BENCH-SCALE RECOVERY OF LEAD USING AN ELECTROMEMBRANE/CHELATION PROCESS

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FOREWORD

Todays's rapidly developing and changing technologies and industrial products and practices frequently carry with them the increased generation of materials that, if improperly dealt with, can threaten both public health and the environment. The U.S. Environmental Protection Agency is charged by Congress with protecting the Nation's land, air, and water resources. Under a mandate of national environmental laws, the agency strives to formulate and implement actions leading to a compatible balance between human activities and the ability of natural systems to support and nurture life. These laws direct the EPA to perform research to define our environmental problems, measure the impacts, and search for solutions.

The Risk Reduction Engineering Laboratory is responsible for planning, implementing, and managing research, development, and demonstration programs to provide an authoritative, defensible engineering basis in support of the policies, programs, and regulations of the EPA with respect to drinking water, wastewater, pesticides, toxic substances, solid and hazardous wastes, and Superfund-related activities. This publication is one of the products of that research and provides a vital communication link between the researcher and the user community.

The remediation of lead compounds from contaminated Superfund or RCRA corrective action sites by reaction with chelating agents may be rendered more cost-effective by an innovative electromembrane/chelation process whereby the lead is separated and the spent chelating agent is regenerated and reused. The data in this report will also be of use to researchers and others with interest in the electrodeposition of lead from its chelated form.

E. Timothy Oppelt, Director
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ABSTRACT

This report presents the results of a bench-scale treatability test to investigate key process parameters influencing an innovative chelation-electrodeposition process for recovery of metals from contaminated soils. IT conducted a series of electro-membrane tests to examine the effects of membranes, chelating agents, electrodes, current density, iron, and lead concentration on lead recovery. The tests were conducted with a synthetic lead solution made of chelating agent and various lead species. In this study, di-sodium ethylenediaminetetraacetic acid (EDTA), tetra-sodium EDTA, and diethylenetriaminepentaacetic acid (DTPA) were used as chelating agents because of the strong lead-chelate compounds that are formed and because of the prevalence with which they are used for soil washing. Lead species used in this study included lead sulfate and basic lead carbonate. This report summarizes the findings of the experimental work that was conducted from July to August 1993.

Results of this study showed that the tests using di-sodium and tetra-sodium EDTA under the same conditions produced approximately the same lead recovery. Regeneration of the di-sodium EDTA, tetra-sodium EDTA, and DTPA solutions was proven feasible because similar lead removals were observed in the tests conducted with fresh solutions. A comparison of the data obtained in the tests performed using initial target lead concentrations of 0.8 and 4 percent showed that a higher percentage of lead was recovered in the 0.8 percent lead solution test, but the amount of lead recovered was greater in the 4 percent lead solution test. Based on data from the tests using the DuPont Nafion® and Ionics membranes, it appeared that the lead removal rates were approximately the same for both membranes. Tests conducted with lead and cadmium electrodes showed that there was no significant increase in lead recovery using the cadmium electrodes in the tests with DTPA solutions.

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SECTION 1

INTRODUCTION

1.1 BACKGROUND

Numerous Superfund sites throughout the United States are contaminated with toxic metals. Battery reclamation, lead smelting, and lead-based paint manufacturing are examples of processes that could result in lead-contaminated soils. Metals, unlike many hazardous organic constituents, cannot be degraded or readily detoxified. Toxic metals represent a long-term threat in the soil environment. The cleanup of metal-contaminated sites has traditionally involved excavation of the wastes and contaminated soils with subsequent disposal at an off-site, RCRA-approved landfill, in accordance with hazardous waste regulations. This approach is expensive because of the special precautions (e.g., double liners) required to prevent leaching of toxic metals from the landfills. In addition to increasing costs and dangers to public safety from large-scale transportation of wastes, long-term environmental liability is also a concern associated with the landfilling approach. Thus, there is great incentive for the development of alternative methods for cleanup of contaminated sites.

Soil characterizations performed on several metal-contaminated soils at battery reclamation sites have shown that the predominant lead species are lead sulfate, lead carbonate, lead dioxide, and elemental lead. Table 1-1 presents metal-contamination concentrations of several battery reclamation site soils. The average lead concentration in these soils is approximately 4 percent. Calcium and iron are also found in appreciable quantities in these soils. Soil screening tests performed on several metal-contaminated soils by soil washing showed that a majority of the metals are adsorbed on the fine soil fraction (less than 250 μm).[1]

TABLE 1-1. TYPICAL METAL CONTAMINATION LEVEL OF BATTERY RECLAMATION SITE SOILS (REFERENCE 1)
(μg/g)

Metal	Old Man's Township	C&R	Shuylkill	Gould	Average
Aluminum	1665	4700	3025	9930	4830.00
Arsenic	92.8	34	5.8	82.4	53.75
Barium	136	67.2	20.3	81.5	76.25
Cadmium	8.5	4.7	1.86	17.6	8.17
Calcium	1020	57,900	43,400	6910	27,307.50
Chromium	33.4	8.9	10.2	18.2	17.68
Copper	771	27.4	6.07	350	288.62
Iron	22,850	5795	1010	32,150	15,451.25
Lead	57,150	75,850	3230	27,150	40,845.00
Magnesium	368	301	301	3205	1043.75
Manganese	114	7.18	7.18	317	111.34
Mercury	0.2	0.2	0.2	3.93	1.13
Nickel	164	5.185	2.235	27.9	49.83
Potassium	169	538	155	516.5	344.63
Silver	1.81	1.2	1	0.5	1.13
Sodium	769	81.45	1080	490	605.11
Vanadium	37.6	11	8.7	42.9	25.05
Zinc	1520	23.8	13	278	458.70

In 1986, PEI Associates conducted a study for the National Science Foundation using an electromembrane reactor (EMR) process to recover lead from an ethylenediamine-tetraacetic acid (EDTA)-lead chelate solution. The bench-scale tests were performed with actual chelate generated from lead-contaminated soils at a battery reclamation site. The PEI study examined the effect of system variables such as electrode potential, current density, pH, current efficiency, and chelate concentration. The chelate solution was adjusted to pH levels of 4, 8, and 11 with sulfuric acid or sodium hydroxide. The amount of lead in solution was varied from 0.2, 1, and 3 percent by weight to determine the effect of lead concentration on plating efficiency.[2]

The purpose of this Work Assignment was to perform a follow-on bench-scale treatability study to the 1986 PEI study by examining the effects of membranes, chelating agents, electrodes, current density, iron levels, and lead concentrations on lead recovery. In this study, a synthetic lead-chelate solution was tested rather than a lead-contaminated soil because soil chelation has been previously studied. The composition of the synthetic lead-chelate solution was similar to one that would be obtained after chelation of soils from typical battery reclamation sites. The primary focus of the bench-scale electromembrane tests performed in this project was on lead recovery. Results of this study may be used to define the parameters that recover the maximum amount of lead from a lead-chelate solution. The lead-chelate solution used in this study represents the amount of lead that can be extracted by a soil washing process.

1.2 REPORT ORGANIZATION

Section 2 of this report presents the experimental design and procedures used to perform the bench-scale electromembrane experiments. Section 3 presents the results of the chelation tests and the electromembrane experiments. Data from the experiments are summarized in this section. Section 4 contains the quality assurance data collected during the study. Section 5 contains the conclusions of this study and recommendations for future study.

SECTION 2

EXPERIMENTAL DESIGN

The purpose of this project was to perform a bench-scale treatability study using an electromembrane reactor to examine the effects of membranes, chelating agents, electrodes, and lead concentrations on lead recovery and chelant regeneration. The experimental work was conducted at the U.S. EPA Test and Evaluation (T&E) Facility in Cincinnati, Ohio. Although soil washing is an important step in the recovery of lead from soil, the main focus of this project was to evaluate the capabilities of the electromembrane process to recover lead and regenerate the chelating agent. Soil washing with chelating agents is well documented in several treatability studies and field applications. These studies have proven that soil washing with chelating agents is a viable method for removing lead from soil. Both di- and tetra-sodium EDTA and diethylenetriamine pentaacetic acid (DTPA) were selected for study because of their relatively high lead stability constants and their previous use in treatability and field studies.

2.1 PROCEDURE

The design of the bench-scale electromembrane reactor (EMR) experiments was derived from the 1986 PEI study and previous studies conducted using chelating agents for soil washing. The bench-scale study conducted by IT examined the ability to recover lead and regenerate the chelating agent using two types of cation-exchange membranes, two chelating agents, two current densities, two types of electrodes, and two lead concentrations in an electromembrane process that uses a cation-transfer membrane to separate the anode and cathode chambers in an electrolytic cell.

The goal of the bench-scale study was to recover the lead on the cathode, while at the same time regenerating the chelating agent in its sodium salt form in the

cathode chamber. The sodium form of the chelating agent was also used for preparing the synthetic lead-chelate solution. In the electromembrane reactor, a sodium salt solution was initially placed in the anode chamber. The sodium ions are transported across the cation-exchange membrane during the electrolytic process to regenerate the sodium salt form of the chelating agent as the lead is plated out.

Figure 2-1 presents a comparison of metal-chelate stability constants for complexes with tetra-sodium EDTA, DTPA, and nitrilotriacetic acid (NTA). Figure 2-1 shows that the highest lead-chelate stability constant for both tetra-sodium EDTA and DTPA occurs at a pH of approximately 9. The optimum lead-chelate stability constant occurs at a pH of approximately 5 for di-sodium EDTA. The cathode chamber was filled with 4 liters of lead-chelate solution adjusted to the experimental pH with sodium hydroxide or sulfuric acid. A 5 percent sodium carbonate solution (Na_2CO_3) was placed in the anode chamber to provide sodium to replenish the sodium-chelate. A pH meter was used to measure the solution pH in both the anode and cathode chambers. A stoichiometric solution of 2 moles of sodium per mole of lead plated is required to regenerate the sodium salt form of the chelating agent. In the bench-scale experiments, twice the stoichiometric quantity of sodium carbonate required was placed in the anode chamber to prevent depletion of sodium ions. The 5 percent by weight sodium carbonate solution in the anode chamber provides enough sodium ions to carry the current across the membrane.

The electrodes were weighed and placed in the anode and cathode chambers approximately one-inch from the membrane. Current densities were adjusted to 15 or 25 ma/cm^2 on the power supply unit, which corresponds to approximately 4.7 or 8.9 amps, respectively. Experiments using the electromembrane reactor were conducted for a total period of 3 hours to 5 hours. Samples of the solutions in the cathode and anode chambers were taken at 30-minute intervals to determine the quantity of lead plated onto the cathode and the depletion of sodium ions in the anode chamber. After the third hour of the reaction, however, the samples were taken at 1-hour intervals. This sampling schedule provided an indication regarding the optimal time required for plating out the lead.

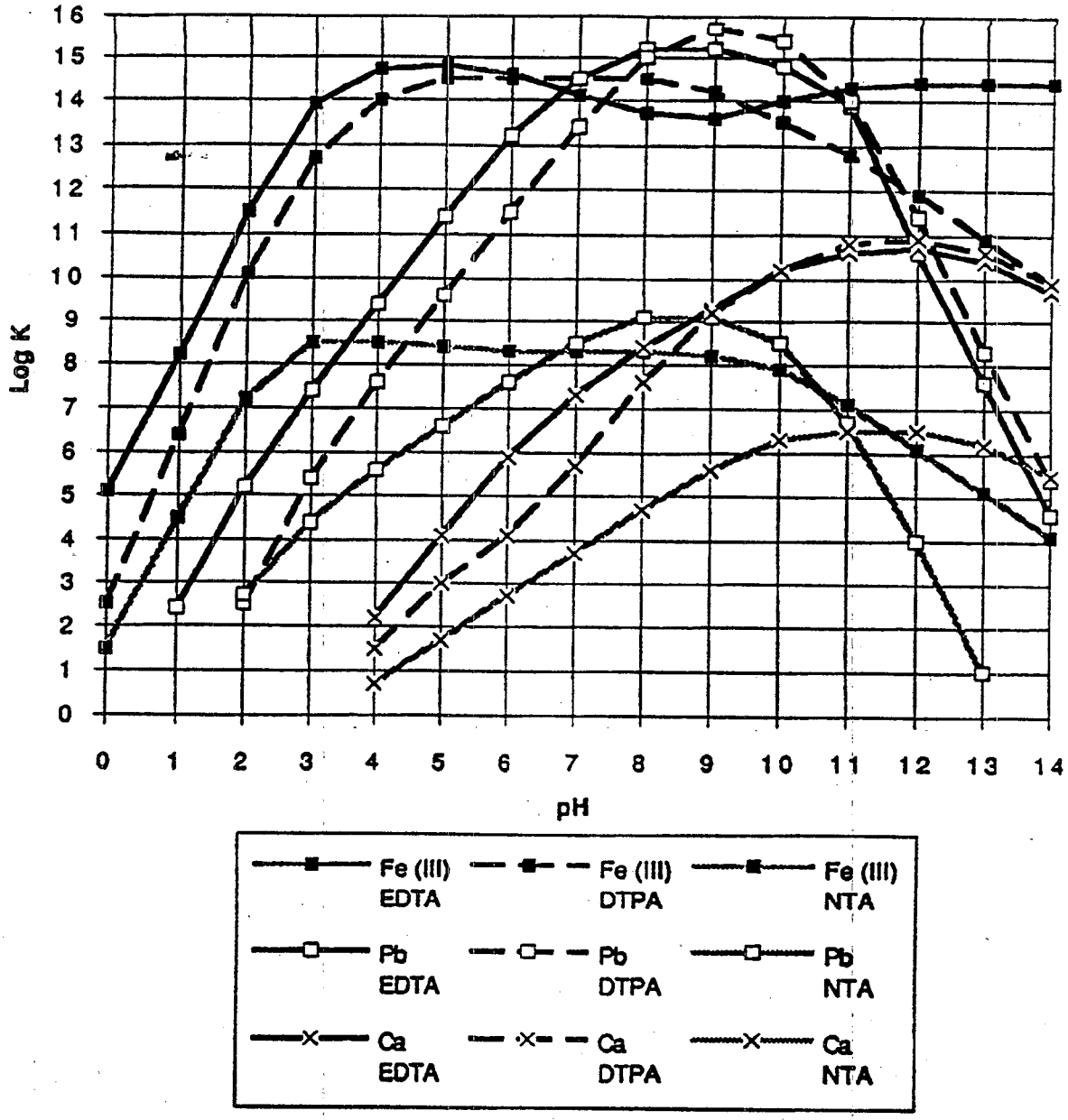


Figure 2-1. Comparison of stability constants for metal-chelate complexes with EDTA, DTPA, and NTA.[3]

2.2 EXPERIMENTAL APPARATUS

The reactor was constructed from a commercial 10-gallon aquarium with 1/4-inch-thick plexiglass. It was divided into two chambers by a plexiglass frame that acted as a support for the cation-exchange membrane. Each chamber was 2 inches long, 12 inches high, and 10 inches deep. A 7-by-7-inch membrane was mounted inside of a frame with gasketing materials and nylon screws and wing nuts. Figure 2-2 presents a diagram of the electromembrane reactor.

Two types of membranes were used in this study: an Ionics 61AZL386 membrane and a DuPont Nafion® membrane. The Ionics membrane is a modacrylic fiber-backed cation-exchange membrane with a specific weight of 14 mg/cm^2, a thickness of 0.6 mm, a burst strength of 8 kg/cm^2, and a 2.7 meq/dry gram resin capacity[4]. The Nafion® membrane is a perfluorosulfonic acid cation-exchange membrane that is reinforced with Teflon, has a weight of 6.3 g/dm,^{2} and is 0.43 mm thick[5]. Both membranes have low electrical resistance, high permselectivity, high burst strength, long-term resistance to aqueous acid, alkaline, and mild oxidizing solutions, and are able to withstand harsh chemical and physical treatment.

One set of electrodes used in the electromembrane tests was made from lead sheet with approximate dimensions of 7 by 10 inches. In a second set of tests, cadmium electrodes were used with the same dimensions as the lead electrodes. Each electrode was supported across the top of the aquarium, approximately 1 inch from the membrane surfaces. The electrodes were wired and connected to a DC power supply with the capabilities for controlling amperage and measuring both current and voltage. The solutions in both the anode and cathode chambers were mixed using magnetic stirrers to create turbulence for enhanced mass transfer. Figure 2-3 presents a photograph of the electromembrane apparatus used to perform the bench-scale electromembrane tests.

2.3 EXPERIMENTAL DESIGN

The type of chelating agent, type of membrane, current density, lead concentration, and reaction time were varied to examine the effects of these parameters on lead recovery. Table 2-1 presents the experimental matrix for the bench-scale electro-

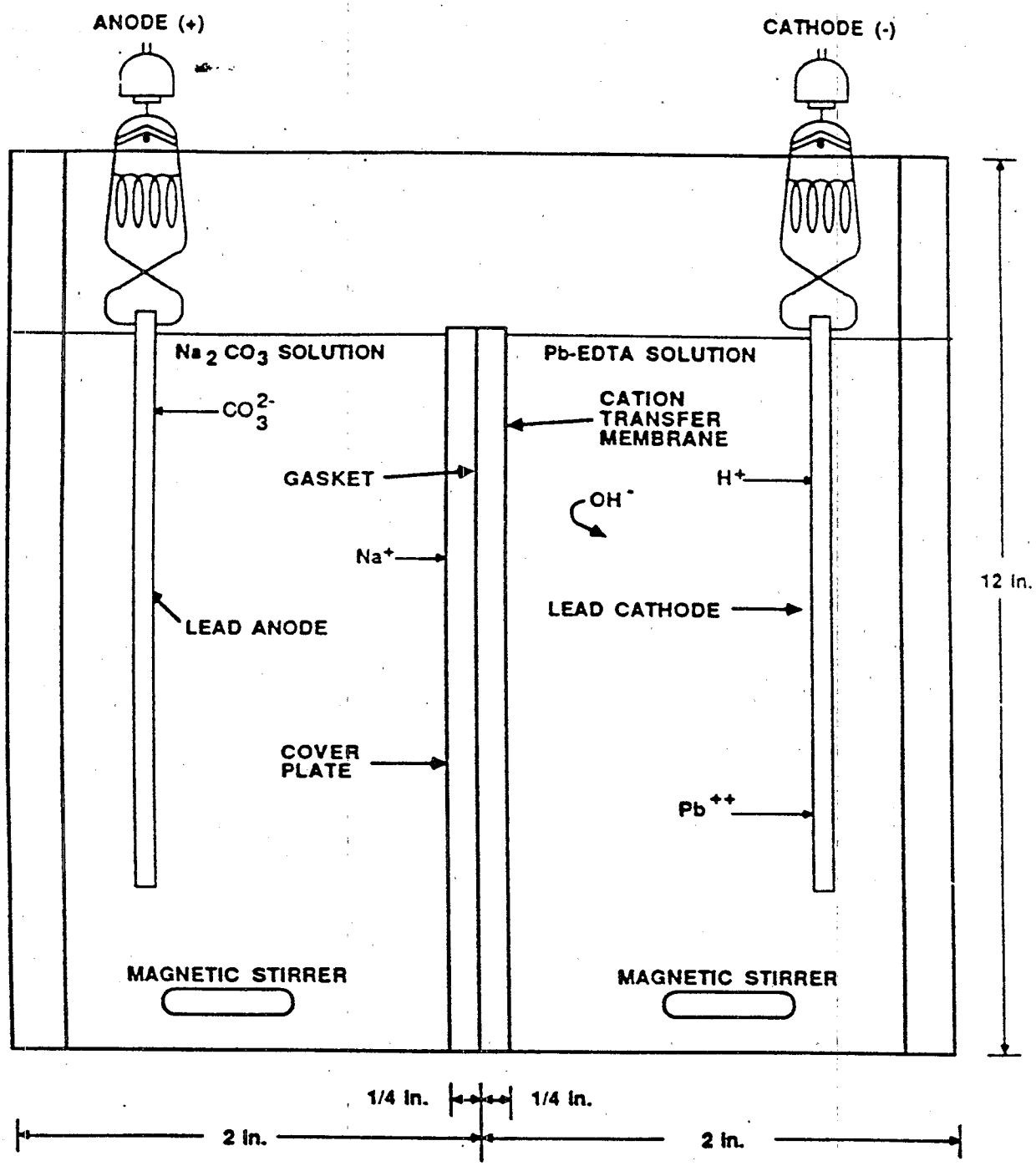


Figure 2-2. Schematic of Electromembrane Reactor.[2]

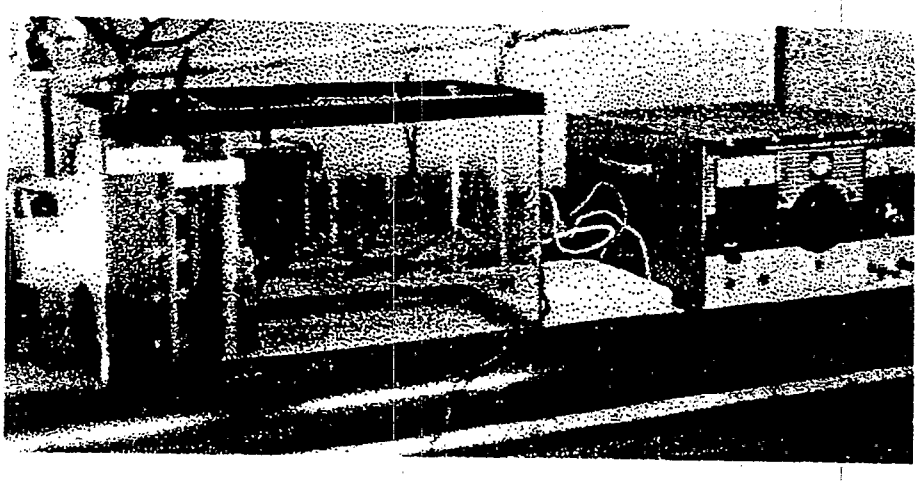


Figure 2-3. Bench-scale Electromembrane Apparatus.

TABLE 2-1. ELECTROMEMBRANE/CHELATION STUDY EXPERIMENTAL MATRIX

Chelating agent	Run No.	Current density, ma/cm^2	Lead conc., %	Membrane	Reaction time, hr	Target pH
Tetra-sodium EDTA	1	25	0.8	DuPont Nafion	3	9
Tetra-sodium EDTA	2	25	0.8	DuPont Nafion	3	9
Tetra-sodium EDTA regenerated solution[a]	3	25	0.8	DuPont Nafion	3	9
DTPA (diethylenetriamine pentaacetic acid)	4	15	0.8	DuPont Nafion	5	9
DTPA	5	25	0.8	DuPont Nafion	5	9
DTPA regenerated solution[b]	6	15	0.8	DuPont Nafion	3	9
DTPA	7	25	0.8	DuPont Nafion	5	9
DTPA	8	25	4	DuPont Nafion	3	9
DTPA	9	25	4	DuPont Nafion	5	9
DTPA regenerated solution[b]	10	25	4	DuPont Nafion	4	9
DTPA	11	15	4	DuPont Nafion	5	9
Tetra-sodium EDTA	12	25	4	DuPont Nafion	3	9
Tetra-sodium EDTA[a]	13	25	4	DuPont Nafion	3	9
Di-sodium EDTA	14	25	0.8	DuPont Nafion	3	5
Di-sodium EDTA regenerated solution[c]	15	25	0.8	DuPont Nafion	3	5
DTPA (Cadmium electrodes)	16	25	4	DuPont Nafion	3	9
DTPA (Cadmium electrodes)	17	15	0.8	DuPont Nafion	3	9
Tetra-sodium EDTA (Cadmium electrodes)	18	25	4	DuPont Nafion	3	9
Tetra-sodium EDTA (1.5% iron)	19	25	4	DuPont Nafion	3	7

(continued)

TABLE 2-1 (continued)

Chelating agent	Run No.	Current density, ma/cm^2	Lead conc., %	Membrane	Reaction time, hr	Target pH
Tetra-sodium EDTA (1.5% iron)	20	25	4	DuPont Nafion	3	9
Tetra-sodium EDTA (1.5% iron)	21	25	4	DuPont Nafion	3	11.5
DTPA (Ionics membrane)	22	25	4	Ionics	3	9
DTPA (Ionics membrane)	23	15	0.8	Ionics	3	9
DTPA (Ionics membrane)	24	15	0.8	Ionics	3	9

[a] Experiment was performed using the Tetra-sodium EDTA solution from runs 2 and 12, respectively.
[b] Experiment was performed using the DTPA solution regenerated from runs 4 and 9, respectively.
[c] Experiment was performed using the Di-sodium EDTA solution regenerated from run 14.

membrane reactor study. Table 2-2 shows the experimental parameters that were compared in different set of experiments.

2.3.1 Chelating Agents

A chelating agent is a compound containing donor atoms that can combine by coordinate bonding with a single metal atom to form a cyclic structure called a chelate. Chelating agents may be organic or inorganic compounds, but are generally organic compounds. The chelating agents selected for this study were di-sodium EDTA, tetra-sodium EDTA, and DTPA. They were selected because of their high lead stability constants and prior-use as soil washing solutions. The structure of EDTA, which is an aminocarboxylic acid, is presented in Figure 2-4[6]. Two sodium salt forms, di-sodium EDTA and tetra-sodium EDTA, were used in the electro-membrane experiments to determine any advantages to the use of one sodium form over another. The lead (II) stability constant for EDTA is 18.0. EDTA is believed to be a sexadentate chelating agent with lead. The highest heat evolved during the formation of the lead complex occurs with EDTA. Di-sodium salts of lead chelates are very soluble. The lead (II) stability constant for DTPA is 18.66 at 25°C and a pH level of 9. DTPA has been proven to form stronger lead-chelate complexes than EDTA[7].

The pH level at which a soil is washed with a chelating agent is important if one or more metals are to be selectively chelated. Iron is of particular interest because of its high concentration in most soils and its relatively similar stability constant as lead for both EDTA and DTPA. Figure 2-1 presents the stability constants as a function of pH for EDTA, DTPA, and nitrilotriacetic acid (NTA). According to this figure, the highest lead-chelate stability constants occur at approximately pH 9 for both tetra-sodium EDTA and DTPA. For EDTA solutions at a pH level above 12, insoluble metal salts are formed; at a pH above 7, the chelation of iron decreases. For DTPA, the chelation of lead decreases at a pH of approximately 9.

2.3.2 Current Density

The current density [calculated as milliamps (ma)/cm^2] that can be applied across a membrane is limited by concentration polarization. The energy requirement

TABLE 2-2. EXPERIMENTAL OBJECTIVES FOR THE ELECTROMEMBRANE STUDY

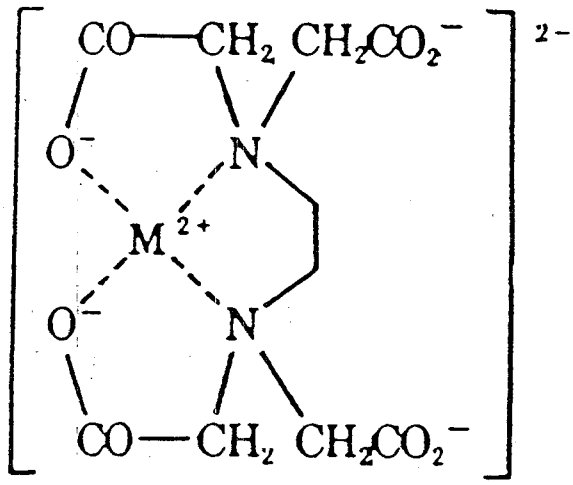
Run No.	Chelating Agent[a]	Current Density, ma/cm^2	Lead Content, %	Objective
14	Di-sodium EDTA	25	0.8	Study effect of two sodium forms of EDTA
1	Tetra-sodium EDTA	25	0.8	
2	Tetra-sodium EDTA	25	0.8	
14	Di-sodium EDTA	25	0.8	Study effect of reusing the chelating solutions
15	Di-sodium EDTA (regenerated)	25	0.8	
12	Tetra-sodium EDTA	25	4	
13	Tetra-sodium EDTA (regenerated)	25	4	
2	Tetra-sodium EDTA	25	0.8	
3	Tetra-sodium EDTA (regenerated)	25	0.8	
4	DTPA	15	0.8	
6	DTPA (regenerated)	15	0.8	
9	DTPA	25	4	
10	DTPA (regenerated)	25	4	
4	DTPA	15	0.8	Study effect of lead concentration
11	DTPA	15	4	
1				
4	DTPA	15	0.8	Study effect of current density
5	DTPA	25	0.8	
7	DTPA	25	0.8	
11	DTPA	15	4	
8	DTPA	25	4	
9	DTPA	25	4	
14	Di-sodium EDTA	25	0.8	Study effect of chelating agents
1	Tetra-sodium EDTA	25	0.8	
2	Tetra-sodium EDTA	25	0.8	
5	DTPA	25	0.8	
7	DTPA	25	0.8	
8	DTPA	25	4	
9	DTPA	25	4	
12	Tetra-sodium EDTA	25	4	

(continued)

TABLE 2-2 (continued)

Run No.	Chelating Agent[a]	Current Density, ma/cm^2	Lead Content, %	Objective
4	DTPA	15	0.8	Study effect of electromembrane
23	DTPA (Ionics membrane)	15	0.8	
24	DTPA (Ionics membrane)	15	0.8	
8	DTPA	25	4	
9	DTPA	25	4	
22	DTPA (Ionics membrane)	25	4	
4	DTPA	15	0.8	Study effect of electrode
17	DTPA (Cadmium electrodes)	15	0.8	
8	DTPA	25	4	
9	DTPA	25	4	
16	DTPA (Cadmium electrodes)	25	4	
18	Tetra-sodium EDTA (cadmium electrodes)	25	4	Study effect of electrode
12	Tetra-sodium EDTA	25	4	
12	Tetra-sodium EDTA	25	4	Study effect of iron in chetating solution
19	Tetra-sodium EDTA (1.5 percent iron at pH 7)	25	4	
20	Tetra-sodium EDTA (1.5 percent iron at pH 9)	25	4	
21	Tetra-sodium EDTA (1.5 percent iron at pH 11.5)	25	4	

[a] A DuPont Nafion® electromembrane was used for the experiments unless otherwise specifically stated; lead electrodes were employed in the electromembrane reactor unless otherwise specifically stated.



(11)

Figure 2-4. Molecular structure of EDTA.[6]

for ionic transport in the electromembrane is a function of the electrical resistance of the solutions and membrane, and the back electromotive forces caused by concentration gradients. Current density is extremely important because it influences the design of full-scale electromembrane reactor units and power supply. The current densities for this set of experiments were kept below 30 ma/cm^2, a level that has been determined to be limiting in full-scale industrial electrodialysis systems[2]. Current densities of 15 and 25 ma/cm^2 were selected for this study. In the electromembrane reactor unit, the voltage was allowed to vary in order to maintain a constant current.

2.3.3 Type of Electrode

Metal dissolution results in the destruction of the electrode. Dissolution may be reduced by using electrodes made of noble metals. Lead was selected for use as electrodes because the standard potential favors spontaneous electron production, and the plated lead and cathode may be smelted together. The selection of cadmium as the alternative electrode material for the electromembrane tests was based on several factors. Chelating agents are corrosive to copper, brass, bronze, and other copper alloys. Chelating agents are also corrosive to zinc, galvanized iron, and nickel. Chelating agents react vigorously with aluminum to produce hydrogen gas, which is a fire and explosion hazard. The ideal metals for use as electrodes in the electromembrane system are noble metals such as gold and silver because the stability of these metals reduces the destruction of the electrode; however, gold and silver are expensive for use as "sacrificial electrodes[2]." Other metals or metal alloys typically used for electrodes are platinum, palladium, cobalt, tin, and magnesium. These metals are also more expensive than cadmium.

Because cadmium is also a contaminant in lead battery waste to a lesser degree than lead, and the stability constant for cadmium-EDTA is similar to that of lead (16.5 for cadmium and 18.0 for lead), cadmium will most likely be present in the metal-chelate solution of an actual battery waste metal-chelate solution. Cadmium also has the added benefit of being relatively volatile for recovery of the cadmium metal from the electrodes by smelting. The standard potential of cadmium favors spontaneous electron production.

2.3.4 Membranes

The cation exchange membranes selected for the bench-scale experiments (Ionics and DuPont Nafion®) are highly permeable to cations and almost impermeable to anions. The resistance of the membrane depends on the transport processes taking place around the membrane. Because the properties of the two membranes are different, the effect of type of membrane on lead removal was studied[2]. The membranes must remain wetted to prevent cracking. Because cracks developed in the membrane when the membrane border was dried for application of a rubber sealant during the 1986 PEI study, the membranes were placed in the reactor with a frame and gasketing material for the IT study. The frame and gasket appeared to adequately seal the membranes to prevent leakage of the lead and sodium solutions.

2.3.5 Lead Content

Soils from battery breaking sites typically contain 1 to 5 percent lead by weight. The IT study focused on the upper and lower ranges of lead contamination. In this study, synthetic lead-chelate solutions were prepared to reflect the lead concentration in contaminated soil. The test solutions were synthetically made from four lead species at total lead concentrations of 0.8 and 4 percent lead, by weight. These percentages represent moderate (0.8 percent lead) and high (4 percent lead) lead concentrations, based on previous EDTA soil washing studies. The forms of lead used in the experiments consisted of a mixture of lead sulfate ($PbSO_4$), basic lead carbonate [$(PbCO_3)_2 \cdot Pb(OH)_2$], lead dioxide ($PbO_2$), and metallic lead; these are the predominant lead species found at battery reclamation sites. The forms of lead initially added in the solution consisted of the following: 60 percent by weight lead sulfate, 20 percent by weight basic lead carbonate, 10 percent by weight elemental lead, and 10 percent by weight lead dioxide[8].

Three bench-scale tests were conducted with 1.5 percent iron/4.0 percent lead solutions to determine the effects of iron on lead-EDTA chelation and lead recoveries in the electromembrane process. Through examination of the chemical analysis of several battery reclamation site soils, the average iron content was estimated to be 1.5 percent. Both ferrous (Fe^{+2}) and ferric (Fe^{+3}) iron may be present in the soil; however, the percentage of each iron species in battery reclamation site soils is unknown

and would be highly dependent on factors such as soil pH and other constituents in the soil. Figure 2-5 presents stability constants for EDTA complexes with Fe^{+2} and Fe^{+3} as a function of pH. From a comparison of Figures 2-1 and 2-5, the EDTA stability constant curve as a function of pH is similar for both ferrous iron and lead and the maximum stability constants for each are associated with a pH level of approximately 9. The stability constant for ferric iron, however, is highest at a pH level of 4. Based on the pH values associated with the maximum EDTA stability constants for Fe^{+2} and Fe^{+3}, it appears that ferrous iron may interfere to a greater extent with lead chelation than ferric iron. Therefore, iron in the ferrous ion form (ferrous sulfate) was used in the bench-scale tests to determine the effect of iron in lead recoveries. The tests were conducted at a pH level of 9, which is the experimental pH used in this study; a pH level of 7, which is the pH that will provide maximum lead chelation while suppressing iron (II) chelation; a pH level of 11.5, which is the pH of the EDTA solution without pH adjustment, and a 4.0 percent lead solution. The electromembrane experiments were conducted with a 25 ma/cm^2 current density, 3-hour reaction time, lead electrodes, and the DuPont Nafion® membrane in order to compare the data from the iron tests with the data from the experiments under the same conditions using lead solution only.

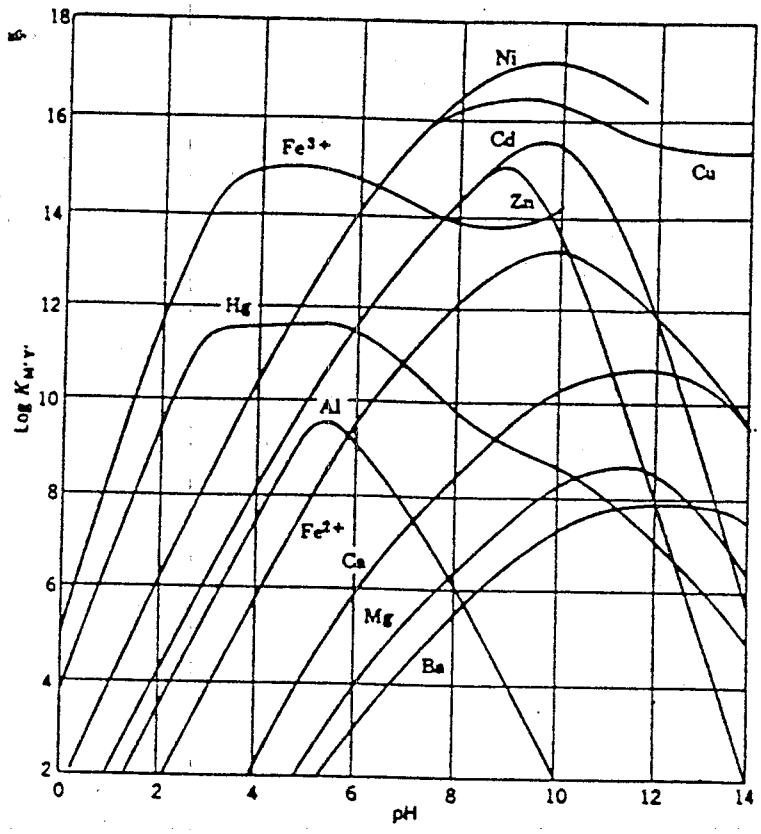


Figure 2-5. Comparison of stability constants for various metal-EDTA complexes as a function of pH.[7]

SECTION 3

CHELATION AND ELECTROMEMBRANE TESTS

This section presents the results of the bench-scale experiments. Preliminary chelation tests were performed (prior to the electromembrane reactor tests) to determine the species of lead that can be chelated by two sodium forms of EDTA and DTPA and the optimum molar ratio of chelating agent and lead to chelate a 4 percent lead solution.

3.1 LEAD CHELATION JAR TESTS

Preliminary jar tests were conducted by IT to determine the ability of the chelating agents to chelate the various species of lead: elemental lead, basic lead carbonate, lead sulfate, and lead dioxide. Jar tests with a 2:1 tetra-sodium EDTA-to-lead molar ratio were performed at a pH level of 9. The following quantities of lead were added to a 4-liter tetra-sodium EDTA and water solution to make a 2:1 EDTA-to-lead molar ratio solution:

14.98 grams of basic lead carbonate (white powder)
2.3 grams of lead dioxide (black powder)
17.56 grams of lead sulfate (white powder)
2.0 grams of elemental lead (silver metal)

The lead and EDTA solution was mixed for one hour and filtered through a Buchner funnel and vacuum flask. Lead that was not chelated remained in the filter and appeared black and silver in color. The solids remaining in the filter weighed 4.3015 grams, which is approximately the combined weight of the elemental lead and lead dioxide. Because the unchelated lead resembled lead dioxide and elemental lead, and the weight of the solids was approximately that of the combined weight of lead dioxide and elemental lead, it was assumed that none of the elemental lead and lead

dioxide was chelated. This is to be expected because EDTA will only chelate with the ionic form of lead. One additional lead-chelation test with tetra-sodium EDTA was performed, and similar results were observed.

Chelation tests, similar to those conducted with tetra-sodium EDTA, were performed with DTPA at a pH level of 9 and with di-sodium EDTA at a pH level of 5. As with the tetra-sodium EDTA tests, none of the lead dioxide or elemental lead was successfully chelated. The inability to completely chelate lead metal or lead dioxide has also been observed in previous soil washing treatability and field tests conducted by the U.S. Bureau of Mines (BOM). However, the BOM study showed that lead dioxide was completely chelated using di-sodium EDTA and a portion of lead metal was chelated by both forms of EDTA. Table 3-1 presents the results of the BOM study.[9] Because the initial form of lead is irrelevant once the lead is chelated, only lead sulfate and basic lead carbonate were used to make the synthetic lead-chelate solution for the electromembrane tests.

TABLE 3-1. BUREAU OF MINES LEAD CHELATION TESTS WITH TWO FORMS OF EDTA

EDTA Form (30 mL, 5% solution)	pH	% Removed		
		5g PbO_2 (1)	5g $PbSO_4$ (1)	5g Pb Metal (2)
Di-sodium	5	100	0	2.7
Tetra-sodium	10	0	100	23.6

Additional jar tests were performed with lead sulfate and basic lead carbonate at a pH level of 9 with tetra-sodium EDTA and DTPA, and at a pH level of 5 with di-sodium EDTA. Chelating-agent-to-lead molar ratios of 1:1, 1.5:1, and 2:1 were used to determine the minimal molar ratio to achieve complete lead chelation. A 4 percent lead concentration solution was prepared with 1:1 and 1.5:1 chelating agent-to-lead molar ratios. Appendix A presents the method for lead and chelating agent solution preparation. The chelating agent manufacturer, Hampshire Chemicals, indicated that a 1.4:1 molar ratio for EDTA and a 1.9:1 molar ratio for DTPA would be adequate for

lead chelation. The solutions consisting of lead and chelating agent were mixed for one hour and filtered through a Buchner funnel and vacuum flask. The lead in both the 1:1 and 1:5:1 di-sodium EDTA-to-lead molar ratio solutions at pH level 5 was completely chelated as indicated by the clear solution and no remaining solid lead. Lead in the 1.5:1 molar tetra-sodium EDTA solution was chelated, but approximately 2.1 grams of the lead compounds was not chelated in the 1:1 molar solution. The DTPA solutions did not chelate 6.1195 grams and 3.776 grams of lead compounds in the 1:1 and 1.5:1 molar ratio solutions, respectively. A 2:1 DTPA-to-lead molar ratio solution was mixed and all of the lead was chelated. Therefore, a 1:1 di-sodium EDTA-to-lead molar ratio, a 1.5:1 tetra-sodium EDTA-to-lead molar ratio, and a 2:1 DTPA-to-lead molar ratio solution would be adequate for lead chelation.

Jar tests were also performed with ferrous sulfate to evaluate lead chelation in the presence of iron. The amount of ferrous sulfate used corresponded to an iron concentration of 1.5 percent, which is typically the concentration of iron encountered in battery breaking site soils. Jar tests were conducted by adding lead sulfate and basic lead carbonate to make a 4 percent lead solution, and 1.5 percent iron in the form of ferrous sulfate to two tetra-sodium EDTA solutions that were adjusted to pH levels of 7 and 9 with sulfuric acid. A third jar test was performed without adjusting the solution pH with sulfuric acid; the pH of this solution was approximately 11.5. During the jar tests, the color of the metal-chelate solution changed from green to brown. This color change was assumed to be the oxidation of iron in ferrous sulfate to ferric hydroxide:

$$4\ FeSO_4 + O_2 + 10\ H_2O \text{ ----> } 4\ Fe(OH)_3 + 4H_2SO_4$$

A drop in pH level was observed after the solutions were thoroughly mixed indicating the formation of sulfuric acid. Once the solutions were filtered through a Buchner funnel and vacuum flask, a transparent reddish filtrate was obtained and a brown residue remained in the filter. Based on laboratory analyses of the metal-chelate solutions, all of the iron (1.5 percent) was chelated in the tests conducted at pH levels of 7 and 9, but only 75 percent of the iron was chelated in the tests performed at a pH of 11.5.

3.2 ELECTROMEMBRANE TESTS

The electromembrane tests were conducted to determine the effects of chelating agent, current density, lead concentration, membrane, and electrode composition on lead recovery and chelant regeneration from lead-chelate solutions. Data collected from observations and measured test parameters were documented and are presented in this section. Table 3-2 contains the raw experimental data for the electromembrane tests. The data include pH and temperature readings in the anode and cathode chambers, and voltage and amperage readings from the DC power supply.

3.2.1 pH

Cathode chamber--

The formation of hydrogen apparently caused bubbles to form at the cathode and anode after the power was supplied to the electrodes by the DC power supply. An increase in the pH of the cathode chamber indicated that hydroxyl ions were being formed from the electrolysis of water:

$$2H_2O + 2e^- \text{-----}> H_2 + 2OH^-$$

A greater increase in cathode chamber pH indicates increased reactivity at the cathode. According to the cathode chamber pH data in Table 3-2, the greatest increase in pH with time occurs for the runs conducted with 0.8 percent lead solutions regardless of which chelating agent was used. The highest increase in pH was observed for the tests conducted with di-sodium EDTA. These tests were performed with a starting pH of approximately 5, and had an approximate 7 pH unit increase within the 3-hour reaction period. The tests conducted with 4 percent lead/1.5 percent iron solutions and a 25 ma/cm^2 current density (Runs 19, 20, and 21) also had a greater than 3 pH unit increase in cathode chamber pH over the 3-hour reaction period. Therefore, it appears that the tests performed with iron solutions and the tests performed with 0.8 percent lead solutions had the greatest reactivity in the cathode chamber.

TABLE 3-2. EXPERIMENTAL DATA FOR ELECTROMEMBRANE TESTS (a)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
1	Na4EDTA	0.8	25	0.00	11.41	9.18	25.9	25.9	7.9	7.0
				0.25	10.49	9.19	26.5	26.5	7.9	7.2
				0.50	10.53	8.67	28.5	29.0	8.0	8.0
				0.75	10.29	8.45	29.5	30.0	8.0	7.0
				1.00	10.20	9.61	30.0	31.0	8.0	7.0
				1.25	10.00	9.00	31.5	32.5	8.0	7.0
				1.50	10.00	9.89	32.0	33.5	8.0	6.5
				1.75	10.00	9.98	34.0	35.0	8.0	6.5
				2.00	9.98	11.23	34.0	34.0	8.0	6.5
				2.25	10.03	10.86	35.0	35.5	8.0	6.5
				2.50	9.49	10.40	36.0	37.0	8.0	6.5
				2.75	9.49	11.10	36.0	37.0	8.0	6.0
				3.00	9.42	12.04	37.0	38.0	8.0	6.0
2	Na4EDTA	0.8	25	0.00	11.40	8.13	26.0	26.0	8.0	7.0
				0.25	10.90	8.75	27.0	27.0	8.0	7.0
				0.50	10.69	9.54	28.0	29.0	8.0	7.0
				0.75	10.59	9.40	29.0	30.0	8.0	7.0
				1.00	10.42	9.43	30.0	31.0	8.0	7.0
				1.25	10.29	9.62	31.5	32.0	8.0	7.0
				1.50	10.21	10.16	32.0	33.0	8.0	7.0
				1.75	10.05	11.05	33.0	34.0	8.5	7.0
				2.00	10.03	11.66	34.0	35.0	8.0	6.5
				2.25	9.85	11.83	34.8	35.0	8.0	5.5
				2.50	9.74	11.89	35.3	35.5	8.0	5.0

(a) A DuPont Nafion electomembrane and lead electrodes were used for the experiments unless otherwise specifically stated.

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
2	Na4EDTA	0.8	25	2.75	9.80	12.02	36.0	37.0	8.0	6.0
				3.00	9.72	11.92	36.5	37.5	8.0	6.0
3	Na4EDTA (Regenerated from Run No. 2)	0.8	25	0.00	11.63	9.15	25.0	25.0	8.0	5.5
				0.25	11.12	9.50	25.5	26.0	7.5	5.5
				0.50	11.24	9.49	26.0	27.5	8.5	6.0
				0.75	10.87	9.61	27.5	28.0	8.0	6.0
				1.00	10.74	9.65	28.0	28.5	8.0	6.0
				1.25	10.56	9.79	29.0	29.0	8.0	6.0
				1.50	10.58	9.96	29.5	30.0	8.0	6.0
				1.75	10.37	10.30	30.5	31.0	8.0	6.0
				2.00	10.28	10.58	30.5	32.0	8.0	6.0
				2.25	10.10	11.76	31.5	32.0	8.5	6.0
				2.50	9.88	12.30	32.5	33.0	8.5	6.0
				2.75	9.95	12.55	33.0	34.0	8.5	6.0
				3.00	9.95	12.57	33.5	34.5	8.0	6.0
4	DTPA	0.8	15	0.00	11.94	7.46	29.0	29.0	4.7	4.0
				0.25	11.39	8.06	29.0	29.0	3.5	4.0
				0.50	11.25	9.13	29.5	30.0	4.8	5.0
				0.75	11.07	9.71	30.0	31.0	4.8	5.0
				1.00	10.72	9.49	31.0	32.0	4.8	5.0
				1.25	10.56	9.68	31.5	32.0	4.8	5.0
				1.50	10.46	10.24	32.0	32.5	4.9	5.0
				1.75	10.70	11.43	33.0	33.5	4.7	4.0
				2.00	10.73	11.79	33.5	33.8	4.7	4.0
				2.25	10.51	11.92	33.5	33.8	4.7	5.0
				2.50	10.41	12.28	33.8	34.0	4.7	6.2
				2.75	10.10	11.91	34.2	34.6	4.7	6.2

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
				3.00	10.00	12.58	37.0	38.0	4.9	5.0
				3.30	10.02	12.55	36.0	37.0	4.9	4.5
				4.00	10.00	12.58	37.0	38.0	4.9	5.0
				5.00	9.76	12.46	37.5	38.0	4.9	5.0
5	DTPA	0.8	25	0.00	10.80	8.07	26.5	26.5	7.9	6.0
				0.25	11.00	8.82	28.0	28.0	7.0	5.5
				0.50	10.60	9.67	29.0	30.0	7.9	7.0
				0.75	10.65	10.34	30.0	31.5	8.0	7.5
				1.00	10.36	10.97	31.0	32.5	8.1	7.5
				1.25	10.59	11.72	32.0	33.5	7.9	7.0
				1.50	10.18	11.78	33.0	34.0	7.9	7.0
				1.75	9.87	11.68	33.8	34.8	7.9	5.8
				2.00	9.65	12.00	34.0	35.2	7.9	5.6
				2.25	10.09	11.88	35.1	36.0	7.9	5.6
				2.50	9.82	12.21	36.0	37.0	8.0	6.5
				2.75	9.72	12.28	36.5	37.0	8.2	6.2
				3.00	9.85	12.57	37.0	38.0	8.1	6.2
6	DTPA (Regenerated from Run No. 4)	0.8	15	0.00	11.33	8.55	28.0	25.5	4.7	3.5
				0.25	11.23	9.05	28.0	27.0	4.7	4.0
				0.50	11.04	9.21	28.5	28.0	4.7	3.5
				0.75	10.98	9.43	29.0	29.0	4.7	4.0
				1.00	10.86	9.64	29.2	29.7	4.7	4.5
				1.25	10.63	9.97	30.0	30.5	5.0	5.0

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
6	DTPA (Regenerated from Run No. 4)	0.8	15	1.50	10.58	10.27	30.5	31.0	5.0	5.0
				1.75	10.61	10.34	31.5	32.0	5.0	5.0
				2.00	10.51	10.54	32.0	33.0	4.7	5.0
				2.25	10.48	10.59	32.5	33.0	4.7	4.5
				2.50	10.25	10.92	33.0	34.0	4.7	4.5
				2.75	10.37	11.49	34.0	34.0	4.7	5.0
				3.00	10.13	11.71	34.0	34.5	4.7	4.5
7	DTPA	0.8	25	0.00	10.94	12.33	25.5	27.0	7.9	5.5
				0.25	10.61	11.87	26.5	27.5	7.9	6.0
				0.50	10.60	11.92	27.0	28.0	7.9	6.5
				0.75	10.54	11.90	28.0	29.0	8.0	6.5
				1.00	10.43	11.82	29.0	29.5	7.9	6.0
				1.25	10.26	11.75	30.0	31.5	8.0	6.5
				1.50	10.05	12.18	31.0	32.0	8.0	6.5
				1.75	9.87	12.11	32.0	33.0	7.9	6.0
				2.00	10.00	12.16	32.5	33.0	7.9	6.0
				2.25	9.82	12.19	32.8	34.3	7.9	6.3
				2.50	9.89	12.08	34.0	34.5	8.0	6.0
				2.75	9.12	12.75	34.5	35.0	8.0	6.0
				3.00	9.42	12.33	35.0	35.5	8.0	6.0
				3.25	9.88	12.44	35.5	36.0	8.0	6.0
				3.50	9.43	12.32	36.0	36.0	7.9	6.0
				3.75	10.12	12.63	36.0	37.0	7.9	6.0
				4.00	9.48	12.43	37.0	37.0	7.9	6.0
				4.25	9.35	12.48	37.0	37.5	7.9	6.0
				4.50	9.00	12.03	37.5	38.0	7.9	6.0
				4.75	10.50	12.73	37.5	38.0	7.9	6.0

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
7	DTPA	0.8	25	5.00	9.22	12.64	38.0	38.0	7.9	6.0
8	DTPA	4	25	0.00	11.00	9.02	22.0	22.0	7.9	5.7
				0.25	10.78	8.93	22.5	23.2	7.9	5.9
				0.50	10.62	9.32	23.2	23.8	7.9	5.9
				0.75	10.43	9.37	25.0	25.0	6.9	4.5
				1.00	10.15	9.32	25.5	26.0	8.0	5.0
				1.25	10.12	9.32	26.0	26.0	7.9	5.0
				1.50	9.93	9.09	26.5	27.0	8.0	5.0
				1.75	9.05	9.33	27.0	27.0	8.0	5.5
				2.00	9.72	9.38	28.0	28.0	8.0	5.5
				2.25	9.51	9.60	28.0	28.5	8.0	5.5
				2.50	9.47	9.47	29.0	29.5	8.0	5.5
				2.75	9.41	9.53	29.0	29.5	8.0	5.5
				3.00	9.20	9.36	30.0	30.0	8.0	5.5
9	DTPA	4	25	0.00	11.16	9.18	24.5	27.5	7.9	6.0
				0.25	11.02	9.26	26.0	27.5	7.9	5.0
				0.50	10.71	9.53	27.0	28.0	7.9	5.0
				0.75	10.79	9.77	28.0	29.0	8.0	5.0
				1.00	10.94	9.68	28.0	29.0	8.0	5.0
				1.25	10.31	9.55	29.5	30.0	8.0	5.0
				1.50	10.03	9.44	30.0	30.0	7.9	5.0
				1.75	10.30	9.62	30.5	31.0	8.0	5.0
				2.00	9.90	9.58	31.0	31.5	7.9	5.0
				2.25	9.90	9.55	32.0	32.0	7.9	5.0
				2.50	9.73	9.54	32.0	32.2	7.9	5.1
				2.75	9.88	9.61	32.0	32.2	7.9	5.0
				3.00	9.42	9.59	32.0	32.3	7.9	5.0

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
9	DTPA	4	25	3.25	9.40	9.62	32.2	32.5	7.9	5.1
				3.50	9.32	9.67	32.3	33.5	7.9	5.1
				3.75	9.28	9.68	34.1	34.4	7.9	5.5
				4.00	9.22	9.74	33.9	34.1	7.9	5.5
				4.25	9.18	9.75	34.0	34.3	7.9	5.5
				4.50	8.86	9.78	34.8	35.0	7.9	5.6
				4.75	8.61	9.73	35.0	35.3	7.9	5.8
				5.00	8.52	9.79	35.2	35.5	7.9	5.8
10	DTPA (Regenerated from Run No. 9)	4	25	0.00	11.27	8.87	22.5	22.5	7.9	4.9
				0.25	11.23	9.48	22.8	22.7	7.9	4.9
				0.50	10.55	9.21	24.5	24.5	8.0	4.5
				0.75	10.66	9.48	25.0	25.0	8.0	4.5
				1.00	10.72	9.89	26.0	26.0	8.0	4.5
				1.25	10.69	9.89	26.5	27.0	7.9	4.5
				1.50	10.42	9.81	27.0	27.5	7.9	4.5
				1.75	10.44	9.75	28.0	28.0	7.9	4.5
				2.00	10.16	9.79	29.0	29.0	7.9	4.5
				2.25	10.04	9.84	29.0	30.0	7.9	5.0
				2.50	10.09	9.90	30.0	30.0	7.9	5.0
				2.75	9.90	10.01	30.6	30.8	7.9	5.0
				3.00	9.80	10.02	31.8	31.7	7.9	5.0
				3.25	9.70	10.03	31.8	32.0	7.9	5.0
				3.50	9.70	10.03	32.2	32.4	7.9	5.0
				3.75	9.38	9.89	32.5	33.0	7.9	5.0
				4.00	9.23	9.86	33.5	34.5	7.9	5.1

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
11	DTPA	4	15	0.00	11.19	9.23	25.0	25.0	4.7	3.0
				0.25	11.09	9.34	26.0	26.0	5.0	3.5
				0.50	11.02	9.55	26.0	26.0	4.5	3.0
				0.75	10.52	9.74	26.0	27.0	4.5	3.0
				1.00	9.87	10.21	27.0	27.0	4.7	3.5
				1.25	10.36	9.76	27.5	28.0	4.5	3.5
				1.50	10.53	9.80	28.0	28.0	4.5	4.0
				1.75	10.21	9.95	28.5	29.0	4.7	4.0
				2.00	10.38	9.98	29.0	29.0	4.7	4.0
				2.25	10.59	9.95	29.5	30.0	4.8	4.0
				2.50	10.06	9.94	30.0	30.0	4.8	4.0
				2.75	9.92	9.99	30.0	30.0	4.7	4.0
				3.00	9.57	9.96	30.5	30.5	5.0	4.0
				3.25	9.57	9.96	31.0	31.0	4.7	4.0
				3.50	9.86	10.03	31.0	31.5	4.9	3.5
				3.75	8.02	10.00	31.5	32.0	4.9	4.0
				4.00	9.72	9.98	32.0	32.0	4.7	3.5
				4.25	9.61	9.99	32.1	32.5	4.7	3.8
				4.50	9.40	9.99	32.3	32.5	4.7	3.9
				4.75	9.48	10.03	32.3	32.5	4.7	3.9
				5.00	9.57	10.07	32.3	32.5	4.7	3.9
12	Na4EDTA	4	25	0.00	11.11	9.13	25.9	25.9	7.9	5.9
				0.25			26.5	26.5	7.9	4.5
				0.50	10.39	9.71	27.5	27.5	7.9	4.9
				0.75			28.5	28.3	7.9	5.0
				1.00	10.17	10.16	28.7	28.8	7.9	5.0
				1.25			29.2	29.5	7.9	5.0

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
12	Na4EDTA	4	25	1.50	9.98	10.31	29.8	30.3	7.9	4.8
				1.75						
				2.00	9.78	10.60	31.3	31.5	7.9	4.3
				2.25						
				2.50	9.66	10.84	32.3	32.5	7.9	4.3
				2.75			32.5	33.0	7.9	4.3
				3.00	9.51	11.17	33.1	33.3	7.9	4.3
13	Na4EDTA (Regenerated From Run No. 12)	4	25	0.00	9.59	8.61	29.0	29.0	7.9	4.8
				0.25			29.5	29.2	7.9	4.1
				0.50	9.45	9.14	30.0	30.0	7.9	4.1
				0.75			30.9	30.9	7.9	4.1
				1.00	9.10	9.30	31.5	31.7	7.9	4.1
				1.25			32 1	32.5	7.9	4.1
				1.50	9.29	9 49	32.6	33.0	7.9	4.1
				1.75			33.2	33.8	7.9	4.1
				2.00	8.90	9 80	34.0	34.2	7.9	4.3
				2.25			34.3	34.6	7.9	4.3
				2.50	8.63	9.86	34.6	34.8	7.9	4.4
				2.75			35.1	35.3	7.9	4.4
				3.00	8.08	9.99	35.9	35.9	7.9	4.5
14	Na2EDTA	0.8	25	0.00	11.48	5.40	24.0	24.0	7.9	8.9
				0.25			24.5	25.2	7.9	8.9
				0.50	10.82	8.01	25.8	27.7	7.9	8.2
				0.75			26.5	28.5	7.9	8.2
				1.00	10.43	9.46	28.5	30.7	7.9	7.9
				1.25			29.8	31.6	7.9	7.9
				1.50	10.33	9.76	30.5	32.5	7.9	8.0

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
14	Na2EDTA	0.8	25	1.75			31.5	33.5	7.9	7.8
				2.00	10.09	10.98	32.6	34.5	7.9	7.7
				2.25			33.5	34.8	7.9	7.2
				2.50	9.97	12.15	34.0	35.5	7.9	7.1
				2.75			34.9	36.0	7.9	6.8
				3.00	9.87	12.45	35.3	36.5	7.9	6.5
15	Na2EDTA (Regenerated From Run No. 14)	0.8	25	0.00	11.39	5.05	26.8	26.8	7.9	7.1
				0.25			27.4	27.9	7.9	7.0
				0.50	10.59	6.82	28.9	29.2	7.9	5.9
				0.75			29.5	30.0	7.9	5.9
				1.00	10.26	9.09	31.0	31.5	7.9	5.9
				1.25			31.3	31.8	7.9	5.8
				1.50	10.11	9.63	32.5	32.8	7.9	5.6
				1.75			33.3	33.8	7.9	5.6
				2.00	9.89	10.31	34.3	34.8	7.9	5.5
				2.25			34.8	35.1	7.9	5.6
				2.50	9.75	11.54	35.5	35.8	7.9	5.7
				2.75			36.2	36.7	7.9	5.8
				3.00	9.63	11.98	36.8	37.2	7.9	5.7
16	DTPA (Cadmium Electrodes)	4	25	0.00	11.41	9.15	25.5	25.5	7.9	6.1
				0.25			26.7	26.5	7.9	6.1
				0.50	10.59	9.25	27.5	27.3	7.9	6.1
				0.75			29.1	28.2	7.9	6.2
				1.00	10.28	9.39	29.7	29.7	7.9	6.1
				1.25			30.3	30.8	7.9	6.1
				1.50	10.13	9.50	31.5	31.4	7.9	6.0

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
16	DTPA (Cadmium Electrodes)	4	25	1.75			32.4	32.1	7.9	6.0
				2.00	9.98	9.48	33.0	32.7	7.9	6.0
				2.25			33.8	33.8	7.9	6.1
				2.50	9.78	9.55	34.3	34.1	7.9	6.0
				2.75			35.3	35.0	7.9	6.0
				3.00	9.69	9.62	35.7	35.5	7.9	6.0
17	DTPA (Cadmium Electrodes)	0.8	15	0.00	11.21	8.40	25.0	25.0	4.7	4.5
				0.25			26.3	26.1	4.7	4.8
				0.50	10.81	9.38	27.5	27.8	4.7	5.3
				1.00	10.55	9.90	29.3	29.5	4.7	5.3
				1.25			29.8	30.3	4.7	5.1
				1.50	10.37	10.47	30.8	31.3	4.7	5.1
				1.75			31.3	31.8	4.7	5.1
				2.00	10.31	11.52	31.8	32.3	4.7	5.1
				2.25			32.5	32.9	4.7	5.1
				2.50	10.12	11.88	33.1	33.5	4.7	5.1
				2.75			33.5	34.0	4.7	4.9
				3.00	10.00	12.13				
18	Na4EDTA (Cadmium Electrodes)	4	25	0.00	11.37	9.28	27.0	27.0	7.9	4.5
				0.25			27.8	28.1	7.9	4.7
				0.50	10.70	9.64	28.5	28.7	7.9	4.9
				0.75			29.1	29.3	7.9	4.9
				1.00	10.36	9.63	30.5	30.5	7.9	4.9
				1.25			31.1	31.4	7.9	4.9
				1.50	10.09	9.59	32.3	32.2	7.9	4.9

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
18	Na4EDTA (Cadmium Electrodes)	4	25	1.75			32.9	32.9	7.9	4.9
				2.00	9.90	9.61			7.9	4.9
				2.25					7.9	4.9
				2.50	9.79	9.72	34.9	34.9	7.9	4.8
				2.75						
				3.00	9.66	9.67	35.8	35.8	7.9	4.8
19	Na4EDTA (1.5% Iron)	4	25	0.00	11.46	6.68	25.8	25.8	7.9	5.8
				0.25			27.1	27.1	7.9	4.9
				0.50	10.60	6.90	28.0	28.1	7.9	4.3
				0.75			28.5	28.8	7.9	4.1
				1.00	10.34	7.21	29.7	29.9	7.9	4.1
				1.25			30.1	30.3	7.9	4.1
				1.50	10.11	8.33	31.5	31.7	7.9	4.0
				1.75			32.1	32.2	7.9	4.0
				2.00	9.95	9.39	32.8	32.9	7.9	4.0
				2.25			33.3	33.5	7.9	4.1
				2.50	9.82	9.83	34.0	34.2	7.9	4.1
				2.75			34.7	34.8	7.9	4.1
				3.00	9.70	10.20	35.2	35.3	7.9	4.1
20	Na4EDTA (1.5% Iron)	4	25	0.00	11.31	6.64	27.2	27.2	7.9	4.0
				0.25			27.8	27.8	7.9	4.2
				0.50	10.58	6.75	28.3	28.1	7.9	4.5
				0.75			29.3	29.4	7.9	4.5
				1.00	10.29	7.53	30.2	30.3	7.9	4.5
				1.25					7.9	
				1.50	10.04	9.08	32.2	32.3	7.9	4.4

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
20	Na4EDTA (1.5% Iron)	4	25	1.75			33.1	33.2	7.9	
				2.00	9.86	9.91	33.8	34.0	7.9	4.3
				2.25					7.9	
				2.50	9.72	10.32	35.1	35.3	7.9	4.2
				2.75					7.9	
				3.00	9.57	10.51	36.5	36.8	7.9	4.4
21	Na4EDTA (1.5% Iron)	4	25	0.00	9.72	6.51	27.9	27.9	7.9	4.2
				0.25			28.7	28.0	7.9	4.3
				0.50	9.49	6.98	29.8	29.8	7.9	4.3
				0.75			30.1	30.2	7.9	4.3
				1.00	9.41	8.02	30.8	31.3	7.9	4.5
				1.25					7.9	
				1.50	9.25	9.52	32.9	33.3	7.9	4.5
				1.75					7.9	
				2.00	9.08	10.35	34.5	34.6	7.9	4.5
				2.25					7.9	
				2.50	8.92	10.45	35.5	35.7	7.9	4.6
				2.75					7.9	
				3.00	8.73	10.51	36.6	36.8	7.9	4.8
22	DTPA (Ionics Membrane)	4	25	0.00	11.36	9.20	27.8	27.8	7.9	4.9
				0.25			28.0	28.0	7.9	4.8
				0.50	10.79	9.50	29.1	29.1	7.9	4.5
				0.75			30.1	30.1	7.9	4.9
				1.00	10.44	9.64	31.0	31.0	7.9	4.7
				1.25			32.0	32.0	7.9	4.8
				1.50	10.15	9.78	33.0	33.0	7.9	4.8

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
22	DTPA (Ionics Membrane)	4	25	1.75			33.5	33.5	7.9	4.8
				2.00	9.96	9.87	34.5	34.5	7.9	4.8
				2.25			35.0	35.0	7.9	4.8
				2.50	9.79	9.93	35.5	35.5	7.9	4.8
				2.75			36.0	36.0	7.9	4.8
				3.00	9.62	9.98	37.0	37.0	7.9	4.8
23	DTPA (Ionics Membrane)	0.8	15	0.00	10.92	8.70	27.6	27.6	4.7	4.0
				0.25			27.7	28.0	4.7	4.0
				0.50	10.56	9.29	28.2	28.5	4.7	3.9
				0.75			28.6	28.8	4.7	4.0
				1.00	10.30	10.72	29.5	29.5	4.7	4.1
				1.25			29.6	30.1	4.7	4.1
				1.50	10.15	11.33	29.7	30.5	4.7	4.1
				1.75			30.4	30.4	4.7	4.1
				2.00	10.06	11.69	31.0	31.0	4.7	4.1
				2.25			31.0	31.0	4.7	4.1
				2.50	9.97	11.86	31.5	31.5	4.7	4.1
				2.75			32.0	32.0	4.7	4.1
				3.00	9.82	12.01	32.5	32.5	4.7	4.1
24	DTPA (Ionics Membrane)	0.8	15	0.00	11.02	8.74	25.6	25.6	3.7	3.8
				0.25			26.0	26.0	3.7	3.8
				0.50	10.65	8.99	27.0	27.0	4.7	4.7
				0.75			28.0	28.0	4.7	4.7
				1.00	10.35	9.82	29.0	29.0	4.7	4.7
				1.25			29.5	29.5	4.7	4.7
				1.50	10.18	10.73	30.0	30.0	4.7	4.7

TABLE 3-2 (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (hr)	pH Anode	pH Cathode	Temperature of Anode Chamber (C)	Temperature of Cathode Chamber (C)	Amp	Volt
24	DTPA	0.8	15	1.75			31.0	31.0	4.7	4.7
	(Ionics			2.00	10.04	11.58	32.0	32.0	4.7	4.7
	Membrane)			2.25			32.5	32.5	4.7	4.7
				2.50	9.92	11.80	33.0	33.0	4.7	4.5
				2.75			33.5	33.5	4.7	4.5
				3.00	9.84	11.98	34.0	34.0	4.7	4.4

Anode chamber--

The production of oxygen is believed to cause bubbles to form at the anode. Because the anode chamber pH decreased with time, the formation of hydrogen ions is believed to be occurring in the anode chamber:

$$2H_2O \text{ ------> } O_2 + 4H^+ + 4e^-$$

According to the anode chamber pH data presented in Table 3-2, the initial pH of the anode chamber was approximately 11.5, but decreased to approximately pH 10 after a 3-hour reaction period in almost all of the tests. Only one difference was noted; the tests conducted with the regenerated DTPA solutions (Runs 6 and 13) had a greater decrease in anode chamber pH than the tests conducted with regenerated tetra-sodium EDTA and di-sodium EDTA (Runs 3, 13, and 15). No other differences were noted for the following test comparisons: 1) two forms of sodium EDTA, 2) two lead concentrations, 3) two current densities, 4) three chelating agents, 5) two membranes, 6) two electrodes, 7) and tests with iron.

3.2.2 Temperature

The temperature in the anode and cathode chambers increased with time, indicating that exothermic reactions were taking place in both chambers. The temperature apparently increased more rapidly in the cathode chamber than in the anode chamber in most of the experiments. Temperature data for the 24 electro-membrane experiments are provided in Table 3-2.

According to the data presented in Table 3-2, both the anode and cathode chamber temperatures increased by more than 10°C for Runs 1 and 2 conducted with tetra-sodium EDTA solutions containing 0.8 percent lead and with a 25 ma/cm^2 current density; Run 5 performed with DTPA solution containing 0.8 percent lead, and with a 25 ma/cm^2 current density; Runs 14 and 15 conducted with di-sodium EDTA solution containing 0.8 percent lead, and with a 25 ma/cm^2 current density; and Run 16 performed with DTPA solution containing 4 percent lead, 25 ma/cm^2 current density, and with cadmium electrodes. The higher temperature increase in the anode chamber suggests an increased reactivity resulting in a greater generation

of electrons, oxygen, and hydrogen ions. The higher temperature increase in the cathode chamber may be a result of the increased reactivity resulting in a greater generation of hydroxyl ions and hydrogen.

3.2.3 Lead recovery

Anode Chamber--

The quantity of lead in the anode chamber was measured for samples of Runs 1, 4, and 8. Because in all of the anode chamber samples no lead was detected, lead analysis of the anode chamber samples was discontinued. The color of the anode changed from silver to an orange-brown indicating that oxidation of the anode was occurring rather than lead dissolution. Anode oxidation occurs according to the following formula:

$$Pb + 2\ OH^- \rightarrow Pb(OH)_2 + 2\ x\ e^-$$

The weight of the anode before and after each test varied by no more than ±0.5 gram from the original weight of 1784.5 grams.

Cathode Chamber--

The amount of lead recovered on the cathode is critical to making the electromembrane process cost-effective because the sale of the lead product is expected to offset some of the capital and operating costs of this lead-recovery process. Lead recovery was measured in two ways: 1) the decrease in the lead content of the cathode chamber solution, and 2) the quantity and quality of lead plated onto the cathode. This section presents a comparison of the lead recoveries achieved by the three chelating agents at varying conditions (e.g., current density, lead concentration, membrane, electrode, iron). A summary of the lead removals for the 24 electromembrane experiments is presented in Table 3-3.

Lead removal rates are compared for tetra-sodium and di-sodium EDTA in Figure 3-1. Run 1 was conducted with tetra-sodium EDTA, 0.8 percent lead content,

TABLE 3-3. PERCENT LEAD REMOVALS OBTAINED IN BENCH-SCALE ELECTROMEMBRANE EXPERIMENTS (a)

Run Number	Chelating Agent	Initial Starting pH	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (min)	Cathode Chamber Sample No.	Dilution Ratio For Analysis	Measured Lead Concentration In Cathode Chamber (mg/L) Sample # 1	Sample # 2	Sample # 3	Avg Lead Concentration In Cathode Chamber (mg/L)	Amount of Lead Removed (%)
1	Na4 EDTA	9	0.8	25	0	EC-1-1-C	250	31.8	32.3	32.5	8050.0	
					30	EC-1-2-C	250	17.2	17.3	17.4	4325.0	46.3
					60	EC-1-3-C	250	14.4	14.3	14.3	3583.3	55.5
					90	EC-1-4-C	250	7.2	7.2	7.2	1800.0	77.6
					120	EC-1-5-C	10	25.1	25.1	25.2	251.3	96.9
					150	EC-1-6-C	5	22.2	22.3	22.3	111.3	98.6
					180	EC-1-7-C	1	50.3	50.2	49.8	50.1	99.4
2	Na4 EDTA	9	0.8	25	0	EC-2-1-C	250	34.2	33.7	34.3	8516.7	
					30	EC-2-2-C	125	53.0	53.2	53.3	6646.8	22.0
					60	EC-2-3-C	125	32.0	32.0	31.9	3995.8	53.1
					90	EC-2-4-C	125	14.6	14.5	14.6	1820.8	78.6
					120	EC-2-5-C	125	5.0	5.0	4.9	620.8	92.7
					150	EC-2-6-C	50	8.6	8.7	8.8	435.0	94.9
					180	EC-2-7-C	25	6.9	7.0	6.9	173.3	98.0
3	Na4 EDTA (Regenerated From Run No. 2)	9	0.8	25	0	EC-3-1-C	125	62.4	62.7	63.3	7850.0	
					30	EC-3-2-C	125	50.6	50.7	50.7	6333.3	19.3
					60	EC-3-3-C	125	32.7	32.7	32.7	4087.5	47.9
					90	EC-3-4-C	125	14.7	14.6	14.6	1829.2	76.7
					120	EC-3-5-C	75	9.5	9.5	9.4	710.0	91.0
					150	EC-3-6-C	10	19.1	19.0	18.9	190.0	97.6
					180	EC-3-7-C	1	53.8	53.9	54.2	54.0	99.3
12	Na4EDTA	9	4	25	0	EC-12-1-C	2000	22.0	22.0	22.0	44000.0	
					30	EC-12-2-C	2000	23.0	23.0	23.0	46000.0	-4.5
					60	EC-12-3-C	2000	23.0	23.0	23.0	46000.0	-4.5
					90	EC-12-4-C	2000	22.0	22.0	22.0	44000.0	0.0
					120	EC-12-5-C	2000	20.0	20.0	20.0	40000.0	9.1
					150	EC-12-6-C	2000	19.0	19.0	19.0	38000.0	13.6
					180	EC-12-7-C	2000	18.0	18.0	18.0	36000.0	18.2
13	Na4EDTA (Regenerated From Run No. 12)	9	4	25	0	EC-13-1-C	2500	27.0	27.0	27.0	67500.0	
					30	EC-13-2-C	2500	27.0	27.0	27.0	67500.0	0.0
					60	EC-13-3-C	2500	26.0	26.0	26.0	65000.0	3.7
					90	EC-13-4-C	2500	25.0	25.0	25.0	62500.0	7.4
					120	EC-13-5-C	2500	23.0	23.0	23.0	57500.0	14.8
					150	EC-13-6-C	2500	21.0	21.0	21.0	52500.0	22.2
					180	EC-13-7-C	2500	19.0	19.0	19.0	47500.0	29.6

(a) All runs were conducted using the DuPont Nafion Membrane and lead electrodes unless otherwise noted.

TABLE 3-3. (cont.)

Run Number	Chelating Agent	Initial Starting pH	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (min)	Cathode Chamber Sample No.	Dilution Ratio For Analysis	Measured Lead Concentration In Cathode Chamber (mg/L) Sample # 1	Sample # 2	Sample # 3	Avg Lead Concentration In Cathode Chamber (mg/L)	Amount of Lead Removed (%)
18	Na4 EDTA (Cadmium Electrodes)	9	4	25	0	EC-18-1-C	800	54.0	53.0	53.0	42666.7	
					30	EC-18-2-C	800	59.0	60.0	61.0	48000.0	-12.5
					60	EC-18-3-C	800	52.0	52.0	52.0	41600.0	2.5
					90	EC-18-4-C	800	54.0	54.0	54.0	43200.0	-1.3
					120	EC-18-5-C	800	58.0	57.0	57.0	45866.7	-7.5
					150	EC-18-6-C	800	38.0	38.0	38.0	30400.0	28.7
					180	EC-18-7-C	800	35.0	35.0	35.0	28000.0	34.4
19	Na4 EDTA (1.5% Iron)	7	4	25	0	EC-19-1-C	2000	19.0	19.0	19.0	38000.0	
					30	EC-19-2-C	2000	17.0	17.0	17.0	34000.0	10.5
					60	EC-19-3-C	2000	17.0	17.0	17.0	34000.0	10.5
					90	EC-19-4-C	2000	14.0	14.0	14.0	28000.0	26.3
					120	EC-19-5-C	2000	13.0	13.0	13.0	26000.0	31.6
					150	EC-19-6-C	2000	11.0	11.0	11.0	22000.0	42.1
					180	EC-19-7-C	2000	9.0	9.0	9.0	18000.0	52.6
20	Na4 EDTA (1.5% Iron)	9	4	25	0	EC-20-1-C	2400	17.0	18.0	18.0	42400.0	
					30	EC-20-2-C	2400	17.0	17.0	17.0	40800.0	3.8
					60	EC-20-3-C	2400	16.0	16.0	16.0	38400.0	9.4
					90	EC-20-4-C	2400	14.0	14.0	14.0	33600.0	20.8
					120	EC-20-5-C	2400	12.0	12.0	12.0	28800.0	32.1
					150	EC-20-6-C	2400	14.0	14.0	14.0	33600.0	20.8
					180	EC-20-7-C	2400	8.0	8.0	8.0	19200.0	54.7
21	Na4 EDTA (1.5% Iron)	11.5	4	25	0	EC-21-1-C	2000	20.0	20.0	20.0	40000.0	
					30	EC-21-2-C	2000	20.0	20.0	20.0	40000.0	0.0
					60	EC-21-3-C	2000	19.0	19.0	19.0	38000.0	5.0
					90	EC-21-4-C	2000	18.0	18.0	17.0	35333.3	11.7
					120	EC-21-5-C	2000	16.0	16.0	16.0	32000.0	20.0
					150	EC-21-6-C	2000	15.0	15.0	15.0	30000.0	25.0
					180	EC-21-7-C	2000	13.0	13.0	13.0	26000.0	35.0
14	Na2 EDTA	5	0.8	25	0	EC-14-1-C	150	58.1	56.0	56.3	8520.0	
					30	EC-14-2-C	150	45.2	44.5	44.7	6720.0	21.1
					60	EC-14-3-C	150	34.9	33.9	33.2	5100.0	40.1
					90	EC-14-4-C	150	23.8	23.7	23.7	3560.0	58.2
					120	EC-14-5-C	150	7.8	7.7	7.6	1155.0	86.4
					150	EC-14-6-C	150	5.0	4.4	5.0	720.0	91.5
					180	EC-14-7-C	1	59.4	59.3	59.7	59.5	99.3

TABLE 3-3. (cont.)

Run Number	Chelating Agent	Initial Starting pH	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (min)	Cathode Chamber Sample No.	Dilution Ratio For Analysis	Measured Lead Concentration In Cathode Chamber (mg/L) Sample # 1	Sample # 2	Sample # 3	Avg Lead Concentration In Cathode Chamber (mg/L)	Amount of Lead Removed (%)
15	Na2 EDTA	5	0.8	25	0	EC-15-1-C	250	33 0	32.0	32.0	8083.3	
	(Regenerated From				30	EC-15-2-C	250	34 0	35.0	35.0	8666.7	-7.2
	Run No. 14)				60	EC-15-3-C	250	29 0	29.0	29.0	7250.0	10 3
					90	EC-15-4-C	250	24 0	23.0	23.0	5833.3	27.8
					120	EC-15-5-C	250	15 0	15.0	15.0	3750.0	53.6
					150	EC-15-6-C	250	7 0	7.0	7.0	1750.0	78.4
					180	EC-15-7-C	125	6 0	6.0	6 0	750.0	90.7
4	DTPA	9	0.8	15	0	EC-4-1-C	250	32.6	32.8	32.8	8183.3	
					30	EC-4-2-C	125	59.0	59 4	59.7	7420.8	9.3
					60	EC-4-3-C	125	53.5	53.7	54.1	6720.8	17.9
					90	EC-4-4-C	125	25.3	25.4	25.5	3175.0	61.2
					120	EC-4-5-C	125	42.9	43.0	43.1	5375.0	34.3
					180	EC-4-6-C	125	31.4	31.5	31.5	3933.3	51.9
					240	EC-4-7-C	125	21 2	21.2	21.1	2645.8	67.7
					300	EC-4-8-C	125	6 9	6.9	6.9	862.5	89.5
6	DTPA	9	0.8	15	0	EC-6-1-C	250	35.1	35.2	35.2	8791.7	
	(Regenerated From				30	EC-6-2-C	125	61 9	61.7	62.0	7733.3	12.0
	Run No. 4)				60	EC-6-3-C	125	57 9	57.8	58.4	7254.2	17.5
					90	EC-6-4-C	125	49 9	49 5	49 4	6200.0	29.5
					120	EC-6-5-C	125	42 1	42 1	42.2	5266.7	40.1
					150	EC-6-6-C	125	33 3	33.5	33.4	4175.0	52.5
					180	EC-6-7-C	125	24 4	24 7	24.7	3075.0	65.0
17	DTPA	9	0.8	15	0	EC-17-1-C	250	32 0	32.0	32 0	8000.0	
	(Cadmium				30	EC-17-2-C	250	30 0	29 0	29 0	7333.3	8 3
	Electrodes)				60	EC-17-3-C	250	26 0	26 0	26 0	6500 0	18 8
					90	EC-17-4-C	250	24 0	24 0	24.0	6000 0	25.0
					120	EC-17-5-C	250	21 0	21.0	21 0	5250.0	34 4
					150	EC-17-6-C	250	17 0	18.0	18.0	4416.7	44.8
					180	EC-17-7-C	250	15.0	15.0	15.0	3750.0	53.1

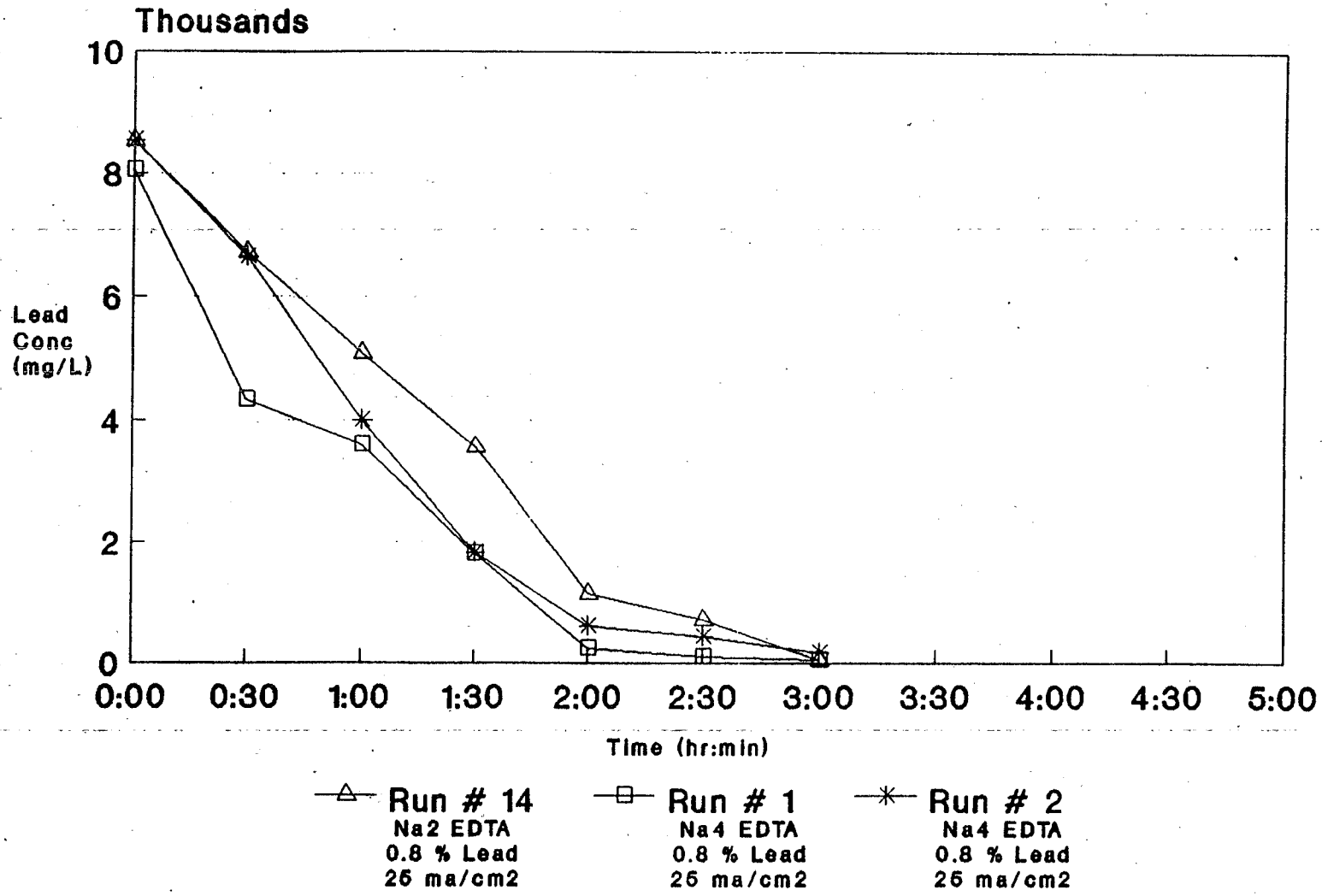
TABLE 3.3 (cont.)

Run Number	Chelating Agent	Initial Starting pH	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (min)	Cathode Chamber Sample No.	Dilution Ratio For Analysis	Measured Lead Concentration In Cathode Chamber (mg/L) Sample # 1	Sample # 2	Sample # 3	Avg Lead Concentration In Cathode Chamber (mg/L)	Amount of Lead Removed (%)
23	DTPA	9	0.8	15	0	EC-23-1-C	250	32.0	32.0	32.0	8000.0	
	(Ionics				30	EC-23-2-C	250	35.0	34.0	34.0	8583.3	-7.3
	Membrane)				60	EC-23-3-C	250	32.0	33.0	33.0	8166.7	-2.1
					90	EC-23-4-C	250	33.0	33.0	33.0	8250.0	-3.1
					120	EC-23-5-C	250	29.0	29.0	29.0	7250.0	9.4
					150	EC-23-6-C	250	26.0	26.0	26.0	6500.0	18.8
					180	EC-23-7-C	250	23.0	23.0	23.0	5750.0	28.1
24	DTPA	9	0.8	15	0	EC-24-1-C	250	31.0	30.0	30.0	7583.3	
	(Ionics				30	EC-24-2-C	250	29.0	29.0	29.0	7250.0	4.4
	Membrane)				60	EC-24-3-C	250	26.0	26.0	26.0	6500.0	14.3
					90	EC-24-4-C	250	24.0	24.0	24.0	6000.0	20.9
					120	EC-24-5-C	250	21.0	21.0	21.0	5250.0	30.8
					150	EC-24-6-C	250	18.0	18.0	18.0	4500.0	40.7
					180	EC-24-7-C	250	16.0	16.0	16.0	4000.0	47.3
5	DTPA	9	0.8	25	0	EC-5-1-C	250	33.3	33.2	33.6	8341.7	
					30	EC-5-2-C	125	57.9	57.7	57.8	7225.0	13.4
					60	EC-5-3-C	125	46.3	46.4	46.4	5795.8	30.5
					90	EC-5-4-C	125	37.2	36.8	37.1	4629.2	44.5
					120	EC-5-5-C	125	28.2	28.0	27.9	3504.2	58.0
					150	EC-5-6-C	75	43.1	43.3	43.1	3237.5	61.2
					180	EC-5-7-C	25	47.3	47.4	47.2	1182.5	85.8
7	DTPA	12	0.8	25	0	EC-7-1-C	150	56.2	56.4	56.5	8455.0	
					30	EC-7-2-C	150	53.9	53.8	53.5	8060.0	4.7
					60	EC-7-3-C	150	9.1	9.0	9.2	1365.0	83.9
					90	EC-7-4-C	150	33.4	33.0	33.3	4985.0	41.0
					120	EC-7-5-C	150	25.2	25.3	25.3	3790.0	55.2
					150	EC-7-6-C	150	18.9	18.6	18.6	2805.0	66.8
					180	EC-7-7-C	150	23.7	23.7	23.7	3555.0	58.0
					240	EC-7-8-C	150	5.4	5.4	5.4	810.0	90.4
					300	EC-7-9-C	75	6.2	6.3	6.3	470.0	94.4
11	DTPA	9	4	15	0	EC-11-1-C	1500	28.3	28.3	28.2	42400.0	
					30	EC-11-2-C	1500	28.4	28.5	28.4	42650.0	-0.6
					60	EC-11-3-C	1500	27.8	27.8	27.8	41700.0	1.7
					90	EC-11-4-C	1500	28.1	28.2	28.4	42350.0	0.1
					120	EC-11-5-C	1500	27.4	27.6	27.6	41300.0	2.6
					150	EC-11-6-C	1500	25.4	25.4	25.4	38100.0	10.1
					180	EC-11-7-C	1500	30.8	30.8	30.7	46150.0	-8.8
					210	EC-11-8-C	1500	22.9	22.9	23.2	34500.0	18.6
					240	EC-11-9-C	1500	21.7	21.8	21.8	32650.0	23.0
					300	EC-11-10-C	1000	27.9	27.0	27.1	27333.3	35.5
8	DTPA	9	4	25	0	EC-8-1-C	600	64.8	65.0	65.2	39000.0	
					30	EC-8-2-C	600	63.3	62.0	63.0	37660.0	3.4
					60	EC-8-3-C	600	59.6	59.2	59.6	35680.0	8.5
					90	EC-8-4-C	600	56.1	56.1	56.1	33660.0	13.7

TABLE 3-3. (cont.)

Run Number	Chelating Agent	Initial Starting pH	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (min)	Cathode Chamber Sample No.	Dilution Ratio For Analysis	Measured Lead Concentration In Cathode Chamber (mg/L) Sample # 1	Sample # 2	Sample # 3	Avg Lead Concentration In Cathode Chamber (mg/L)	Amount of Lead Removed (%)
8	DTPA	9	4	25	120	EC-8-5-C	600	55.1	55.0	55.1	33040.0	15.3
					150	EC-8-6-C	600	46.7	46.8	46.6	28020.0	28.2
					180	EC-8-7-C	600	43.0	43.0	43.1	25820.0	33.8
9	DTPA	9	4	25	0	EC-9-1-C	600	64.1	63.9	64.4	38480.0	
					30	EC-9-2-C	600	63.8	63.9	64.2	38380.0	0.3
					60	EC-9-3-C	600	59.4	59.6	59.4	35680.0	7.3
					90	EC-9-4-C	600	48.7	48.8	48.8	29260.0	24.0
					120	EC-9-5-C	600	46.2	46.0	46.1	27660.0	28.1
					150	EC-9-6-C	600	45.7	45.9	45.9	27500.0	28.5
					180	EC-9-7-C	600	38.1	38.5	38.4	23000.0	40.2
					240	EC-9-8-C	?	?	?	?	0.0	
					300	EC-9-9-C	600	23.6	23.6	23.5	14140.0	63.3
10	DTPA (Regenerated From Run No. 9)	9	4	25	0	EC-10-1-C	1000	60.0	59.8	59.9	59900.0	
					30	EC-10-2-C	1000	59.9	60.1	60.0	60000.0	-0.2
					60	EC-10-3-C	1000	59.2	59.0	59.2	59133.3	1.3
					90	EC-10-4-C	1000	57.9	57.7	57.5	57700.0	3.7
					120	EC-10-5-C	1000	52.6	52.7	52.7	52666.7	12.1
					150	EC-10-6-C	1000	48.9	49.1	49.2	49066.7	18.1
					180	EC-10-7-C	1000	43.9	43.8	44.1	43933.3	26.7
					240	EC-10-8-C	1000	35.0	34.0	35.0	34666.7	42.1
16	DTPA (Cadmium Electrodes)	9	4	25	0	EC-16-1-C	800	51.0	52.0	53.0	41600.0	
					30	EC-16-2-C	800	57.0	57.0	57.0	45600.0	-9.6
					60	EC-16-3-C	800	46.0	47.0	47.0	37866.7	9.0
					90	EC-16-4-C	800	43.0	43.0	43.0	34400.0	17.3
					120	EC-16-5-C	800	39.0	39.0	39.0	31200.0	25.0
					150	EC-16-6-C	800	36.0	35.0	35.0	28266.7	32.1
					180	EC-16-7-C	800	31.0	30.0	31.0	24533.3	41.0
22	DTPA (Ionics Membrane)	9	4	25	0	EC-22-1-C	1800	23.0	23.0	23.0	41400.0	
					30	EC-22-2-C	1800	26.0	25.0	25.0	45600.0	-10.1
					60	EC-22-3-C	1800	22.0	22.0	23.0	40200.0	2.9
					90	EC-22-4-C	1800	21.0	20.0	20.0	36600.0	11.6
					120	EC-22-5-C	1800	19.0	19.0	19.0	34200.0	17.4
					150	EC-22-6-C	1800	17.0	17.0	17.0	30600.0	26.1
					180	EC-22-7-C	1800	15.0	15.0	15.0	27000.0	34.8

Figure 3-1. Comparison of Lead Removal Rates for the Two Sodium Forms of EDTA



and with 25 ma/cm^2 current density. Run 2 is a duplicate run conducted with the same lead content, current density, and chelating agent as Run 1. Run 14 was conducted with di-sodium EDTA, 0.8 percent lead content, and 25 ma/cm^2 current density. Figure 3-1 shows similar lead removal rates for di-sodium and tetra-sodium EDTA solutions. Most lead removal occurs within 2 hours for both forms of EDTA, and little additional lead removal is observed after 2 hours. According to the data in Table 3-3, the lead removal for tetra-sodium EDTA in Run 1 was 96.9 percent after 2 hours of plating, and 99.4 percent after 3 hours. In Run 2, the lead removal was 92.7 percent after two hours of plating, and 98.0 percent after 3 hours. Di-sodium EDTA produced an 86.4 percent lead removal rate after 2 hours of plating, and 99.3 percent after 3 hours.

The lead removal efficiencies for regenerated chelating agent solutions are compared with those of fresh chelating agent solutions in Figures 3-2 and 3-3. Figure 3-2 shows the lead removal rates for tests conducted with di-sodium EDTA solution containing 0.8 percent lead and with 25 ma/cm^2 current density. Based on this figure, it is noteworthy that the regenerated lead di-sodium EDTA solution achieved a lead removal rate that is comparable to the original di-sodium EDTA solution. Based on the data in Table 3-3, the test performed with di-sodium EDTA produced 99.3 percent lead removal in the original solution (Run 14) and 90.7 percent lead removal in the regenerated solution (Run 15). The slightly greater lead removal in Run 14 may be attributed to the higher lead concentration in the starting solution (8,520 mg/L lead in Run 14 solution and 8,083 mg/L lead in Run 15). Figure 3-3 depicts the lead removal rates for tests performed with fresh (Run 2) and regenerated (Run 3) tetra-sodium EDTA solutions containing 0.8 percent lead and with 25 ma/cm^2 current density. This figure shows that very similar lead removal rates were achieved for the original tetra-sodium EDTA solution and the regenerated tetra-sodium EDTA solution. The original tetra-sodium EDTA solution (Run 2) produced 98 percent lead removal, compared to 99.3 percent lead removal in the regenerated solution (Run 3). Figure 3-4 shows the lead removal rates for tests conducted with tetra-sodium EDTA containing 4 percent lead and with 25 ma/cm^2 current density. The overall lead removal rate was only 18.2 percent for the test conditions associated with Run 12. For the test run involving the generated solution resulting from Run 12 (i.e., Run 13), the lead removal rate

Figure 3-2. Comparison of Lead Removal Rates for Fresh and Regenerated Solutions of Di-Sodium EDTA

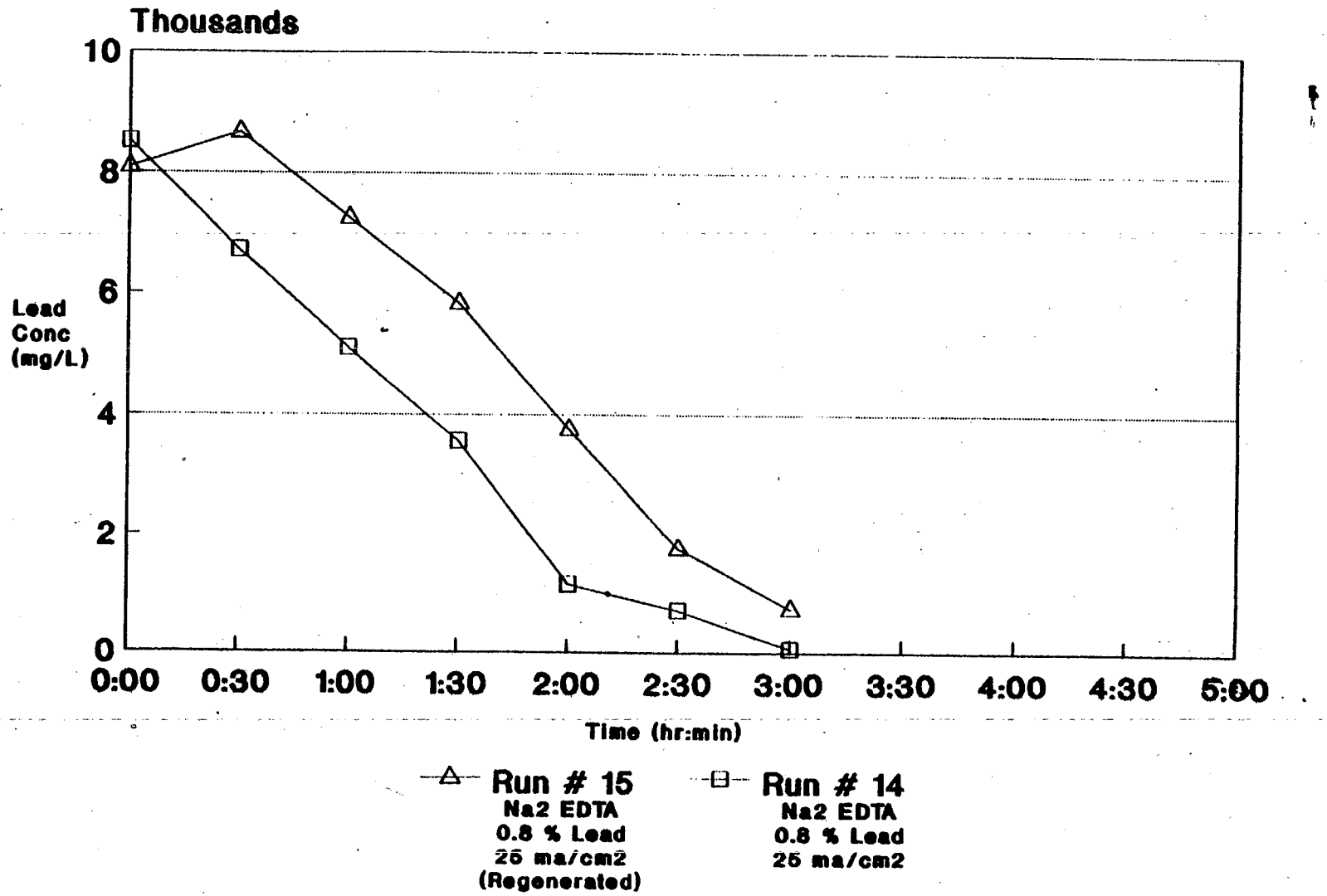


Figure 3-3. Comparison of Lead Removal Rates for Fresh and Regenerated Solutions of Tetra-Sodium EDTA Containing 0.8 Percent Lead

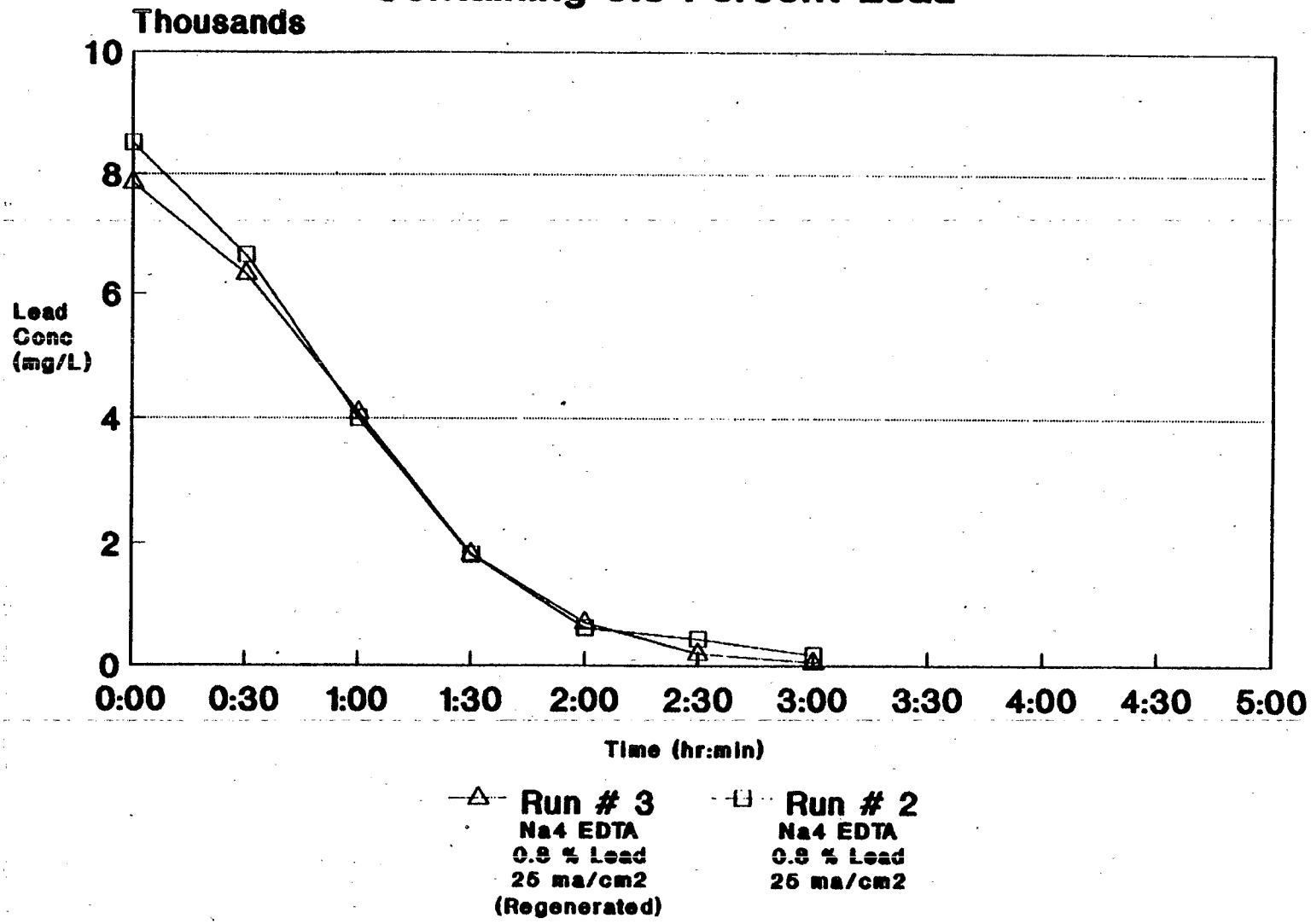
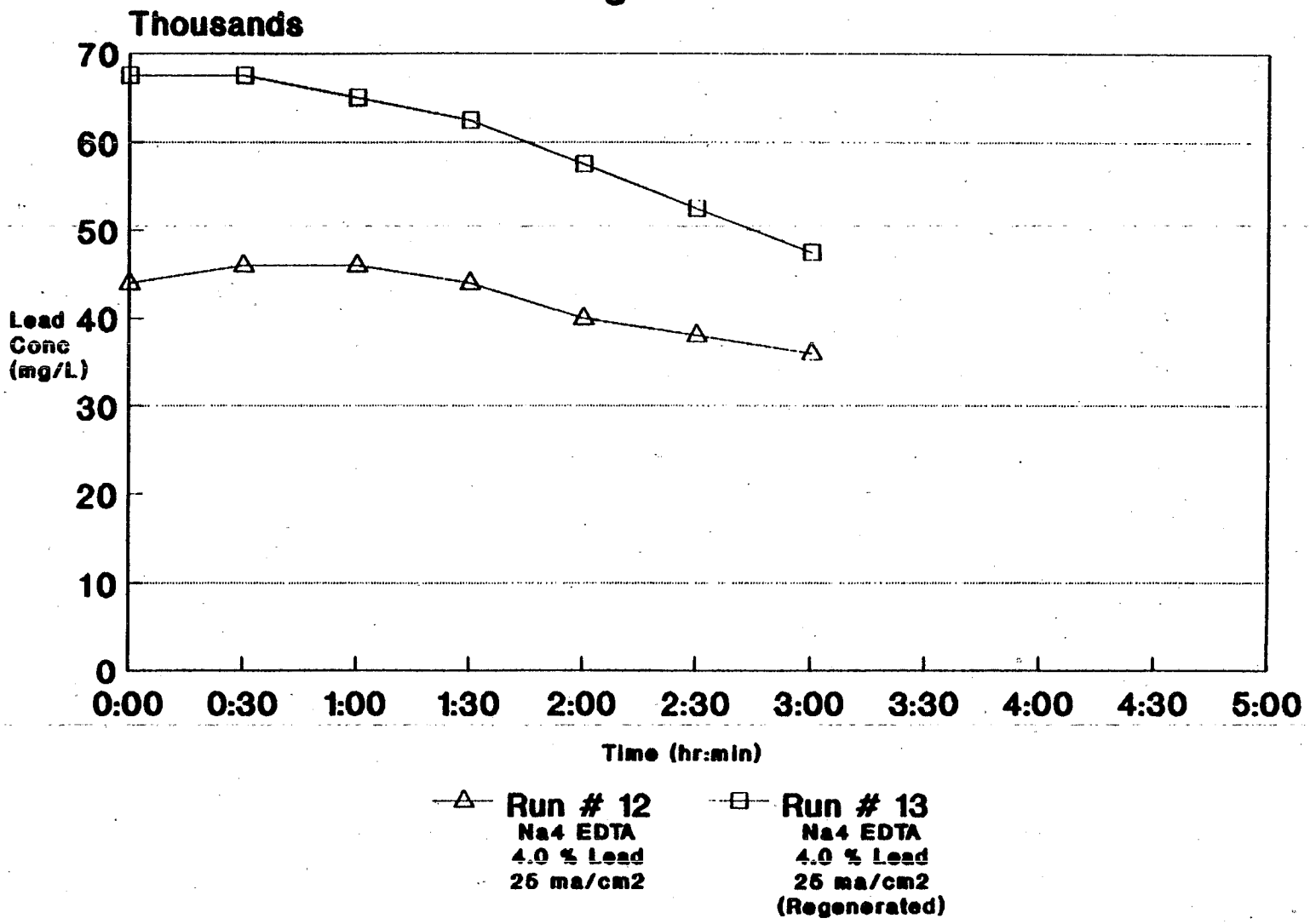


Figure 3-4. Comparison of Lead Removal Rates for Fresh and Regenerated Solutions of Tetra-Sodium EDTA Containing 4 Percent Lead



increased to 29.6 percent. Based on Figure 3-4, it appears that the lead removal rate was faster for the regenerated solution than in the original solution. The faster lead removal rate for the regenerated solution is attributed to a higher lead concentration gradient, and therefore faster lead plating.

Figure 3-5 presents a comparison of the lead removal rates for the tests using DTPA solutions containing 0.8 percent lead and with 15 ma/cm^2 current density. According to this figure and the data presented in Table 3-3, the lead removal rates for the original and regenerated DTPA solutions (Runs 4 and 6, respectively) are very similar. In Run 4, 52 percent lead removal was observed after 3 hours plating time; 65.0 percent lead removal was observed in Run 6 after a 3-hour plating time. Figure 3-6 presents a comparison of the lead removal rates for the tests using DTPA solutions containing 4 percent lead and with 25 ma/cm^2 current density. According to this figure, the percent lead removal is 40 percent for Run 9 and 27 percent for Run 10 after 3 hours plating time. The data obtained using the regenerated solutions shows the potential of the electromembrane reactor to regenerate the sodium form of the chelating agent for reuse. Figure 3-6 presents a comparison of the lead removal rates for the tests using DTPA solutions containing 4 percent lead and 25 ma/cm^2 current density. According to this figure and the data presented in Table 3-3, the percent lead removal after a 3-hour plating time was 40 percent for the test conducted with fresh solution (Run 9) and 27 percent for the test conducted with regenerated solution (Run 10). Because 37 percent of the lead was not removed in Run 9 (after a run time of 5 hours), the initial concentration of the regenerated solution for Run 10 was higher than the target concentration of 4 percent. Similarly, the initial lead concentration of the regenerated solution in Run 13 was also higher than the target concentration of 4 percent.

Figure 3-7 shows a comparison of lead concentration and lead removal rate for tests conducted with DTPA solutions containing an initial concentration of 0.8 (Run 4) and 4 (Run 11) percent lead and with 15 ma/cm^2 current density. A higher percentage of lead was removed after 5 hours for the 0.8 percent lead solution (90 percent for 0.8 percent lead concentration solution versus 36 percent lead removal for the 4 percent lead solution); however, a higher quantity of lead was plated from the 4 percent lead solution.

Figure 3-5. Comparison of Lead Removal Rates for Fresh and Regenerated Solutions of DTPA Containing 0.8 Percent Lead

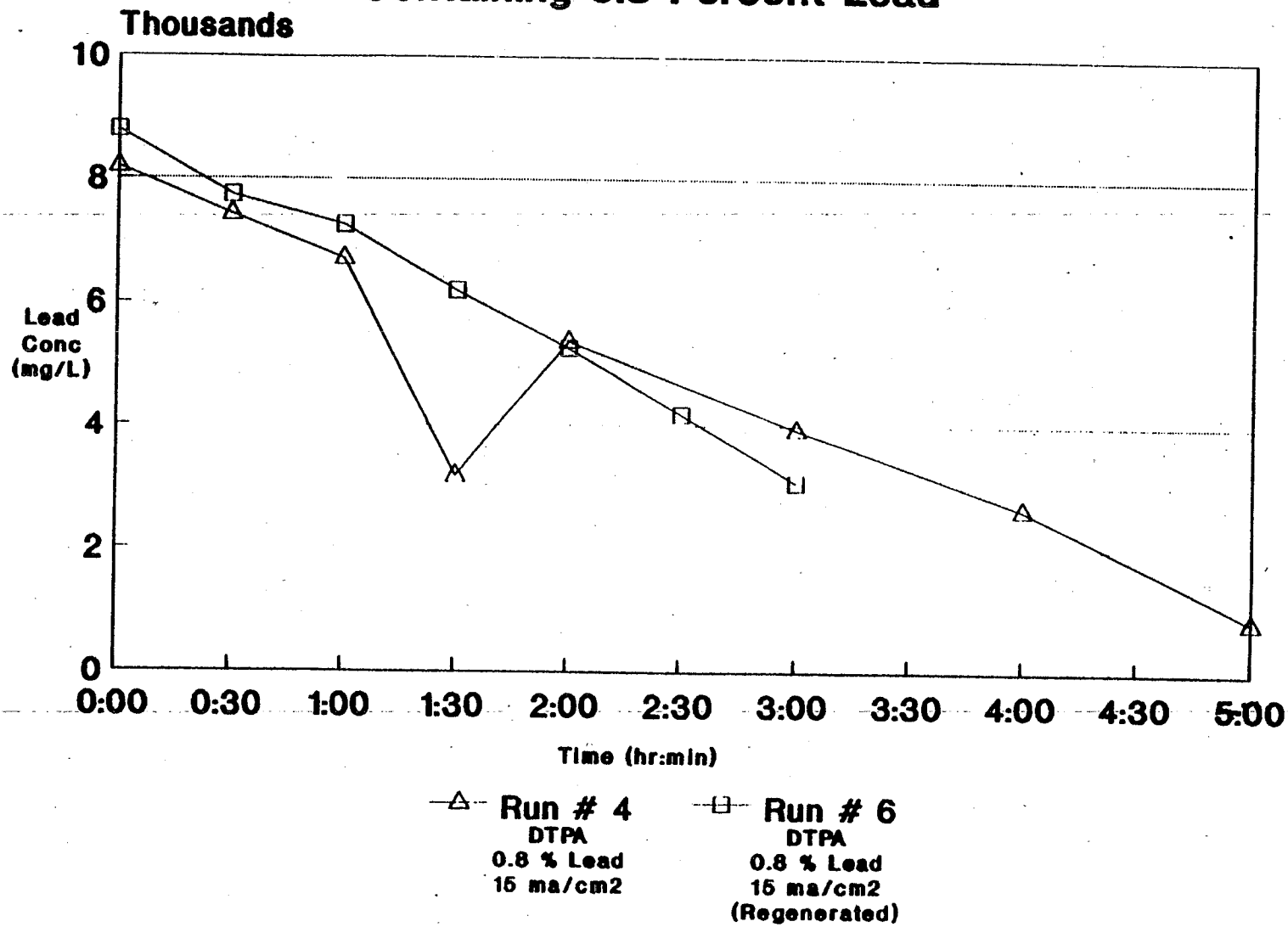


Figure 3-6. Comparison of Lead Removal Rates for Fresh and Regenerated Solutions of DTPA Containing 4 Percent Lead

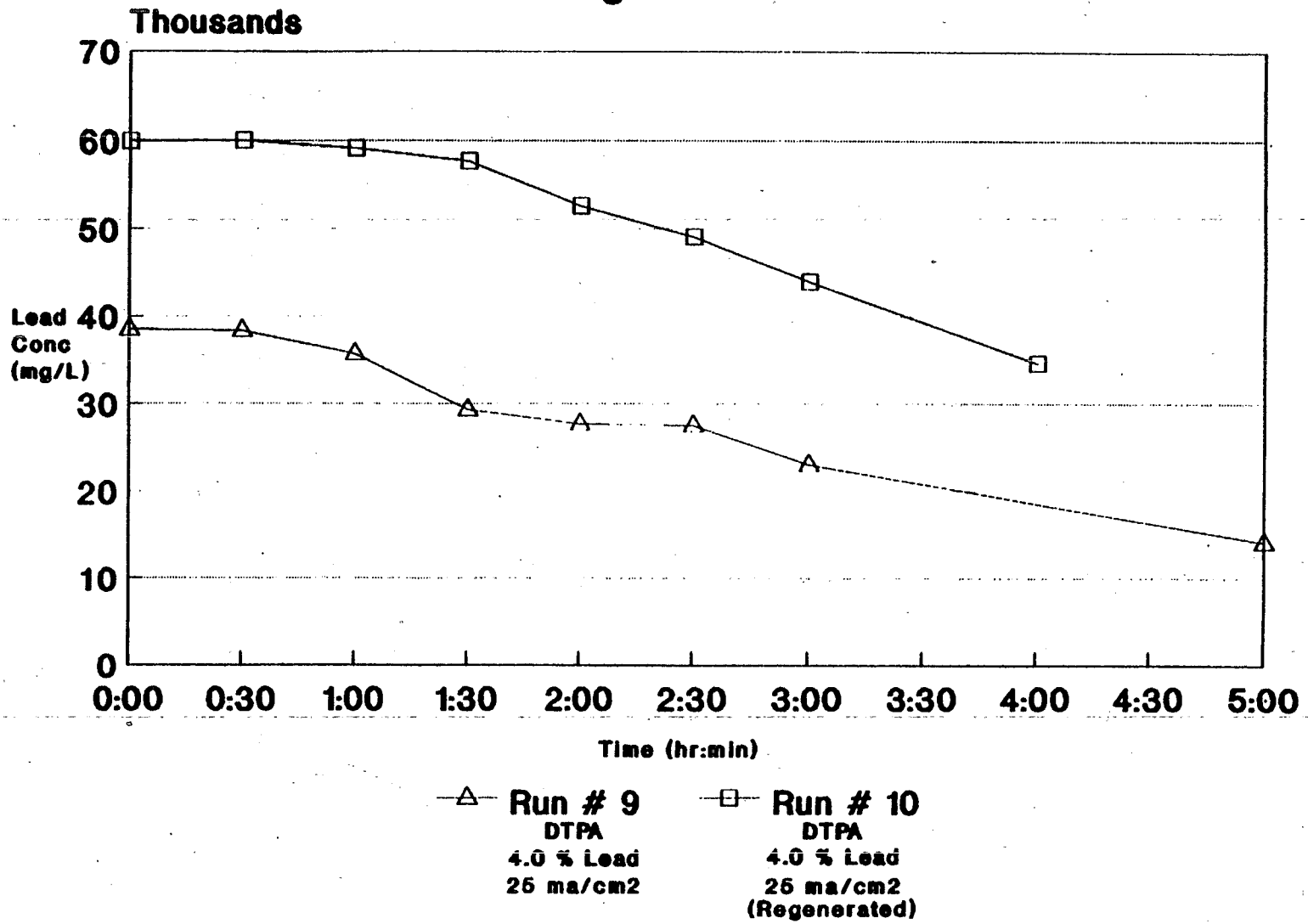
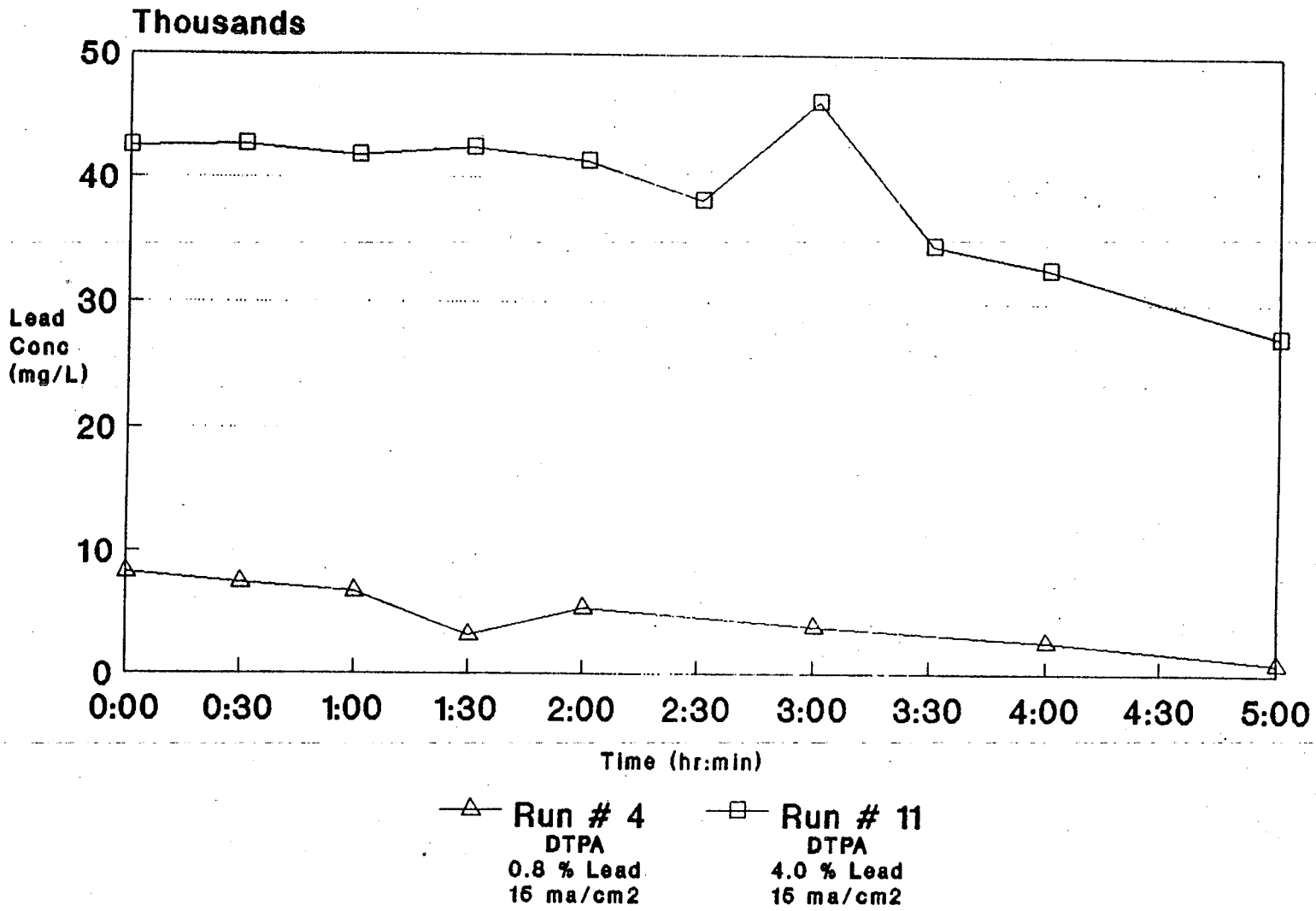


Figure 3-7. Comparison of Lead Removal Rates for DTPA Using Two Initial Lead Concentrations



In Figure 3-8, the two current densities are compared for tests performed using DTPA solutions containing 0.8 percent lead and with 15 and 25 ma/cm^2 current densities. As expected, the lead recovery rate for the tests using the 25 ma/cm^2 current density were higher than for the 15 ma/cm^2 current density test. The lead recovery rate in the run with a starting lead concentration of 0.8 percent and a 15 ma/cm^2 current density (Run 4) was 51.9 percent after a 3-hour plating time, and 85.8 and 58.0 percent lead recoveries were attained in the 25 ma/cm^2 current density test (Runs 5 and 7). Figure 3-9 presents a comparison of lead removal rates for DTPA solutions containing 4 percent lead and with 15 and 25 ma/cm^2 current densities. A higher lead removal percentage was also observed in the test conducted with a 25 ma/cm^2 current density and 4 percent lead DTPA solution. A 5-hour reaction period was required for the 15 ma/cm^2 current density test (35.5 percent lead removal in Run 11) to equal the lead removal of the 25 ma/cm^2 test at 3 hours (33.8 and 40.2 percent lead removal in Runs 8 and 9, respectively). Therefore, as expected, a higher current density will produce higher lead recoveries than the lower current density.

In Figure 3-10, the lead removal rates for the tests using the three chelating agents (tetra-sodium EDTA, di-sodium EDTA, and DTPA) are compared using a 0.8 percent lead concentration solution and a 25 ma/cm^2 current density. Based on this figure and the data presented in Table 3-3, it appears that the highest lead removals occur for the tests conducted with EDTA solutions after a 3-hour plating time (99.3 percent for di-sodium EDTA in Run 14 and 99.4 percent for tetra-sodium EDTA in Run 1). Run 2 was conducted under the same conditions as Run 1 and produced similar results (98.0 percent removal after a 3-hour plating time). The tests with DTPA showed lower lead removals (85.8 percent in Run 5). Run 7 was performed under the same conditions, but produced a lower percent lead removal than Run 5. Because DTPA forms a more stable complex with lead, particularly in alkaline solutions, this may inhibit the removal of lead from the DTPA complex, and hence less lead is recovered in the same reaction period as with EDTA.

Lead removal rates for tests conducted with 4 percent lead concentration solutions of tetra-sodium EDTA and DTPA and 25 ma/cm^2 current density are

Figure 3-8. Comparison of Lead Removal Rates for DTPA Using Two Current Densities and 0.8 Percent Lead Concentrations

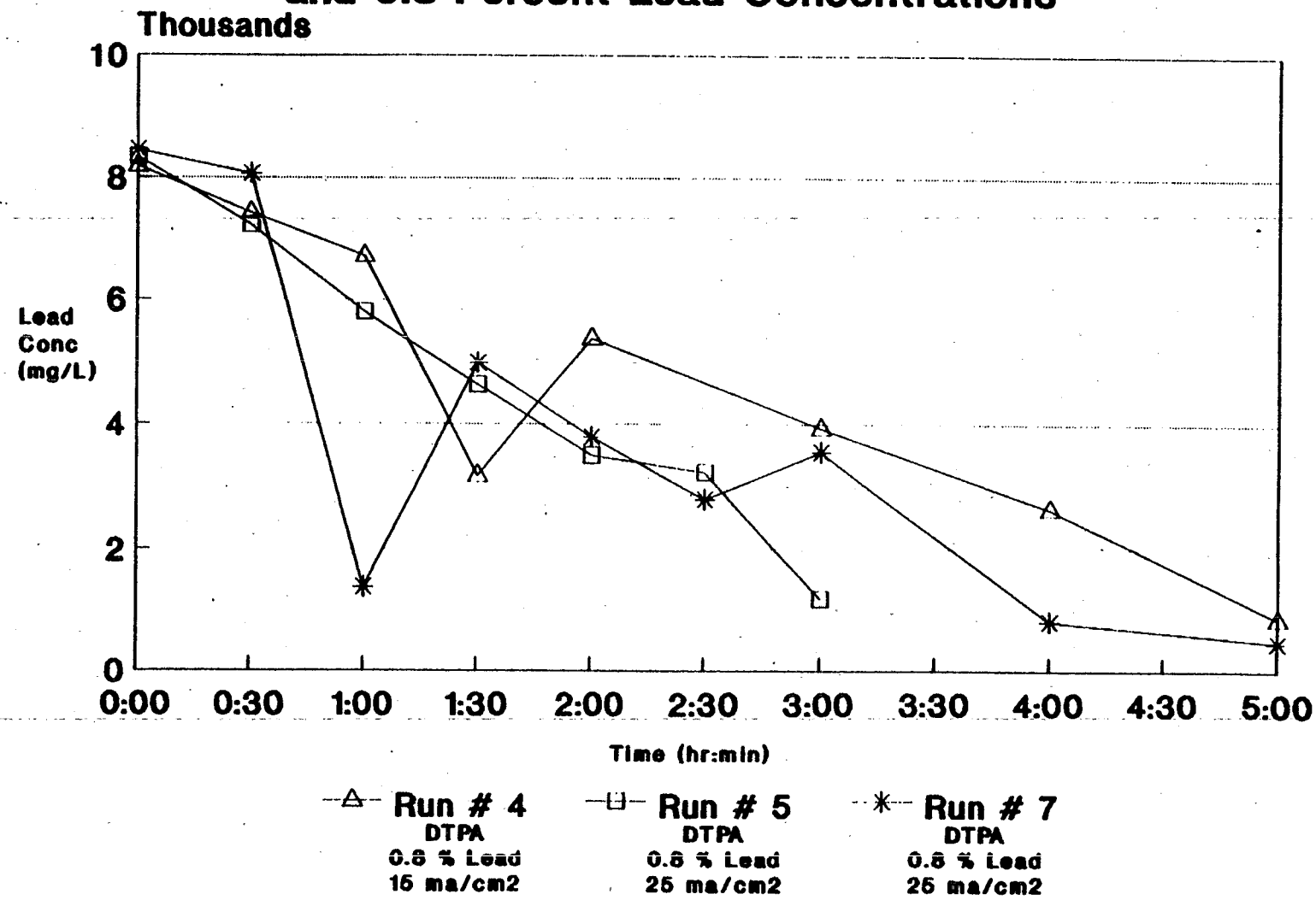


Figure 3-9. Comparison of Lead Removal Rates for DTPA Using Two Current Densities and 4 Percent Lead Concentrations

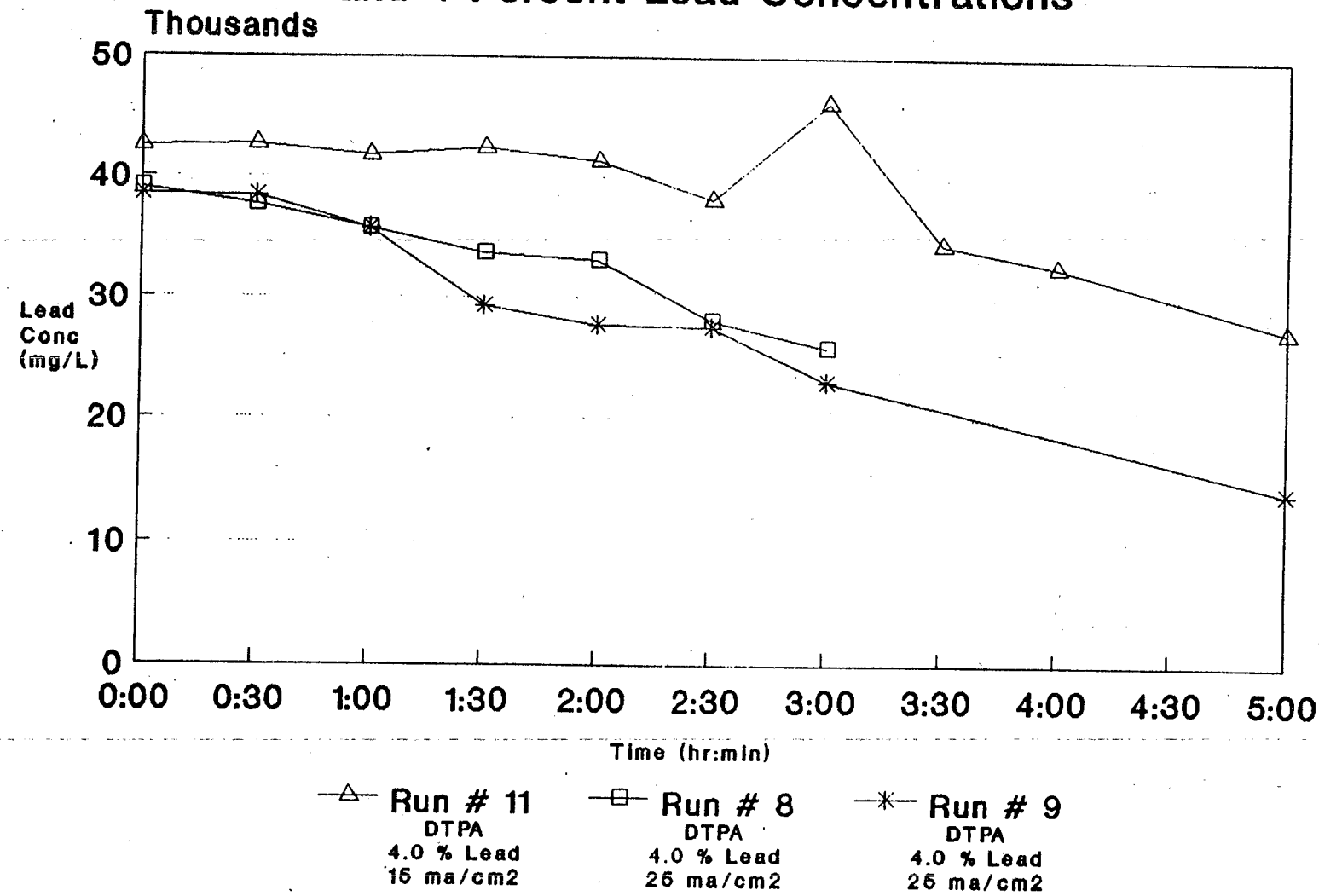
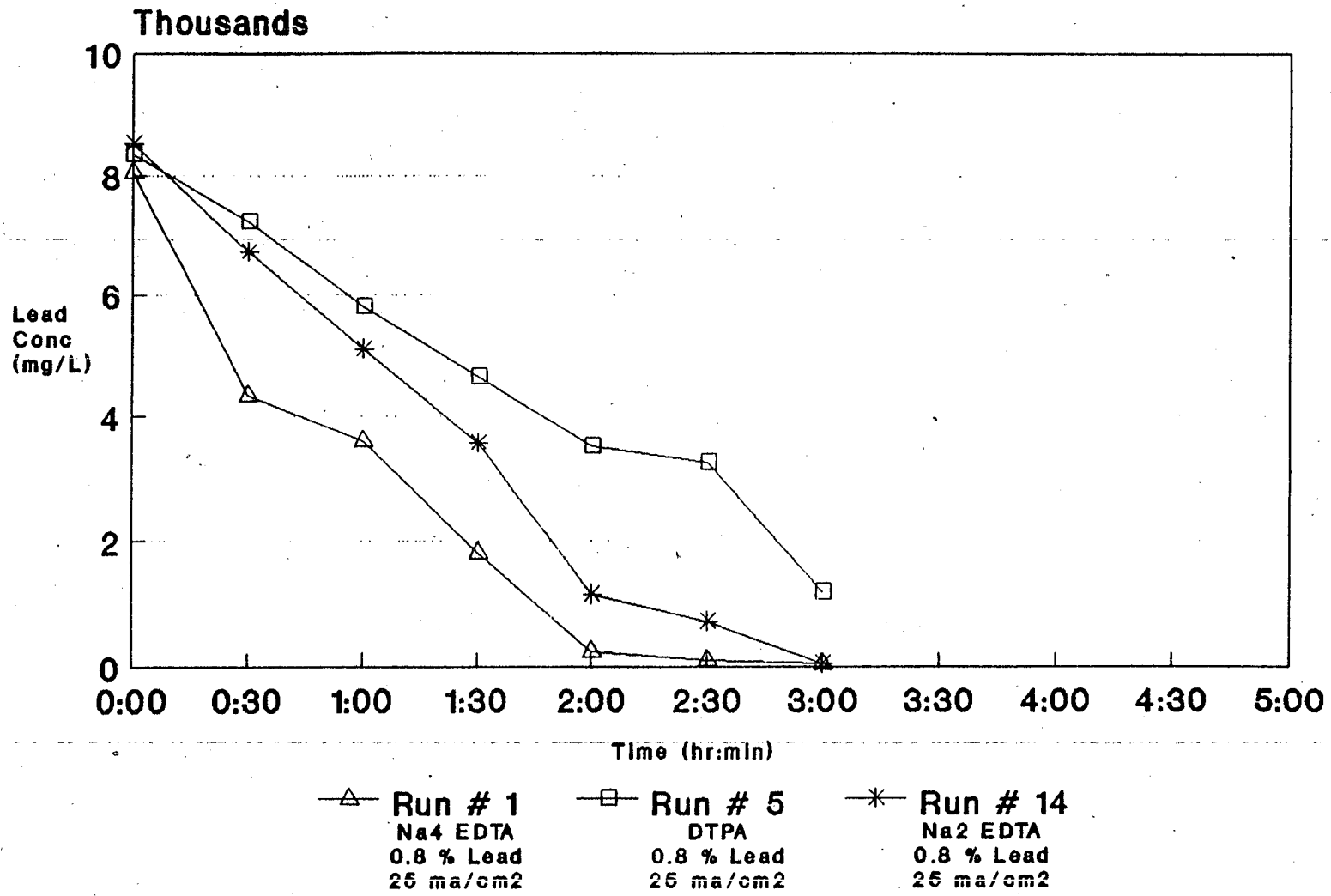


Figure 3-10. Comparison of Lead Removal Rates for Three Chelating Agents and 0.8 Percent Lead Concentrations



compared in Figure 3-11. According to this figure and the data presented in Table 3-3, the overall lead removal rates after a 3-hour plating time are 18.2 percent for tetra-sodium EDTA, Run 12, and 33.8 and 40.2 percent for DTPA, for Runs 8 and 9, respectively.

Figure 3-12 compares the lead removal rates for tests with the Nafion® and Ionics membranes using 0.8 percent lead concentration, DTPA solutions, and 15 ma/cm^2 current densities. The tests produced a 51.9, 28.1, and 47.3 percent lead removal rate after a 3-hour plating time with the Nafion® (Run 4) and Ionics (Runs 23 and 24) membranes, respectively. Figure 3-13 presents a comparison of lead removal rates for the Nafion® and Ionics membranes using 4 percent lead concentration DTPA solutions and 25 ma/cm^2 current densities. For the 4 percent lead concentration and 25 ma/cm^2 current density tests, the lead removal rates were 33.8, 40.2, and 34.8 percent for the Nafion® (Runs 8 and 9) and Ionics (Run 22) membranes, respectively. The data indicate that for both lead concentrations and current densities, the lead recovery for the tests conducted using the DuPont Nafion® membrane is comparable to the lead recovery for the Ionics membrane.

Figure 3-14 compares the lead removal rates for the tests conducted with the lead and cadmium electrodes using DTPA solutions with 0.8 and 4 percent lead concentrations and 15 and 25 ma/cm^2 current densities. Based on this figure, it appears that there is no appreciable difference in the lead removal rates for the tests performed with a 15 ma/cm^2 current density, and 0.8 percent lead concentration DTPA solutions. The lead electrode produced a 51.9 percent lead recovery (Run 4), whereas a 53.1 percent lead recovery was observed in the cadmium electrode test (Run 17). The experiments with 4 percent lead DTPA solutions and 25 ma/cm^2 current density also exhibited little difference in lead recovery for the two electrodes. In the tests using the lead electrodes, the lead removal rate was 33.8 percent (Run 8) and 40.2 percent (Run 9). The lead removal rate was 41 percent with the cadmium electrodes Run 16). According to Figure 3-15, however, the lead removal rate is greater using the cadmium electrodes than the lead electrodes for the 25 ma/cm^2 current density, and 4 percent lead concentration tetra-sodium EDTA solutions.

Figure 3-11. Comparison of Lead Removal Rates for Tetra-Sodium EDTA and DTPA and 4 Percent Lead Concentrations

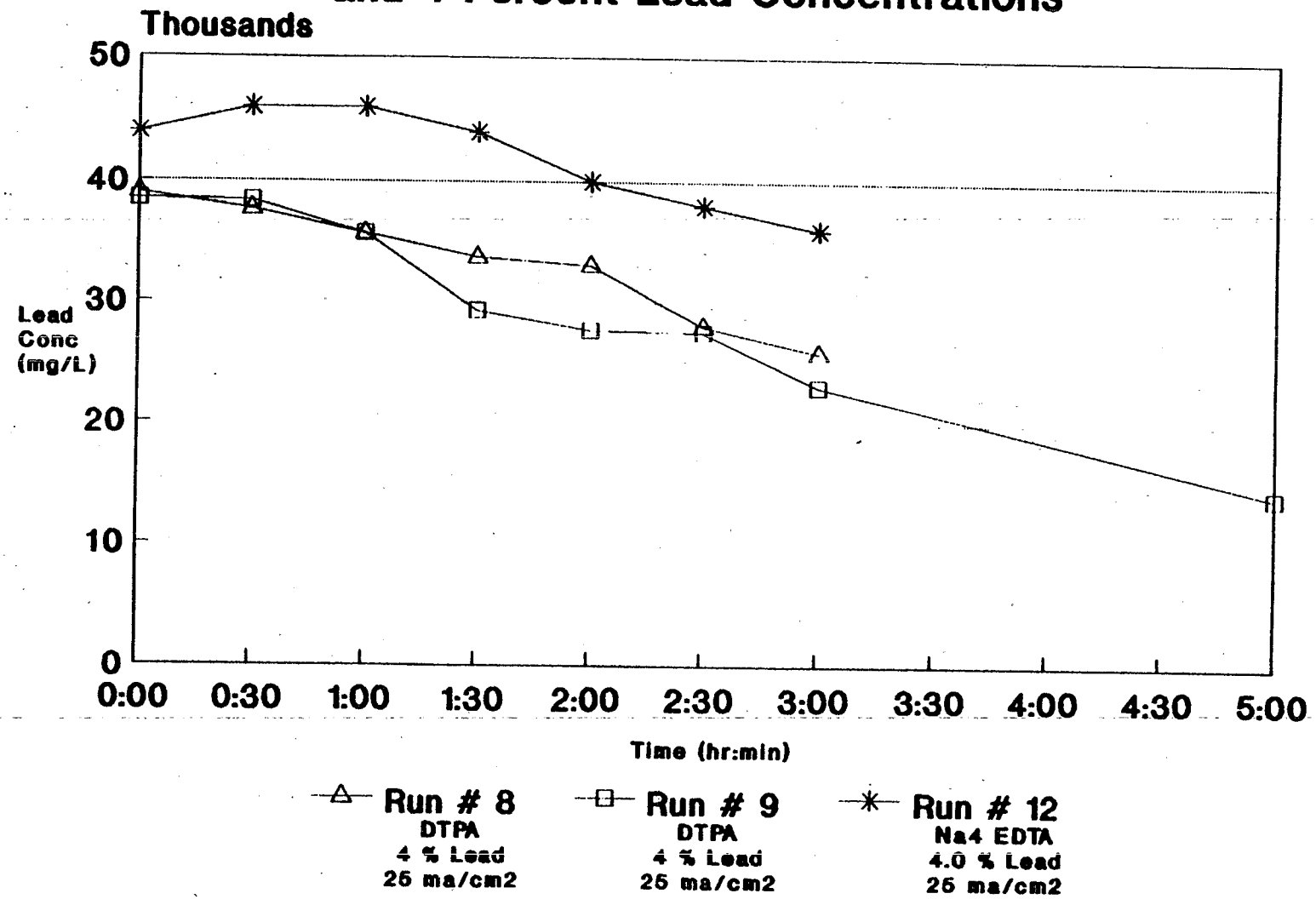


Figure 3-12. Comparison of Lead Removal Rates for Two Membranes and 0.8 Percent Lead Concentrations

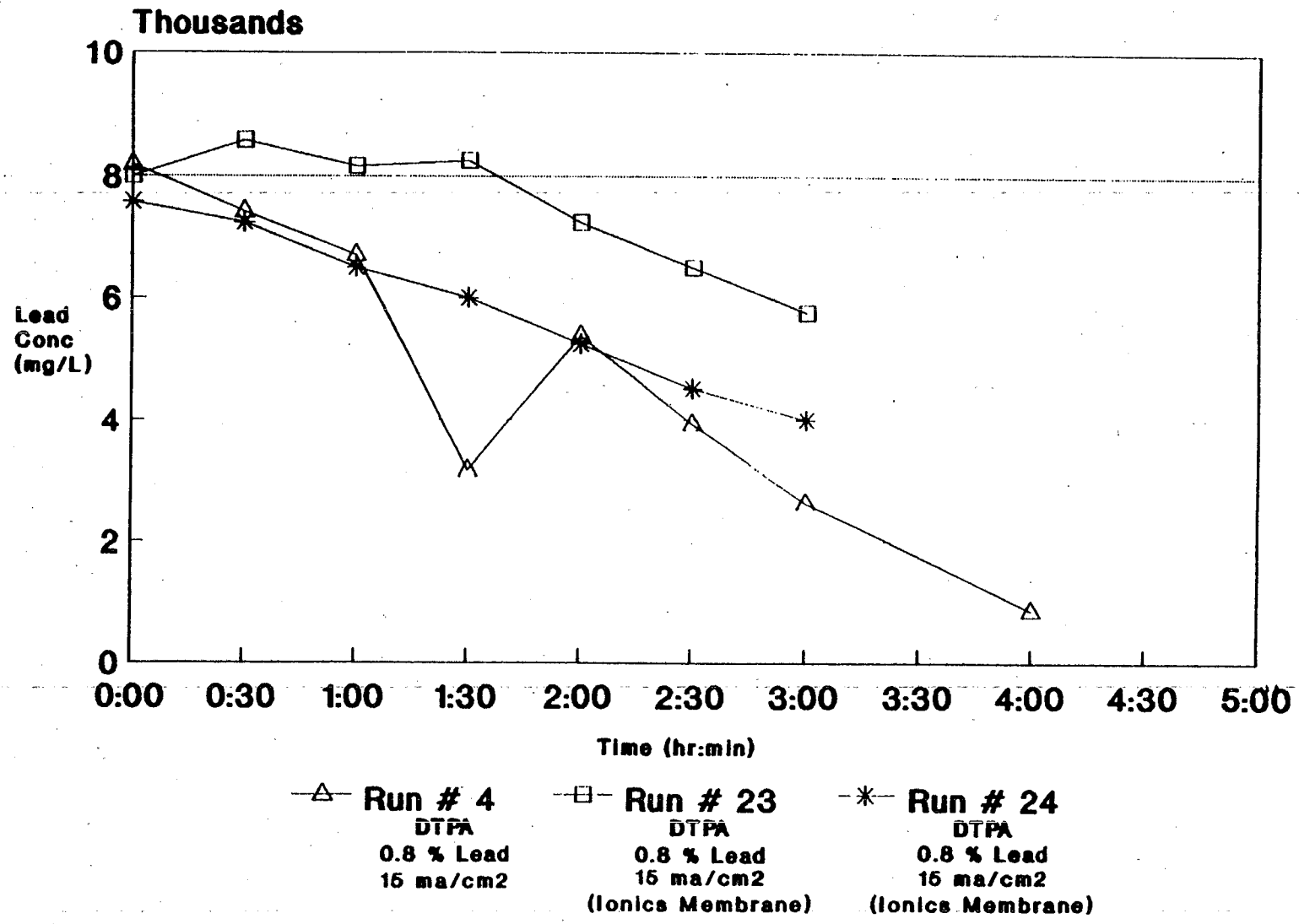


Figure 3-13. Comparison of Lead Removal Rates for Two Membranes and 4 Percent Lead Concentrations

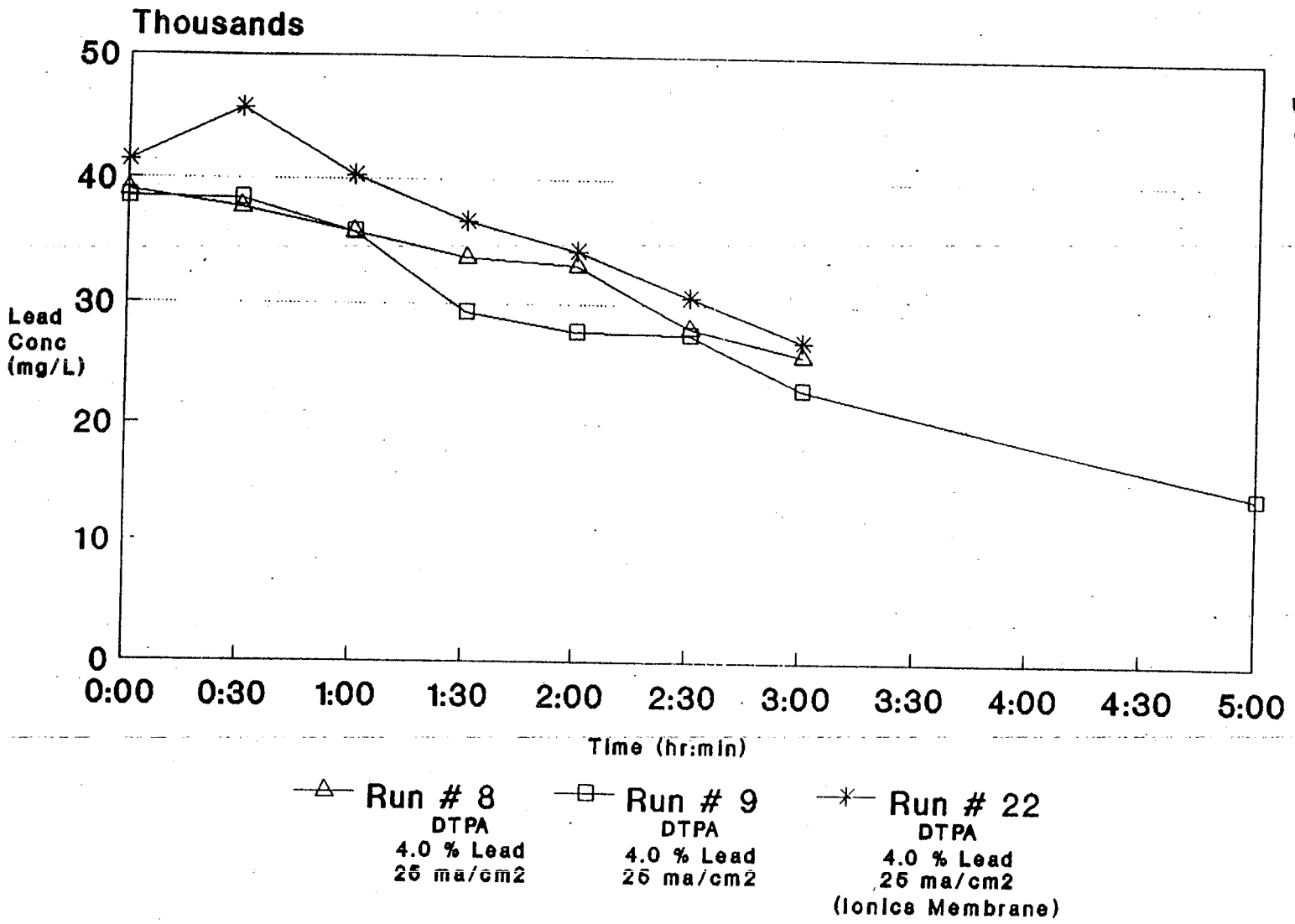


Figure 3-14. Comparison of Lead Removal Rates for DTPA Using Two Electrodes

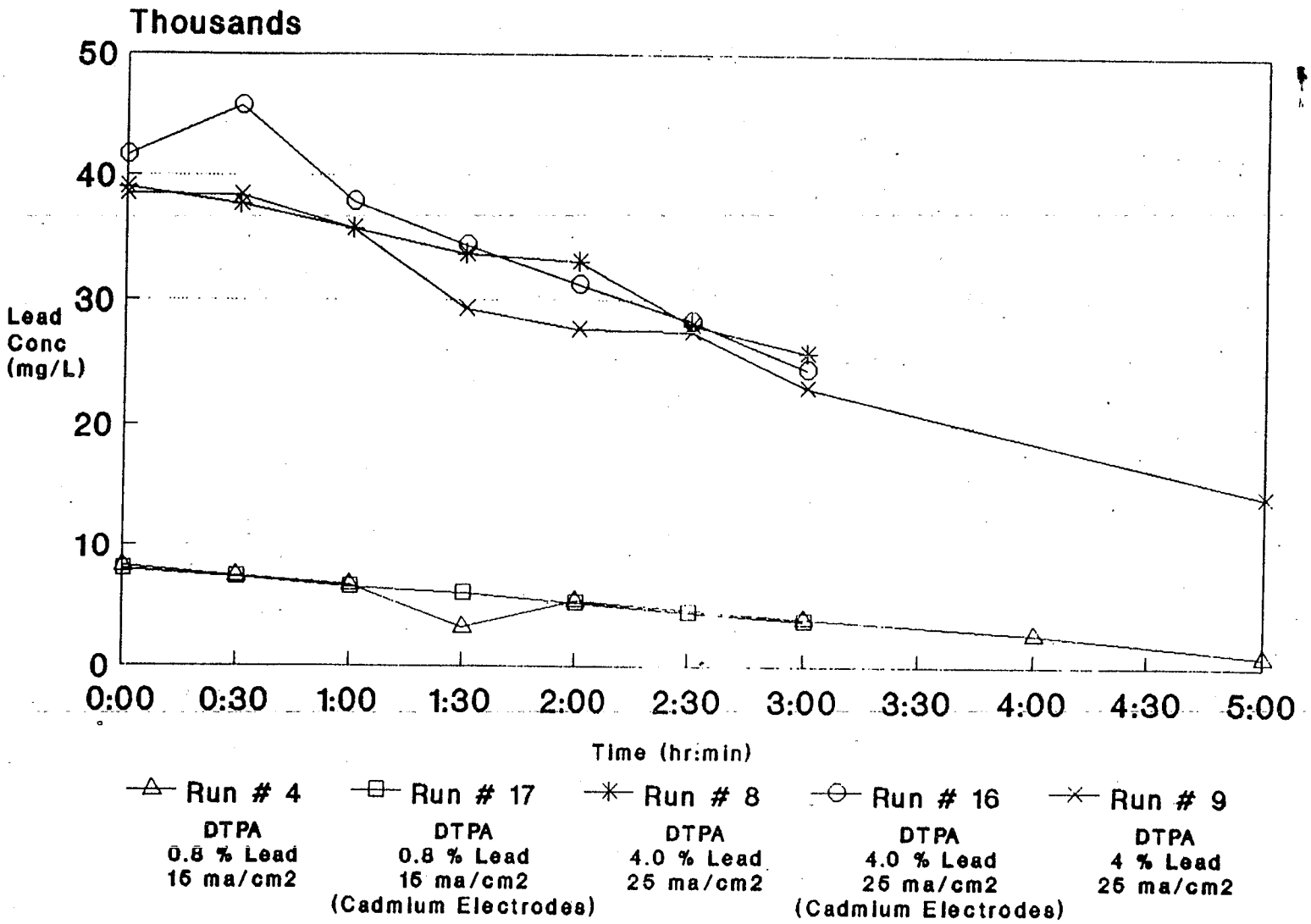
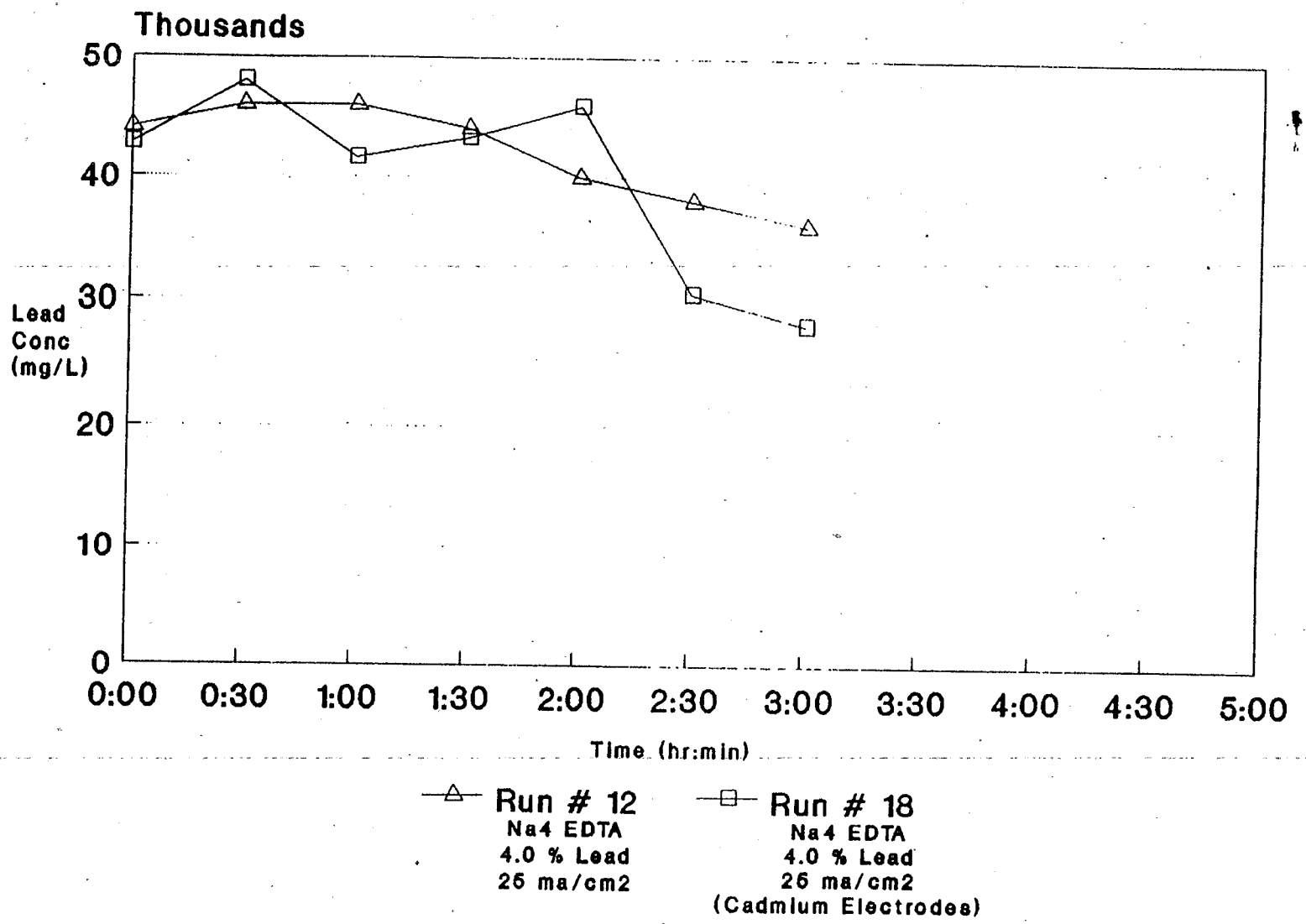


Figure 3-15. Comparison of Lead Removal Rates for EDTA Using Two Electrodes



The lead removal rate was 18.2 percent using the lead electrodes (Run 12), and 34.4 percent using the cadmium electrodes (Run 18). The increased amount of lead plated may result from the greater standard potential of cadmium than lead. The increased lead removal rate may be observed with the tetra-sodium EDTA solution because the stability constant of lead-EDTA is less than lead-DTPA, and therefore the lead is more easily stripped from the lead-chelate. The electrode potential required to plate metal on the cathode is affected by the type of complex formed when chelating agents are employed.

Figure 3-16 shows a comparison of lead removal rates for tests performed with tetra-sodium EDTA containing 4 percent lead concentration, 1.5 percent iron, 25 ma/cm^2 current density, and initial target pH levels of 7, 9, and 11.5 for Runs 19, 20, and 21, respectively. Based on this figure, it appears that the lead removal rate is slightly greater for solutions containing 1.5 percent iron than for the lead-EDTA solution without iron (Run 12). The lead removal rate was only 18.2 percent in Run 12, and 52.6, 54.7 and 35.0 percent in Runs 19, 20, and 21 with 1.5 percent iron, respectively. Table 3-4 shows that the percent iron removed was 4.27, 7.08, and 25 percent in Runs 19, 20, and 21 respectively. The standard potential for ferrous iron is -0.44 volt, whereas ferric iron has a standard potential of +0.77 volt. In Runs 19 and 20, the iron in solution was assumed to be mostly ferric iron because of the oxidized form of iron (Fe^{+3}) anticipated from the formation of sulfuric acid. In Run 21, the iron that was chelated was assumed to be comprised of mostly ferrous iron because the amount of ferric iron chelation drops off sharply after a pH level of 11. This may account for the higher removal of iron (and lower removal of lead) from the chelate in Run 21, as compared to Runs 19 and 20.

The amount of lead recovered from the lead chelate solution was determined from the concentration and quantity of lead found in the cathode scrapings and by the amount of lead remaining in the cathode chamber. Table 3-5 presents the amount of lead plated onto the cathode in all 24 electromembrane experiments. According to this data, the highest quantities of lead recovered were in Runs 13, 16, 18, 19, and 21. All of these experiments were conducted with 4 percent lead solutions and a 25 ma/cm^2 current density. The following differences were noted in these experiments:

Figure 3-16. Comparison of Lead Removal Rates for Experiments Performed with Iron

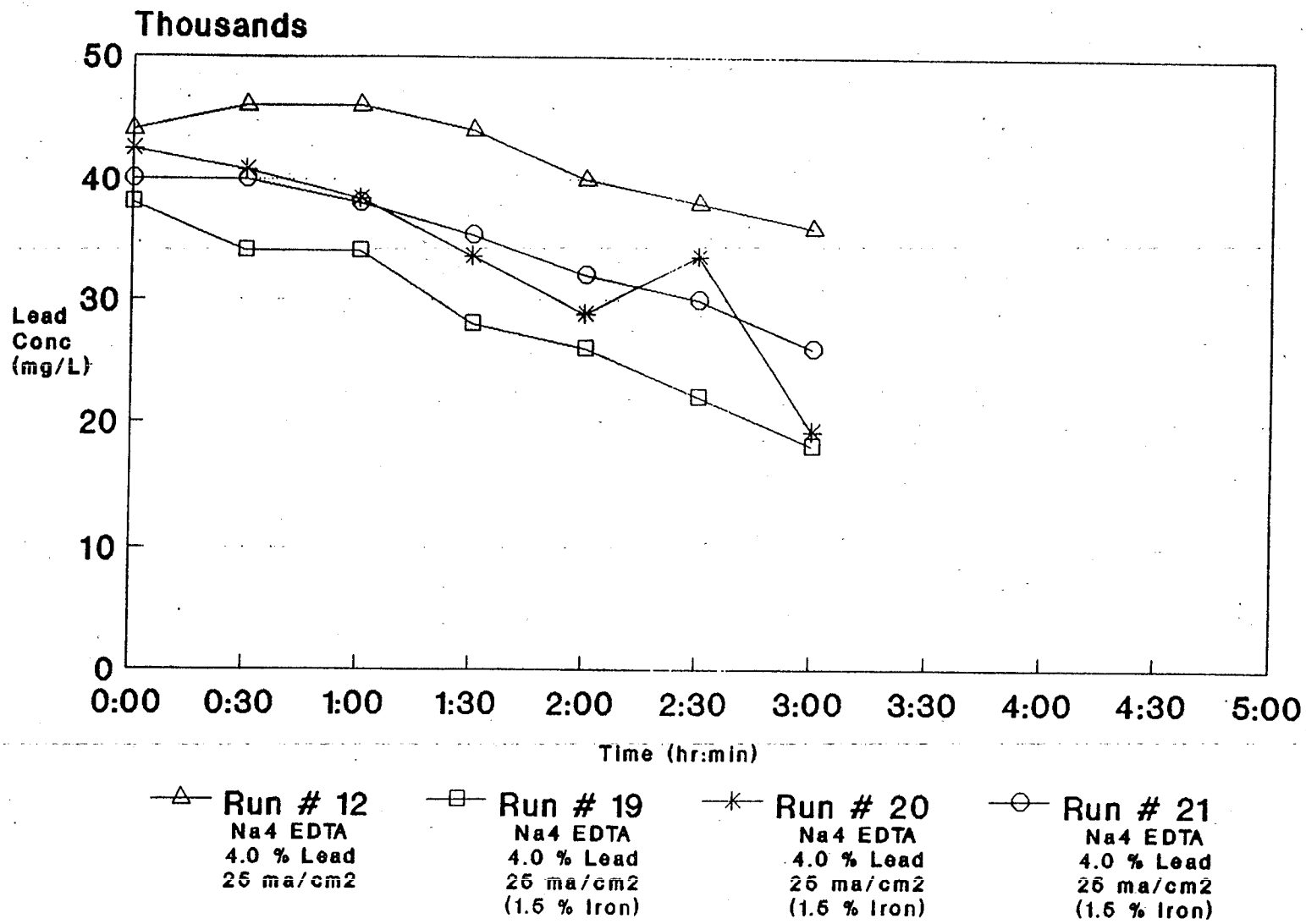


TABLE 3-4. PERCENT IRON REMOVALS FOR ELECTROMEMBRANE EXPERIMENTS

Run Number	Chelating Agent	Target Lead Concentration (%)	Target Starting pH	Reaction Time (hr)	Current Density (ma/cm2)	Cathode Chamber Sample No	Dilution Ratio for Analysis	Measured Iron Concentration In Cathode Chamber (mg/L) Sample # 1	Sample # 2	Sample # 3	Average Iron Concentration (mg/L)	Percent Iron Removal (%)
19	Na4 EDTA	4	7	3	25	EC-19-1-C	400	39	39	39	15600.0	
						EC-19-2-C	400	38	38	38	15200.0	2.56
						EC-19-3-C	400	38	38	38	15200.0	2.56
						EC-19-4-C	400	37	37	37	14800.0	5.13
						EC-19-5-C	400	40	39	39	15733.3	-0.85
						EC-19-6-C	400	37	38	37	14933.3	4.27
						EC-19-7-C	400	37	37	38	14933.3	4.27
20	Na4 EDTA	4	9	3	25	EC-20-1-C	400	37	38	38	15066.7	
						EC-20-2-C	400	37	37	38	14933.3	0.88
						EC-20-3-C	400	36	36	37	14533.3	3.54
						EC-20-4-C	400	34	34	34	13600.0	9.73
						EC-20-5-C	400	34	34	34	13600.0	9.73
						EC-20-6-C	400	34	34	34	13600.0	9.73
						EC-20-7-C	400	35	35	35	14000.0	7.08
21	Na4 EDTA	4	11.5	3	25	EC-21-1-C	400	28	28	28	11200.0	
						EC-21-2-C	400	28	28	28	11200.0	0.00
						EC-21-3-C	400	24	24	24	9600.0	14.29
						EC-21-4-C	400	23	23	23	9200.0	17.86
						EC-21-5-C	400	23	23	23	9200.0	17.86
						EC-21-6-C	400	22	22	22	8800.0	21.43
						EC-21-7-C	400	21	21	21	8400.0	25.00

TABLE 3-5. MASS OF LEAD DEPOSITED ON CATHODE

Run Number	Chelating Agent	Target Lead Concentration (%)	Current Density (ma/cm2)	Weight of Solid Sample (g)	Concentration of Lead in Solid Samples (mg/L) Sample # 1	Sample # 2	Sample # 3	Average Concentration of Lead in Solid Sample (mg/L)	Dilution Ratio for Analysis	Weight of Lead in Sample (g)	Percent Lead (%)	Initial Weight of Cathode (g)	Final Weight of Cathode (g)	Weight of Deposit (g)	Total Mass of Lead Deposited (g)
1	Na4 EDTA	0.8	25	0.9975	12.1	12.2	12.3	12.2	200	0.2	24.5	1737.5	1850.4	112.9	27.6
2	Na4 EDTA	0.8	25	0.9992	23.8	23.8	23.7	23.8	100	0.2	23.8	1744.1	1856.0	111.9	26.6
3	Na4 EDTA*	0.8	25	1.0053	20.6	20.9	20.9	20.8	100	0.2	20.7	1749.2	1865.8	116.6	24.1
12	Na4 EDTA	4	25	0.9992	25.0	25.0	25.0	25.0	400	1.0	100.1	1999.1	2042.0	42.9	42.9
13	Na4 EDTA*	4	25	1.0121	19.0	19.0	20.0	19.3	400	0.8	76.4	2013.6	2127.7	114.1	87.2
18	Na4 EDTA	4	25	1.0336	25.0	25.0	25.0	25.0	400	1.0	96.7	743.9	831.4	87.5	84.7
19	Na4 EDTA	4	25	1.0540	26.0	26.0	26.0	26.0	400	1.0	98.7	2017.5	2112.4	94.9	93.6
20	Na4 EDTA	4	25	0.6711	22.0	22.0	22.0	22.0	400	0.9	131.1	2072.6	2139.3	66.7	87.5
21	Na4 EDTA	4	25	1.0288	26.0	26.0	26.0	26.0	400	1.0	101.1	2035.0	2123.5	88.5	89.5
14	Na2 EDTA	0.8	25	1.0425	17.0	17.0	17.0	17.0	400	0.7	65.2	2047.4	2106.5	59.1	38.5
15	Na2 EDTA*	0.8	25	1.0465	17.0	17.0	17.0	17.0	400	0.7	65.0	2027.1	2117.8	90.7	58.9
4	DTPA	0.8	15	0.2044	8.0	7.0	8.0	7.7	400	0.3	150.0	1751.0	1781.7	30.7	46.1
6	DTPA*	0.8	15	0.5694	35.7	36.5	36.6	36.3	100	0.4	63.7	1818.0	1865.6	47.6	30.3
17	DTPA	0.8	15	1.0145	23.0	23.0	23.0	23.0	400	0.9	90.7	721.8	743.9	22.1	20.0
23	DTPA	0.8	15	1.0859	19.0	19.0	19.0	19.0	400	0.8	70.0	2085.5	2111.8	26.3	18.4
24	DTPA	0.8	15	1.0733	21.0	21.0	21.0	21.0	400	0.8	78.3	2097.8	2117.2	19.4	15.2
5	DTPA	0.8	25	0.7528	19.0	16.0	16.0	17.0	400	0.7	90.3	1784.7	1823.3	38.6	34.9
7	DTPA	0.8	25	0.6506	18.0	15.0	15.0	16.0	400	0.6	98.4	1824.9	1866.0	41.1	40.4
11	DTPA	4	15	1.0783	24.0	24.0	24.0	24.0	400	1.0	89.0	1955.1	2031.3	76.2	67.8
8	DTPA	4	25	0.5136	9.0	8.0	8.0	8.3	400	0.3	64.9	1855.1	1959.1	104.0	67.5
9	DTPA	4	25	0.6212	6.0	6.0	6.0	6.0	400	0.2	38.6	1885.4	2033.0	147.6	57.0
10	DTPA*	4	25	1.0144	22.0	22.0	22.0	22.0	400	0.9	86.8	1956.5	2033.2	76.7	66.5
16	DTPA	4	25	1.0221	22.0	22.0	22.0	22.0	400	0.9	88.1	683.3	820.9	137.6	118.5
22	DTPA	4	25	1.0029	18.0	18.0	18.0	18.0	400	0.7	71.8	2056.5	2136.7	80.2	57.6

* Regenerated chelating agent solutions

1) Run 13 was conducted with regenerated tetra-sodium EDTA solution, 2) Run 16 was conducted with DTPA using cadmium electrodes, 3) Run 18 was conducted with tetra-sodium EDTA using cadmium electrodes, and 4) Runs 19 and 21 were conducted with tetra-sodium EDTA containing 1.5 percent iron. Figure 3-17 shows the lead plated onto the cathode. One observation made during the electromembrane tests was that the lead only plated on the side of the cathode facing the membrane.

The lead recovered on the cathode varied in moisture content depending on the chelating agent used. The lead recovered from the di-sodium EDTA and tetra-sodium EDTA solution was more "spongy" than the lead recovered from the DTPA solutions. Lead plated from the DTPA solutions was typically a "harder" lead than lead plated from EDTA solutions. The reason the lead is "spongy" is that the plating solution is trapped within the fine hair-like threads of the lead. The moisture content of the lead scrapings from the cathode is presented in Table 3-6 for all 24 bench-scale electromembrane experiments. These data show that the lead from the tetra-sodium EDTA contained higher percentages of moisture than the lead from the di-sodium EDTA and DTPA experiments. The 0.8 percent lead had higher moisture content than the 4 percent lead for tetra-sodium EDTA. On average, the moisture content of lead from the 4 percent lead solutions were lower for 15 ma/cm^2 current density than for 25 ma/cm^2 current density. This is to be expected because of the faster plating rate for the 25 ma/cm^2 current density resulting in a spongier lead form.

3.2.4 Current efficiency

Current efficiencies varied with time during the experiments. Table 3-7 presents the final current efficiencies for the 24 experiments. The current efficiencies were calculated by the following formula:

$$\text{Current efficiency} = \frac{\text{Metal ion removed (meq)} \times 96.5\ \text{(C/meq)}}{\text{Time (s)} \times \text{Applied current (C/s)}} \times 100$$

where meq = milliequivalents
C = Coulombs (ampere-seconds)
s = seconds

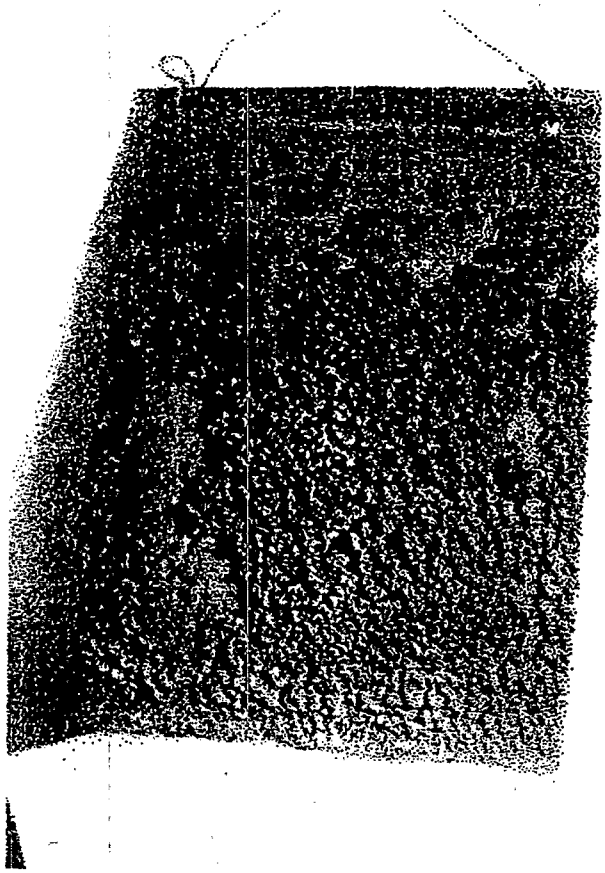


Figure 3-17. As-Plated Lead on Cathode.

TABLE 3-6. Moisture Content of Plated Lead from the Electromembrane Experiments.

Run No.	Chelating Agent	Initial Target Lead Concentration (%)	Current Density, (ma/cm2)	Initial Weight of Lead Sample, (g)	Final Weight of Lead Sample, (g)	Percent Moisture (%)
1	Na4EDTA	0.8	25	1.2607	0.6913	45.2
2	Na4EDTA	0.8	25	4.8815	1.9698	59.6
3	Na4EDTA (Regenerated from Run No. 2)	0.8	25	2.1604	1.0605	50.9
12	Na4EDTA	4	25	0.9915	0.9212	7.1
13	Na4EDTA (Regenerated from Run No. 12)	4	25	2.8032	1.983	29.3
18	Na4EDTA (Cadmium Electrodes)	4	25	1.1934	1.0442	12.5
19	Na4EDTA (1.5% Iron)	4	25	0.9999	0.8797	12
20	Na4EDTA (1.5% Iron)	4	25	0.9753	0.7811	19.9
21	Na4EDTA (1.5% Iron)	4	25	1.0955	0.8953	18.3
14	Na2EDTA	0.8	25	3.6458	2.2724	37.7

*A Ac appropriate amount [illegible]

[illegible]

TABLE 3-6. (cont.)

Run No.	Chelating Agent	Initial Target Lead Concentration (%)	Current Density, (ma/cm2)	Initial Weight of Lead Sample, (g)	Final Weight of Lead Sample, (g)	Percent Moisture (%)
15	Na2EDTA (Regenerated from Run No. 14)	0.8	25	8.497	5.0457	40.6
4	DTPA	0.8	15	NA	NA	NA
6	DTPA (Regenerated from Run No. 4)	0.8	15	1.5215	1.0858	28.6
17	DTPA (Cadmium Electrodes)	0.8	15	2.5129	2.1292	15.3
23	DTPA (Ionics Membrane)	0.8	15	1.017	0.7684	24.4
24	DTPA (Ionics Membrane)	0.8	15	1.0118	0.9044	10.6
5	DTPA	0.8	25	1.3716	1.179	14
7	DTPA	0.8	25	2.3251	2.2962	1.2
11	DTPA	4	15	1.0506	0.9741	7.3
8	DTPA	4	25	2.0356	1.7545	13.8
9	DTPA	4	25	2.1523	1.9045	11.5
10	DTPA (Regenerated from Run No. 9)	4	25	1.0188	0.9824	3.6

TABLE 3-6. (cont.)

Run No.	Chelating Agent	Initial Target Lead Concentration (%)	Current Density, (ma/cm2)	Initial Weight of Lead Sample, (g)	Final Weight of Lead Sample, (g)	Percent Moisture (%)
16	DTPA (Cadmium Electrodes)	4	25	1.0266	0.8025	21.8
22	DTPA (Ionics Membrane)	4	25	1.0473	0.8299	20.8

TABLE 3-7. FINAL CURRENT EFFICIENCIES FOR THE ELECTROMEMBRANE TESTS

Run No.	Chelating Agent	Initial Target Lead Concentration, (mg/L)	Current Density (ma/cm2)	Target Current, (amps)	Reaction Time, (min)	Current Efficiency, (%)
1	Na4EDTA	0.8	25	7.9	180	35.0
2	Na4EDTA	0.8	25	8.0	180	36.6
3	Na4EDTA	0.8	25	8.0	180	34.1
4	DTPA	0.8	15	4.7	180	33.6
					300	32.7
5	DTPA	0.8	25	7.9	180	32.0
6	DTPA	0.8	15	4.7	180	45.2
7	DTPA	0.8	25	7.9	180	23.5
					300	21.2
8	DTPA	4	25	7.9	180	66.1
9	DTPA	4	25	7.9	300	67.5
10	DTPA	4	25	7.9	180	84.1
					240	92.7
11	DTPA	4	15	4.7	180	--
					300	80.0
12	Na4EDTA	4	25	7.9	180	46.8
13	Na4EDTA	4	25	7.9	180	102.9
14	Na2EDTA	0.8	25	7.9	180	37.0
15	Na2EDTA	0.8	25	7.9	180	32.2
16	DTPA	4	25	7.9	180	82.6
17	DTPA	0.8	15	4.7	180	33.3
18	Na4EDTA	4	25	7.9	180	73.3
19	Na4EDTA	4	25	7.9	180	93.3
20	Na4EDTA	4	25	7.9	180	108.7
21	Na4EDTA	4	25	7.9	180	69.7
22	DTPA	4	25	7.9	180	71.8
23	DTPA	0.8	15	4.7	180	19.7
24	DTPA	0.8	15	4.7	180	28.5

Figure 3-18 shows that the test conducted with 4 percent lead concentration DTPA solution (Run 11) had a much higher current efficiency than the run with 0.8 percent lead concentration solution (Run 4). This conclusion is expected because a higher plating efficiency is achieved with a higher initial lead concentration.

Current density and current efficiency for DTPA solutions containing 0.8 percent lead are compared in Figure 3-19. This graph shows that for the lower lead concentration experiments (Run 4, 5, and 7), there is virtually no difference in current efficiency with current density. The current efficiency remains steady throughout all three experiments. Figure 3-20 presents a comparison of current density and current efficiency for DTPA solutions containing 4 percent lead. In the 4 percent lead experiments (Runs 8, 9, and 11), the current efficiency after a 3-hour plating time is slightly higher for the tests conducted at a 25 ma/cm^2 current density (Runs 8 and 9) than the test conducted at 15 ma/cm^2 current density (Run 11). After a 5-hour plating time, the current efficiency is slightly higher for the 15 ma/cm^2 current density (Run 11) than for the 25 ma/cm^2 current density (Run 9). According to a similar study conducted by IT (formerly PEI Associates), the current efficiency was higher for the lower current density (15 ma/cm^2 than for the higher current density (25 ma/cm^2). Major differences in the two studies that may be contributing factors to this difference are that synthetic EDTA and DTPA solutions were used in the IT study, and actual lead-EDTA solutions from soil washing tests were used in the PEI study.[2]

In Figure 3-21, the current efficiencies are compared for tests with 0.8 percent lead and 15 ma/cm^2 current density using lead and cadmium electrodes. In the 0.8 percent lead experiments (Run 4 and 17), there appears to be no difference in the current efficiency for lead and cadmium electrodes; the current efficiencies remain constant in both experiments. Figure 3-22 presents a comparison of the current efficiencies for tests with 4 percent lead and 25 ma/cm^2 current density using lead and cadmium electrodes. In the 4 percent lead experiments (Run 8, 9, and 16), the current efficiency seems to be greater for the cadmium electrodes (Run 16) than for the lead electrodes (Runs 8 and 9). Figure 3-23 also shows a comparison of cadmium and lead electrodes for the tests conducted with tetra-sodium EDTA containing 4 percent lead and with 25 ma/cm^2 current density. As with the DTPA

Figure 3-18. Comparison of Current Efficiency for DTPA Using Two Initial Lead Concentrations

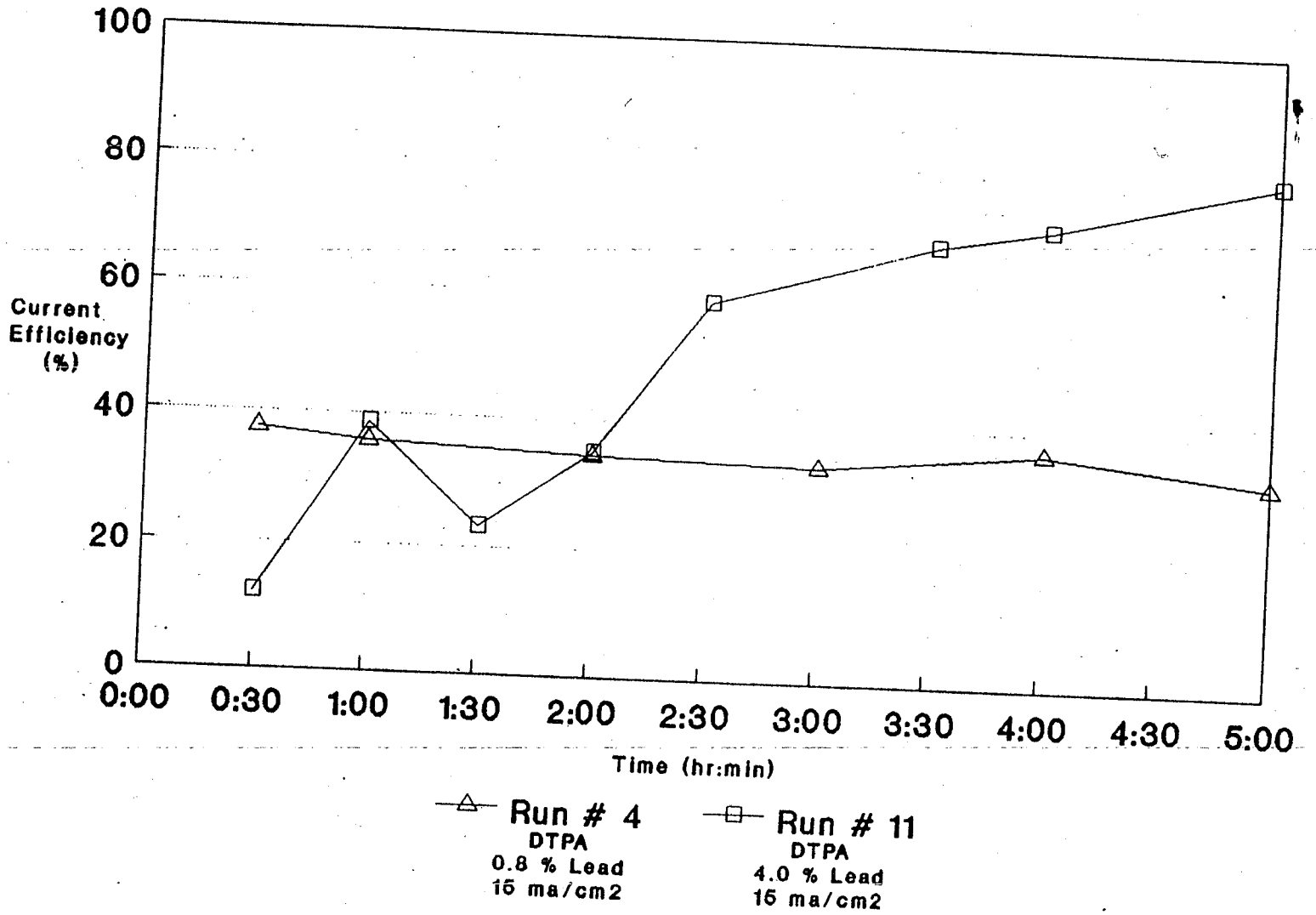


Figure 3-19. Comparison of Current Density and Current Efficiency for 0.8 Percent Lead Concentrations

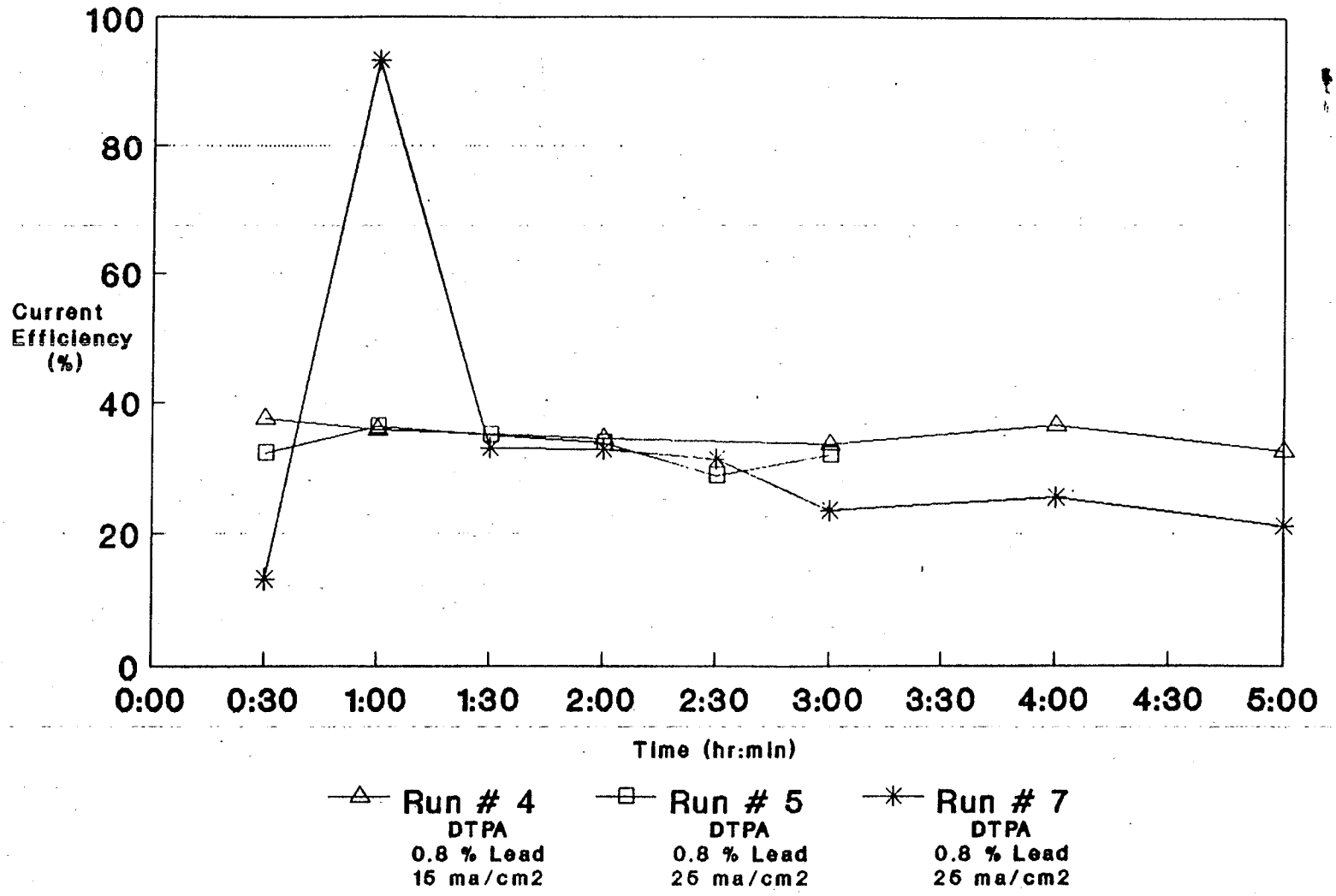


Figure 3-20. Comparison of Current Density and Current Efficiency for 4 Percent Lead Concentrations

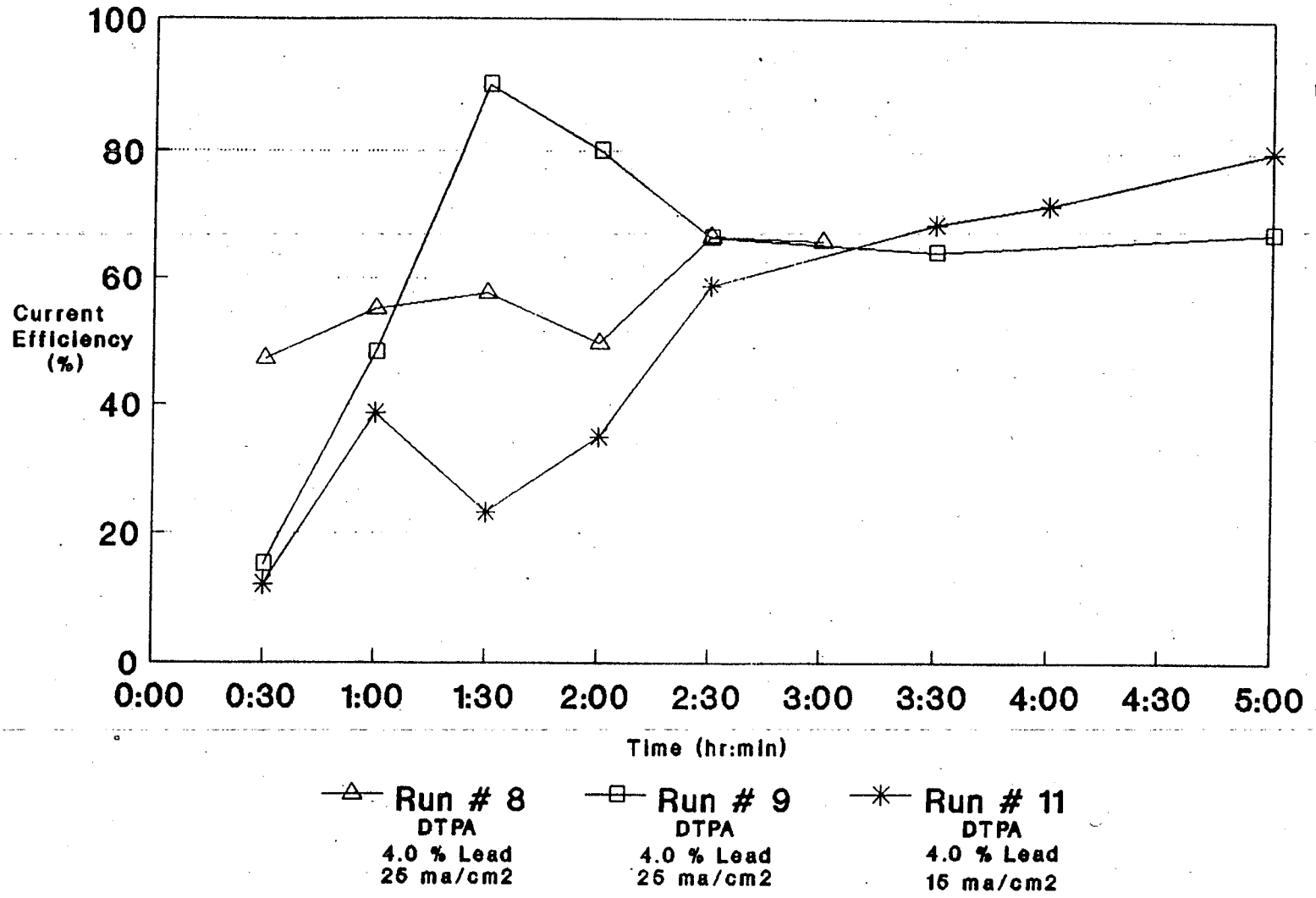


Figure 3-21. Comparison of Type of Electrode and Current Efficiency for DTPA and 0.8 Percent Lead Concentrations

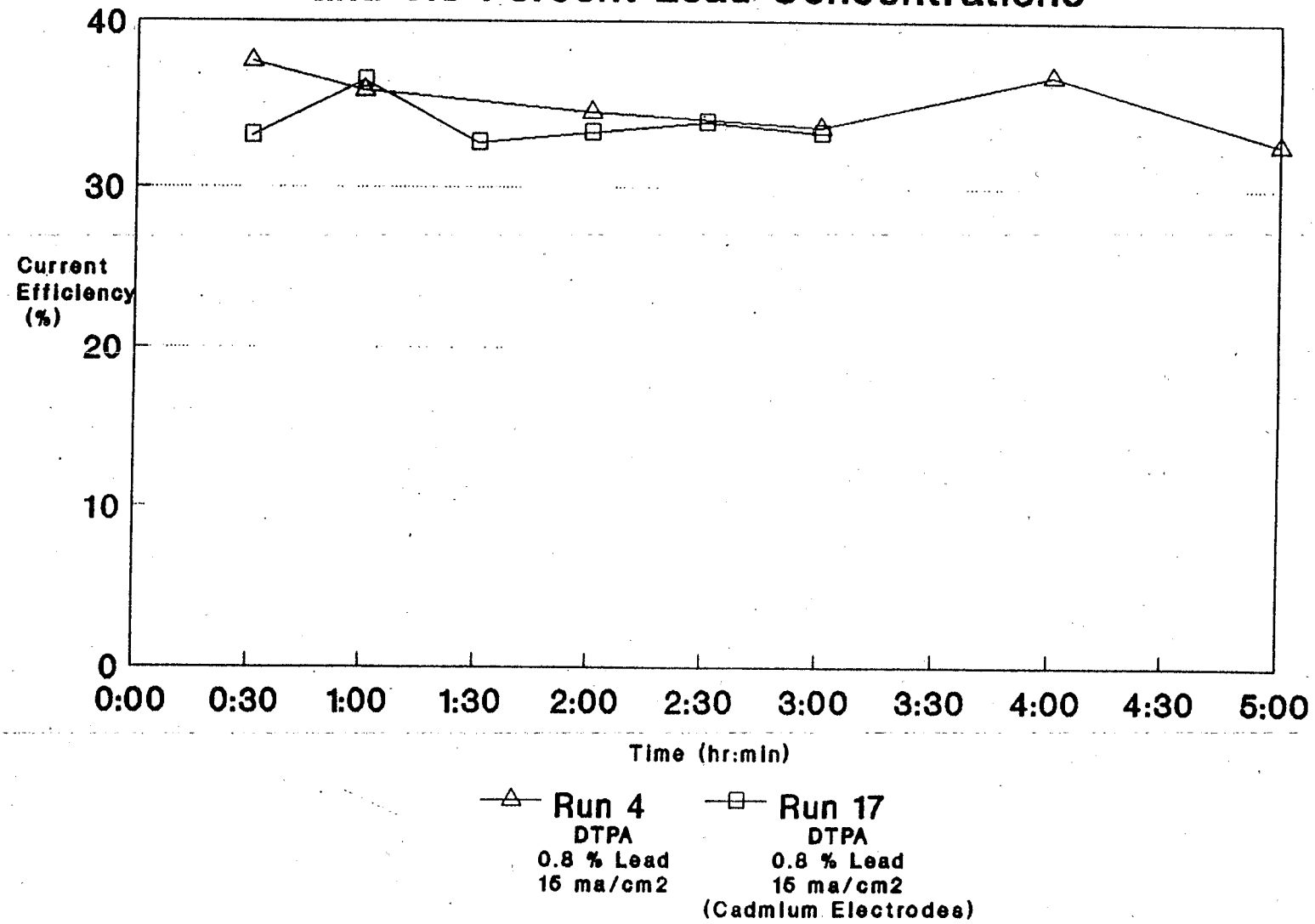


Figure 3-22. Comparison of Type of Electrode and Current Efficiency for DTPA and 4 Percent Lead Concentrations

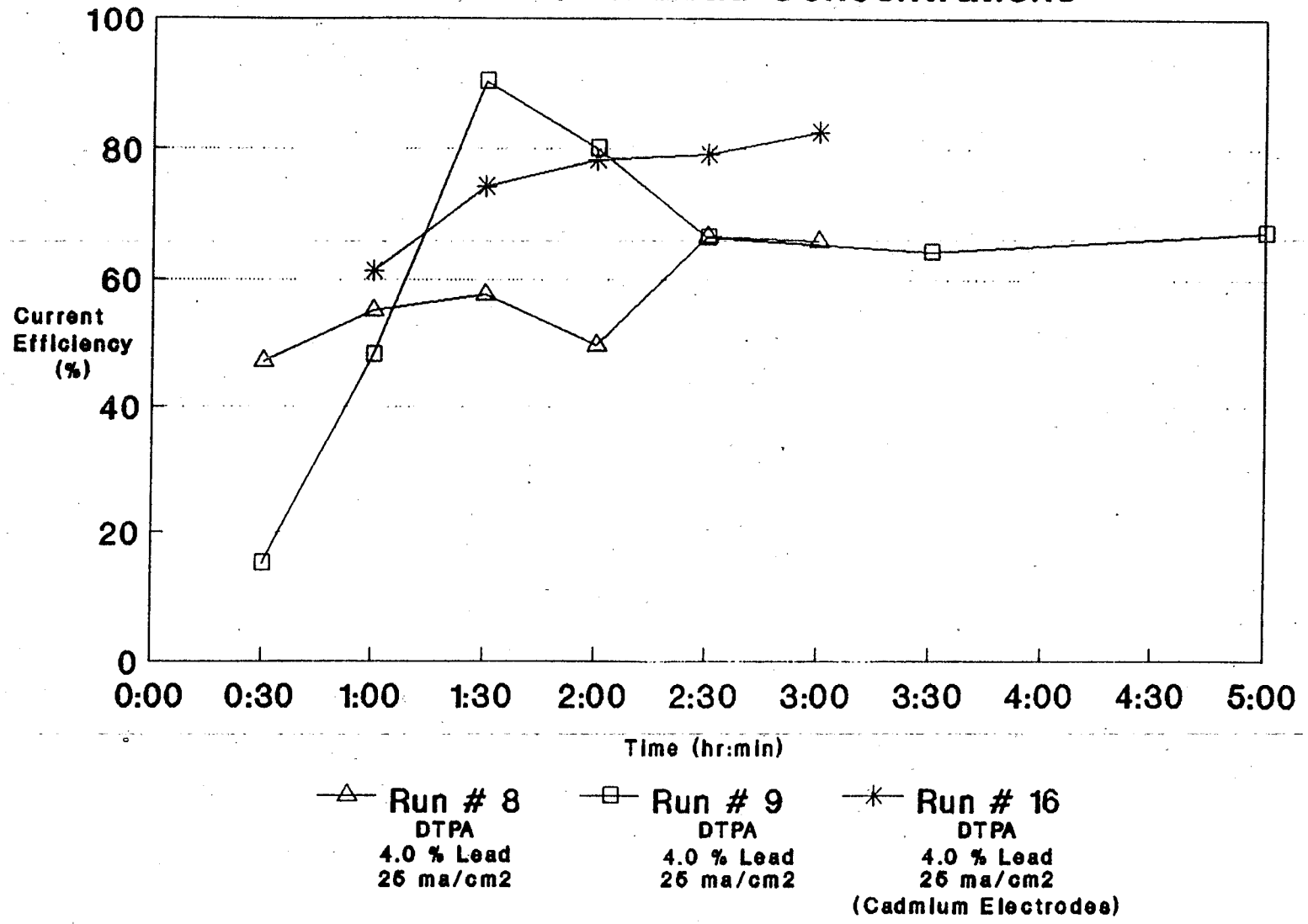
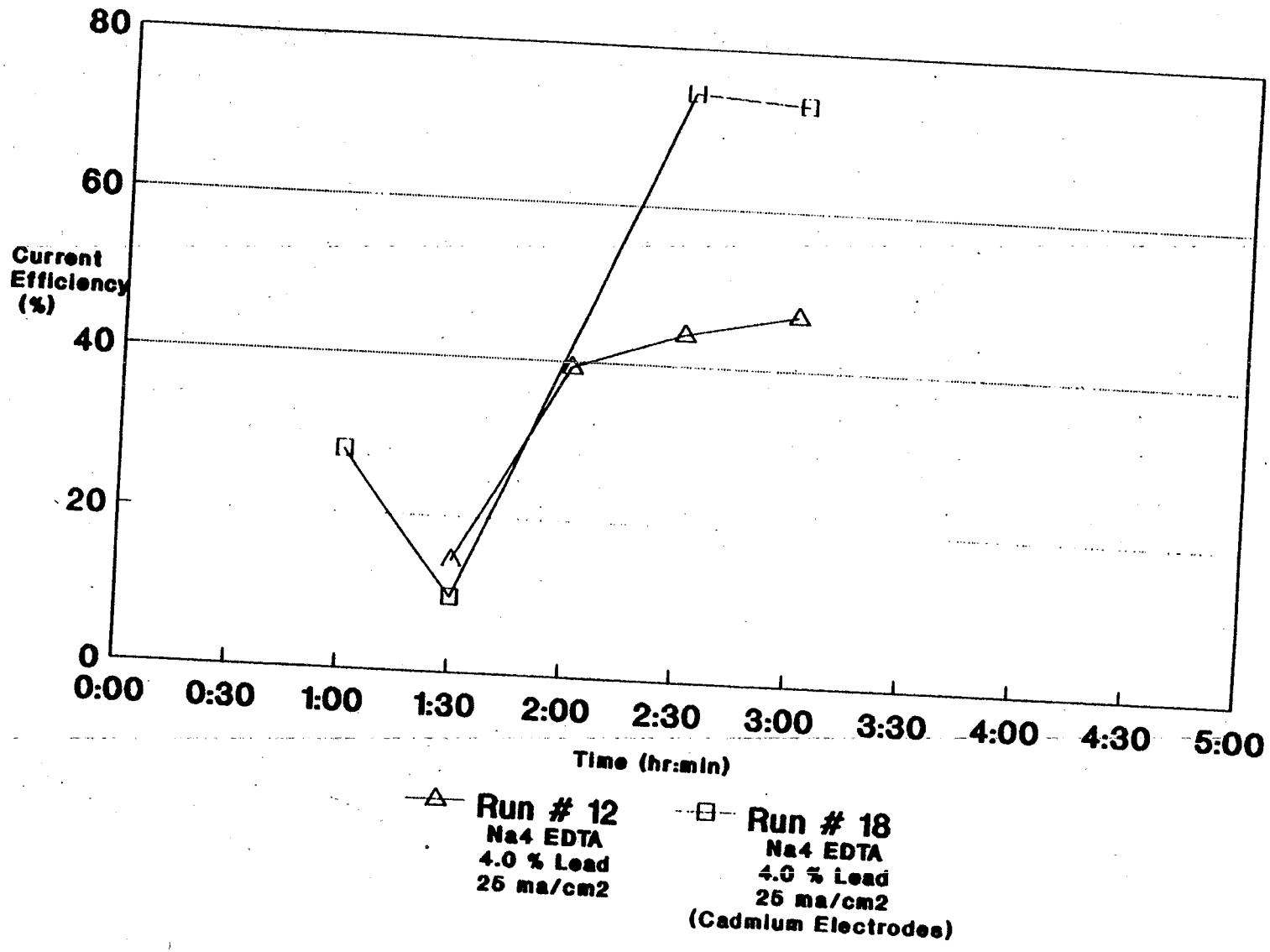


Figure 3-23. Comparison of Type of Electrode and Current Efficiency for Tetra-sodium EDTA



experiments, the cadmium electrodes exhibit higher current efficiencies than the lead electrodes. The higher current efficiencies for the cadmium electrodes may be attributed to the higher standard potential for cadmium.

The current efficiencies for tests conducted with iron in the chelating agent solution are compared in Figure 3-24. According to this graph and the data presented in Table 3-7, the current efficiencies were approximately 100 percent for Runs 19 and 20 conducted with 1.5 percent iron at a solution pH of 7 and 9, respectively. In Run 21, conducted with iron, the solution pH was 11.5, and the current efficiency was 69.7 percent after a 3-hour reaction period. Run 12 was conducted under the same conditions except without iron. The current efficiency for this test was only 46.8 percent after a 3-hour reaction period.

3.2.5 Sodium Concentration

The amount of sodium in the cathode and anode chambers was measured throughout the experiments to determine the depletion of sodium in the anode chamber solution and the increase in sodium in the cathode chamber solution as the sodium passes through the cation-exchange membrane to regenerate the chelating agent. The amount of sodium that is transported across the membrane from the anode to the cathode chamber is important to determine if the sodium form of the chelating agent is being regenerated and to determine if an adequate quantity of sodium has been supplied for the system.

Table 3-8 presents the initial and final sodium concentrations in the cathode chamber for the 24 electromembrane experiments. The difference in sodium concentration in the cathode chamber from the initial sample to the final sample was greatest in Runs 1, 2, 12, and 18. Runs 1 and 2 also showed greater than 98 percent lead removal. Based on the data presented in Table 3-9, the difference in sodium concentration in the anode chamber from the initial sample to the final sample was greatest in Runs 1, 20, and 21.

Figure 3-24. Comparison of Current Efficiencies for Tests with Iron

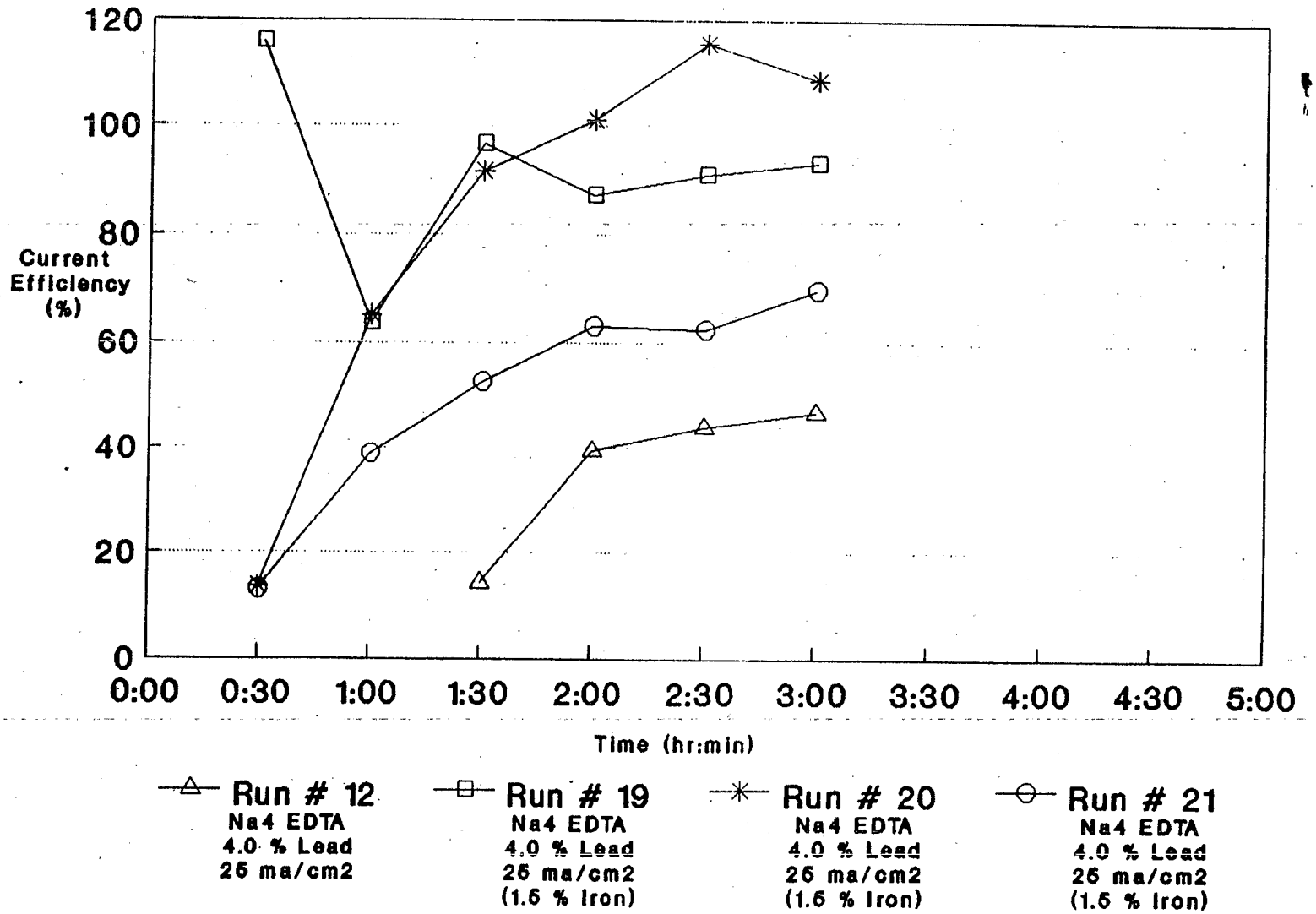


TABLE 3-8. SODIUM CONCENTRATIONS IN CATHODE CHAMBER

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (min)	Cathode Chamber Sample No.	Dilution Factor for Analysis	Measured Sodium Concentration In Cathode Chamber (mg/L) Sample # 1	Sample # 2	Sample # 3	Avg. Sodium Concentration In Cathode Chamber (mg/L)
1	Na4 EDTA	0.8	25	0	EC-1-1-C	400	21	21	22	8533
				180	EC-1-7-C	400	50	51	50	20133
2	Na4 EDTA	0.8	25	0	EC-2-1-C	400	12	14	14	5333
				180	EC-2-7-C	400	43	43	42	17067
3	Na4 EDTA	0.8	25	0	EC-3-1-C	400	24	24	24	9600
				180	EC-3-7-C	400	47	47	47	18800
12	Na4EDTA	4	25	0	EC-12A-1-C	800	51	52	52	41333
				180	EC-12A-7-C	800	70	69	71	56000
13	Na4EDTA	4	25	0	EC-13A-1-C	800	67	67	68	53867
				180	EC-13A-7-C	800	66	66	66	52800
18	Na4 EDTA	4	25	0	EC-18-1-C	800	56	56	56	44800
				180	EC-18-7-C	800	91	92	94	73867
19	Na4 EDTA	4	25	0	EC-19-1-C	800	52	52	52	41600
				180	EC-19-7-C	800	65	66	65	52267
20	Na4 EDTA	4	25	0	EC-20-1-C	800	53	54	54	42933
				180	EC-20-7-C	800	67	66	67	53333
21	Na4 EDTA	4	25	0	EC-21-1-C	800	49	50	49	39467
				180	EC-21-7-C	800	55	55	55	44000
14	Na2 EDTA	0.8	25	0	EC-14-1-C	400	13	14	13	5333
				180	EC-14-7-C	200	54	54	53	10733
15	Na2 EDTA	0.8	25	0	EC-15-1-C	400	31	30	29	12000
				180	EC-15-7-C	400	43	42	43	17067
4	DTPA	0.8	15	0	EC-4-1-C	200	11	11	12	2267
				180	EC-4-6-C	100	18	18	18	1800
				300	EC-4-8-C	100	22	22	21	2167
6	DTPA	0.8	15	0	EC-6-1-C	400	24	24	23	9467
				180	EC-6-7-C	400	29	30	30	11867

TABLE 3-8. (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (min)	Cathode Chamber Sample No.	Dilution Factor for Analysis	Measured Sodium Concentration In Cathode Chamber (mg/L) Sample # 1	Sample # 2	Sample # 3	Avg. Sodium Concentration In Cathode Chamber (mg/L)
17	DTPA	0.8	15	0	EC-17-1-C	400	25	25	25	10000
				180	EC-17-7-C	400	29	30	30	11867
23	DTPA	0.8	15	0	EC-23-1-C	200	53	53	53	10600
				180	EC-23-7-C	200	69	69	71	13933
24	DTPA	0.8	15	0	EC-24-1-C	200	43	44	44	8733
				180	EC-24-7-C	200	66	65	65	13067
5	DTPA	0.8	25	0	EC-5-1-C	400	32	33	33	13067
				180	EC-5-7-C	400	52	52	52	20800
7	DTPA	0.8	25	0	EC-7-1-C	400	28	27	28	11067
				180	EC-7-7-C	400	42	41	42	16667
				300	EC-7-9-C	400	47	46	47	18667
11	DTPA	4	15	0	EC-11-1-C	800	50	48	49	39200
				180	EC-11-7-C	800	56	57	57	45333
				300	EC-11-10-C	800	61	61	61	48800
8	DTPA	4	25	0	EC-8-1-C	1000	48	49	49	48667
				180	EC-8-7-C	1000	54	54	54	54000
9	DTPA	4	25	0	EC-9-1-C	1000	48	48	48	48000
				180	EC-9-7-C	1000	19	19	19	19000
				300	EC-9-9-C	1000	55	55	55	55000
10	DTPA	4	25	0	EC-10-1-C	1000	48	48	47	47667
				180	EC-10-7-C	1000	52	53	53	52667
				240	EC-10-8-C	1000	54	54	54	54000
16	DTPA	4	25	0	EC-16-1-C	800	59	59	59	47200
				180	EC-16-7-C	800	64	65	65	51733
22	DTPA	4	25	0	EC-22-1-C	800	56	56	56	44800
				180	EC-22-7-C	800	59	59	59	47200

TABLE 3-9. SODIUM CONCENTRATIONS IN ANODE CHAMBER

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (min)	Anode Chamber Sample No.	Dilution Factor for Analysis	Measured Sodium Concentration In Anode Chamber (mg/L) Sample # 1	Sample # 2	Sample # 3	Avg. Sodium Concentration In Anode Chamber (mg/L)
1	Na4 EDTA	0.8	25	0	EC-1-1-A	400	31	29	30	12000
				180	EC-1-7-A	200	30	30	31	6067
2	Na4 EDTA	0.8	25	0	EC-2-1-A	100	53	51	51	5167
				180	EC-2-7-A	100	30	29	30	2967
3	Na4 EDTA	0.8	25	0	EC-3-1-A	200	39	39	39	7800
				180	EC-3-7-A	200	37	36	36	7267
12	Na4EDTA	4	25	0	EC-12A-1-A	400	47	48	48	19067
				180	EC-12A-7-A	400	40	40	40	16000
13	Na4EDTA	4	25	0	EC-13A-1-A	400	39	39	39	15600
				180	EC-13A-7-A	400	30	31	30	12133
18	Na4 EDTA	4	25	0	EC-18-1-A	400	47	46	47	18667
				180	EC-18-7-A	400	37	37	37	14800
19	Na4 EDTA	4	25	0	EC-19-1-A	400	48	48	47	19067
				180	EC-19-7-A	400	39	40	39	15733
20	Na4 EDTA	4	25	0	EC-20-1-A	800	62	62	61	49333
				180	EC-20-7-A	800	45	45	45	36000
21	Na4 EDTA	4	25	0	EC-21-1-A	400	45	45	45	18000
				180	EC-21-7-A	400	32	33	33	13067
14	Na2 EDTA	0.8	25	0	EC-14-1-A	400	47	47	46	18667
				180	EC-14-7-A	400	34	34	36	13867
15	Na2 EDTA	0.8	25	0	EC-15-1-A	400	42	40	40	16267
				180	EC-15-7-A	400	36	35	36	14267
4	DTPA	0.8	15	0	EC-4-1-A	100	30	28	30	2933
				180	EC-4-6-A	100	22	21	22	2167
				360	EC-4-8-A	100	35	36	36	3567
6	DTPA	0.8	15	0	EC-6-1-A	400	19	19	19	7600
				180	EC-6-7-A	200	34	32	32	6533

TABLE 3-9. (cont.)

Run Number	Chelating Agent	Initial Target Lead Concentration (%)	Current Density (ma/cm2)	Reaction Time (min)	Anode Chamber Sample No.	Dilution Factor for Analysis	Measured Sodium Concentration In Anode Chamber (mg/L) Sample # 1	Sample # 2	Sample # 3	Avg. Sodium Concentration In Anode Chamber (mg/L)
17	DTPA	0.8	15	0	EC-17-1-A	400	47	47	47	18800
				180	EC-17-7-A	400	44	43	43	17333
23	DTPA	0.8	15	0	EC-23-1-A	400	55	55	55	22000
				180	EC-23-7-A	400	47	47	47	18800
24	DTPA	0.8	15	0	EC-24-1-A	200	105	102	102	20600
				180	EC-24-7-A	200	98	98	98	19600
5	DTPA	0.8	25	0	EC-5-1-A	200	37	37	37	7400
				180	EC-5-7-A	200	32	31	31	6267
7	DTPA	0.8	25	0	EC-7-1-A	200	38	38	37	7533
				180	EC-7-7-A	200	45	42	39	8400
				300	EC-7-9-A	200	23	24	23	4667
11	DTPA	4	15	0	EC-11-1-A	400	47	46	44	18267
				180	EC-11-7-A	400	42	42	42	16800
				300	EC-11-10-A	400	36	35	35	14133
8	DTPA	4	25	0	EC-8-1-A	800	25	25	25	20000
				180	EC-8-7-A	800	22	22	22	17600
9	DTPA	4	25	0	EC-9-1-A	800	26	25	26	20533
				180	EC-9-7-A	800	55	54	54	43467
				300	EC-9-9-A	800	18	18	18	14400
10	DTPA	4	25	0	EC-10-1-A	1000	22	21	22	21667
				180	EC-10-7-A	1000	17	18	17	17333
16	DTPA	4	25	0	EC-16-1-A	400	47	46	47	18667
				180	EC-16-7-A	400	42	40	41	16400
22	DTPA	4	25	0	EC-22-1-A	400	54	53	54	21467
				180	EC-22-7-A	400	43	43	43	17200

SECTION 4

QUALITY ASSURANCE

4.1 SAMPLE DIGESTION

Samples of the anode chamber sodium carbonate solutions were to be digested in accordance with EPA SW846 Method 3010. This method involves mixing a portion of the sample with nitric acid, refluxing the sample, and repeating these steps until the digestate is light in color or until the color has stabilized. The digestate was then to be brought to a low volume, refluxed with hydrochloric acid, and brought up to volume with water. Because a small quantity of white precipitate formed when the sample was cooled after first digestion with nitric acid, however, this method was discontinued. There are several possible theories regarding the composition of the white precipitate. One possibility is the formation of lead chloride or lead nitrate; another possibility is the formation of the hydrated form of EDTA which is insoluble in acidic solutions. The white precipitant was not analyzed. The samples of the anode chamber solutions were analyzed without the use of digestion by EPA SW846 Method 7770, Atomic Absorption, Direct Aspiration[10]. The sample did not require digestion to solubilize the sodium because sodium carbonate is highly soluble in water and the original concentration of sodium was known (5 percent sodium carbonate).

Samples of the cathode chamber solutions containing lead and sodium were to be digested by EPA SW 846 Method 3010. Lead was analyzed by EPA SW846 Method 7420, Atomic Absorption, Direct Aspiration. The digested cathode chamber solution sample also formed a white precipitate when the sample was heated and acidified with nitric acid. The precipitate is believed to be an insoluble lead complex with the chelating agent. Because all of the lead and sodium were in solution (chelated), digestion of the cathode chamber solution samples was determined to be unnecessary based on recommendations from the chelating agent manufacturer, Hampshire Chemical[11].

The lead scrapings from the cathode were digested according to EPA SW846 Method 3050, Acid Digestion of Sediments, Sludges, and Soils. This method involves digesting a 1- to 2-gram portion of the sample in nitric acid and hydrogen peroxide, and then refluxing the sample with nitric acid. A final reflux with dilute hydrochloric acid is performed and the sample is diluted to 100 ml with water[9]. A white precipitant formed during the nitric acid digestion step, but was dissolved after the addition of hydrogen peroxide. Another white precipitant formed during the last step of the digestion, which involved acidifying the sample with hydrochloric acid after the sample was cooled. The precipitant is believed to be a lead complex formed upon acidification. The digestion procedure was modified by eliminating the hydrochloric acidification step to prohibit precipitant formation.

4.2 SAMPLE QUALITY ASSURANCE

Method blanks, matrix spikes, and matrix spike duplicates were analyzed according to the procedures outlined in the Quality Assurance Project Plan. Table 4-1 presents the precision and accuracy results of the lead, sodium, and iron analysis. Method precision was measured by determining the relative percent difference (RPD) between the recoveries of the matrix spike and the matrix spike duplicate. The RPD was calculated by the following equation:

$$RPD = \frac{(Rms - Rmsd)}{\frac{(Rms + Rmsd)}{2}} \times 100\%$$

where Rms = Recovery of matrix spike
Rmsd = Recovery of matrix spike duplicate

According to the Quality Assurance Project Plan, the precision for the lead and sodium samples was to be less than 35 percent. IT's analytical precision was within 9.5 percent for all samples.

TABLE 4-1. QUALITY CONTROL CHECKS FOR LEAD, SODIUM, AND IRON ANAYLSES BY ATOMIC ABSORPTION

Analyte	Sample No.	Sample Conc. (mg/L)	Matrix Spike Conc. (mg/L)	Matrix Spike Duplicate Conc. (mg/L)	Relative Percent Difference (%) (Precision)	Dilution Ratio for Analysis	Spike Solution Conc. (mg/L)	Percent Recovery (%) (Accuracy)
Lead								
	EC-2-7-C	173.3	1202.5	1206.7	0.35	25	40	102.92
	EC-3-6-C	190	539	541	0.37	10	40	87.25
	EC-4-8-C	862.5	6254.2	6291.7	0.60	125	40	107.83
	EC-9-9-C	14140	38720	38900	0.46	600	40	102.42
	EC-12-7-C	36000	56000	56000	0.00	2000	10	100.00
	EC-14-6-C	720	6685	6645	0.60	150	40	99.42
	EC-15-7-C	750	6333.3	6458	1.95	125	30	148.89
	EC-17-7-C	3750	14250	14500	1.74	250	50	84.00
	EC-20-7-C	19200	43200	43200	0.00	2400	10	100.00
	EC-22-7-C	27000	45000	46800	3.92	1800	10	100.00
	EC-23-7-C	5750	7833.3	8000	2.11	250	10	83.33
Sodium								
	EC-3-6-A	6800	23600	23600	0.00	400	40	105.00
	EC-6-1-A	7600	22800	22520	1.24	400	40	95.00
	EC-9-6-A	17600	40800	40000	1.98	800	30	96.67
	EC-9-7-C	18400	40267	40000	0.67	800	30	91.11
	EC-11-10-A	14133.3	26533	26266	1.01	400	30	103.33
	EC-12-7-A	16000	27066	27733	2.43	400	30	92.22
	EC-13-7-A	12133	23067	22533	2.34	400	30	91.12
	EC-14-3-C	7600	14133	14867	5.06	200	30	108.88
	EC-15-7-C	17067	28533	30400	6.34	400	30	95.55
	EC-16-7-A	16400	26800	26667	0.50	400	30	86.67
	EC-17-7-C	11866.7	23467	23467	0.00	400	30	96.67
	EC-18-7-A	14800	26667	26667	0.00	400	30	98.89

TABLE 4-1. (cont.)

Analyte	Sample No.	Sample Conc. (mg/L)	Matrix Spike Conc. (mg/L)	Matrix Spike Duplicate Conc. (mg/L)	Relative Percent Difference (%) (Precision)	Dilution Ratio for Analysis	Spike Solution Conc. (mg/L)	Percent Recovery (%) (Accuracy)
Sodium (cont'd)	EC-19-7-A	15733.3	27600	27067	1.95	400	30	98.89
	EC-20-7-A	18000	33200	33600	1.20	400	40	95.00
	EC-21-7-A	13080	29468	29733	0.90	400	40	102.43
	EC-22-7-A	17200	35867	35333	1.50	400	50	93.34
	EC-23-1-C	10600	15200	15933	4.71	200	30	76.67
	EC-24-1-C	8733.3	15067	14533	3.61	200	30	105.56
Iron								
	EC-19-7-C	14933.3	22133	22400	1.20	400	20	90.00
	EC-21-7-C	8400	16400	16800	2.41	400	20	100.00

The accuracy of the method was measured by the percent recovery of the spiked and unspiked sample concentration as calculated by the following equation:

$$\% \text{ Recovery} = 100 \times (Cm/Csrm)$$

where Cm = measured concentration of standard reference material (srm)
Csrm = actual concentration of srm

According to the Quality Assurance Objectives outlined in the quality assurance project plan, the accuracy of the matrix spike samples is to be 80 to 120 percent of the actual value. All but two samples were outside of this limit for accuracy. The completeness of the matrix spike and matrix spike duplicate samples were calculated by the following equations:

$$C = 100 \cdot (V/T)$$

where

C = percent completeness
V = number of measurements judged valid
T = total number of measurements

The completeness was determined to be 96.7 percent which is well within the 90 percent completeness specified for this study. The most likely reason the two samples were outside of the limit for accuracy was the error in the matrix spike addition.

Method blanks were also analyzed with each batch of samples to determine the amount of target analyte in the blank samples. Table 4-2 presents the method blank data obtained through the analysis of the lead, sodium, and iron samples.

TABLE 4-2. METHOD BLANK ANALYSES BY ATOMIC ABSORPTION

Analyte	Date of sample analysis	Sample concentration (mg/L)
Lead	8/2/93	ND[a]
	8/3/93	ND
	8/11/93	ND
	8/17/93	ND
	8/19/93	ND
	8/24/93	ND
	9/1/93	ND
	9/13/93	ND
Sodium	8/7/93	ND
	8/11/93	ND
	8/16/93	2
	8/19/93	ND
	8/31/93	ND
Iron	8/24/93	ND
	9/1/93	ND

[a] Nondetectable.

All but one of the method blanks were below the method detection limit of 1 mg/L for lead, sodium, and iron.

4.3 MASS BALANCES

Mass balances were performed on lead and sodium in all 24 bench-scale electromembrane experiments. The percent closure of the mass balance also indicates the reliability of the atomic absorption data and the procedures used to perform the experiments. Table 4-3 presents the mass balance closures for lead. The lead mass balances were calculated by the following formula:

$$Mi = \Sigma (C \times V) + M_c$$

where

M_i = Mass of lead used in test solution, g

$\Sigma(C \times V)$ = $(C_L \times V_L) + (C_{S1} \times V_{S1}) + (C_{S2} \times V_{S2}) + ...$

TABLE 4-3. LEAD MASS BALANCES FOR THE ELECTROMEMBRANE TESTS

				INPUT		OUTPUTS				
Run No.	Chelating Agent	Target Lead Concentration (%)	Current Density (ma/cm2)	Initial Quantity of Lead (g)	Cathode Chamber Sample No.	Quantity of Lead in Samples (g)	Quantity of Lead Deposited on Cathode (g)	Final Quantity of Lead in Cathode Chamber (g)	Total Quantity of Lead in Outputs (g)	Percent Mass Balance Closure (%)
1	Na4EDTA	0.8	25	32.2	EC-1-1-C	0.4	27.6	0.2	28.0	112
					EC-1-2-C	0.2				
					EC-1-3-C	0.2				
					EC-1-4-C	0.2				
					EC-1-5-C	0.03				
					EC-1-6-C	0.01				
					EC-1-7-C	0.001				
2	Na4EDTA	0.8	25	34.1	EC-2-1-C	0.4	26.6	0.6	28.7	119
					EC-2-2-C	0.3				
					EC-2-3-C	0.4				
					EC-2-4-C	0.2				
					EC-2-5-C	0.1				
					EC-2-6-C	0.04				
					EC-2-7-C	0.02				
3	Na4EDTA (Regenerated from Run No. 2)	0.8	25	31.4	EC-3-1-C	0.4	24.1	0.2	25.7	122
					EC-3-2-C	0.3				
					EC-3-3-C	0.4				
					EC-3-4-C	0.2				
					EC-3-5-C	0.1				
					EC-3-6-C	0.02				
					EC-3-7-C	0.01				
12	Na4EDTA	4	25	170.0	EC-12-1-C	2.2	42.9	133.2	180.8	92
					EC-12-2-C	2.3				
					EC-12-3-C	2.3				
					EC-12-4-C	2.2				
					EC-12-5-C	2.0				
					EC-12-6-C	1.9				
					EC-12-7-C	1.8				
13	Na4EDTA (Regenerated from Run No. 12)	4	25	270.0	EC-13-1-C	3.4	87.2	175.8	284.1	85
					EC-13-2-C	3.4				
					EC-13-3-C	3.3				
					EC-13-4-C	3.1				
					EC-13-5-C	2.9				
					EC-13-6-C	2.6				
					EC-13-7-C	2.4				
18	Na4EDTA (Cadmium Electrodes)	4	25	170.7	EC-18-1-C	2.1	84.7	100.6	202.3	84
					EC-18-2-C	2.4				
					EC-18-3-C	2.1				
					EC-18-4-C	2.2				
					EC-18-5-C	2.3				
					EC-18-6-C	1.5				
					EC-18-7-C	1.4				

TABLE 4-3: (cont.)

Run No.	Chelating Agent	Target Lead Concentration (%)	Current Density (ma/cm2)	INPUT: Initial Quantity of Lead (g)	Cathode Chamber Sample No.	OUTPUTS: Quantity of Lead in Samples (g)	Quantity of Lead Deposited on Cathode (g)	Final Quantity of Lead in Cathode Chamber (g)	Total Quantity of Lead in Outputs (g)	Percent Mass Balance Closure (%)
19	Na4EDTA (1.8% Iron)	4	25	152.0	EC-19-1-C	1.9	84.9	88.6	171.8	89
					EC-19-2-C	1.7				
					EC-19-3-C	1.7				
					EC-19-4-C	1.4				
					EC-19-5-C	1.3				
					EC-19-6-C	1.1				
					EC-19-7-C	0.9				
20	Na4EDTA (1.8% Iron)	4	25	169.6	EC-20-1-C	2.1	87.5	70.1	169.9	100
					EC-20-2-C	2.1				
					EC-20-3-C	1.9				
					EC-20-4-C	1.7				
					EC-20-5-C	1.4				
					EC-20-6-C	1.1				
					EC-20-7-C	1.0				
21	Na4EDTA (1.8% Iron)	4	25	160.0	EC-21-1-C	2.0	89.5	95.2	187.6	01
					EC-21-2-C	2.0				
					EC-21-3-C	1.9				
					EC-21-4-C	1.8				
					EC-21-5-C	1.6				
					EC-21-6-C	1.5				
					EC-21-7-C	1.3				
14	Na2EDTA	0.8	25	34.1	EC-14-1-C	0.4	39.9	0.2	40.1	85
					EC-14-2-C	0.3				
					EC-14-3-C	0.3				
					EC-14-4-C	0.2				
					EC-14-5-C	0.1				
					EC-14-6-C	0.04				
					EC-14-7-C	0.008				
15	Na2EDTA (Regenerated from Run No. 14)	0.8	25	22.3	EC-15-1-C	0.4	59.9	2.8	63.5	51
					EC-15-2-C	0.4				
					EC-15-3-C	0.4				
					EC-15-4-C	0.3				
					EC-15-5-C	0.2				
					EC-15-6-C	0.1				
					EC-15-7-C	0.04				
4	DTPA	0.8	15	32.7	EC-4-1-C	0.4	48.1	3.0	51.3	64
					EC-4-2-C	0.4				
					EC-4-3-C	0.3				
					EC-4-4-C	[illegible]				
					EC-4-5-C	0.3				
					EC-4-6-C	0.4				
					EC-4-7-C	0.3				
					EC-4-8-C	0.1				

TABLE 4–3. (cont.)

Run No.	Chelating Agent	Target Lead Concentration (%)	Current Density (amps/m2)	INPUT: Initial Quantity of Lead (g)	Cathode Chamber Sample No.	OUTPUTS: Quantity of Lead in Samples (g)	Quantity of Lead Deposited on Cathode (g)	Final Quantity of Lead in Cathode Chamber (g)	Total Quantity of Lead in Outputs (g)	Percent Mass Balance Closure (%)
6	DTPA (Regenerated from Run No. 4)	0.8	15	35.2	EC-6-1-C	0.4	30.3	10.6	44.2	80
					EC-6-2-C	0.4				
					EC-6-3-C	0.7				
					EC-6-4-C	0.6				
					EC-6-5-C	0.5				
					EC-6-6-C	0.4				
					EC-6-7-C	0.3				
17	DTPA (Cadmium Electrodes)	0.8	15	32.0	EC-17-1-C	0.4	20.0	13.9	38.0	89
					EC-17-2-C	0.4				
					EC-17-3-C	0.3				
					EC-17-4-C	0.3				
					EC-17-5-C	0.3				
					EC-17-6-C	0.2				
					EC-17-7-C	0.2				
23	DTPA (Ionics Membrane)	0.8	15	32.0	EC-23-1-C	0.4	18.4	21.5	42.3	78
					EC-23-2-C	0.4				
					EC-23-3-C	0.4				
					EC-23-4-C	0.4				
					EC-23-5-C	0.4				
					EC-23-6-C	0.3				
					EC-23-7-C	0.3				
24	DTPA (Ionics Membrane)	0.8	15	30.3	EC-24-1-C	0.4	15.2	14.9	32.1	94
					EC-24-2-C	0.4				
					EC-24-3-C	0.3				
					EC-24-4-C	0.3				
					EC-24-5-C	0.3				
					EC-24-6-C	0.2				
					EC-24-7-C	0.2				
5	DTPA	0.8	25	33.4	EC-5-1-C	0.4	34.8	4.1	41.7	80
					EC-5-2-C	0.4				
					EC-5-3-C	0.6				
					EC-5-4-C	0.5				
					EC-5-5-C	0.4				
					EC-5-6-C	0.3				
					EC-5-7-C	0.1				
7	DTPA	0.8	25	33.8	EC-7-1-C	0.4	40.4	1.5	44.5	76
					EC-7-2-C	0.4				
					EC-7-3-C	0.1				
					EC-7-4-C	0.5				
					EC-7-5-C	0.4				
					EC-7-6-C	0.3				
					EC-7-7-C	0.4				

TABLE 4-3. (cont.)

Run No.	Chelating Agent	Target Lead Concentration (%)	Current Density (ma/cm2)	INPUT: Initial Quantity of Lead (g)	Cathode Chamber Sample No.	OUTPUTS: Quantity of Lead in Samples (g)	Quantity of Lead Deposited on Cathode (g)	Final Quantity of Lead in Cathode Chamber (g)	Total Quantity of Lead in Outputs (g)	Percent Mass Balance Closure (%)
					EC-7-8-C	0.1				
					EC-7-9-C	0.1				
11	DTPA	4	18	169.6	EC-11-1-C	2.1	67.6	67.6	164.2	98
					EC-11-2-C	2.1				
					EC-11-3-C	2.1				
					EC-11-4-C	2.1				
					EC-11-5-C	2.1				
					EC-11-6-C	1.8				
					EC-11-7-C	2.3				
					EC-11-8-C	1.7				
					EC-11-9-C	1.6				
					EC-11-10-C	1.4				
8	DTPA	4	28	168.0	EC-8-1-C	2.0	67.5	69.5	174.6	89
					EC-8-2-C	1.8				
					EC-8-3-C	1.6				
					EC-8-4-C	1.7				
					EC-8-5-C	1.7				
					EC-8-6-C	1.4				
					EC-8-7-C	1.3				
9	DTPA	4	25	153.9	EC-9-1-C	1.9	57.0	60.9	119.7	120
					EC-9-2-C	1.9				
					EC-9-3-C	1.8				
					EC-9-4-C	1.5				
					EC-9-5-C	1.4				
					EC-9-6-C	1.4				
					EC-9-7-C	1.2				
					EC-9-8-C	-				
					EC-9-9-C	0.7				
10	DTPA (Regenerated from Run No. 8)	4	28	239.6	EC-10-1-C	3.0	86.5	125.6	213.0	112
					EC-10-2-C	3.0				
					EC-10-3-C	3.0				
					EC-10-4-C	2.6				
					EC-10-5-C	2.6				
					EC-10-6-C	2.5				
					EC-10-7-C	2.2				
					EC-10-8-C	1.7				
16	DTPA (Cadmium Electrodes)	4	25	160.4	EC-16-1-C	2.1	118.5	80.8	221.5	78
					EC-16-2-C	2.3				
					EC-16-3-C	1.8				
					EC-16-4-C	1.7				
					EC-16-5-C	1.6				
					EC-16-6-C	1.4				
					EC-16-7-C	1.2				

TABLE 4-3. (cont.)

Run No.	Chelating Agent	Target Lead Concentration (%)	Current Density (ma/cm2)	INPUT: Initial Quantity of Lead (g)	Cathode Chamber Sample No.	OUTPUTS: Quantity of Lead in Samples (g)	Quantity of Lead Deposited on Cathode (g)	Final Quantity of Lead in Cathode Chamber (g)	Total Quantity of Lead in Outputs (g)	Percent Mass Balance Closure (%)
22	DTPA (Ionics Membrane)	4	28	165.6	EC-22-1-C	2.1	57.6	99.9	170.3	97
					EC-22-2-C	2.3				
					EC-22-3-C	2.0				
					EC-22-4-C	1.8				
					EC-22-5-C	1.7				
					EC-22-6-C	1.5				
					EC-22-7-C	1.4				

C_L = Final concentration of lead in cathode chamber, g/L
V_L = Volume of liquid in cathode chamber, L
C_{S1}, C_{S2}... = Concentration of sample 1, 2..., g/L
V_{S1}, V_{S2}... = Volume of sample 1, 2..., L
M_c = Mass of lead deposited on cathode, g

Seven of the 24 electromembrane experiment lead mass balance results were outside of the quality control range of 80 to 120 percent established in the quality assurance project plan; five of the seven experiments were only slightly outside of this range. The percent mass balance closure was outside of the quality control range by more than 5 percent for Runs 4 and 15. The closures were below 100 percent indicating that either the initial quantity of lead was measured low, or the outputs (quantity of lead in the samples, the lead deposited on the cathode, and the final quantity of lead in the cathode) were measured too high. Because the initial quantity of lead is known and is approximately equivalent to 0.8 percent lead solution for Runs 4 and 15, it was assumed that the quantity of lead in the outputs was measured too high. Thus, the percent lead removal may be lower than originally reported.

The mass balance for sodium was calculated for each electromembrane experiment by the following formula:

$$M_j = \frac{\Delta M_C}{\Delta M_A} x\, 100$$

where:

M_j = percent mass balance closure for sodium, %
ΔMc = difference in initial and final mass of sodium in the cathode chamber, g
ΔMa = difference in initial and final mass of sodium in the anode chamber, g

The percent closure of the mass balance was calculated by first determining the amount of sodium lost from the anode chamber. This calculation was made by subtracting the sum of the final mass of sodium in the anode chamber solution and the total mass of sodium in the samples collected at various intervals during the tests, from the initial mass of sodium in the anode chamber solution. The difference in the amount of sodium is the amount transferred to the cathode chamber. The amount of

sodium received by the cathode chamber was calculated in the same manner. Table 4-4 presents the sodium mass balance data determined from the 24 electro-membrane tests. All but five of the mass balances were outside of the 80 to 120 percent quality control range. Based on the precision and accuracy data presented in Table 4-1, the method of sodium analysis appears to be adequate. However, the sample matrix appears to pose some difficulties in analyzing sodium as indicated by the mass balance data. Because the material balance closures for sodium were outside of the quality control limits, the data does not appear to be useful for comparison of sodium concentrations.

4.4 DUPLICATE TESTS

Several tests were performed with the same test parameters to determine if the test results could be duplicated. The following tests are duplicates:

Run No.	Chelating Agent	Target lead concentration, %	Current Density, ma/cm^2
1	Na_4EDTA	0.8	25
2	Na_4EDTA	0.8	25
5	DTPA	0.8	25
7	DTPA	0.8	25
8	DTPA	4	25
9	DTPA	4	25
23	DTPA (Ionics membrane)	0.8	15
24	DTPA (Ionics membrane)	0.8	15

The percent lead removal rates in Runs 1 and 2 were 99.4 and 98.0, respectively. The quantities of lead deposited on the cathode were 27.6 and 26.6 grams in Runs 1 and 2, respectively. Other parameters measured during the electromembrane tests (e.g., temperature of the anode and cathode chambers, pH of the anode and cathode chambers) were similar for the two tests. Current efficiencies after a 180-minute

TABLE 4-4. SODIUM MASS BALANCES FOR THE ELECTROMEMBRANE TESTS

Run Number	Chelating Agent	Current Density (ma/cm2)	Initial Target Lead Concentration (%)	Initial Cathode Chamber Quantity of Sodium (g)	Final Cathode Chamber Quantity of Sodium (g)	Cathode Chamber Sample No.	Sample Volume (ml)	Average Sodium Concentration In Cathode Chamber (mg/L)	Sodium Content of Cathode Chamber Samples (g)	Sum of Outputs In Cathode Chamber (a) (g)	Difference In Amount of Sodium In Cathode Chamber (b) (g)	Initial Quantity of Sodium In Anode Chamber (g)	Final Quantity of Sodium In Anode Chamber (g)	Anode Chamber Sample No.	Sample Volume (ml)	Average Sodium Concentration In Anode Chamber (mg/L)	Sodium Content of Anode Chamber Samples (g)	Sum of Outputs In Anode Chamber (a) (g)	Difference In Amount of Sodium In Anode Chamber (b) (g)	Percent Mass Balance Closure (%)
1	Na4 EDTA	25	0.6	84.13	72.48	EC-1-1-C	50	9533	0.43	78.47	44.34	48.00	20.63	EC-1-1-A	50	12000	0.60	28.71	22.29	100
						EC-1-2-C	50	9667	0.48					EC-1-2-A	50	8667	0.41			
						EC-1-3-C	100	11667	1.19					EC-1-3-A	100	11600	1.16			
						EC-1-4-C	100	13200	1.32					EC-1-4-A	100	6733	0.67			
						EC-1-5-C	50	14533	0.73					EC-1-5-A	100	7000	0.70			
						EC-1-6-C	50	16533	0.83					EC-1-6-A	100	6067	0.61			
						EC-1-7-C	50	20133	1.01					EC-1-7-A	100	6067	0.61			
2	Na4 EDTA	25	0.6	21.39	88.88	EC-2-1-C	50	5333	0.27	66.59	46.28	20.67	9.79	EC-2-1-A	50	5167	0.26	11.24	9.46	480
						EC-2-2-C	100	6133	0.61					EC-2-2-A	50	5200	0.26			
						EC-2-3-C	100	6733	0.67					EC-2-3-A	50	4400	0.22			
						EC-2-4-C	100	11600	1.16					EC-2-4-A	50	3667	0.18			
						EC-2-5-C	100	13067	1.31					EC-2-5-A	50	3600	0.18			
						EC-2-6-C	100	14800	1.48					EC-2-6-A	50	3667	0.18			
						EC-2-7-C	100	17067	1.71					EC-2-7-A	50	3667	0.18			
3	Na4 EDTA (Regenerated from Run No. 2)	25	0.6	38.40	64.88	EC-3-1-C	50	9600	0.48	74.45	38.05	31.20	28.88	EC-3-1-A	50	7800	0.39	29.10	2.10	1717
						EC-3-2-C	100	10933	1.09					EC-3-2-A	50	7600	0.38			
						EC-3-3-C	100	12933	1.29					EC-3-3-A	50	6733	0.44			
						EC-3-4-C	100	14667	1.47					EC-3-4-A	50	6800	0.34			
						EC-3-5-C	100	16000	1.60					EC-3-5-A	50	6867	0.34			
						EC-3-6-C	100	17733	1.77					EC-3-6-A	50	6866	0.36			
						EC-3-7-C	100	19800	1.88					EC-3-7-A	50	7267	0.36			
12	Na4EDTA	25	4	165.38	207.20	EC-12A-1-C	50	41333	2.07	224.93	59.60	76.27	88.40	EC-12A-1-A	50	16067	0.80	64.69	11.68	510
						EC-12A-2-C	50	39467	1.97					EC-12A-2-A	50	16000	0.80			
						EC-12A-3-C	50	42133	2.11					EC-12A-3-A	50	16066	0.80			
						EC-12A-4-C	50	55467	2.77					EC-12A-4-A	50	17800	0.89			
						EC-12A-5-C	50	66667	3.33					EC-12A-5-A	50	16036	0.80			
						EC-12A-6-C	50	53600	2.68					EC-12A-6-A	50	16000	0.80			
						EC-12A-7-C	50	56000	2.80					EC-12A-7-A	50	16000	0.80			
13	Na4EDTA (Regenerated from Run No. 12)	25	4	215.47	195.86	EC-13A-1-C	50	53867	2.69	213.48	1.99	62.40	44.28	EC-13A-1-A	50	15600	0.78	46.62	13.66	15
						EC-13A-2-C	50	60533	3.03					EC-13A-2-A	50	14133	0.71			
						EC-13A-3-C	50	57333	2.87					EC-13A-3-A	50	13800	0.69			
						EC-13A-4-C	50	45067	2.25					EC-13A-4-A	50	12800	0.64			
						EC-13A-5-C	50	45600	2.28					EC-13A-5-A	50	11733	0.59			
						EC-14A-6-C	50	47200	2.36					EC-14A-6-A	50	10667	0.53			
						EC-13A-7-C	50	52800	2.64					EC-13A-7-A	50	12133	0.61			

(a) Sum of outputs equals the sum of the quantity of sodium in the samples and the final quantity of sodium in the anode or cathode chamber.

(b) Difference in the amount of sodium in the anode or cathode chamber equals the initial quantity of sodium minus the sum of the outputs.

TABLE 4-4. (cont.)

Run Number	Chelating Agent	Current Density (ma/cm2)	Initial Target Lead Concentration (%)	Initial Cathode Chamber Quantity of Sodium (g)	Final Cathode Chamber Quantity of Sodium (g)	Cathode Chamber Sample No.	Sample Volume (ml)	Average Sodium Concentration in Cathode Chamber (mg/L)	Sodium Content of Cathode Chamber Samples (g)	Sum of Outputs in Cathode Chamber (a) (g)	Difference in Amount of Sodium in Cathode Chamber (b) (g)	Initial Quantity of Sodium in Anode Chamber (g)	Final Quantity of Sodium in Anode Chamber (g)	Anode Chamber Sample No.	Sample Volume (ml)	Average Sodium Concentration in Anode Chamber (mg/L)	Sodium Content of Anode Chamber Samples (g)	Sum of Outputs in Anode Chamber (a) (g)	Difference in Amount of Sodium in Anode Chamber (b) (g)	Percent Mass Balance Closure (%)
18	Na4 EDTA (Cadmium Electrodes)	25	4	179.20	273.81	EC-18-1-C	50	44800	2.24	294.66	115.46	74.67	84.02	EC-18-1-A	50	16667	0.83	89.68	14.71	765
						EC-18-2-C	50	62133	3.11					EC-18-2-A	50	16267	0.81			
						EC-18-3-C	50	60533	3.03					EC-18-3-A	50	17733	0.89			
						EC-18-4-C	50	72533	3.63					EC-18-4-A	50	16667	0.83			
						EC-18-5-C	50	53333	2.67					EC-18-5-A	50	16533	0.83			
						EC-18-6-C	50	59733	2.99					EC-18-6-A	50	16133	0.81			
						EC-18-7-C	50	73867	3.69					EC-18-7-A	50	14800	0.74			
19	Na4 EDTA (1.5 % Iron)	25	4	168.40	199.39	EC-19-1-C	50	41600	2.08	210.55	44.15	76.27	57.43	EC-19-1-A	50	19067	0.95	69.44	12.03	344
						EC-19-2-C	50	41600	2.08					EC-19-2-A	50	20000	1.00			
						EC-19-3-C	50	42400	2.12					EC-19-3-A	50	17333	0.87			
						EC-19-4-C	50	42667	2.13					EC-19-4-A	50	17333	0.87			
						EC-19-5-C	50	78933	3.95					EC-19-5-A	50	15733	0.79			
						EC-19-6-C	50	43733	2.19					EC-19-6-A	50	15067	0.75			
						EC-19-7-C	50	52267	2.61					EC-19-7-A	50	15733	0.79			
20	Na4 EDTA (1.5 % Iron)	25	4	171.73	194.67	EC-20-1-C	50	42933	2.15	210.51	38.79	157.33	151.40	EC-20-1-A	50	49333	2.47	148.66	51.49	75
						EC-20-2-C	50	40000	2.00					EC-20-2-A	50	43200	2.16			
						EC-20-3-C	50	43200	2.16					EC-20-3-A	50	42133	2.11			
						EC-20-4-C	50	44000	2.20					EC-20-4-A	50	41600	2.08			
						EC-20-5-C	50	44533	2.23					EC-20-5-A	50	38200	1.96			
						EC-20-6-C	50	48800	2.44					EC-20-6-A	50	37600	1.88			
						EC-20-7-C	50	53333	2.67					EC-20-7-A	50	36000	1.80			
21	Na4 EDTA (1.5 % Iron)	25	4	157.86	162.80	EC-21-1-C	50	39467	1.97	177.44	19.36	72.00	47.89	EC-21-1-A	50	16000	0.80	69.17	19.69	104
						EC-21-2-C	50	41600	2.08					EC-21-2-A	50	16896	0.85			
						EC-21-3-C	50	40933	2.05					EC-21-3-A	50	16800	0.84			
						EC-21-4-C	50	42400	2.12					EC-21-4-A	50	15467	0.77			
						EC-21-5-C	50	42133	2.11					EC-21-5-A	50	14933	0.75			
						EC-21-6-C	50	42667	2.13					EC-21-6-A	50	14400	0.72			
						EC-21-7-C	50	44000	2.20					EC-21-7-A	50	13067	0.65			
14	Na2 EDTA	25	0.9	21.89	59.71	EC-14-1-C	50	5333	0.27	42.54	21.21	74.67	50.81	EC-14-1-A	50	16667	0.83	66.45	16.22	116
						EC-14-2-C	50	6000	0.30					EC-14-2-A	50	17667	0.88			
						EC-14-3-C	50	7800	0.39					EC-14-3-A	50	17200	0.86			
						EC-14-4-C	50	7667	0.38					EC-14-4-A	50	16400	0.82			
						EC-14-5-C	50	9133	0.46					EC-14-5-A	50	17200	0.86			
						EC-14-6-C	50	10067	0.50					EC-14-6-A	50	15600	0.78			
						EC-14-7-C	50	10733	0.54					EC-14-7-A	50	13667	0.68			
15	Na2 EDTA (Regenerated from Run No. 14)	25	0.9	46.00	63.15	EC-15-1-C	50	12000	0.60	66.32	20.32	65.07	52.10	EC-15-1-A	50	16428	0.82	57.08	7.39	275
						EC-15-2-C	50	13067	0.65					EC-15-2-A	50	19733	0.99			
						EC-15-3-C	50	14000	0.70					EC-15-3-A	50	16267	0.81			
						EC-15-4-C	50	15067	0.75					EC-15-4-A	50	15733	0.79			
						EC-15-5-C	50	16267	0.81					EC-15-5-A	50	14933	0.75			
						EC-15-6-C	50	16000	0.80					EC-15-6-A	50	14267	0.71			
						EC-15-7-C	50	17067	0.85					EC-15-7-A	50	14267	0.71			

TABLE 4-4. (cont.)

Run Number	Chelating Agent	Current Density (ma/cm2)	Initial Target Lead Concentration (%)	Initial Cathode Chamber Quantity of Sodium (g)	Final Cathode Chamber Quantity of Sodium (g)	Cathode Chamber Sample No.	Sample Volume (ml)	Average Sodium Concentration in Cathode Chamber (mg/L)	Sodium Content of Cathode Chamber Samples (g)	Sum of Outputs in Cathode Chamber (a) (g)	Difference in Amount of Sodium in Cathode Chamber (b) (g)	Initial Quantity of Sodium in Anode Chamber (g)	Final Quantity of Sodium in Anode Chamber (g)	Anode Chamber Sample No.	Sample Volume (ml)	Average Sodium Concentration in Anode Chamber (mg/L)	Sodium Content of Anode Chamber Samples (g)	Sum of Outputs in Anode Chamber (a) (g)	Difference in Amount of Sodium in Anode Chamber (b) (g)	Percent Mass Balance Closure (%)
4	DTPA	15	0.8	8.67	7.47	EC-4-1-C	100	2267	0.23	8.90	0.17	11.73	13.02	EC-4-1-A	100	2933	0.29	16.08	3.06	8
						EC-4-2-C	100	2733	0.27					EC-4-2-A	100	3033	0.30			
						EC-4-3-C	100	1133	0.11					EC-4-3-A	100	2933	0.29			
						EC-4-4-C	100	1600	0.16					EC-4-4-A	100	3233	0.32			
						EC-4-5-C	100	1233	0.12					EC-4-5-A	100	4600	0.46			
						EC-4-6-C	80	1800	0.09					EC-4-6-A	80	2167	0.11			
						EC-4-7-C	100	2267	0.23					EC-4-7-A	80	2800	0.14			
						EC-4-8-C	100	2167	0.22					EC-4-8-A	80	3067	0.16			
6	DTPA (Regenerated from Run No. 4)	15	0.8	37.67	40.04	EC-6-1-C	80	9467	0.47	47.85	10.08	30.40	23.86	EC-6-1-A	80	7600	0.38	28.28	4.11	245
						EC-6-2-C	100	10000	1.00					EC-6-2-A	80	7200	0.36			
						EC-6-3-C	100	10267	1.03					EC-6-3-A	80	7000	0.35			
						EC-6-4-C	100	10667	1.07					EC-6-4-A	80	6933	0.35			
						EC-6-5-C	100	11200	1.12					EC-6-5-A	80	6667	0.34			
						EC-6-6-C	100	11333	1.13					EC-6-6-A	80	6800	0.35			
						EC-6-7-C	100	11867	1.19					EC-6-7-A	80	6933	0.35			
17	DTPA (Cadmium Electrodes)	15	0.8	40.00	43.01	EC-17-1-C	50	10000	0.50	47.60	7.60	75.20	69.27	EC-17-1-A	50	16800	0.84	69.84	75.20	10
						EC-17-2-C	50	10000	0.50					EC-17-2-A	50	17667	0.88			
						EC-17-3-C	50	9600	0.48					EC-17-3-A	50	18133	0.91			
						EC-17-4-C	50	10400	0.52					EC-17-4-A	50	18400	0.92			
						EC-17-5-C	50	10800	0.54					EC-17-5-A	50	17467	0.87			
						EC-17-6-C	50	11067	0.55					EC-17-6-A	50	17467	0.87			
						EC-17-7-C	50	11867	0.59					EC-17-7-A	50	17333	0.87			
23	DTPA (Ionics Membrane)	15	0.8	42.40	51.55	EC-23-1-C	50	10600	0.53	55.74	13.34	89.00	68.62	EC-23-1-A	50	22000	1.10	73.71	12.29	109
						EC-23-2-C	50	11467	0.57					EC-23-2-A	50	21067	1.05			
						EC-23-3-C	50	11200	0.56					EC-23-3-A	50	20800	1.04			
						EC-23-4-C	50	11533	0.58					EC-23-4-A	50	20133	1.01			
						EC-23-5-C	50	12308	0.62					EC-23-5-A	50	19467	0.97			
						EC-23-6-C	50	12800	0.64					EC-23-6-A	50	19800	0.99			
						EC-23-7-C	50	13933	0.70					EC-23-7-A	50	18800	0.94			
24	DTPA (Ionics Membrane)	15	0.8	34.84	46.36	EC-24-1-C	50	8733	0.44	52.21	17.27	82.40	71.84	EC-24-1-A	50	20800	1.04	78.68	3.72	464
						EC-24-2-C	50	9800	0.49					EC-24-2-A	50	20200	1.01			
						EC-24-3-C	50	9800	0.49					EC-24-3-A	50	21067	1.05			
						EC-24-4-C	50	10733	0.54					EC-24-4-A	50	21133	1.06			
						EC-24-5-C	50	11800	0.59					EC-24-5-A	50	20933	1.05			
						EC-24-6-C	50	13267	0.66					EC-24-6-A	50	19200	0.96			
						EC-24-7-C	50	13067	0.65					EC-24-7-A	50	19800	0.99			
5	DTPA	25	0.8	52.27	71.76	EC-5-1-C	50	13067	0.65	62.73	30.46	29.60	22.67	EC-5-1-A	50	7400	0.37	25.30	4.30	708
						EC-5-2-C	100	13067	1.31					EC-5-2-A	50	8600	0.43			
						EC-5-3-C	100	14000	1.40					EC-5-3-A	50	6733	0.34			
						EC-5-4-C	100	16267	1.63					EC-5-4-A	50	6733	0.34			
						EC-5-5-C	100	16933	1.69					EC-5-5-A	50	6133	0.31			
						EC-5-6-C	100	20133	2.01					EC-5-6-A	50	6667	0.33			
						EC-5-7-C	100	20800	2.08					EC-5-7-A	50	6267	0.31			

TABLE 4-4. (cont.)

Run Number	Chelating Agent	Current Density (ma/cm2)	Initial Target Lead Concentration (%)	Initial Cathode Chamber Quantity of Sodium (g)	Final Cathode Chamber Quantity of Sodium (g)	Cathode Chamber Sample No.	Sample Volume (ml)	Average Sodium Concentration in Cathode Chamber (mg/L)	Sodium Content of Cathode Chamber Samples (g)	Sum of Outputs in Cathode Chamber (a) (g)	Difference in Amount of Sodium in Cathode Chamber (b) (g)	Initial Quantity of Sodium in Anode Chamber (g)	Final Quantity of Sodium in Anode Chamber (g)	Anode Chamber Sample No.	Sample Volume (ml)	Average Sodium Concentration in Anode Chamber (mg/L)	Sodium Content of Anode Chamber Samples (g)	Sum of Outputs in Anode Chamber (a) (g)	Difference in Amount of Sodium in Anode Chamber (b) (g)	Percent Mass Balance Closure (%)
7	DTPA	25	0.8	44.27	60.67	EC-7-1-C	50	11067	0.55	73.48	28.18	90.14	17.08	EC-7-1-A	50	7000	0.35	20.12	10.02	201
						EC-7-2-C	100	11667	1.19					EC-7-2-A	50	6800	0.46			
						EC-7-3-C	100	12667	1.27					EC-7-3-A	50	6800	0.34			
						EC-7-4-C	100	13700	1.37					EC-7-4-A	50	7600	0.38			
						EC-7-5-C	100	15067	1.51					EC-7-5-A	50	6933	0.35			
						EC-7-6-C	100	16133	1.61					EC-7-6-A	50	6800	0.35			
						EC-7-7-C	100	16667	1.67					EC-7-7-A	50	8400	0.42			
						EC-7-8-C	100	17800	1.78					EC-7-8-A	50	5067	0.25			
						EC-7-9-C	100	18667	1.87					EC-7-9-A	50	4667	0.23			
11	DTPA	15	4	158.80	173.24	EC-11-1-C	50	38200	1.96	187.27	40.47	73.07	51.80	EC-11-1-A	50	16267	0.81	60.09	12.69	312
						EC-11-2-C	50	37067	1.85					EC-11-2-A	50	16133	0.81			
						EC-11-3-C	50	46400	2.32					EC-11-3-A	50	17800	0.89			
						EC-11-4-C	50	51467	2.57					EC-11-4-A	50	17800	0.89			
						EC-11-5-C	50	46133	2.31					EC-11-5-A	50	17600	0.88			
						EC-11-6-C	50	41600	2.08					EC-11-6-A	50	17200	0.86			
						EC-11-7-C	50	45333	2.27					EC-11-7-A	50	16800	0.84			
						EC-11-8-C	50	48800	2.44					EC-11-8-A	50	17200	0.86			
						EC-11-9-C	50	55733	2.79					EC-11-9-A	50	15467	0.77			
						EC-11-10-C	50	48800	2.44					EC-11-10-A	50	14133	0.71			
8	DTPA	25	4	194.67	199.80	EC-8-1-C	50	48667	2.43	217.53	22.86	60.00	84.24	EC-8-1-A	50	20000	1.00	71.06	8.66	256
						EC-8-2-C	50	47667	2.38					EC-8-2-A	50	21667	1.09			
						EC-8-3-C	50	46667	2.33					EC-8-3-A	50	20267	1.01			
						EC-8-4-C	50	52333	2.62					EC-8-4-A	50	19467	0.97			
						EC-8-5-C	50	53333	2.67					EC-8-5-A	50	19067	0.95			
						EC-8-6-C	50	52000	2.60					EC-8-6-A	50	18400	0.92			
						EC-8-7-C	50	54000	2.70					EC-8-7-A	50	17800	0.89			
9	DTPA	25	4	192.00	198.00	EC-9-1-C	50	48000	2.40	216.75	24.75	62.13	52.56	EC-9-1-A	50	20533	1.05	61.20	20.93	116
						EC-9-2-C	50	49333	2.47					EC-9-2-A	50	20000	1.00			
						EC-9-3-C	50	50000	2.50					EC-9-3-A	50	19467	0.97			
						EC-9-4-C	50	50333	2.52					EC-9-4-A	50	18867	0.94			
						EC-9-5-C	50	52000	2.60					EC-9-5-A	50	18400	0.92			
						EC-9-6-C	50	51333	2.57					EC-9-6-A	50	17800	0.89			
						EC-9-7-C	50	19000	0.95					EC-9-7-A	50	43467	2.17			
						EC-9-8-C	50							EC-9-8-A	50					
						EC-9-9-C	50	55000	2.75					EC-9-9-A	50	14400	0.72			
10	DTPA (Regenerated from Run No. 9)	25	4	193.67	197.10	EC-10-1-C	50	47667	2.38	217.60	28.93	66.67	50.97	EC-10-1-A	50	21667	1.09	68.47	28.20	116
						EC-10-2-C	50	51000	2.55					EC-10-2-A	50	21000	1.05			
						EC-10-3-C	50	50333	2.52					EC-10-3-A	50	19333	0.97			
						EC-10-4-C	50	50000	2.50					EC-10-4-A	50	18667	0.93			
						EC-10-5-C	50	50667	2.53					EC-10-5-A	50	19000	0.95			
						EC-10-6-C	50	51667	2.58					EC-10-6-A	50	17667	0.88			
						EC-10-7-C	50	52667	2.63					EC-10-7-A	50	17333	0.87			
						EC-10-8-C	50	54000	2.70					EC-10-8-A	50	15333	0.77			

TABLE 4–4. (cont.)

Run Number	Chelating Agent	Current Density (ma/cm2)	Initial Target Lead Concentration (%)	Initial Cathode Chamber Quantity of Sodium (g)	Final Cathode Chamber Quantity of Sodium (g)	Cathode Chamber Sample No.	Sample Volume (ml)	Average Sodium Concentration in Cathode Chamber (mg/L)	Sodium Content of Cathode Chamber Samples (g)	Sum of Outputs in Cathode Chamber (a) (g)	Difference in Amount of Sodium in Cathode Chamber (b) (g)	Initial Quantity of Sodium in Anode Chamber (g)	Final Quantity of Sodium in Anode Chamber (g)	Anode Chamber Sample No.	Sample Volume (ml)	Average Sodium Concentration in Anode Chamber (mg/L)	Sodium Content of Anode Chamber Samples (g)	Sum of Outputs in Anode Chamber (a) (g)	Difference in Amount of Sodium in Anode Chamber (b) (g)	Percent Mass Balance Closure (%)
16	DTPA (Cadmium Electrodes)	25	4	169.60	161.41	EC-16-1-C	50	47200	2.36	209.64	19.84	74.67	59.86	EC-16-1-A	50	16667	0.85	65.68	[illegible]	281
						EC-16-2-C	50	50667	2.53					EC-16-2-A	50	17200	0.89			
						EC-16-3-C	50	48533	2.43					EC-16-3-A	50	14267	0.81			
						EC-16-4-C	50	46133	2.31					EC-16-4-A	50	15533	0.77			
						EC-16-5-C	50	46667	2.33					EC-16-5-A	50	17467	0.87			
						EC-16-6-C	50	53600	2.68					EC-16-6-A	50	15067	0.75			
						EC-16-7-C	50	51733	2.59					EC-16-7-A	50	16400	0.82			
22	DTPA (Ionics Membrane)	25	4	179.20	174.64	EC-22-1-C	50	44800	2.24	190.60	11.40	60.67	62.76	EC-22-1-A	50	21467	1.07	69.67	16.20	70
						EC-22-2-C	50	44800	2.24					EC-22-2-A	50	21200	1.06			
						EC-22-3-C	50	44000	2.20					EC-22-3-A	50	20400	1.02			
						EC-22-4-C	50	45800	2.29					EC-22-4-A	50	19467	0.97			
						EC-22-5-C	50	45800	2.29					EC-22-5-A	50	20000	1.00			
						EC-22-6-C	50	47200	2.36					EC-22-6-A	50	18133	0.91			
						EC-22-7-C	50	47200	2.36					EC-22-7-A	50	17200	0.86			

reaction period were 35 and 36.6 percent in Runs 1 and 2, respectively. Therefore, the tests with tetra-sodium EDTA show good replication.

The percent lead removals in Runs 5 and 7 were 85.8 and 58.0 percent, respectively. These percentages are quite different; however, the percentage of lead removal after 150 minutes was 66.8 percent in Run 7, which is a higher percentage than for 180 minutes. Therefore, it is assumed that the analysis of the last lead sample (180 minutes) was below what the actual lead concentration should have been. The actual lead removal should be approximately 77 percent for the 180-minute sample in Run 7. The amount of lead deposited on the cathode was similar: 34.9 grams as deposited in Run 5, and 31.1 grams as deposited in Run 7. All other data (e.g., pH of anode and cathode chamber, temperature of anode and cathode chamber) were similar in both tests. Current efficiencies for Runs 5 and 7 were 32 and 23.5, respectively. Therefore, the runs show good replication.

In Runs 8 and 9, the percent lead removals were 33.8 and 40.2 percent, respectively after a 3-hour reaction period. The amount of lead deposited on the cathode was 67.5 grams in Run 8 and 36.2 grams in Run 9. The experimental data from Table 3-2 shows that the anode and cathode chamber temperatures, and anode and cathode chamber pH levels were very similar for the two tests. Current efficiencies in Runs 8 and 9 were 66.1 and 64.4 (after 210 minutes) percent, respectively.

In Runs 23 and 24, the percent lead removals were 28.1 and 47.3 percent, respectively, after a 3-hour reaction period. The quantities of lead deposited on the cathode was 18.4 grams in Run 23 and 15.2 grams in Run 24. The initial quantity of lead in the cathode chamber was 32.0 and 30.3 grams for Runs 23 and 24, respectively. According to the experimental data presented in Table 3-2, the anode and cathode chamber pH levels were similar, the anode chamber temperatures were sirnilar, but the change in cathode chamber temperature was higher for Run 24 (8.4°C) than for Run 23 (4.9°C). The current efficiencies for Runs 23 and 24 were 19.7 and 28.5 (after 3 hours) percent, respectively. Based on the data for Runs 23 and 24, the results are comparable.

Based on the comparisons of the duplicate tests, it appears that better replication of results occurred for the tests conduced with 0.8 percent lead solutions than for

4 percent lead solutions. Only a selected number of conditions were duplicated. Other test conditions such as use of cadmium electrodes and use of the Ionics membrane were performed with a minimum number of tests in order to approximate the percent lead recoveries under these conditions. Further testing would need to be performed if these parameters were to be selected for pilot- or full-scale application.

4.5 Impact on Quality

The bench-scale treatability program was designed as a screening study to identify effects of various experimental parameters. Conclusions have been made in the report based on trends that are intuitively apparent from the data. These conclusions are not interpreted in light of analytical and field QC samples that were analyzed in conjunction with the field samples. The results from the bench-scale treatability program provide useful conclusions regarding the effects of various parameters investigated in this study, but may not be compared directly to results obtained outside of this study.

The following subsection presents a discussion of certain parameters that were outside of the limits specified in the quality objectives and how this impacts the results of this study.

4.5.1 pH

The data quality objectives for this study were to adjust the initial lead-chelate solution pH levels to ± 0.5 pH unit of the prescribed experimental pH level. For Runs 2, 4, 5, 7, 17, 19, 20, and 21, the pH level of the starting lead-chelate solution was greater than ±0.5 pH unit from the experimental pH. In Runs 2, 4, 5, 7, and 17, the solutions were made with 0.8 percent lead.

Runs 19, 20, and 21 were conducted with 1.5 percent iron in the 4 percent starting lead solutions. While the lead/iron-chelate solution was being mixed, the iron was oxidized to Fe^{+3} and sulfuric acid was generated, which decreased the solution pH to below pH level 7. According to Tables 3-3 and 3-4, almost all of the lead was chelated in the solutions for Runs 19, 20, and 21. Therefore, there appears to be no appreciable impact to the results of these tests due to the low starting solution pH levels.

4.5.2 Lead Removal

In Runs 10, 11, 12, 13, 15, 16, 18, 21, 22, and 23, the lead concentration in the initial cathode sample was lower than in subsequent samples. Therefore, either the subsequent sample analyses were high or the initial quantity of lead in the cathode chamber should have been greater. If the initial quantity of lead was higher than actually measured, then the percent lead removal should have been higher than reported. This may be the case for Runs 11, 12, and 23 where more than one subsequent sample contained more lead than the initial cathode chamber sample.

The percent lead removal from Run 11 was used to compare the lead removal rates for 0.8 and 4 percent lead solutions of DTPA, and for 15 and 25 ma/cm^2 tests with 4 percent lead solutions. Based on the data presented in Table 3-3, the initial lead solution concentration is assumed to be 42,650 mg/L. By extrapolation, the lead solution concentration for 180 minutes plating time was calculated to be 36,300 mg/L. Using these new lead solution concentrations, it is estimated that the percent lead removal was 14.9 percent after a 3-hour plating time. Because this new percent lead removal is lower than the original percent removal of 35.5 percent, the conclusions for Run 11 are correct.

Based on the data presented in Table 3-3, the initial lead solution concentration in the cathode chamber is assumed to be 46,000 mg/L, which would yield an overall percent lead removal of 21.7. The percent lead removal from Run 12 was used to compare the lead removal for 1) 4 percent lead solutions of EDTA and DTPA, 2) lead and cadmium electrodes, and 3) lead and lead/iron chelate solutions. In all three cases, the 21.7 percent lead removal is still lower than the lead removals of the other runs compared to Run 12. Therefore, it is assumed that the conclusions for Run 12 are correct.

The percent lead removal from Run 23 was compared to Runs 4 and 24 to determine if the type of membrane affects lead removal. Based on the data presented in Table 3-3, the initial lead solution concentration for Run 23 is assumed to be 8583.3 mg/L, which yields a 33.0 percent lead removal. Because the new percent lead removal for Run 23 is still less than that of Runs 4 and 24, the conclusion for Run 23 is correct.

SECTION 5.0

CONCLUSIONS AND RECOMMENDATIONS

The purpose of this study was to examine the ability of an innovative electromembrane process (which uses a cation-transfer membrane separating the anode and cathode chambers in an electrolytic cell) to recover lead from a synthetic lead solution. The synthetic lead solution was used to simulate a soil washing solution (obtained after chelation) from a typical battery reclamation site. Process parameters that were studied included the following: type of chelating agent, type of membrane, current density, lead concentration, and reaction time. The bench-scale treatability program was designed as a screening study and was not intended to enable development of rigorous conclusions regarding the various experimental parameters. Thus, no quantitative criteria were established to determine significant differences between or among runs. The conclusions that have been made in the report are those that are intuitively apparent from different sets of data. Certain conclusions are not fully supported by all the data collected for the report.

Preliminary jar tests performed in this study determined that lead dioxide and elemental lead could not be chelated by any of the chelating agents studied (di-sodium EDTA, tetra-sodium EDTA, and DTPA), but lead sulfate and basic lead carbonate could be completely chelated by all three chelating agents. The optimal chelating-agent-to-lead molar ratios were determined to be 1:1 for di-sodium EDTA, 1:1.5 for tetra-sodium EDTA, and 1:2 for DTPA.

A comparison of the tests using di-sodium EDTA and tetra-sodium EDTA under the same conditions showed that both forms of EDTA produced approximately the same lead recovery. Based on the treatability study data, there appears to be no advantage to using one sodium form of EDTA over the other. The use of DTPA as the chelating agent resulted in lower lead recoveries based on data using a solution containing 0.8 percent initial lead concentration and higher lead recoveries for the solutions containing 4 percent initial lead concentration. Because the test with 4

percent lead concentration tetra-sodium EDTA solution and 25 ma/cm^2 current density (Run 12) appeared to have a lower-than-expected lead recovery, it is uncertain whether DTPA is superior. Additional tests with actual soil and a cost analysis should be performed to compare the capabilities and cost-effectiveness of EDTA and DTPA for removing lead from soil.

The data from the regenerated chelating agent solution tests showed that the regenerated chelating agent solutions resulted in lead removals comparable to those from the original solutions.

A comparison of the data obtained in the tests performed using initial target lead concentrations of 0.8 and 4 percent showed that a higher percentage of lead was recovered in the 0.8 percent lead solution test, but the amount of lead recovered was greater in the 4 percent lead solution test. One possible reason the lead removal percentages were not higher in the electromembrane tests conducted with 4 percent lead-chelate was the limited surface area of the cathode. The cathode appeared to be "saturated" with lead, and therefore the lead may have been inhibited from plating onto the cathode and thus remained in the solution. It is suggested that future tests be run to investigate how surface area and shape of the electrode affect lead recovery. This data also indicates that the use of a higher percentage lead solution results in more lead recovery and higher current efficiencies.

A comparison of the tests conducted with 15 and 25 ma/cm^2 current densities showed that the lead recovery rates and current efficiencies were higher for the 25 ma/cm^2 current density tests.

Lead recovery efficiencies of the Nafion® and Ionics membranes were compared to determine if the type of membrane used had any effect on lead recovery. Based on the data from the tests in the 0.8 and 4 percent lead solutions, it appears that there is no difference in lead recovery for both the Nafion® and Ionics membranes. A cost analysis would need to be performed to determine which membrane is more cost-effective.

The tests with lead and cadmium electrodes were compared using DTPA solutions as well as tetra-sodium EDTA solutions. In tests conducted with tetra-

sodium EDTA, the cadmium electrodes were definitely superior to the lead electrodes with respect to lead recovery rates. However, the initial lead concentration for the test with 4 percent lead concentration EDTA solution, 25 ma/cm^2 current density, and lead electrodes (Run 12) was in question because the lead concentration was higher in subsequent samples. The tests with DTPA solutions, however, did not reveal a significant increase in lead recovery when using the cadmium electrodes. A cost analysis should be performed to determine if the lower cost of cadmium outweighs the benefit of lead product purity. With the lead electrodes, it was anticipated that the entire electrode would be smelted instead of scraping the lead from the cathode. The use of cadmium as the cathode requires a comparison of the cost of scraping the lead from the cathode versus the saleable product produced from the lead deposited onto the cadmium electrode.

In the tests conducted with 1.5 percent iron, a slightly higher lead recovery was observed in the solutions containing iron.

In future testing, it is recommended that actual lead-contaminated soil be used in the study to determine if exclusive lead chelation is possible and to determine the degree to which other metals in the soil are chelated. Various types of soils ranging from clay to sandy should be tested to determine the effect of particle size on separation of soil and chelating agent solution. Previous soil washing tests have shown that a colloid of soil and chelating agent may form which can carry-over to the electromembrane process and plug the membrane.

In this study, the chelating agent solutions were regenerated once; however, it is unknown whether there is a limit to regeneration that will produce an unusable chelating agent solution. Multiple regenerations of the chelating agent should be investigated, especially with soil, to determine the extent of regeneration of the chelating agent.

SECTION 6

REFERENCES

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APPENDIX A

SOLUTION PREPARATION METHOD FOR THE ELECTROMEMBRANE TESTS

Jar Tests

The lead solutions containing di-sodium EDTA, tetra-sodium EDTA, and DTPA were prepared by adding deionized water, chelating agent, and lead to a 250 ml beaker. The contents of the beaker were stirred using a magnetic stirrer. While the solutions were being stirred, the solution pHs were adjusted to a pH level of 9 with sulfuric acid for the tetra-sodium and DTPA solutions, and to a pH level of 5 with sodium hydroxide for the di-sodium EDTA solutions. The solutions were allowed to mix for one hour after pH adjustment. The tetra-sodium EDTA and DTPA chelating agents were commercial solutions purchased from the Hampshire Chemical Company. The tetra-sodium EDTA solution was a mixture of approximately 38 percent by weight chelating agent and the remaining 62 percent by weight was water, trisodium nitrilotriacetic acid (<2 percent) and sodium hydroxide (1 to 2 percent). Di-sodium EDTA was supplied as a solid containing 10 percent water. The DTPA solution was a mixture of 40.1 percent chelating agent, and the remaining 59.9 percent by weight was water, trisodium nitriloacetic acid, and sodium hydroxide. The following presents the quantities of lead, water, and chelating agent solution used to prepare the 4 percent by weight lead jar test solutions.

	Molar ratio (chelating agent:lead)		
	1:1	1.5:1	2:1
Di-sodium EDTA			
Lead sulfate, g	8.1	8.1	8.1
Basic lead carbonate, g	6.3	6.3	6.3
Di-sodium EDTA, g	13.1	19.8	26.1
Deionized water, ml	180	180	180
Tetra-sodium EDTA			
Lead sulfate, g	8.1	8.1	8.1
Basic lead carbonate, g	6.3	6.3	6.3
Di-sodium EDTA, g	26.9	40.4	53.8
Deionized water, ml	153.1	139.6	126.2
DTPA			
Lead sulfate, g	8.1	8.1	8.1
Basic lead carbonate, g	6.3	6.3	6.3
Di-sodium EDTA, g	33.8	50.7	67.5
Deionized water, ml	146.2	129.3	112.5

Bench-Scale Tests

The 0.8 and 4 percent by weight lead solutions were prepared by adding the chelating agent, water, and lead to a 4-liter flask. The contents were stirred using a magnetic stirrer. While the solutions were being stirred, the solution pHs were adjusted to the experimental pH with sulfuric acid for tetra-sodium EDTA and DTPA solutions, and with sodium hydroxide for di-sodium EDTA solutions. The lead chelate solutions were allowed to mix for one hour to complete the lead chelation. The solutions were prepared with the following quantities of lead, chelating agent, and water.

	0.8 percent lead	4 percent lead
Di-sodium EDTA		
Lead sulfate, g	35.0	--
Basic lead carbonate, g	26.8	--
Di-sodium EDTA, g	56.0	--
Deionized water, ml	4,000	--
(1:1 Di-sodium EDTA-to-lead molar ratio)		
Tetra-sodium EDTA*		
Lead sulfate, g	35.0	179.8
Basic lead carbonate, g	26.8	137.4
Tetra-sodium EDTA, ml	173.1	888.5
Deionized water, ml	3,826.9	3,111.5
(1.5:1 Tetra-sodium EDTA-to-lead molar ratio)		
DTPA		
Lead sulfate, g	35.0	179.8
Basic lead carbonate, g	26.8	137.4
DTPA, ml	289.5	1,485.9
Deionized water, ml	3,710.5	2,514.1
(2:1 DTPA-to-lead molar ratio)		

* For the solutions prepared with 1.5 percent by weight iron, 298.7 grams ferrous sulfate ($FeSO_4 \cdot 7H_2O$) was added to the lead solution prior to pH adjustment.