B-112 Experimental data and current efficiency for Ex. No. 112

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.94	0.0484	0.1216	39.78
0.5	192.19	0.0469	0.2149	21.80
1.0	181.40	0.1116	0.2431	45.90
	180.00	0.1200	0.4299	27.91
1.5	168.27	0.1904	0.3647	52.21
	164.10	0.2154	0.6448	33.40
2.0	152.99	0.2821	0.4862	58.01
	147.82	0.3131	0.8598	36.41

 Table B-113 Experimental data and current efficiency for Ex. No. 113

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	191.57	0.0506	0.1216	41.61
0.5	191.58	0.0505	0.2149	23.50
1.0	180.67	0.1160	0.2431	47.71
	178.72	0.1277	0.4299	29.70
1.5	167.12	0.1973	0.3647	54.10
	162.06	0.2276	0.6448	35.30
2.0	151.62	0.2903	0.4862	59.70
	140.10	0.3594	0.8598	41.80

 Table B-114 Experimental data and current efficiency for Ex. No. 114

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.19	0.0529	0.1216	43.49
0.3	190.90	0.0546	0.2149	25.40
1.0	179.86	0.1208	0.2431	49.71
1.0	177.36	0.1358	0.4299	31.60
1.5	165.96	0.2042	0.3647	56.01
	160.13	0.2392	0.6448	37.10
20	150.08	0.2995	0.4862	61.60
2.0	137.09	0.3775	0.8598	43.90

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.89	0.0487	0.1305	37.30
0.5	192.00	0.0480	0.2307	20.81
1.0	181.13	0.1132	0.2609	43.40
1.0	179.24	0.1246	0.4614	27.00
1.5	167.78	0.1933	0.3914	49.40
	162.52	0.2249	0.6920	32.50
20	151.73	0.2896	0.5218	55.50
2.0	145.89	0.3247	0.9227	35.19

 Table B-115 Experimental data and current efficiency for Ex. No. 115

 Table B-116 Experimental data and current efficiency for Ex. No. 116

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.48	0.0511	0.1305	39.19
0.5	191.31	0.0521	0.2307	22.60
1.0	180.39	0.1177	0.2609	45.10
	177.78	0.1333	0.4614	28.90
1.5	166.54	0.2008	0.3914	51.30
	160.44	0.2374	0.6920	34.30
2.0	150.08	0.2995	0.5218	57.40
	137.87	0.3728	0.9227	40.40

**Table B-117** Experimental data and current efficiency for Ex. No. 117

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.06	0.0536	0.1305	41.12
0.5	190.54	0.0568	0.2307	24.61
1.0	179.56	0.1226	0.2609	47.01
	176.24	0.1426	0.4614	30.90
1.5	165.30	0.2082	0.3914	53.20
	158.25	0.2505	0.6920	36.20
2.0	148.43	0.3094	0.5218	59.30
	134.80	0.3912	0.9227	42.40

 Table B-118 Experimental data and current efficiency for Ex. No. 118

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.80	0.0492	0.1393	35.31
0.5	191.95	0.0483	0.2464	19.60
1.0	180.72	0.1157	0.2787	41.51
1.0	178.65	0.1281	0.4928	25.99
1.5	166.91	0.1985	0.4180	47.49
	160.95	0.2343	0.7392	31.70
2.0	150.11	0.2993	0.5574	53.70
	143.99	0.3361	0.9856	34.10

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.36	0.0518	0.1393	37.20
0.5	191.25	0.0525	0.2464	21.31
1.0	179.89	0.1207	0.2787	43.30
1.0	177.08	0.1375	0.4928	27.91
1.5	165.51	0.2069	0.4180	49.50
	158.60	0.2484	0.7392	33.60
2.0	148.54	0.3088	0.5574	55.39
2.0	135.28	0.3883	0.9856	39.40

 Table B-119 Experimental data and current efficiency for Ex. No. 119

 Table B-120 Experimental data and current efficiency for Ex. No. 120

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	190.94	0.0544	0.1393	39.01
0.5	190.47	0.0572	0.2464	23.21
1.0	178.96	0.1262	0.2787	45.30
	175.61	0.1463	0.4928	29.70
1.5	164.05	0.2157	0.4180	51.60
	156.14	0.2632	0.7392	35.60
2.0	146.86	0.3188	0.5574	57.20
	132.32	0.4061	0.9856	41.20

 Table B-121 Experimental data and current efficiency for Ex. No. 121

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	291.40	0.0516	0.1038	49.73
0.5	192.66	0.0440	0.1835	24.00
1.0	280.66	0.1160	0.2075	55.91
	181.47	0.1112	0.3670	30.30
1.5	267.68	0.1939	0.3113	62.29
	167.29	0.1963	0.5505	35.65
2.0	252.33	0.2860	0.4151	68.91
	149.68	0.3019	0.7340	41.14

 Table B-122 Experimental data and current efficiency for Ex. No. 122

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	291.08	0.0535	0.1038	51.58
0.5	192.14	0.0472	0.1835	25.70
1.0	280.08	0.1195	0.2075	57.59
1.0	180.71	0.1157	0.3670	31.54
1.5	266.69	0.1999	0.3113	64.20
	165.25	0.2085	0.5505	37.88
20	251.02	0.2939	0.4151	70.80
2.0	147.56	0.3146	0.7340	42.87

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	290.75	0.0555	0.1038	53.48
0.5	191.59	0.0505	0.1835	27.50
1.0	279.42	0.1235	0.2075	59.50
	179.31	0.1241	0.3670	33.83
1.5	265.65	0.2061	0.3113	66.21
	163.32	0.2201	0.5505	39.98
20	249.63	0.3022	0.4151	72.81
2.0	145.19	0.3289	0.7340	44.81

 Table B-123 Experimental data and current efficiency for Ex. No. 123

 Table B-124 Experimental data and current efficiency for Ex. No. 124

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	291.12	0.0533	0.1127	47.29
0.5	192.10	0.0474	0.1992	23.79
1.0	279.80	0.1212	0.2253	53.79
	180.27	0.1184	0.3984	29.71
1.5	265.92	0.2045	0.3380	60.50
	164.33	0.2140	0.5977	35.81
2.0	249.90	0.3006	0.4507	66.70
	144.33	0.3340	0.7969	41.92

**Table B-125** Experimental data and current efficiency for Ex. No. 125

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	290.78	0.0553	0.1127	49.10
0.5	191.47	0.0512	0.1992	25.69
1.0	279.12	0.1253	0.2253	55.60
	179.08	0.1255	0.3984	31.50
1.5	264.85	0.2109	0.3380	62.40
	162.63	0.2242	0.5977	37.52
2.0	248.55	0.3087	0.4507	68.50
	142.48	0.3451	0.7969	43.31

 Table B-126 Experimental data and current efficiency for Ex. No. 126

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	290.39	0.0577	0.1127	51.18
0.5	190.83	0.0550	0.1992	27.62
1.0	278.33	0.1300	0.2253	57.70
1.0	177.68	0.1339	0.3984	33.61
1.5	263.78	0.2173	0.3380	64.30
	161.74	0.2296	0.5977	38.41
2.0	247.27	0.3164	0.4507	70.21
2.0	140.09	0.3595	0.7969	45.11

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	290.92	0.0545	0.1216	44.82
0.5	191.65	0.0501	0.2149	23.31
1.0	279.42	0.1235	0.2431	50.79
	178.60	0.1284	0.4299	29.87
1.5	265.30	0.2082	0.3647	57.09
	161.82	0.2291	0.6448	35.52
2.0	247.47	0.3152	0.4862	64.82
2.0	139.39	0.3637	0.8598	42.30

 Table B-127 Experimental data and current efficiency for Ex. No. 127

 Table B-128 Experimental data and current efficiency for Ex. No. 128

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	290.58	0.0565	0.1216	46.50
0.5	190.84	0.0550	0.2149	25.57
1.0	278.69	0.1279	0.2431	52.59
	177.48	0.1351	0.4299	31.43
1.5	264.20	0.2148	0.3647	58.90
	159.59	0.2425	0.6448	37.60
2.0	245.99	0.3241	0.4862	66.65
2.0	136.11	0.3833	0.8598	44.59

**Table B-129** Experimental data and current efficiency for Ex. No. 129

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	290.19	0.0589	0.1216	48.42
0.5	190.10	0.0594	0.2149	27.63
1.0	277.92	0.1325	0.2431	54.49
	175.79	0.1453	0.4299	33.79
1.5	263.11	0.2213	0.3647	60.70
	157.45	0.2553	0.6448	39.59
2.0	244.25	0.3345	0.4862	68.79
	133.53	0.3988	0.8598	46.39

 Table B-130 Experimental data and current efficiency for Ex. No. 130

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	290.85	0.0549	0.1305	42.08
0.3	191.46	0.0512	0.2307	22.21
1.0	278.95	0.1263	0.2609	48.41
	177.45	0.1353	0.4614	29.33
1.5	264.52	0.2129	0.3914	54.40
	160.13	0.2392	0.6920	34.57
2.0	247.47	0.3152	0.5218	60.40
	136.64	0.3802	0.9227	41.20

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	290.43	0.0574	0.1305	44.02
0.5	190.87	0.0548	0.2307	23.75
1.0	278.13	0.1312	0.2609	50.29
	176.25	0.1425	0.4614	30.89
1.5	263.28	0.2203	0.3914	56.30
	157.75	0.2535	0.6920	36.63
2.0	245.99	0.3241	0.5218	62.10
	133.67	0.3980	0.9227	43.13

 Table B-131 Experimental data and current efficiency for Ex. No. 131

 Table B-132 Experimental data and current efficiency for Ex. No. 132

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	290.04	0.0598	0.1305	45.81
0.5	189.80	0.0612	0.2307	26.53
1.0	277.35	0.1359	0.2609	52.09
	174.46	0.1532	0.4614	33.22
1.5	261.97	0.2282	0.3914	58.31
	155.46	0.2672	0.6920	38.62
2.0	244.25	0.3345	0.5218	64.10
	131.10	0.4134	0.9227	44.80

**Table B-133** Experimental data and current efficiency for Ex. No. 133

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	290.68	0.0559	0.1393	40.13
0.5	191.30	0.0522	0.2464	21.18
1.0	278.54	0.1288	0.2787	46.20
	176.84	0.1390	0.4928	28.20
1.5	263.35	0.2199	0.4180	52.60
	158.68	0.2479	0.7392	33.54
2.0	245.47	0.3272	0.5574	58.70
	135.12	0.3893	0.9856	39.50

 Table B-134 Experimental data and current efficiency for Ex. No. 134

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	290.29	0.0583	0.1393	41.81
0.3	190.42	0.0575	0.2464	23.33
1.0	277.66	0.1340	0.2787	48.10
1.0	174.86	0.1508	0.4928	30.61
1.5	262.1	0.2274	0.4180	54.40
	156.08	0.2635	0.7392	35.65
2.0	243.89	0.3367	0.5574	60.40
	131.5	0.4110	0.9856	41.70

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	289.85	0.0609	0.1393	43.70
0.5	189.54	0.0628	0.2464	25.47
1.0	276.68	0.1399	0.2787	50.21
1.0	173.42	0.1595	0.4928	32.36
1.5	260.64	0.2362	0.4180	56.49
	153.74	0.2776	0.7392	37.55
2.0	242.22	0.3467	0.5574	62.20
	127.61	0.4343	0.9856	44.07

 Table B-135 Experimental data and current efficiency for Ex. No. 135

 Table B-136 Experimental data and current efficiency for Ex. No. 136

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	92.94	0.0424	0.1038	40.82
0.5	195.48	0.0271	0.0958	28.31
	193.3	0.0402	0.1835	21.91
	83.92	0.0965	0.2075	46.49
1.0	188.95	0.0663	0.1916	34.60
	183.18	0.1009	0.3670	27.50
	72.76	0.1634	0.3113	52.50
1.5	180.41	0.1175	0.2874	40.90
	169.91	0.1805	0.5505	32.80
2.0	59.6	0.2424	0.4151	58.40
	169.92	0.1805	0.3832	47.10
	154.13	0.2752	0.7340	37.50

Table B-137 Experimental data and current efficiency for Ex. No. 137

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	92.62	0.0443	0.1038	42.67
0.5	195.19	0.0289	0.0958	30.12
	192.75	0.0435	0.1835	23.71
	83.29	0.1003	0.2075	48.31
1.0	188.34	0.0700	0.1916	36.51
	182.08	0.1075	0.3670	29.30
	71.88	0.1687	0.3113	54.20
1.5	179.55	0.1227	0.2874	42.69
	168.35	0.1899	0.5505	34.50
2.0	58.42	0.2495	0.4151	60.11
	168.77	0.1874	0.3832	48.90
	151.93	0.2884	0.7340	39.30

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
	92.30	0.0462	0.1038	44.52
0.5	194.92	0.0305	0.0958	31.82
	192.17	0.0470	0.1835	25.60
1.0	82.64	0.1042	0.2075	50.19
	187.77	0.0734	0.1916	38.30
	181.04	0.1138	0.3670	31.00
	70.95	0.1743	0.3113	55.99
1.5	178.64	0.1282	0.2874	44.59
	166.70	0.1998	0.5505	36.30
2.0	57.18	0.2569	0.4151	61.90
	167.68	0.1939	0.3832	50.60
	149.72	0.3017	0.7340	41.10

 Table B-138 Experimental data and current efficiency for Ex. No. 138

 Table B-139 Experimental data and current efficiency for Ex. No. 139

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	92.92	0.0425	0.1127	37.71
0.5	195.35	0.0279	0.1040	26.82
	193.03	0.0418	0.1992	20.99
	83.63	0.0982	0.2253	43.59
1.0	188.56	0.0686	0.2080	33.00
	182.34	0.1060	0.3984	26.59
	71.72	0.1697	0.3380	50.20
1.5	179.51	0.1229	0.3120	39.40
	168.22	0.1907	0.5977	31.90
2.0	57.86	0.2528	0.4507	56.11
	168.38	0.1897	0.4161	45.60
	149.53	0.3028	0.7969	38.00

Table B-140 Experimental data and current efficiency for Ex. No. 140

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
	92.58	0.0445	0.1127	39.52
0.5	195.06	0.0296	0.1040	28.50
	192.43	0.0454	0.1992	22.80
	82.99	0.1021	0.2253	45.29
1.0	187.93	0.0724	0.2080	34.81
	181.27	0.1124	0.3984	28.21
	70.71	0.1757	0.3380	52.00
1.5	178.63	0.1282	0.3120	41.09
	166.63	0.2002	0.5977	33.50
2.0	56.51	0.2609	0.4507	57.90
	167.27	0.1964	0.4161	47.20
	147.14	0.3172	0.7969	39.80

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
	92.32	0.0461	0.1127	40.90
0.5	194.77	0.0314	0.1040	30.17
	191.83	0.0490	0.1992	24.61
1.0	82.27	0.1064	0.2253	47.21
	187.31	0.0761	0.2080	36.60
	180.01	0.1199	0.3984	30.10
	69.64	0.1822	0.3380	53.90
1.5	177.69	0.1339	0.3120	42.90
	164.84	0.2110	0.5977	35.30
	55.08	0.2695	0.4507	59.81
2.0	166.02	0.2039	0.4161	49.00
	144.62	0.3323	0.7969	41.70

 Table B-141 Experimental data and current efficiency for Ex. No. 141

**Table B-142** Experimental data and current efficiency for Ex. No. 142

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	92.87	0.0428	0.1216	35.19
0.5	195.25	0.0285	0.1122	25.40
	192.62	0.0443	0.2149	20.60
	83.19	0.1009	0.2431	41.49
1.0	188.10	0.0714	0.2245	31.81
	181.23	0.1126	0.4299	26.20
	71.07	0.1736	0.3647	47.60
1.5	178.57	0.1286	0.3367	38.19
	166.47	0.2012	0.6448	31.20
2.0	56.24	0.2626	0.4862	54.00
	166.71	0.1997	0.4489	44.50
	146.84	0.3190	0.8598	37.10

Table B-143 Experimental data and current efficiency for Ex. No. 143

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	92.52	0.0449	0.1216	36.92
0.5	194.91	0.0305	0.1122	27.21
	192.01	0.0479	0.2149	22.30
	82.50	0.1050	0.2431	43.19
1.0	187.43	0.0754	0.2245	33.60
	180.01	0.1199	0.4299	27.90
	69.98	0.1801	0.3647	49.39
1.5	177.61	0.1343	0.3367	39.90
	164.53	0.2128	0.6448	33.00
2.0	54.78	0.2713	0.4862	55.80
	165.44	0.2074	0.4489	46.19
	144.11	0.3353	0.8598	39.00

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	92.14	0.0472	0.1216	38.80
0.5	194.56	0.0326	0.1122	29.08
	191.33	0.0520	0.2149	24.20
1.0	81.73	0.1096	0.2431	45.09
	186.76	0.0794	0.2245	35.39
	178.79	0.1273	0.4299	29.60
	68.82	0.1871	0.3647	51.30
1.5	176.49	0.1411	0.3367	41.90
	162.60	0.2244	0.6448	34.80
	53.32	0.2801	0.4862	57.60
2.0	164.01	0.2159	0.4489	48.10
	141.53	0.3508	0.8598	40.80

 Table B-144 Experimental data and current efficiency for Ex. No. 144

 Table B-145 Experimental data and current efficiency for Ex. No. 145

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	92.78	0.0433	0.1305	33.21
0.5	195.10	0.0294	0.1204	24.41
	192.31	0.0461	0.2307	20.00
	83.04	0.1018	0.2609	39.00
1.0	187.76	0.0734	0.2409	30.49
	180.24	0.1186	0.4614	25.70
	70.52	0.1769	0.3914	45.20
1.5	177.72	0.1337	0.3613	37.00
	164.71	0.2117	0.6920	30.60
2.0	55.04	0.2698	0.5218	51.70
	164.83	0.2110	0.4817	43.80
	143.87	0.3368	0.9227	36.50

Table B-146 Experimental data and current efficiency for Ex. No. 146

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	92.41	0.0455	0.1305	34.91
0.5	194.74	0.0316	0.1204	26.20
	191.66	0.0500	0.2307	21.69
	82.26	0.1064	0.2609	40.80
1.0	187.07	0.0776	0.2409	32.21
	179.01	0.1259	0.4614	27.30
	69.41	0.1835	0.3914	46.90
1.5	176.64	0.1402	0.3613	38.79
	162.75	0.2235	0.6920	32.30
2.0	53.47	0.2792	0.5218	53.50
	163.47	0.2192	0.4817	45.50
	141.26	0.3524	0.9227	38.20

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	91.99	0.0481	0.1305	36.84
0.5	194.36	0.0338	0.1204	28.10
	190.97	0.0542	0.2307	23.49
1.0	81.43	0.1114	0.2609	42.71
	186.35	0.0819	0.2409	34.00
	177.62	0.1343	0.4614	29.11
	68.17	0.1910	0.3914	48.80
1.5	175.49	0.1471	0.3613	40.70
	160.67	0.2360	0.6920	34.10
	51.82	0.2891	0.5218	55.40
2.0	162.02	0.2279	0.4817	47.30
	138.33	0.3700	0.9227	40.10

 Table B-147 Experimental data and current efficiency for Ex. No. 147

**Table B-148** Experimental data and current efficiency for Ex. No. 148

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	92.68	0.0439	0.1393	31.52
0.5	194.90	0.0306	0.1286	23.79
	192.12	0.0473	0.2464	19.19
	82.54	0.1048	0.2787	37.59
1.0	187.22	0.0767	0.2573	29.80
	179.38	0.1237	0.4928	25.11
	69.48	0.1831	0.4180	43.80
1.5	176.71	0.1397	0.3859	36.21
	162.79	0.2233	0.7392	30.20
2.0	53.83	0.2770	0.5574	49.70
	163.04	0.2218	0.5146	43.09
	140.86	0.3548	0.9856	36.00

Table B-149 Experimental data and current efficiency for Ex. No. 149

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	92.29	0.0463	0.1393	33.20
0.5	194.55	0.0327	0.1286	25.42
	191.42	0.0515	0.2464	20.89
	81.70	0.1098	0.2787	39.40
1.0	186.49	0.0811	0.2573	31.50
	177.91	0.1325	0.4928	26.89
	68.23	0.1906	0.4180	45.60
1.5	175.62	0.1463	0.3859	37.90
	160.58	0.2365	0.7392	32.00
2.0	52.25	0.2865	0.5574	51.40
	161.49	0.2311	0.5146	44.90
	138.07	0.3716	0.9856	37.70

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	91.85	0.0489	0.1393	35.09
0.5	194.17	0.0350	0.1286	27.19
	190.72	0.0557	0.2464	22.60
1.0	80.82	0.1151	0.2787	41.29
	185.76	0.0854	0.2573	33.21
	176.35	0.1419	0.4928	28.79
	66.91	0.1985	0.4180	47.49
1.5	174.53	0.1528	0.3859	39.60
	158.24	0.2506	0.7392	33.90
	50.49	0.2971	0.5574	53.30
2.0	159.95	0.2403	0.5146	46.70
	134.95	0.3903	0.9856	39.60

 Table B-150 Experimental data and current efficiency for Ex. No. 150

 Table B-151 Experimental data and current efficiency for Ex. No. 151

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	192.06	0.0476	0.1038	45.91
0.5	195.24	0.0286	0.0958	29.81
	192.81	0.0431	0.1835	23.51
	181.98	0.1081	0.2075	52.10
1.0	188.25	0.0705	0.1916	36.79
	181.83	0.1090	0.3670	29.71
	169.91	0.1805	0.3113	57.99
1.5	178.97	0.1262	0.2874	43.90
	166.97	0.1982	0.5505	36.00
2.0	155.86	0.2648	0.4151	63.81
	167.49	0.1951	0.3832	50.90
	148.38	0.3097	0.7340	42.20

Table B-152 Experimental data and current efficiency for Ex. No. 152

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	191.73	0.0496	0.1038	47.82
0.5	194.97	0.0302	0.0958	31.50
	192.29	0.0463	0.1835	25.21
	181.39	0.1117	0.2075	53.80
1.0	187.74	0.0736	0.1916	38.39
	180.79	0.1153	0.3670	31.41
	169.03	0.1858	0.3113	59.69
1.5	178.11	0.1313	0.2874	45.70
	165.23	0.2086	0.5505	37.90
2.0	154.82	0.2711	0.4151	65.31
	166.34	0.2020	0.3832	52.70
	146.30	0.3222	0.7340	43.90

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
	191.42	0.0515	0.1038	49.61
0.5	194.67	0.0320	0.0958	33.38
	191.71	0.0497	0.1835	27.11
1.0	180.73	0.1156	0.2075	55.71
	187.10	0.0774	0.1916	40.40
	179.69	0.1219	0.3670	33.21
	168.20	0.1908	0.3113	61.29
1.5	177.20	0.1368	0.2874	47.60
	163.67	0.2180	0.5505	39.60
	153.65	0.2781	0.4151	67.00
2.0	165.32	0.2081	0.3832	54.30
	144.10	0.3354	0.7340	45.70

 Table B-153 Experimental data and current efficiency for Ex. No. 153

 Table B-154 Experimental data and current efficiency for Ex. No. 154

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	191.83	0.0490	0.1127	43.51
0.5	195.09	0.0295	0.1040	28.32
	192.56	0.0446	0.1992	22.41
	181.34	0.1120	0.2253	49.69
1.0	187.76	0.0734	0.2080	35.30
	180.94	0.1144	0.3984	28.70
	168.62	0.1883	0.3380	55.71
1.5	177.79	0.1333	0.3120	42.71
	165.04	0.2098	0.5977	35.10
2.0	153.50	0.2790	0.4507	61.91
	165.33	0.2080	0.4161	50.00
	145.02	0.3299	0.7969	41.40

Table B-155 Experimental data and current efficiency for Ex. No. 155

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
	191.53	0.0508	0.1127	45.11
0.5	194.77	0.0314	0.1040	30.17
	191.97	0.0482	0.1992	24.18
	180.66	0.1160	0.2253	51.50
1.0	187.10	0.0774	0.2080	37.21
	179.75	0.1215	0.3984	30.49
	167.67	0.1940	0.3380	57.39
1.5	176.86	0.1388	0.3120	44.49
	163.44	0.2194	0.5977	36.70
2.0	152.08	0.2875	0.4507	63.80
	164.15	0.2151	0.4161	51.70
	142.63	0.3442	0.7969	43.20

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	191.18	0.0529	0.1127	46.97
0.5	194.45	0.0333	0.1040	32.01
	191.40	0.0516	0.1992	25.90
1.0	180.02	0.1199	0.2253	53.20
	186.44	0.0814	0.2080	39.11
	178.48	0.1291	0.3984	32.41
	166.60	0.2004	0.3380	59.29
1.5	175.92	0.1445	0.3120	46.30
	161.55	0.2307	0.5977	38.60
2.0	150.65	0.2961	0.4507	65.71
	162.90	0.2226	0.4161	53.50
	140.10	0.3594	0.7969	45.10

 Table B-156 Experimental data and current efficiency for Ex. No. 156

**Table B-157** Experimental data and current efficiency for Ex. No. 157

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	191.59	0.0505	0.1216	41.51
0.5	194.93	0.0304	0.1122	27.11
	192.15	0.0471	0.2149	21.91
	180.67	0.1160	0.2431	47.71
1.0	187.36	0.0758	0.2245	33.79
	180.01	0.1199	0.4299	27.90
	167.12	0.1973	0.3647	54.10
1.5	177.05	0.1377	0.3367	40.90
	162.92	0.2225	0.6448	34.50
2.0	151.13	0.2932	0.4862	60.31
	164.24	0.2146	0.4489	47.80
	141.53	0.3508	0.8598	40.80

Table B-158 Experimental data and current efficiency for Ex. No. 158

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	191.21	0.0527	0.1216	43.39
0.5	194.61	0.0323	0.1122	28.82
	191.55	0.0507	0.2149	23.59
	179.98	0.1201	0.2431	49.41
1.0	186.68	0.0799	0.2245	35.61
	178.65	0.1281	0.4299	29.80
	166.15	0.2031	0.3647	55.69
1.5	176.04	0.1438	0.3367	42.70
	161.10	0.2334	0.6448	36.19
2.0	149.76	0.3014	0.4862	62.00
	163.04	0.2218	0.4489	49.40
	138.95	0.3663	0.8598	42.60

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
	190.86	0.0548	0.1216	45.11
0.5	194.31	0.0341	0.1122	30.42
	190.97	0.0542	0.2149	25.21
1.0	179.21	0.1247	0.2431	51.31
	186.01	0.0839	0.2245	37.40
	177.36	0.1358	0.4299	31.60
	164.99	0.2101	0.3647	57.60
1.5	175.03	0.1498	0.3367	44.50
	159.05	0.2457	0.6448	38.10
2.0	148.22	0.3107	0.4862	63.90
	161.69	0.2299	0.4489	51.20
	136.52	0.3809	0.8598	44.30

 Table B-159 Experimental data and current efficiency for Ex. No. 159

**Table B-160** Experimental data and current efficiency for Ex. No. 160

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	191.61	0.0503	0.1305	38.59
0.5	194.80	0.0312	0.1204	25.91
	191.85	0.0489	0.2307	21.20
	180.43	0.1174	0.2609	45.01
1.0	186.79	0.0793	0.2409	32.91
	179.24	0.1246	0.4614	27.00
	166.47	0.2012	0.3914	51.41
1.5	175.55	0.1467	0.3613	40.60
	161.48	0.2311	0.6920	33.40
2.0	149.65	0.3021	0.5218	57.90
	161.70	0.2298	0.4817	47.70
	138.49	0.3691	0.9227	40.00

Table B-161 Experimental data and current efficiency for Ex. No. 161

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	191.22	0.0527	0.1305	40.38
0.5	194.42	0.0335	0.1204	27.80
	191.23	0.0526	0.2307	22.81
	179.69	0.1219	0.2609	46.71
1.0	186.07	0.0836	0.2409	34.70
	177.85	0.1329	0.4614	28.81
	165.24	0.2086	0.3914	53.29
1.5	174.47	0.1532	0.3613	42.40
	159.29	0.2443	0.6920	35.30
2.0	148.17	0.3110	0.5218	59.60
	160.26	0.2384	0.4817	49.49
	135.57	0.3866	0.9227	41.90

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	190.80	0.0552	0.1305	42.31
0.5	194.06	0.0356	0.1204	29.59
	190.58	0.0565	0.2307	24.50
1.0	178.87	0.1268	0.2609	48.59
	185.35	0.0879	0.2409	36.49
	176.55	0.1407	0.4614	30.50
	164.06	0.2156	0.3914	55.10
1.5	173.32	0.1601	0.3613	44.31
	157.21	0.2567	0.6920	37.10
	146.51	0.3209	0.5218	61.51
2.0	158.81	0.2471	0.4817	51.30
	132.95	0.4023	0.9227	43.60

 Table B-162 Experimental data and current efficiency for Ex. No. 162

**Table B-163** Experimental data and current efficiency for Ex. No. 163

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	191.57	0.0506	0.1393	36.30
0.5	194.60	0.0324	0.1286	25.18
	191.46	0.0512	0.2464	20.80
	180.21	0.1187	0.2787	42.61
1.0	186.23	0.0826	0.2573	32.11
	177.58	0.1345	0.4928	27.30
	166.14	0.2032	0.4180	48.60
1.5	174.46	0.1532	0.3859	39.71
	158.36	0.2498	0.7392	33.80
2.0	149.28	0.3043	0.5574	54.60
	159.86	0.2408	0.5146	46.80
	133.64	0.3982	0.9856	40.40

Table B-164 Experimental data and current efficiency for Ex. No. 164

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	191.15	0.0531	0.1393	38.11
0.5	194.23	0.0346	0.1286	26.91
	190.68	0.0559	0.2464	22.69
	179.42	0.1235	0.2787	44.31
1.0	185.46	0.0872	0.2573	33.91
	176.02	0.1439	0.4928	29.20
	164.89	0.2107	0.4180	50.39
1.5	173.37	0.1598	0.3859	41.40
	156.14	0.2632	0.7392	35.60
2.0	147.61	0.3143	0.5574	56.40
	158.40	0.2496	0.5146	48.50
	130.51	0.4169	0.9856	42.30

**B-40** 

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	190.71	0.0557	0.1393	40.00
0.5	193.85	0.0369	0.1286	28.68
	189.90	0.0606	0.2464	24.59
1.0	178.54	0.1288	0.2787	46.20
	184.65	0.0921	0.2573	35.80
	174.62	0.1523	0.4928	30.90
	163.56	0.2186	0.4180	52.30
1.5	172.21	0.1667	0.3859	43.20
	154.05	0.2757	0.7392	37.30
	146.03	0.3238	0.5574	58.10
2.0	156.86	0.2588	0.5146	50.30
	127.39	0.4357	0.9856	44.20

 Table B-165 Experimental data and current efficiency for Ex. No. 165

 Table B-166 Experimental data and current efficiency for Ex. No. 166

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	291.18	0.0529	0.1038	51.00
0.5	195.10	0.0294	0.0958	30.69
	192.28	0.0463	0.1835	25.24
	280.21	0.1187	0.2075	57.21
1.0	187.87	0.0728	0.1916	37.98
	180.77	0.1154	0.3670	31.44
	267.11	0.1973	0.3113	63.39
1.5	178.06	0.1316	0.2874	45.80
	164.89	0.2107	0.5505	38.27
2.0	251.71	0.2897	0.4151	69.80
	166.28	0.2023	0.3832	52.80
	145.97	0.3242	0.7340	44.17

Table B-167 Experimental data and current efficiency for Ex. No. 167

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	290.87	0.0548	0.1038	52.79
0.5	194.83	0.0310	0.0958	32.38
	191.71	0.0497	0.1835	27.11
	279.63	0.1222	0.2075	58.89
1.0	187.26	0.0764	0.1916	39.89
	179.67	0.1220	0.3670	33.24
	266.17	0.2030	0.3113	65.20
1.5	177.15	0.1371	0.2874	47.70
	163.87	0.2168	0.5505	39.38
2.0	250.47	0.2972	0.4151	71.60
	165.19	0.2089	0.3832	54.50
	144.77	0.3314	0.7340	45.15

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
	290.54	0.0568	0.1038	54.70
0.5	194.54	0.0328	0.0958	34.20
	191.16	0.0530	0.1835	28.91
1.0	279.00	0.1260	0.2075	60.71
	186.68	0.0799	0.1916	41.71
	178.43	0.1294	0.3670	35.27
	265.19	0.2089	0.3113	67.09
1.5	176.29	0.1423	0.2874	49.50
	162.19	0.2269	0.5505	41.21
	249.15	0.3051	0.4151	73.51
2.0	164.04	0.2158	0.3832	56.30
	141.79	0.3493	0.7340	47.59

 Table B-168 Experimental data and current efficiency for Ex. No. 168

 Table B-169 Experimental data and current efficiency for Ex. No. 169

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	290.84	0.0550	0.1127	48.78
0.5	194.97	0.0302	0.1040	29.02
	191.53	0.0508	0.1992	25.51
	279.23	0.1246	0.2253	55.31
1.0	187.35	0.0759	0.2080	36.49
	179.51	0.1229	0.3984	30.86
	265.02	0.2099	0.3380	62.10
1.5	176.96	0.1382	0.3120	44.30
	162.43	0.2254	0.5977	37.72
2.0	248.62	0.3083	0.4507	68.41
	164.56	0.2126	0.4161	51.11
	142.41	0.3455	0.7969	43.36

Table B-170 Experimental data and current efficiency for Ex. No. 170

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	290.5	0.0570	0.1127	50.59
0.5	194.68	0.0319	0.1040	30.69
	191.23	0.0526	0.1992	26.41
	278.56	0.1286	0.2253	57.09
1.0	186.76	0.0794	0.2080	38.19
	178.61	0.1283	0.3984	32.21
	264.00	0.2160	0.3380	63.91
1.5	176.08	0.1435	0.3120	45.99
	161.64	0.2302	0.5977	38.51
2.0	247.27	0.3164	0.4507	70.21
	163.38	0.2197	0.4161	52.81
	140.02	0.3599	0.7969	45.16

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	290.14	0.0592	0.1127	52.51
0.5	194.37	0.0338	0.1040	32.48
	190.73	0.0556	0.1992	27.92
1.0	277.81	0.1331	0.2253	59.09
	186.10	0.0834	0.2080	40.09
	176.82	0.1391	0.3984	34.91
	262.93	0.2224	0.3380	65.81
1.5	175.14	0.1492	0.3120	47.80
	159.55	0.2427	0.5977	40.61
	245.77	0.3254	0.4507	72.20
2.0	162.07	0.2276	0.4161	54.70
	136.63	0.3802	0.7969	47.71

 Table B-171 Experimental data and current efficiency for Ex. No. 171

**Table B-172** Experimental data and current efficiency for Ex. No. 172

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	290.6	0.0564	0.1216	46.40
0.5	194.86	0.0308	0.1122	27.48
	191.60	0.0504	0.2149	23.45
	278.73	0.1276	0.2431	52.49
1.0	186.80	0.0792	0.2245	35.29
	177.80	0.1332	0.4299	30.98
	264.14	0.2152	0.3647	59.00
1.5	175.93	0.1444	0.3367	42.90
	160.96	0.2342	0.6448	36.33
2.0	247.33	0.3160	0.4862	64.99
	162.67	0.2240	0.4489	49.90
	139.04	0.3658	0.8598	42.54

Table B-173 Experimental data and current efficiency for Ex. No. 173

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	290.22	0.0587	0.1216	48.27
0.5	194.52	0.0329	0.1122	29.30
	190.69	0.0559	0.2149	25.99
	278.04	0.1318	0.2431	54.20
1.0	186.08	0.0835	0.2245	37.21
	177.11	0.1373	0.4299	31.95
	263.05	0.2217	0.3647	60.79
1.5	174.92	0.1505	0.3367	44.70
	158.64	0.2482	0.6448	38.48
2.0	245.95	0.3243	0.4862	66.70
	161.47	0.2312	0.4489	51.50
	135.46	0.3872	0.8598	45.04

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	289.83	0.0610	0.1216	50.20
0.5	194.18	0.0349	0.1122	31.12
	190.07	0.0596	0.2149	27.72
1.0	277.27	0.1364	0.2431	56.10
	185.45	0.0873	0.2245	38.89
	175.85	0.1449	0.4299	33.71
	261.89	0.2287	0.3647	62.70
1.5	173.91	0.1565	0.3367	46.50
	156.10	0.2634	0.6448	40.85
	244.41	0.3335	0.4862	68.60
2.0	160.12	0.2393	0.4489	53.30
	132.24	0.4066	0.8598	47.29

 Table B-174 Experimental data and current efficiency for Ex. No. 174

**Table B-175** Experimental data and current efficiency for Ex. No. 175

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	290.48	0.0571	0.1305	43.79
0.5	194.82	0.0311	0.1204	25.81
	190.89	0.0547	0.2307	23.70
	278.30	0.1302	0.2609	49.90
1.0	186.43	0.0814	0.2409	33.80
	177.40	0.1356	0.4614	29.39
1.5	263.47	0.2192	0.3914	56.01
	175.13	0.1492	0.3613	41.30
	158.98	0.2461	0.6920	35.57
	246.34	0.3220	0.5218	61.70
2.0	161.30	0.2322	0.4817	48.20
	135.10	0.3894	0.9227	42.20

Table B-176 Experimental data and current efficiency for Ex. No. 176

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	290.09	0.0595	0.1305	45.58
0.5	194.48	0.0331	0.1204	27.50
	190.56	0.0566	0.2307	24.55
	277.52	0.1349	0.2609	51.70
1.0	185.75	0.0855	0.2409	35.50
	176.31	0.1421	0.4614	30.81
1.5	262.30	0.2262	0.3914	57.80
	173.98	0.1561	0.3613	43.21
	157.02	0.2579	0.6920	37.26
	244.86	0.3308	0.5218	63.40
2.0	159.77	0.2414	0.4817	50.10
	130.89	0.4147	0.9227	44.94

**B-44** 

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
	289.67	0.0620	0.1305	47.51
0.5	194.08	0.0355	0.1204	29.49
	189.87	0.0608	0.2307	26.35
1.0	276.69	0.1399	0.2609	53.61
	185.03	0.0898	0.2409	37.29
	175.01	0.1499	0.4614	32.50
1.5	261.13	0.2332	0.3914	59.59
	172.84	0.1630	0.3613	45.10
	154.54	0.2728	0.6920	39.41
	243.21	0.3407	0.5218	65.30
2.0	158.17	0.2510	0.4817	52.10
	130.42	0.4175	0.9227	45.25

 Table B-177 Experimental data and current efficiency for Ex. No. 177

**Table B-178** Experimental data and current efficiency for Ex. No. 178

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	290.36	0.0578	0.1393	41.51
0.5	194.73	0.0316	0.1286	24.58
	190.76	0.0554	0.2464	22.50
	277.80	0.1332	0.2787	47.79
1.0	186.15	0.0831	0.2573	32.30
	175.56	0.1466	0.4928	29.76
1.5	262.52	0.2249	0.4180	53.79
	173.88	0.1567	0.3859	40.61
	156.22	0.2627	0.7392	35.54
2.0	244.82	0.3311	0.5574	59.40
	159.00	0.2460	0.5146	47.80
	130.38	0.4177	0.9856	42.38

Table B-179 Experimental data and current efficiency for Ex. No. 179

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
	289.94	0.0604	0.1393	43.32
0.5	194.36	0.0338	0.1286	26.30
	189.80	0.0612	0.2464	24.84
	277.01	0.1379	0.2787	49.50
1.0	185.38	0.0877	0.2573	34.09
	173.98	0.1561	0.4928	31.68
1.5	261.33	0.2320	0.4180	55.50
	172.98	0.1621	0.3859	42.01
	153.48	0.2791	0.7392	37.76
	243.15	0.3411	0.5574	61.20
2.0	157.55	0.2547	0.5146	49.50
	127.12	0.4373	0.9856	44.37

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
	289.5	0.0630	0.1393	45.21
0.5	193.95	0.0363	0.1286	28.22
	188.98	0.0661	0.2464	26.83
1.0	276.13	0.1432	0.2787	51.39
	184.52	0.0929	0.2573	36.10
	172.52	0.1649	0.4928	33.46
1.5	260.08	0.2395	0.4180	57.30
	171.76	0.1694	0.3859	43.90
	151.14	0.2932	0.7392	39.66
	241.29	0.3523	0.5574	63.20
2.0	156.00	0.2640	0.5146	51.30
	122.66	0.4640	0.9856	47.08

 Table B-180 Experimental data and current efficiency for Ex. No. 180

**Table B-181** Experimental data and current efficiency for Ex. No. 181

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	196.15	0.0231	0.0356	64.93
1.0	191.50	0.0510	0.0712	71.67
2.0	179.74	0.1216	0.1423	85.42

**Table B-182** Experimental data and current efficiency for Ex. No. 182

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	196.03	0.0238	0.0356	66.95
1.0	191.25	0.0525	0.0712	73.78
2.0	179.25	0.1245	0.1423	87.48

 Table B-183 Experimental data and current efficiency for Ex. No. 183

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.89	0.0247	0.0356	69.31
1.0	190.96	0.0542	0.0712	76.23
2.0	178.68	0.1279	0.1423	89.89

Table B-184 Experimental data and current efficiency f	or Ex.	No. 184
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Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.73	0.0496	0.0593	83.68
1.0	182.13	0.1072	0.1186	90.41
2.0	158.14	0.2512	0.2372	105.89

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	191.53	0.0508	0.0593	85.71
1.0	181.76	0.1094	0.1186	92.28
2.0	157.27	0.2564	0.2372	108.09

 Table B-185 Experimental data and current efficiency for Ex. No. 185

 Table B-186 Experimental data and current efficiency for Ex. No. 186

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	191.31	0.0521	0.0593	87.93
1.0	181.34	0.1120	0.1186	94.41
2.0	156.32	0.2621	0.2372	110.50

Table B-187 Experimental data and current efficiency for Ex. No. 187

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	182.76	0.1034	0.2253	45.91
1.0	160.79	0.2353	0.4507	52.20
2.0	100.41	0.5975	0.9013	66.30

Table B-188 Experimental data and current efficiency for Ex. No. 188

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	182.05	0.1077	0.2253	47.80
1.0	159.22	0.2447	0.4507	54.29
2.0	97.40	0.6156	0.9013	68.30

Table B-189 Experimental data and current efficiency for Ex. No. 189

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	181.30	0.1122	0.2253	49.79
1.0	157.49	0.2551	0.4507	56.60
2.0	94.10	0.6354	0.9013	70.50

**Table B-190** Experimental data and current efficiency for Ex. No. 190

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	191.00	0.0540	0.1588	34.01
1.0	178.62	0.1283	0.3175	40.40
2.0	144.01	0.3359	0.6350	52.90

 Table B-191 Experimental data and current efficiency for Ex. No. 191

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	190.50	0.0570	0.1588	35.90
1.0	177.56	0.1346	0.3175	42.40
2.0	142.00	0.3480	0.6350	54.80

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	189.92	0.0605	0.1588	38.10
1.0	176.45	0.1413	0.3175	44.50
2.0	139.67	0.3620	0.6350	57.00

 Table B-192 Experimental data and current efficiency for Ex. No. 192

 Table B-193 Experimental data and current efficiency for Ex. No. 193

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	190.55	0.0567	0.1916	29.59
1.0	176.88	0.1387	0.3832	36.20
2.0	137.79	0.3733	0.7664	48.70

Table B-194 Experimental data and current efficiency for Ex. No. 194

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	189.88	0.0607	0.1916	31.69
1.0	175.60	0.1464	0.3832	38.20
2.0	134.98	0.3901	0.7664	50.90

Table B-195 Experimental data and current efficiency for Ex. No. 195

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	189.21	0.0647	0.1916	33.79
1.0	174.13	0.1552	0.3832	40.51
2.0	132.30	0.4062	0.7664	53.00

Table B-196 Experimental data and current efficiency for Ex. No. 196

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	189.71	0.0617	0.2245	27.51
1.0	174.49	0.1531	0.4489	34.10
2.0	131.32	0.4121	0.8978	45.90

**Table B-197** Experimental data and current efficiency for Ex. No. 197

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	188.97	0.0662	0.2245	29.49
1.0	173.07	0.1616	0.4489	35.99
2.0	128.48	0.4291	0.8978	47.80

 Table B-198 Experimental data and current efficiency for Ex. No. 198

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	188.14	0.0712	0.2245	31.70
1.0	171.35	0.1719	0.4489	38.29
2.0	125.48	0.4471	0.8978	49.80

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	184.95	0.0903	0.3670	24.61
1.0	162.32	0.2261	0.7340	30.80
2.0	98.47	0.6092	1.4679	41.50

 Table B-199 Experimental data and current efficiency for Ex. No. 199

 Table B-200 Experimental data and current efficiency for Ex. No. 200

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	183.79	0.0973	0.3670	26.50
1.0	159.75	0.2415	0.7340	32.90
2.0	93.82	0.6371	1.4679	43.40

Table B-201 Experimental data and current efficiency for Ex. No. 201

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	182.57	0.1046	0.3670	28.50
1.0	157.06	0.2576	0.7340	35.10
2.0	88.93	0.6664	1.4679	45.40

Table B-202 Experimental data and current efficiency for Ex. No. 202

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	184.02	0.0959	0.4299	22.30
1.0	159.16	0.2450	0.8598	28.50
2.0	89.95	0.6603	1.7196	38.40

Table B-203 Experimental data and current efficiency for Ex. No. 203

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	182.66	0.1040	0.4299	24.20
1.0	156.15	0.2631	0.8598	30.60
2.0	84.22	0.6947	1.7196	40.40

Table B-204 Experimental data and current efficiency for Ex. No. 204

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	181.09	0.1135	0.4299	26.39
1.0	153.28	0.2803	0.8598	32.60
2.0	78.20	0.7308	1.7196	42.50

 Table B-205 Experimental data and current efficiency for Ex. No. 205

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	183.00	0.1020	0.4928	20.70
1.0	155.48	0.2671	0.9856	27.10
2.0	79.10	0.7254	1.9712	36.80

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	181.27	0.1124	0.4928	22.80
1.0	152.36	0.2858	0.9856	29.00
2.0	72.20	0.7668	1.9712	38.90

 Table B-206 Experimental data and current efficiency for Ex. No. 206

 Table B-207 Experimental data and current efficiency for Ex. No. 207

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	179.55	0.1227	0.4928	24.90
1.0	149.24	0.3046	0.9856	30.90
2.0	65.30	0.8082	1.9712	41.00

Table B-208 Experimental data and current efficiency for Ex. No. 208

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	85.58	0.0865	0.1720	50.31
1.0	67.39	0.1957	0.3439	56.89
2.0	20.56	0.4766	0.6878	69.30
0.5	190.79	0.0553	0.1588	34.81
1.0	177.51	0.1349	0.3175	42.50
2.0	139.57	0.3626	0.6350	57.10

**Table B-209** Experimental data and current efficiency for Ex. No. 209

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	85.04	0.0898	0.1720	52.20
1.0	66.24	0.2026	0.3439	58.90
2.0	18.26	0.4904	0.6878	71.30
0.5	190.26	0.0584	0.1588	36.81
1.0	176.40	0.1416	0.3175	44.60
2.0	137.24	0.3766	0.6350	59.30

 Table B-210 Experimental data and current efficiency for Ex. No. 210

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %	
0.5	84.41	0.0935	0.1720	54.40	
1.0	1.0 64.98		0.3439	61.10	
2.0	2.0 15.86		0.6878	73.40	
0.5	189.71	0.0617	0.1588	38.89	
1.0	175.34	0.1480	0.3175	46.60	
2.0	135.02	0.3899	0.6350	61.40	

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	84.71	0.0917	0.2075	44.20
1.0	64.79	0.2113	0.4151	50.90
2.0	11.45	0.5313	0.8301	64.00
0.5	190.01	0.0599	0.1916	31.28
1.0	175.16	0.1490	0.3832	38.89
2.0	133.83	0.3970	0.7664	51.80

 Table B-211 Experimental data and current efficiency for Ex. No. 211

 Table B-212 Experimental data and current efficiency for Ex. No. 212

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %	
0.5	83.99	0.0961	0.2075	46.29	
1.0	63.40	0.2196	0.4151	52.91	
2.0	8.55	0.5487	0.8301	66.10	
0.5	189.37	0.0638	0.1916	33.29	
1.0	173.94	0.1564	0.3832	40.80	
2.0	131.41	0.4115	0.7664	53.70	

 Table B-213 Experimental data and current efficiency for Ex. No. 213

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	83.26	0.1004	0.2075	48.40
1.0	61.88	0.2287	0.4151	55.10
2.0	5.78	0.5653	0.8301	68.10
0.5	188.66	0.0680	0.1916	35.51
1.0	172.60	0.1644	0.3832	42.90
2.0	128.72	0.4277	0.7664	55.80

 Table B-214 Experimental data and current efficiency for Ex. No. 214

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	84.12	0.0953	0.2431	39.19
1.0	62.89	0.2227	0.4862	45.79
2.0	4.54	0.5728	0.9725	58.90
0.5	189.41	0.0635	0.2245	28.31
1.0	173.74	0.1576	0.4489	35.10
2.0	127.43	0.4354	0.8978	48.50

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	83.27	0.1004	0.2431	41.29
1.0	61.35	0.2319	0.4862	47.69
2.0	1.29	0.5923	0.9725	60.90
0.5	188.59	0.0685	0.2245	30.50
1.0	172.24	0.1666	0.4489	37.10
2.0	125.03	0.4498	0.8978	50.10

Deposition Time (h)	Remaining. Conc.(ppm)	Remaining. Conc.(ppm)Wt. Exp.(g)		С.Е. %
0.5	82.42	0.1055	0.2431	43.39
1.0	59.56	0.2426	0.4862	49.90
2.0	2.0 0.00		0.9725	61.70
0.5	0.5 187.81		0.2245	32.59
1.0	170.52	0.1769	0.4489	39.40
2.0	121.74	0.4696	0.8978	52.30

 Table B-216 Experimental data and current efficiency for Ex. No. 216

 Table B-217 Experimental data and current efficiency for Ex. No. 217

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	184.07	0.0956	0.1720	55.58
1.0	164.46	0.2132	0.3439	62.00
2.0	114.71	0.5117	0.6878	74.40
0.5	190.47	0.0572	0.1588	36.02
1.0	176.45	0.1413	0.3175	44.50
2.0	137.34	0.3760	0.6350	59.20

**Table B-218** Experimental data and current efficiency for Ex. No. 218

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	183.46	0.0992	0.1720	57.71
1.0	163.32	0.2201	0.3439	63.99
2.0	112.53	0.5248	0.6878	76.30
0.5	189.95	0.0603	0.1588	37.98
1.0	175.45	0.1473	0.3175	46.39
2.0	135.12	0.3893	0.6350	61.30

Table B-219 Experimental data and current efficiency for Ex. No. 219

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	182.89	0.1027	0.1720	59.70
1.0	162.05	0.2277	0.3439	66.21
2.0	110.01	0.5399	0.6878	78.50
0.5	189.39	0.0637	0.1588	40.10
1.0	174.33	0.1540	0.3175	48.51
2.0	132.90	0.4026	0.6350	63.40

Table B-220 Ex	perimental da	ita and cur	rent efficiency	y for Ex	. No. 220
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Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g) Wt. Theo.(g)		C.E. %	
0.5	182.67	0.1040 0.2075		50.10	
1.0	160.56	0.2366	0.4151	57.01	
2.0	105.50	0.5670	0.8301	68.30	
0.5	189.53	0.0628	0.1916	32.79	
1.0	174.96	0.1502	0.3832	39.21	
2.0	130.77	0.4154	0.7664	54.20	

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g) Wt. Theo.(g)		С.Е. %	
0.5	181.91	0.1085 0.2075		52.30	
1.0	159.18	0.2449	0.4151	59.01	
2.0	2.0 102.87	0.5828	0.8301	70.20	
0.5	188.92	2 0.0665 0.1916	665 0.1916 34.	34.70	
1.0	173.62	0.1583	0.3832	41.30	
2.0	128.08	0.4315	0.7664	56.30	

Table B-221 Experimental data and current efficiency for Ex. No. 221

 Table B-222 Experimental data and current efficiency for Ex. No. 222

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	181.18	0.1129	0.2075	54.41
1.0	157.73	0.2536	0.4151	61.10
2.0	99.83	0.6010	0.8301	72.40
0.5	188.25	0.0705	0.1916	36.79
1.0	172.22	0.1667	0.3832	43.50
2.0	125.40	0.4476	0.7664	58.40

 Table B-223 Experimental data and current efficiency for Ex. No. 223

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	181.97	0.1082	0.2431	44.50
1.0	158.59	0.2485	0.4862	51.10
2.0	97.57	0.6146	0.9725	63.20
0.5	188.93	0.0664	0.2245	29.59
1.0	172.17	0.1670	0.4489	37.20
2.0	121.74	0.4696	0.8978	52.30

Table B-224 Experimental data and current efficiency for Ex. No. 224

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %	
0.5	181.12	0.1133	0.2431	46.60	
1.0	156.97	0.2582	0.4862	53.10	
2.0	94.16	0.6350	0.9725	65.30	
0.5	188.18	0.0709	0.2245	31.60	
1.0	170.60	0.1764	0.4489	39.30	
2.0	118.45	0.4893	0.8978	54.50	

Table B-225 Experimental data and current efficiency for H	Ex. No. 225
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Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g) Wt. Theo.(g)		C.E. %
0.5	180.23	0.1186	0.2431	48.79
1.0	155.19	0.2689 0.4862 53	55.30	
2.0	90.76	0.6554	0.9725	67.40
0.5	187.39	0.0757	0.2245	33.71
1.0	168.88	0.1867	0.4489	41.59
2.0	115.01	0.5099	0.8978	56.80

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g) Wt. Theo.(g)		С.Е. %
0.5	282.72	0.1037	0.1720	60.29
1.0	261.77	0.2294	0.3439	66.70
2.0	209.21 0.5447	0.6878	79.20	
0.5	189.81	0.0611	0.1588	38.51
1.0	175.45	0.1473	0.3175	46.39
2.0	135.44	0.3874	0.6350	61.00

Table B-226 Experimental data and current efficiency for Ex. No. 226

 Table B-227 Experimental data and current efficiency for Ex. No. 227

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	282.15	0.1071	0.1720	62.28
1.0	260.68	0.2359	0.3439	68.60
2.0	206.80	0.5592	0.6878	81.30
0.5	189.26	0.0644	0.1588	40.59
1.0	174.39	0.1537	0.3175	48.39
2.0	133.32	0.4001	0.6350	63.00

**Table B-228** Experimental data and current efficiency for Ex. No. 228

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	281.51	0.1109	0.1720	64.52
1.0	259.42	0.2435	0.3439	70.80
2.0	204.39	0.5737	0.6878	83.40
0.5	188.70	0.0678	0.1588	42.71
1.0	173.28	0.1603	0.3175	50.49
2.0	130.99	0.4141	0.6350	65.20

Table B-229 Experimental data and current efficiency for Ex. No. 229

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Exp.(g) Wt. Theo.(g)	
0.5	281.11	0.1133	0.2075	54.61
1.0	257.46	0.2552	0.4151	61.49
2.0	197.48	0.6151	0.8301	74.10
0.5	188.66	0.0680	0.1916	35.51
1.0	172.66	0.1640	0.3832	42.81
2.0	127.32	0.4361	0.7664	56.90

Table B-230 Experimenta	l data and current	efficiency	y for Ex.	No. 230
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Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	280.35	0.1179	0.2075	56.81
1.0	256.07	0.2636	0.4151	63.50
2.0	194.43	0.6334	0.8301	76.30
0.5	188.03	0.0718	0.1916	37.48
1.0	171.45	0.1713	0.3832	44.70
2.0	124.76	0.4514	0.7664	58.90

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	279.63	0.1222	0.2075	58.89
1.0	254.62	0.2723	0.4151	65.60
2.0	191.53	0.6508	0.8301	78.40
0.5	187.32	0.0761	0.1916	39.71
1.0	170.05	0.1797	0.3832	46.89
2.0	121.95	0.4683	0.7664	61.10

 Table B-231 Experimental data and current efficiency for Ex. No. 231

 Table B-232 Experimental data and current efficiency for Ex. No. 232

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	279.98	0.1201	0.2431	49.41
1.0	254.70	0.2718	0.4862	55.90
2.0	188.98	0.6661	0.9725	68.50
0.5	188.03	0.0718	0.2245	32.00
1.0	170.22	0.1787	0.4489	39.80
2.0	119.20	0.4848	0.8978	54.00

 Table B-233 Experimental data and current efficiency for Ex. No. 233

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	279.21	0.1247	0.2431	51.31
1.0	253.08	0.2815	0.4862	57.90
2.0	185.41	0.6875	0.9725	70.70
0.5	187.24	0.0766	0.2245	34.11
1.0	168.80	0.1872	0.4489	41.70
2.0	116.06	0.5036	0.8978	56.10

Table B-234 Experimental data and current efficiency for Ex. No. 234

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	278.32	0.1301	0.2431	53.51
1.0	251.38	0.2917	0.4862	60.00
2.0	182.01	0.7079	0.9725	72.80
0.5	186.46	0.0812	0.2245	36.19
1.0	167.23	0.1966	0.4489	43.80
2.0	113.06	0.5216	0.8978	58.10

Table <b>F</b>	<b>3-235</b> ]	Experimental	data ar	d current	efficiency	y for	Ex. No	. 235
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Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	84.30	0.0942	0.2075	45.39
1.0	64.17	0.2150	0.4151	51.79
2.0	11.73	0.5296	0.8301	63.80
0.5	184.52	0.0929	0.3670	25.31
1.0	160.98	0.2341	0.7340	31.90
2.0	97.24	0.6166	1.4679	42.00
Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
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0.5	83.64	0.0982	0.2075	47.30
1.0	62.71	0.2237	0.4151	53.90
2.0	9.24	0.5446	0.8301	65.60
0.5	183.19	0.1009	0.3670	27.48
1.0	158.65	0.2481	0.7340	33.80
2.0	92.35	0.6459	1.4679	44.00

Table B-236 Experimental data and current efficiency for Ex. No. 236

 Table B-237 Experimental data and current efficiency for Ex. No. 237

Deposition Time (h)	Remaining. Conc.(ppm)	Remaining. Conc.(ppm)Wt. Exp.(g)		C.E. %
0.5	82.88	0.1027	0.2075	49.49
1.0	61.26	0.2324	0.4151	56.00
2.0	6.47	0.5612	0.8301	67.60
0.5	182.11	0.1073	0.3670	29.25
1.0	156.08	0.2635	0.7340	35.90
2.0	87.21	0.6767	1.4679	46.10

 Table B-238 Experimental data and current efficiency for Ex. No. 238

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	83.91	0.0965	0.2431	39.71
1.0	62.40	0.2256	0.4862	46.40
2.0	4.21	0.5747	0.9725	59.10
0.5	183.81	0.0971	0.4299	22.60
1.0	158.73	0.2476	0.8598	28.80
2.0	85.08	0.6895	1.7196	40.10

Table B-239 Experimental data and current efficiency for Ex. No. 239

Deposition Time (h)	on Time Remaining. h) Conc.(ppm) Wt. Exp.(g)		Wt. Theo.(g)	C.E. %
0.5	83.06	0.1016	0.2431	41.81
1.0	60.86	0.2348	0.4862	48.30
2.0	0.81	0.5951	0.9725	61.20
0.5	182.23	0.1066	0.4299	24.80
1.0	155.86	0.2648	0.8598	30.80
2.0	78.77	0.7274	1.7196	42.30

Table B-240 Experimental data and current efficiency for	Ex. No. 240
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Deposition Time (h)	e Remaining. Conc.(ppm) Wt. Exp.(g)		Wt. Theo.(g)	C.E. %
0.5	82.25	0.1065	0.2431	43.81
1.0	59.16	0.2450	0.4862	50.40
2.0	0.00	0.6000	0.9725	61.70
0.5	180.73	0.1156	0.4299	26.89
1.0	152.71	0.2837	0.8598	33.00
2.0	72.75	0.7635	1.7196	44.40

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	83.79	0.0973	0.2787	34.90
1.0	61.17	0.2330	0.5574	41.80
2.0	1.63	0.5902	1.1148	52.95
0.5	182.67	0.1040	0.4928	21.10
1.0	154.99	0.2701	0.9856	27.40
2.0	73.84	0.7570	1.9712	38.40

Table B-241 Experimental data and current efficiency for Ex. No. 241

 Table B-242 Experimental data and current efficiency for Ex. No. 242

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %	
0.5	82.77	0.1034	0.2787	37.09	
1.0	59.31	0.2441	0.5574	43.80	
2.0	0.00	0.6000	1.1148	53.82	
0.5	181.03	0.1138	0.4928	23.10	
1.0	151.87	0.2888	0.9856	29.30	
2.0	66.94	0.7984	1.9712	40.50	

 Table B-243 Experimental data and current efficiency for Ex. No. 243

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	81.79	0.1093	0.2787	39.20
1.0	57.17	0.2570	0.5574	46.10
2.0	0.00	0.6000	1.1148	53.82
0.5	179.22	0.1247	0.4928	25.30
1.0	148.26	0.3104	0.9856	31.50
2.0	60.04	0.8398	1.9712	42.60

Table B-244 Experimental data and current efficiency for Ex. No. 244

Deposition Time (h)	Remaining. Conc.(ppm)	Remaining. Conc.(ppm) Wt. Exp.(g)		С.Е. %
0.5	182.53	0.1048	0.2075	50.51
1.0	160.22	0.2387	0.4151	57.50
2.0	104.95	0.5703	0.8301	68.70
0.5	183.73	0.0976	0.3670	26.60
1.0	158.78	0.2473	0.7340	33.70
2.0	90.88	0.6547	1.4679	44.60

Table B-	-245 Ex	perimental	data	and	current	efficie	ency	for	Ex.	No.	245	
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Deposition Time (h)	on Time Remaining. a) Conc.(ppm) Wt. Exp.(g)		Wt. Theo.(g)	C.E. %
0.5	181.81	0.1091	0.2075	52.59
1.0	158.91	0.2465	0.4151	59.40
2.0	102.18	0.5869	0.8301	70.70
0.5	182.51	0.1049	0.3670	28.60
1.0	156.08	0.2635	0.7340	35.90
2.0	85.75	0.6855	1.4679	46.70

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	181.08	0.1135	0.2075	54.70
1.0	157.39	0.2557	0.4151	61.59
2.0	99.28	0.6043	0.8301	72.80
0.5	181.16	0.1130	0.3670	30.80
1.0	153.52	0.2789	0.7340	38.00
2.0	80.36	0.7178	1.4679	48.90

Table B-246 Experimental data and current efficiency for Ex. No. 246

 Table B-247 Experimental data and current efficiency for Ex. No. 247

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	181.73	0.1096	0.2431	45.09
1.0	158.10	0.2514	0.4862	51.70
2.0	96.60	0.6204	0.9725	63.80
0.5	182.23	0.1066	0.4299	24.80
1.0	155.29	0.2683	0.8598	31.20
2.0	78.48	0.7291	1.7196	42.40

 Table B-248 Experimental data and current efficiency for Ex. No. 248

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	180.96	0.1142	0.2431	46.99
1.0	156.48	0.2611	0.4862	53.70
2.0	93.03	0.6418	0.9725	66.00
0.5	180.73	0.1156	0.4299	26.89
1.0	152.14	0.2872	0.8598	33.40
2.0	72.46	0.7652	1.7196	44.50

Table B-249 Experimental data and current efficiency for Ex. No. 249

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	180.07	0.1196	0.2431	49.19
1.0	154.70	0.2718	0.4862	55.90
2.0	89.63	0.6622	0.9725	68.10
0.5	179.22	0.1247	0.4299	29.00
1.0	148.99	0.3061	0.8598	35.60
2.0	66.16	0.8030	1.7196	46.70

Table B-250 Experimental data and current efficiency for Ex. N	o. 250
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Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	181.51	0.1109	0.2787	39.81
1.0	156.62	0.2603	0.5574	46.70
2.0	89.45	0.6633	1.1148	59.50
0.5	180.86	0.1148	0.4928	23.30
1.0	150.56	0.2966	0.9856	30.10
2.0	64.64	0.8122	1.9712	41.20

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	180.58	0.1165	0.2787	41.81
1.0	154.57	0.2726	0.5574	48.90
2.0	85.92	0.6845	1.1148	61.40
0.5	179.14	0.1252	0.4928	25.40
1.0	147.27	0.3164	0.9856	32.10
2.0	57.74	0.8536	1.9712	43.30

 Table B-251 Experimental data and current efficiency for Ex. No. 251

 Table B-252 Experimental data and current efficiency for Ex. No. 252

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	179.52	0.1229	0.2787	44.09
1.0	152.53	0.2848	0.5574	51.10
2.0	81.83	0.7090	1.1148	63.60
0.5	177.33	0.1360	0.4928	27.60
1.0	143.82	0.3371	0.9856	34.20
2.0	50.84	0.8950	1.9712	45.40

 Table B-253 Experimental data and current efficiency for Ex. No. 253

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	281.22	0.1127	0.2075	54.29
1.0	257.73	0.2536	0.4151	61.10
2.0	196.65	0.6201	0.8301	74.70
0.5	182.32	0.1061	0.3670	28.91
1.0	157.19	0.2569	0.7340	35.00
2.0	82.08	0.7075	1.4679	48.20

Table B-254 Experimental data and current efficiency for Ex. No. 254

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	280.46	0.1172	0.2075	56.49
1.0	256.35	0.2619	0.4151	63.10
2.0	194.02	0.6359	0.8301	76.60
0.5	179.89	0.1207	0.3670	32.88
1.0	154.49	0.2731	0.7340	37.20
2.0	76.94	0.7384	1.4679	50.30

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	279.73	0.1216	0.2075	58.60
1.0	254.83	0.2710	0.4151	65.29
2.0	191.11	0.6533	0.8301	78.70
0.5	179.94	0.1204	0.3670	32.80
1.0	151.93	0.2884	0.7340	39.30
2.0	72.04	0.7678	1.4679	52.30

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	279.78	0.1213	0.2431	49.90
1.0	254.38	0.2737	0.4862	56.29
2.0	188.17	0.6710	0.9725	69.00
0.5	180.44	0.1174	0.4299	27.30
1.0	150.71	0.2957	0.8598	34.40
2.0	70.46	0.7772	1.7196	45.20

Table B-256 Experimental data and current efficiency for Ex. No. 256

 Table B-257 Experimental data and current efficiency for Ex. No. 257

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	278.89	0.1267	0.2431	52.10
1.0	252.84	0.2830	0.4862	58.19
2.0	184.60	0.6924	0.9725	71.20
0.5	178.94	0.1264	0.4299	29.39
1.0	147.70	0.3138	0.8598	36.50
2.0	64.44	0.8134	1.7196	47.30

**Table B-258** Experimental data and current efficiency for Ex. No. 258

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	278.04	0.1318	0.2431	54.20
1.0	251.05	0.2937	0.4862	60.40
2.0	181.36	0.7118	0.9725	73.20
0.5	177.50	0.1350	0.4299	31.40
1.0	144.69	0.3319	0.8598	38.60
2.0	58.42	0.8495	1.7196	49.40

Table B-259 Experimental data and current efficiency for Ex. No. 259

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	278.73	0.1276	0.2787	45.79
1.0	250.86	0.2948	0.5574	52.90
2.0	178.50	0.7290	1.1148	65.39
0.5	179.06	0.1256	0.4928	25.49
1.0	147.43	0.3154	0.9856	32.00
2.0	57.74	0.8536	1.9712	43.30

Table B-260 Expe	erimental data	and current	efficiency	for Ex.	No. 260
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Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	277.75	0.1335	0.2787	47.90
1.0	248.91	0.3065	0.5574	55.00
2.0	174.40	0.7536	1.1148	67.60
0.5	177.50	0.1350	0.4928	27.39
1.0	143.98	0.3361	0.9856	34.10
2.0	51.50	0.8910	1.9712	45.20

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	276.82	0.1391	0.2787	49.90
1.0	247.05	0.3177	0.5574	57.00
2.0	170.50	0.7770	1.1148	69.70
0.5	175.69	0.1459	0.4928	29.60
1.0	140.37	0.3578	0.9856	36.30
2.0	44.27	0.9344	1.9712	47.40

 Table B-261 Experimental data and current efficiency for Ex. No. 261

 Table B-262 Experimental data and current efficiency for Ex. No. 262

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	84.50	0.0930	0.2075	44.81
1.0	64.51	0.2129	0.4151	51.30
2.0	11.17	0.5330	0.8301	64.20
0.5	189.94	0.0604	0.1916	31.50
1.0	174.71	0.1517	0.3832	39.60
2.0	132.81	0.4031	0.7664	52.60
0.5	184.28	0.0943	0.3670	25.70
1.0	158.90	0.2466	0.7340	33.60
2.0	91.24	0.6526	1.4679	44.45

Table B-263 Experimental data and current efficiency for Ex. No. 263

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	83.74	0.0976	0.2075	47.01
1.0	63.06	0.2216	0.4151	53.40
2.0	7.99	0.5521	0.8301	66.50
0.5	189.27	0.0644	0.1916	33.60
1.0	173.37	0.1598	0.3832	41.70
2.0	130.00	0.4200	0.7664	54.80
0.5	183.12	0.1013	0.3670	27.60
1.0	156.45	0.2613	0.7340	35.60
2.0	86.72	0.6797	1.4679	46.30

 Table B-264 Experimental data and current efficiency for Ex. No. 264

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	83.05	0.1017	0.2075	49.00
1.0	61.54	0.2308	0.4151	55.60
2.0	5.09	0.5695	0.8301	68.60
0.5	188.57	0.0686	0.1916	35.79
1.0	172.03	0.1678	0.3832	43.79
2.0	127.45	0.4353	0.7664	56.80
0.5	181.83	0.1090	0.3670	29.71
1.0	153.76	0.2774	0.7340	37.80
2.0	81.34	0.7120	1.4679	48.50

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	83.75	0.0975	0.2431	40.10
1.0	62.16	0.2270	0.4862	46.69
2.0	3.24	0.5806	0.9725	59.70
0.5	189.15	0.0651	0.2245	29.00
1.0	172.92	0.1625	0.4489	36.19
2.0	125.33	0.4480	0.8978	49.90
0.5	182.73	0.1036	0.4299	24.10
1.0	156.29	0.2623	0.8598	30.50
2.0	78.77	0.7274	1.7196	42.30

 Table B-265 Experimental data and current efficiency for Ex. No. 265

Table B-266 Experimental data and current efficiency for Ex. No. 266

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	82.90	0.1026	0.2431	42.20
1.0	60.62	0.2363	0.4862	48.59
2.0	0.00	0.6000	0.9725	61.70
0.5	188.29	0.0703	0.2245	31.30
1.0	171.27	0.1724	0.4489	38.40
2.0	122.04	0.4678	0.8978	52.10
0.5	181.30	0.1122	0.4299	26.10
1.0	153.28	0.2803	0.8598	32.60
2.0	72.18	0.7669	1.7196	44.60

**Table B-267** Experimental data and current efficiency for Ex. No. 267

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	82.09	0.1075	0.2431	44.20
1.0	58.75	0.2475	0.4862	50.90
2.0	0.00	0.6000	0.9725	61.70
0.5	187.47	0.0752	0.2245	33.50
1.0	169.70	0.1818	0.4489	40.50
2.0	118.90	0.4866	0.8978	54.20
0.5	179.80	0.1212	0.4299	28.19
1.0	150.42	0.2975	0.8598	34.60
2.0	65.87	0.8048	1.7196	46.80

Table B-268 Ext	perimental data an	d current efficienc	y for Ex.	No. 268
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Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	83.60	0.0984	0.2787	35.31
1.0	60.61	0.2363	0.5574	42.40
2.0	0.00	0.6000	1.1148	53.82
0.5	188.77	0.0674	0.2573	26.19
1.0	171.61	0.1703	0.5146	33.10

#### Table B-268 Cont.

2.0	119.21	0.4847	1.0292	47.10
0.5	181.36	0.1118	0.4928	22.69
1.0	152.03	0.2878	0.9856	29.20
2.0	66.61	0.8003	1.9712	40.60

 Table B-269 Experimental data and current efficiency for Ex. No. 269

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	82.63	0.1042	0.2787	37.40
1.0	58.75	0.2475	0.5574	44.40
2.0	0.00	0.6000	1.1148	53.82
0.5	187.86	0.0728	0.2573	28.31
1.0	169.98	0.1801	0.5146	35.00
2.0	115.26	0.5084	1.0292	49.40
0.5	179.55	0.1227	0.4928	24.90
1.0	148.58	0.3085	0.9856	31.30
2.0	59.39	0.8437	1.9712	42.80

 Table B-270 Experimental data and current efficiency for Ex. No. 270

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	81.56	0.1106	0.2787	39.70
1.0	56.80	0.2592	0.5574	46.50
2.0	0.00	0.6000	1.1148	53.82
0.5	187.01	0.0779	0.2573	30.29
1.0	168.10	0.1914	0.5146	37.19
2.0	111.66	0.5300	1.0292	51.50
0.5	177.82	0.1331	0.4928	27.00
1.0	145.30	0.3282	0.9856	33.30
2.0	52.49	0.8851	1.9712	44.90

Table B-271 Experimental data and current efficiency for Ex. No. 271

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	182.46	0.1052	0.2075	50.71
1.0	159.95	0.2403	0.4151	57.89
2.0	104.53	0.5728	0.8301	69.00
0.5	189.33	0.0640	0.1916	33.41
1.0	174.45	0.1533	0.3832	40.00
2.0	130.00	0.4200	0.7664	54.80
0.5	182.94	0.1024	0.3670	27.89
1.0	157.31	0.2561	0.7340	34.90
2.0	85.01	0.6899	1.4679	47.00

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	181.70	0.1098	0.2075	52.91
1.0	158.56	0.2486	0.4151	59.90
2.0	101.63	0.5902	0.8301	71.10
0.5	188.70	0.0678	0.1916	35.39
1.0	173.11	0.1613	0.3832	42.10
2.0	127.57	0.4346	0.7664	56.70
0.5	181.77	0.1094	0.3670	29.81
1.0	154.62	0.2723	0.7340	37.10
2.0	79.87	0.7208	1.4679	49.10

 Table B-272 Experimental data and current efficiency for Ex. No. 272

 Table B-273 Experimental data and current efficiency for Ex. No. 273

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	180.98	0.1141	0.2075	54.99
1.0	157.11	0.2573	0.4151	62.00
2.0	98.99	0.6061	0.8301	73.01
0.5	187.99	0.0721	0.1916	37.61
1.0	171.77	0.1694	0.3832	44.20
2.0	125.02	0.4499	0.7664	58.70
0.5	180.49	0.1171	0.3670	31.90
1.0	151.93	0.2884	0.7340	39.30
2.0	74.50	0.7530	1.4679	51.30

Table B-274 Experimental data and current efficiency for Ex. No. 274

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	181.36	0.1118	0.2431	46.00
1.0	157.70	0.2538	0.4862	52.20
2.0	96.11	0.6233	0.9725	64.10
0.5	188.48	0.0691	0.2245	30.80
1.0	171.27	0.1724	0.4489	38.40
2.0	120.84	0.4750	0.8978	52.90
0.5	181.16	0.1130	0.4299	26.29
1.0	152.42	0.2855	0.8598	33.20
2.0	69.31	0.7841	1.7196	45.60

Table B-275 Experimental data and current efficiency for Ex. No. 275

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	180.51	0.1169	0.2431	48.10
1.0	156.08	0.2635	0.4862	54.20
2.0	93.03	0.6418	0.9725	66.00
0.5	187.77	0.0734	0.2245	32.69

Table D-275 Cont.						
1.0	169.70	0.1818	0.4489	40.50		
2.0	117.85	0.4929	0.8978	54.90		
0.5	179.65	0.1221	0.4299	28.40		
1.0	149.70	0.3018	0.8598	35.10		
2.0	63.60	0.8184	1.7196	47.59		

## Table B-275 Cont.

 Table A-276 Experimental data and current efficiency for Ex. No. 276

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	179.66	0.1220	0.2431	50.20
1.0	154.30	0.2742	0.4862	56.39
2.0	89.46	0.6632	0.9725	68.20
0.5	187.02	0.0779	0.2245	34.70
1.0	168.13	0.1912	0.4489	42.60
2.0	114.56	0.5126	0.8978	57.10
0.5	178.08	0.1315	0.4299	30.59
1.0	146.84	0.3190	0.8598	37.10
2.0	57.56	0.8546	1.7196	49.70

Table B-277 Experimental data and current efficiency for Ex. No. 277

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	181.23	0.1126	0.2787	40.41
1.0	156.15	0.2631	0.5574	47.20
2.0	88.89	0.6667	1.1148	59.80
0.5	187.74	0.0736	0.2573	28.59
1.0	168.61	0.1883	0.5146	36.60
2.0	111.83	0.5290	1.0292	51.40
0.5	179.63	0.1222	0.4928	24.80
1.0	148.26	0.3104	0.9856	31.50
2.0	55.77	0.8654	1.9712	43.90

Table B-278 Experimental data and current efficiency for Ex. No. 278

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	180.21	0.1187	0.2787	42.61
1.0	154.29	0.2743	0.5574	49.20
2.0	85.36	0.6878	1.1148	61.70
0.5	186.84	0.0790	0.2573	30.69
1.0	166.98	0.1981	0.5146	38.50
2.0	108.23	0.5506	1.0292	53.50
0.5	177.91	0.1325	0.4928	26.89
1.0	144.64	0.3322	0.9856	33.70
2.0	49.53	0.9028	1.9712	45.80

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	179.28	0.1243	0.2787	44.61
1.0	152.34	0.2860	0.5574	51.30
2.0	81.46	0.7112	1.1148	63.80
0.5	185.89	0.0847	0.2573	32.90
1.0	165.09	0.2095	0.5146	40.70
2.0	104.80	0.5712	1.0292	55.50
0.5	176.35	0.1419	0.4928	28.79
1.0	141.19	0.3529	0.9856	35.80
2.0	42.30	0.9462	1.9712	48.00

 Table B-279 Experimental data and current efficiency for Ex. No. 279

 Table B-280 Experimental data and current efficiency for Ex. No. 280

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	280.94	0.1144	0.2075	55.10
1.0	257.18	0.2569	0.4151	61.90
2.0	195.68	0.6259	0.8301	75.40
0.5	188.22	0.0707	0.1916	36.89
1.0	172.09	0.1675	0.3832	43.70
2.0	125.79	0.4453	0.7664	58.10
0.5	181.71	0.1097	0.3670	29.90
1.0	154.74	0.2716	0.7340	37.00
2.0	76.45	0.7413	1.4679	50.50

 Table B-281 Experimental data and current efficiency for Ex. No. 281

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	280.22	0.1187	0.2075	57.19
1.0	255.86	0.2648	0.4151	63.81
2.0	192.77	0.6434	0.8301	77.50
0.5	187.55	0.0747	0.1916	38.99
1.0	170.68	0.1759	0.3832	45.91
2.0	123.23	0.4606	0.7664	60.10
0.5	180.49	0.1171	0.3670	31.90
1.0	152.17	0.2870	0.7340	39.10
2.0	71.07	0.7736	1.4679	52.70

**Table B-282** Experimental data and current efficiency for Ex. No. 282

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	279.52	0.1229	0.2075	59.21
1.0	254.34	0.2740	0.4151	66.00
2.0	189.73	0.6616	0.8301	79.70
0.5	186.81	0.0791	0.1916	41.30

Table D-202 Cont.					
1.0	169.41	0.1835	0.3832	47.90	
2.0	120.42	0.4775	0.7664	62.30	
0.5	179.14	0.1252	0.3670	34.11	
1.0	149.48	0.3031	0.7340	41.30	
2.0	65.93	0.8044	1.4679	54.80	

#### Table B-282 Cont.

 Table B-283 Experimental data and current efficiency for Ex. No. 283

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	279.62	0.1223	0.2431	50.30
1.0	253.90	0.2766	0.4862	56.89
2.0	187.20	0.6768	0.9725	69.60
0.5	187.51	0.0749	0.2245	33.39
1.0	169.10	0.1854	0.4489	41.30
2.0	116.36	0.5018	0.8978	55.90
0.5	179.44	0.1234	0.4299	28.70
1.0	148.84	0.3070	0.8598	35.70
2.0	66.45	0.8013	1.7196	46.60

Table B-284 Experimental data and current efficiency for Ex. No. 284

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	278.81	0.1271	0.2431	52.30
1.0	252.19	0.2869	0.4862	59.00
2.0	184.12	0.6953	0.9725	71.50
0.5	186.68	0.0799	0.2245	35.61
1.0	167.53	0.1948	0.4489	43.40
2.0	113.51	0.5189	0.8978	57.80
0.5	178.08	0.1315	0.4299	30.59
1.0	145.69	0.3259	0.8598	37.90
2.0	60.43	0.8374	1.7196	48.70

Table B-285 Experimental data and current efficiency for Ex. No. 285

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	277.88	0.1327	0.2431	54.59
1.0	250.49	0.2971	0.4862	61.09
2.0	180.55	0.7167	0.9725	73.70
0.5	185.90	0.0846	0.2245	37.69
1.0	165.81	0.2051	0.4489	45.70
2.0	110.37	0.5378	0.8978	59.90
0.5	176.57	0.1406	0.4299	32.70
1.0	142.68	0.3439	0.8598	40.00
2.0	54.70	0.8718	1.7196	50.70

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	278.49	0.1291	0.2787	46.31
1.0	250.21	0.2987	0.5574	53.60
2.0	177.19	0.7369	1.1148	66.10
0.5	187.05	0.0777	0.2573	30.20
1.0	167.24	0.1966	0.5146	38.20
2.0	110.12	0.5393	1.0292	52.40
0.5	178.07	0.1316	0.4928	26.70
1.0	145.30	0.3282	0.9856	33.30
2.0	50.52	0.8969	1.9712	45.50

 Table B-286 Experimental data and current efficiency for Ex. No. 286

 Table B-287 Experimental data and current efficiency for Ex. No. 287

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	277.52	0.1349	0.2787	48.40
1.0	248.26	0.3104	0.5574	55.70
2.0	173.47	0.7592	1.1148	68.10
0.5	186.06	0.0836	0.2573	32.51
1.0	165.35	0.2079	0.5146	40.40
2.0	106.51	0.5609	1.0292	54.50
0.5	176.26	0.1424	0.4928	28.90
1.0	142.01	0.3479	0.9856	35.30
2.0	42.96	0.9422	1.9712	47.80

Table B-288 Experimental data and current efficiency for Ex. No. 288

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	276.54	0.1408	0.2787	50.51
1.0	246.40	0.3216	0.5574	57.70
2.0	169.39	0.7837	1.1148	70.30
0.5	185.16	0.0890	0.2573	34.61
1.0	163.55	0.2187	0.5146	42.50
2.0	102.91	0.5825	1.0292	56.60
0.5	174.62	0.1523	0.4928	30.90
1.0	138.40	0.3696	0.9856	37.50
2.0	35.73	0.9856	1.9712	50.00

Table B-289 Experimental data and current efficiency for Ex. No. 289

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	196.06	0.0236	0.0534	44.30
1.0	191.46	0.0512	0.1067	48.01
2.0	179.69	0.1219	0.2135	57.09

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.96	0.0242	0.0534	45.42
1.0	191.22	0.0527	0.1067	49.36
2.0	179.19	0.1249	0.2135	58.49

 Table B-290 Experimental data and current efficiency for Ex. No. 290

 Table B-291 Experimental data and current efficiency for Ex. No. 291

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	195.82	0.0251	0.0534	47.00
1.0	190.88	0.0547	0.1067	51.27
2.0	178.60	0.1284	0.2135	60.15

Table B-292 Experimental data and current efficiency for Ex. No. 292

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	189.67	0.0620	0.2245	27.61
1.0	174.41	0.1535	0.4489	34.20
2.0	131.29	0.4123	0.8978	45.92

Table B-293 Experimental data and current efficiency for Ex. No. 293

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	188.90	0.0666	0.2245	29.67
1.0	173.02	0.1619	0.4489	36.06
2.0	128.43	0.4294	0.8978	47.83

Table B-294 Experimental data and current efficiency for Ex. No. 294

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	188.08	0.0715	0.2245	31.86
1.0	171.30	0.1722	0.4489	38.36
2.0	125.45	0.4473	0.8978	49.82

 Table B-295 Experimental data and current efficiency for Ex. No. 295

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	С.Е. %
0.5	184.90	0.0906	0.5505	16.46
1.0	162.28	0.2263	1.1010	20.56
2.0	98.45	0.6093	2.2019	27.67

 Table B-296 Experimental data and current efficiency for Ex. No. 296

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	183.74	0.0976	0.5505	17.72
1.0	159.70	0.2418	1.1010	21.96
2.0	93.76	0.6374	2.2019	28.95

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	182.53	0.1048	0.5505	19.04
1.0	157.01	0.2579	1.1010	23.43
2.0	88.88	0.6667	2.2019	30.28

 Table B-297 Experimental data and current efficiency for Ex. No. 297

 Table B-298 Experimental data and current efficiency for Ex. No. 298

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	183.99	0.0961	0.2579	37.24
1.0	164.41	0.2135	0.5159	41.39
2.0	114.63	0.5122	1.0318	49.65
0.5	190.45	0.0573	0.2381	24.06
1.0	176.37	0.1418	0.4763	29.77
2.0	137.31	0.3761	0.9525	39.49

Table B-299 Experimental data and current efficiency for Ex. No. 299

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	183.40	0.0996	0.2579	38.61
1.0	163.25	0.2205	0.5159	42.74
2.0	112.48	0.5251	1.0318	50.90
0.5	189.91	0.0605	0.2381	25.42
1.0	175.39	0.1477	0.4763	31.00
2.0	135.03	0.3898	0.9525	40.92

Table B-300 Experimental data and current efficiency for Ex. No. 300

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	182.87	0.1028	0.2579	39.85
1.0	162.00	0.2280	0.5159	44.20
2.0	109.96	0.5402	1.0318	52.36
0.5	189.32	0.0641	0.2381	26.91
1.0	174.30	0.1542	0.4763	32.38
2.0	132.50	0.4050	0.9525	42.52

 Table B-301 Experimental data and current efficiency for Ex. No. 301

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	182.51	0.1049	0.3113	33.71
1.0	160.18	0.2389	0.6226	38.37
2.0	104.90	0.5706	1.2452	45.82
0.5	183.68	0.0979	0.5505	17.79
1.0	158.71	0.2477	1.1010	22.50
2.0	90.83	0.6550	2.2019	29.75

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	181.76	0.1094	0.3113	35.16
1.0	158.90	0.2466	0.6226	39.61
2.0	102.12	0.5873	1.2452	47.16
0.5	182.47	0.1052	0.5505	19.11
1.0	156.00	0.2640	1.1010	23.98
2.0	85.70	0.6858	2.2019	31.15

 Table B-302 Experimental data and current efficiency for Ex. No. 302

Table B-303 Experimental data and current efficiency for Ex. No. 303

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	181.02	0.1139	0.3113	36.58
1.0	157.30	0.2562	0.6226	41.15
2.0	99.22	0.6047	1.2452	48.56
0.5	181.11	0.1133	0.5505	20.59
1.0	153.46	0.2792	1.1010	25.36
2.0	80.33	0.7180	2.2019	32.61

Table B-304 Experimental data and current efficiency for Ex. No. 304

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	182.41	0.1055	0.3113	33.90
1.0	159.90	0.2406	0.6226	38.64
2.0	104.47	0.5732	1.2452	46.03
0.5	189.29	0.0643	0.2874	22.36
1.0	174.44	0.1534	0.5748	26.68
2.0	129.29	0.4243	1.1496	36.90
0.5	182.88	0.1027	0.5505	18.66
1.0	157.29	0.2563	1.1010	23.28
2.0	85.00	0.6900	2.2019	31.34

Table B-305 Experimental data and current efficiency for Ex. No. 305

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	181.65	0.1101	0.3113	35.37
1.0	158.50	0.2490	0.6226	39.99
2.0	101.57	0.5906	1.2452	47.43
0.5	188.66	0.0680	0.2874	23.67
1.0	173.10	0.1614	0.5748	28.08
2.0	127.52	0.4349	1.1496	37.83
0.5	181.75	0.1095	0.5505	19.89
1.0	154.58	0.2725	1.1010	24.75
2.0	79.81	0.7211	2.2019	32.75

Deposition Time (h)	Remaining. Conc.(ppm)	Wt. Exp.(g)	Wt. Theo.(g)	C.E. %
0.5	180.93	0.1144	0.3113	36.76
1.0	157.07	0.2576	0.6226	41.37
2.0	98.96	0.6062	1.2452	48.69
0.5	187.92	0.0725	0.2874	25.22
1.0	171.75	0.1695	0.5748	29.49
2.0	125.00	0.4500	1.1496	39.14
0.5	180.41	0.1175	0.5505	21.35
1.0	151.87	0.2888	1.1010	26.23
2.0	74.48	0.7531	2.2019	34.20

 Table B-306 Experimental data and current efficiency for Ex. No. 306

### الخلاصة

يمتل هذا البحث الدراسة المختبرية لمفاعل كهروكيمياوي ذو حشوة ثابتة يكون فيه الجريان السائل الألكتروليتي عاموديا على اتجاه التيار.

تم إنجاز التجارب في نوعيتين من الخلايا الكهروكيمياوية (١) خلية ذات نظام لاجرياني (٢) خلية ذات نظام جرياني، باستخدام قطب مسامي ذو موصلية عالية لمشبك من سبيكة البراص الذي يتكون من (٥٠×٥٠ فتحة في الإنج المربع)، لتصميم وبناء مفاعل كهروكيمياوي ذو الجريان بإتجاه واحد (الحجرة الواحدة) للبحث في إزالة وإستعادة الأيونات المعدنية للنحاس <sup>2+</sup>، والنيكل الن<sup>+2</sup>، والكادميوم <sup>2+</sup>Cd، وتوزيع التركيز مع وقت الترسيب ، وكذلك حساب كفاءة التيار لإزالة المعادن.

في خلية ذات نظام لاجرياني التفاعل الكهروكيمياوي الذي تم تفاعل إختزال (ترسيب) لآيون النحاس فقط الى معدن النحاس على مشبك مفرد ومزدوج وثلاثي لمختلف التيارات الكلية المسلطة، ووقت الترسيب (٥,٠، ١,٠، و ١,٥ ساعة) بوجود حامض الكبريتيك بتركيز ٥,٠ مولاري كمحلول موصل وبدرجة حرارة المحلول ٤٠ ثم لدراسة تأثير تلك المتغيرات على إزالة الآيونات المعدنية وكفاءة التيار.

أما في الخلية ذات النظام الجرياني التفاعل الكهروكيمياوي الذي تم تفاعل إخترال (ترسيب) لآيون النحاس والنيكل والكادميوم الى معادنها على مشبك مفرد ومزدو ج وثلاثي لمختلف التيارات الكلية المسلطة، سرعة الجريان (١٠٠، ٢٠٠، و ٣٠٠ لتر/ساعة)، ونسبة التركيز نحاس :نيكل، نحاس:كادميوم، نحاس:نيكل:كادميوم للترسيب المشترك، ووقت سحب النموذج (٥,٠، ٠,٥، و ٢,٠ ساعة) لمشبك مفرد (٥,٠، ١,٠ و ٢,٠ ساعة) لمشبك مزدوج وثلاثي بوجود حامض الكبريتيك بتركيز ٥,٠ مولاري كمحلول موصل وبدرجة حرارة المحلول ٤٠ °م.

قدمت نتائج ترسيب النحاس والنيكل والكادميوم الى معادنها بدلالة كفاءة التيار . لقد أثبتت النتائج أنه بزيادة التيار الكلي المسلط للخلية الألكتروليتية يؤدي الى نقصان في قيم كفاءة التيار لكن لآيون النحاس هنالك زيادة بكفاءة التيار بسبب عدم تحرر غاز الهيدروجين. كذلك إن زيادة سرعة الجريان تؤدي الى زيادة في إزالة الايونات المعدنية وبالتالي زيادة بكفاءة التيار . كذلك وجد أن زيادة وقت الترسيب يؤدي الى زيادة في الإزالة وكفاءة التيار .

أن وجود آيون النحاس مع آيونات النيكل والكادميوم للترسيب المشترك يؤدي الى زيادة كفاءة التيار للآيونات المعدنية . وكذلك عند زيادة تركيز آيون النحاس (٢٠٠، ٢٠٠، و ٣٠٠ جزء من المليون) يؤدي الى زيادة نسبية لكفاءة التيار للآيونات المعدنية . بإلاضافة الى ذلك ، وجد إن إضافة آيون النيكل للترسيب المشترك سبب زيادة قليلة في كفاءة التيار لآيون الكادميوم.

استخدم في هذا البحث ثلاثة طبقات من مشبك البراص لإيجاد افضل سمك فعال للحشوة الثابتة. تظهر النتائج أن كفاءة التيار تزداد بزيادة عدد طبقات المشبك المعدني لكن فقط لطبقتين من المشبك المعدني وكفاءة التيار تقل بإستخدام مشبك ثالث بسبب رداءة اختراق التيار الكهربائي وكذلك إن تحلل الآيونات المعدنية هو أكبر من ترسيبها.

لقد تم إستخدام تقنية الإمتصاص الذري بواسطة التصوير الطيفي ( Atomic Absorption لقد تم إستخدام تقنية الإمتصاص الذري بواسطة المعدنية المعدنية المختلف آوقات التوسيب.

# شكر وتقدير

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كما وأشكر كافه منتسبين قسم الهندسة الكيمياوي لمساندتهم لي وفي الأخص د.سرمد طالب نجم في إنجاز هذا البحث.

حيدر إسماعيل إبراهيم الكروي

# الإزالة الكهروكيمياوية لآيونات المواد الثقيلة من المخلفات الصناعية المقلدة

_01271	ربيع الأول
۲۰۱۰	شباط