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AN ELECTROLYTIC PROCEDURE FOR THE REMOVAL OF RUTHENIUM AND NITRATE FROM ALKALINE WASTE SOLUTIONS

A. F. Messing
I. R. Higgins

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CHEMICAL TECHNOLOGY DIVISION

Chemical Development Section B

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ABSTRACT

The flowsheet proposed by KAPL for the treatment of alkaline nitrate radiochemical processing waste has been modified to include an ion-exchange step for the decontamination of cesium, strontium, and other cationic fission products. In laboratory studies of the electrolysis steps in a nitrate reduction cell, synthetic alkaline waste, 0.55 - 4.83 M total electrolyte, was decontaminated from ruthenium by factors of $>$210. The nitrate of 3.0 M waste was reduced to ammonia with current efficiencies of 100%. Power consumption was 4.7 kwh/lb of nitrate reduced. Significant factors affecting the rate of ruthenium decontamination were temperature, cathode area, cathode current density, and electrolyte concentration. Those affecting nitrate reduction current efficiencies were the cathode current density, electrolyte concentration, and stirring rate.

In an acid-base membrane cell, reusable nitric acid as well as sodium hydroxide was regenerated. However, such a cell is less economical to construct and operate than the nitrate reduction cell. At least 5 kwh of power is required to transfer 1 lb of nitrate from ORNL type waste. In addition, the greater complexity of the acid-base cell makes it less adaptable for remote control.

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1.0 INTRODUCTION

The purpose of this investigation was to develop an electrolytic procedure for removing ruthenium and nitrate from alkaline waste solutions. The process is considered to be a step in a general waste treatment method.

Fuel reprocessing wastes, neutralized with caustic, contain nearly 100% of the cesium and nitrate in solution along with about 10% of the ruthenium and lesser quantities of strontium and the rare earths. Because of the biological hazards and long half-lives of these species, direct release of such solutions to the environment is limited. Efficient and economical removal of cesium, strontium, and the rare earths has been demonstrated on a laboratory scale.[1] Thus, ruthenium and nitrate remain as the major deterent to direct environmental disposal.

Ruthenium is significant in that the predominant isotope, Ru-106, is radioactive with a half-life of about one year.[2] In solution, ruthenium exists in a variety of ionic and nonionic species.[3,4] Thus it is not easily fixed in soils[5] and offers a threat to ground water supplies. No radioactivity is associated with nitrate. Its importance lies in the fact that it is not fixed by soils or easily contained in pits[5] and tends to concentrate in ground water. A limit of 10 mg/liter has been set for nitrate nitrogen in ground water supplies.[6]

Electrolytic methods appear to be the most efficient and economical means for solving the ruthenium and nitrate problem. Three types of cells have been suggested for this purpose.[7] The first type is an acid-base membrane cell of the Ionics design[8] which converts sodium nitrate to sodium hydroxide and nitric acid for recycle. The cationic fraction of ruthenium migrates to the cathode where it is deposited while the anionic fraction appears in the nitric acid product. Nonionic species are not removed from the waste solution. The second type is the nitrate reduction cell which produces caustic and ammonia gas, as studied by KAPL.[9-11] Ruthenium is plated onto the cathode. The third type is the ruthenium plating cell which is similar in design to the nitrate reduction cell but is operated at a low current density so that a minimum quantity of the nitrate is reduced.

All experimental work discussed in this report was on a laboratory scale; a detailed description of the cells studied is included in the appendix. Ruthenium plating and the majority of the nitrate reduction studies, with the production of sodium hydroxide, were performed in a small cell with a capacity of 500 ml. Investigation included the effect of varying solution temperature, applied potential, electrode current density, electrode area, solution concentration, and stirring rate. Additional nitrate reduction studies were made in a KAPL design cell with a capacity of 1800 ml. Nitrate and sodium migration studies with the production of nitric acid and sodium hydroxide were made with an Ionics design and a less complex, simple design acid-base membrane cell. Comparison of the cell types was made, based on simplicity and economy of construction and efficiency and economy of operation.

In order to deposit ruthenium, cathodic evolution of hydrogen is required.[10-13] Once the hydrogen over-voltage is exceeded, ruthenium is deposited at a first order rate with a positive temperature coefficient.[10] Present in at least two forms, ruthenium divides itself between a "platable" and an "unplatable" fraction.[13] The platable fraction readily deposits on bright platinum while the unplatable fraction does not. However, when the cathode is platinum black, a hydrogenation catalyst, all the ruthenium is deposited. Similarly, after a monomolecular layer of ruthenium has been formed on the cathode, the characteristics are like those of platinum black and essentially all the ruthenium is deposited.[12]

The chief products of electrolytic nitrate reduction in alkaline solution are ammonia and sodium hydroxide.[14] Reduction goes through the oxidation states, +3, +1, and -1 with the intermediate products being nitrite, hyponitrite, and hydroxylamine. Anodic reoxidation of nitrite to nitrate is favored in dilute hydroxide solutions but does not occur to a measurable degree in solutions of high hydroxide concentration.

The advent of the ion exchange membranes introduced an economical way of producing acids and bases from salts by migration. Under the influence of an electric current, cations of a salt, placed in the central compartment of a three-compartment cell, migrate through a cation exchange membrane into the cathode compartment while anions migrate through an anion exchange membrane into the anode compartment. With the decomposition of water to hydroxide ion and hydrogen gas at the cathode and to hydrogen ion and oxygen gas at the anode, base and acid are formed as respective products.

Synthetic waste solutions were used in this investigation; studies on actual waste in a shielded facility remain to be done. Nitrate reduction and ruthenium plating studies were carried out batchwise. Design and evaluation of a nitrate reduction cell for continuous operation is needed. While ruthenium decontamination factors of >210 were demonstrated, it is uncertain whether the remaining trace activity was ruthenium or not. Experimentation is thus required on solutions containing higher levels of ruthenium activity.

The authors are indebted to G. R. Wilson and to W. R. Laing and staffs of the ORNL Analytical Chemistry Division for analyses performed.

2.0 PROCESS DESCRIPTION

The flowsheet (Fig. 2.1) for the treatment of fuel reprocessing waste is identical to that proposed and described by KAPL[9] with the exception that an ion-exchange step and an additional evaporation step have been added (steps 2 and 3). Continuous ion exchange has been inserted for the purpose of removing cesium, strontium, and other cationic species from solution, thus preventing their build-up in the system. Operational data for the ion-exchange step may be found elsewhere.[1] An evaporation step has been inserted prior to electrolysis for concentration of waste solutions with low salt content. This is directed specifically to the type of waste found at ORNL.

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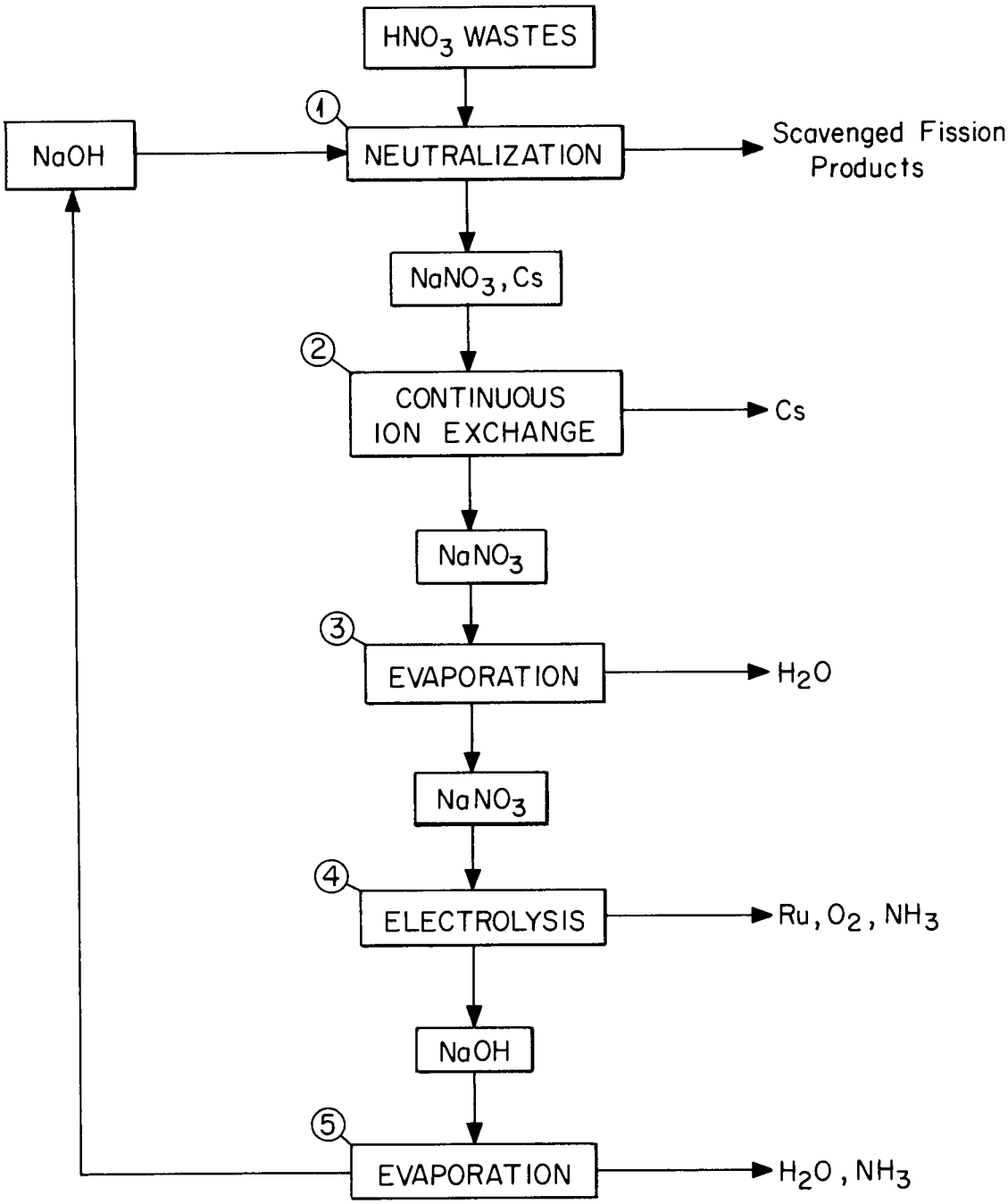

Fig. 2.1. Flowsheet for treatment of fuel reprocessing wastes.

It is estimated that a 250-gal KAPL design cell containing six types 347 stainless steel anodes and five Armco iron cathodes, 48 by 12 in., would process 1,000 liters of 2.0 M $NaNO_3$--0.7 M NaOH waste in 24 hr when operated at a current density of 3 amp/in.^{2} at 100°C. Nitrate levels would be reduced below 0.1 M with decontamination factors of > 200 for ruthenium. Because of the simplicity of its construction, this cell is adapted to remote handling and maintenance. It is considered the most practical electrolytic method for the treatment of alkaline waste with regard to nitrate and ruthenium. Not only are current efficiencies high, but, in addition, at higher concentrations the current density may be increased without harming the efficiency or power requirements. The sodium hydroxide produced would be reused. Trace activity and unreduced nitrate would remain in the system to be recycled, none being released to the environment.

In a second electrolytic process ruthenium is removed in a plating cell. The effluent, containing sodium nitrate, is passed through an acid-base membrane cell, with the production of sodium hydroxide and nitric acid. Recycle of both the nitric acid and sodium hydroxide products is assumed. However, even at the low current densities at which the ruthenium plating cell operates, approximately 20% of the nitrate is reduced in ORNL type wastes in the time required to obtain maximum ruthenium decontamination. Thus much of the nitric acid product is lost. The time required for maximum decontamination from ruthenium increased from about 17 hr to about 25 hr from 500 ml of ORNL type waste at 60°C when the current density was decreased on 3-in.^{2} electrodes from 1.0 to 0.13 amp/in.^{2} Results further indicate that the time of maximum ruthenium decontamination would be increased still more in solution of higher salt content, other factors remaining constant.

The acid-base membrane cell operated at 60-70% efficiency on ORNL type solutions with the production of about 1.0 M HNO_3 and 1.5 M NaOH. At least two cells connected in series would be necessary to reduce nitrate concentrations to acceptable levels for environmental release. Power consumption to transfer 1 lb of nitrate from the waste solution was > 5 kwh, depending on the type of cell used and the stage under consideration. Previously, it was found that the efficiency of nitrate migration decreased rapidly to about 20% as the solution concentration was increased to about 2.5 M $NaNO_3$ and then remained essentially constant.[15] An increase in power requirements would be expected as the solution salt content is increased. Finally, because of the number of cells required in the process and because of their complex construction, remote operation and maintenance problems are expected to be great in comparison to those of the nitrate reduction cell.

3.0 RUTHENIUM PLATING AND NITRATE REDUCTION STUDIES

The rate of ruthenium plating was influenced by temperature, cathode area, cathode current density, and concentration of solution electrolyte but was independent of variations in applied potential, anode area, and anode current density. From 500 ml of 0.25 M $NaNO_3$--0.12 M NaOH solution, with a cathode area of 3 in.^{2} and a cathode current density of 1 amp/in.^{2}, the half-life for ruthenium plating decreased from 3.3 to 1.3 hr as the temperature was increased from 46 to 80°C. An activation energy of 6.0 ± 1.0 kcal/mole was calculated for the plating reaction. The ruthenium plating half-life decreased from 1.5 to 1.1 hr as the current density on a 3-in.^{2} cathode was increased from 1.0 to 3.0 amp/in.^{2} from a 500 ml volume of this solution at a temperature of 80°C. Maintaining a cathode current density of 3 amp/in.^{2} but reducing the area to 1 in.^{2} resulted in a decrease in the plating half-life from 1.1 to 1.6 hr. Finally, at 5 volts, 80°C, a cathode area of 3 in.^{2}, and a cathode current density of 1 amp/in.^{2}, the ruthenium deposition half-life increased from 1.4 to 1.7 hr when the solution concentration was increased from 0.25 M $NaNO_3$--0.12 M NaOH to 0.70 M $NaNO_3$--0.20 M NaOH. Decontamination factors from ruthenium were >210, corresponding to 99.8% ruthenium removal. It must be borne in mind that the ruthenium plating half-lives, as stated, are a function of all the above variables. They are therefore valid only under the conditions stated.

Current efficiencies, based on nitrate reduction, were dependent only on the cathode current density, the molarity of the solution, and the rate of solution stirring. On electrolysis of a 0.25 M $NaNO_3$--0.12 M NaOH solution, an increase in the cathode current density from 1.0 to 3.0 amp/in.^{2} resulted in a decrease in current efficiency from 53 to 21%. As the electrolyte concentration of the solution was increased from 0.25 M $NaNO_3$--0.12 M NaOH to 3.58 M $NaNO_3$--1.23 M NaOH the current efficiency was increased from 25 to 61% at a current density of 2.5 amp/in.^{2} From a solution 4.0 M in total electrolyte and at a current density of 4.0 amp/in.^{2}, the nitrate reduction current efficiency was increased from about 35 to 55% as the stirring rate was increased from a slow to a fast rate.

No weight loss was observed from a type 347 stainless steel anode over a 24 hr period at a current density of 2 amp/in.^{2} in a solution 1.8 M $NaNO_3$--0.6 M NaOH, containing 1000 ppm of chloride or sulfate.

3.1 Effect of Temperature

The half-life for ruthenium plating was reduced from 3.3 to 1.3 hr when the temperature of the solution was increased from 46 to 80°C. An activation energy of 6.0 ± 1.0 kcal/mole was calculated from the Arrhenius equation. Current efficiencies, based on nitrate reduction, remained constant at about 54% (Table 3.1). The influence of temperature was studied at 46, 60, and 80°C. Conditions maintained constant throughout were: applied potential, 5 volts; current density, 1 amp/in.^{2}, electrode areas, 3 in.^{2}; initial solution concentration, 0.25 M $NaNO_3$--0.12 M NaOH + Ru-106 tracer; and a slow stirring rate. Initial solution volumes were 500 ml.

Table 3.1. Effect of Temperature on the Rate of Ruthenium Plating and Nitrate Reduction Efficiency

Initial solution concentrations: 0.25 M $NaNO_3$, 0.12 M NaOH, Ru-106 tracer
Initial solution volumes: 500 ml
Anode and cathode areas: 3 in.^{2} each
Potential: 5 volts
Anode and cathode current densities: 1 amp/in.^{2}
Constant slow stirring rate
Current: 3 amp

Temperature, °C	Ruthenium Plating		Nitrate Reduction Current Efficiency, %
	k, hr^{-1}	$T_{1/2}$, hr	
46	0.208	3.3	54
60	0.302	2.3	53
80	0.523	1.3	54

3.2 Effect of Cell Potential

With an increase in the cell potential from 4 to 7 volts, the ruthenium plating half-life and the nitrate reduction current efficiency remained constant at 1.4 hr and about 54%, respectively (Table 3.2). Current density was maintained constant at 1 amp/in.^{2}, as were the electrode area, 3 in.^{2}, the solution temperature, 80°C, the initial solution concentration, 0.25 M $NaNO_3$--0.12 M NaOH + Ru-106 tracer, and the slow stirring rate. Initial solution volumes were 500 ml.

Table 3.2. Effect of Applied Potential on the Rate of Ruthenium Plating and Nitrate Reduction Efficiency

Conditions same as in Table 3.1 except constant temperature of of 80°C and variable potential

Potential, volts	Ruthenium Plating		Nitrate Reduction Current Efficiency, %
	k, hr^{-1}	$T_{1/2}$, hr	
4	0.479	1.4	54
5	0.503	1.4	54
7	0.473	1.4	53

3.3 Effect of Electrode Area and Current Density

The rate of ruthenium plating was increased by an increase in either the cathode area or cathode current density but was unaffected by changes in anode area or anode current density. Nitrate reduction current efficiencies were decreased by an increase in the cathode current density but were not changed by variations in cathode area, anode area or anode current density.

With electrode areas of 3 in.^{2}, increasing the current density from 1.0 to 3.0 amp/in.^{2} decreased the half-life for ruthenium plating from 1.5 to 1.1 hr, and the current efficiency for nitrate reduction from 53 to 22% (Table 3.3). With a cathode area of 3 in.^{2} and a cathode current density of 1 amp/in.^{2}, reducing the anode area from 3.0 to 1.0 in.^{2} and increasing the anode current density from 1.0 to 3.0 amp/in.^{2} produced no change in ruthenium deposition rate or nitrate reduction current efficiency. At an anode area of 3 in.^{2} and current density of 1 amp/in.^{2}, reducing cathode area to 1 in.^{2} and increasing its current density to 3 amp/in.^{2} produced no change in ruthenium deposition half-life, but decreased nitrate reduction current efficiencies from 53 to 21% (Fig. 3.1). The increasing effect on ruthenium plating half-life of increasing current density was cancelled by the effect of decreasing area.

In the above experiments the initial concentrations of the 500 ml solution were 0.25 M $NaNO_3$--0.12 M NaOH + Ru-106 tracer. Applied potential of 7 volts, a solution temperature of 80°C, and a constant slow stirring rate were maintained throughout.

Table 3.3. Effect of Varying Electrode Area and Current Density on the Rate of Ruthenium Plating and Nitrate Reduction

Initial solution concentration: 0.25 M $NaNO_3$, 0.12 M NaOH, Ru-106 tracer
Initial solution volumes: 500 ml
Temperature: 80°C
Potential: 7 volts
Constant slow stirring rate

Electrode Area, in.^{2}		Current Density, amp/in.^{2}			Ruthenium Plating		Nitrate Reduction
Anode	Cathode	Anode	Cathode	Current, amp	k, hr^{-1}	$T_{1/2}$, hr	Current Efficiency, %
3	3	1.0	1.0	3.0	0.473	1.5	53
3	3	1.67	1.67	5.0	0.535	1.3	41
3	3	2.5	2.5	7.5	0.595	1.2	25
3	3	3.0	3.0	9.0	0.640	1.1	22
3	1	1.0	3.0	3.0	0.425	1.6	21
1	3	3.0	1.0	3.0	0.460	1.5	55

3.4 Effect of Varying Solution Concentration

The rate of ruthenium plating decreased slightly but nitrate reduction current efficiencies increased as the total electrolyte concentration was increased. In the small 500-ml cell at an applied potential of 5 volts, a current density of 1 amp/in.^{2} and a solution temperature of 80°C, the ruthenium plating half-life was increased from 1.3 to 1.7 hr when the solution concentration was increased from 0.25 M $NaNO_3$--0.12 M NaOH to 0.70 M $NaNO_3$--0.20 M NaOH. Similarly, at 7 volts, 2.5 amp/in.^{2}, and 80°C the half-life increased from 1.2 to 1.4 hr as the solution concentration was increased from 0.25 M $NaNO_3$--0.12 M NaOH to 0.91 M $NaNO_3$--0.28 M NaOH (Table 3.4). All solutions contained Ru-106 tracer. At a current density of 2.5 amp/in.^{2} the nitrate reduction current efficiency increased from 25 to 61% as the solution concentration was increased from 0.37 to 4.83 M total electrolyte. Likewise, at 4.0 amp/in.^{2}, the efficiency increased from 23 to 44% with a concentration increase from 2.41 to 4.83 M total electrolyte (Table 3.4 and Fig. 3.2). Current efficiencies at 4 amp/in.^{2} were approximately 20% below those at 2.5 amp/in.^{2}.

In the KAPL design cell, with an increase in the solution total electrolyte concentration from 0.38 to 3.16 M, current efficiencies increased from 43 to 105% at a cathode current density of 2 amp/in.^{2} (Table 3.5 and Fig. 3.2). These current efficiencies appear to be out of line with those in the small cell; however, this is due to the effect of the greater rate of stirring, which is discussed in the next section. The power required to reduce 1 lb of nitrate to ammonia decreased from 19.0 to 4.7 kwh with the increase in the solution concentration.

3.5 Effect of Stirring Rate

The effect of solution stirring rate on current efficiencies was studied at a cathode current density of 4 amp/in.^{2}. The results are qualitative in that accurate stirring rates were unknown; however, it is sufficient to note that an increased rate of stirring resulted in an increase in the current efficiency (Table 3.6 and Fig. 3.3). The effect appeared to become more pronounced as the concentration of the solution was increased. In the 2 M total electrolyte solution the current efficiency increased from 20 to 30% while in the 4 M total electrolyte solution the increase was from about 35 to 55%.

Stirring effects on the rate of ruthenium plating was not studied.

3.6 Decontamination from Ruthenium

In order to determine what degree decontamination of ruthenium could be obtained, a 500 ml volume solution, 0.55 M total electrolyte containing Ru-106 tracer, was electrolyzed at a potential of 3 volts, a current density of 0.13 amp/in.^{2}, and a temperature of 60°C. The half-life for the ruthenium deposition was 3.4 hr (Table 3.7 and Fig. 3.4). At 25 hr, a sharp break occurred in the plot of log gross β vs time, which represented a decontamination factor of >250. Continued electrolysis for an additional 46 hr resulted in an increase in the decontamination factor to >430. Similarly,

Table 3.4. Effect of Varying Solution Total Electrolyte Concentration on the Rate of Ruthenium Plating and Nitrate Reduction Current Efficiencies

Initial solution volumes: 500 ml
Anode and cathode areas: 3 in.^{2} each
Constant slow stirring rate

Electrolyte Concentrations, M			Potential, volts	Temp. °C	Current, amp	Anode and Cathode Current Density, amp/in.^{2}	Ruthenium Plating		Nitrate Reduction Current Efficiency, %
$NaNO_3$	NaOH	Total					k, hr^{-1}	$T_{1/2}$, hr	
0.25	0.12	0.37	5.0	80	3.0	1.0	0.503	1.4	53
0.25	0.12	0.37	7.0	80	7.5	2.5	0.594	1.2	25
0.70	0.20	0.90	5.0	80	3.0	1.0	0.410	1.7	62
0.91	0.28	1.19	7.0	80	7.5	2.5	0.514	1.4	35
1.37	0.91	2.38	5.6	80	7.5	2.5	---	---	44
1.78	0.62	2.40	5.8	80	7.5	2.5	---	---	43
1.61	0.70	2.41	7.0	75	12.0	4.0	---	---	23
2.73	1.08	3.81	5.2	72	7.5	2.5	---	---	53
2.75	1.08	3.83	5.9	92	12.0	4.0	---	---	36
3.58	1.23	4.81	4.5	65	7.5	2.5	---	---	61
3.43	1.40	4.83	6.2	89	12.0	4.0	---	---	44

Table 3.5. Electrolytic Reduction in Alkaline Solution by the KAPL Design Cell

Power requirements based on amount of nitrate reduced to ammonia, and do not include that reduced to nitrite.

Current: 284 amp
Cathode area: 142 in.^{2}
Cathode current density: 2 amp/in.^{2}
Anode area: 95 in.^{2}
Initial solution volume: 1800 ml
Circulation rate: 3 gpm

Potential, volts	Initial Solution Concentrations, M			Nitrate Reduction Current Efficiency, %	Power, kwh/lb NO_3^-
	$NaNO_3$	NaOH	Total		
4.4	0.24	0.10	0.38	43	19.0
3.4	0.54	0.24	0.78	53	14.4
3.0	1.13	0.44	1.57	71	9.0
2.9	2.38	0.78	3.16	105	4.7

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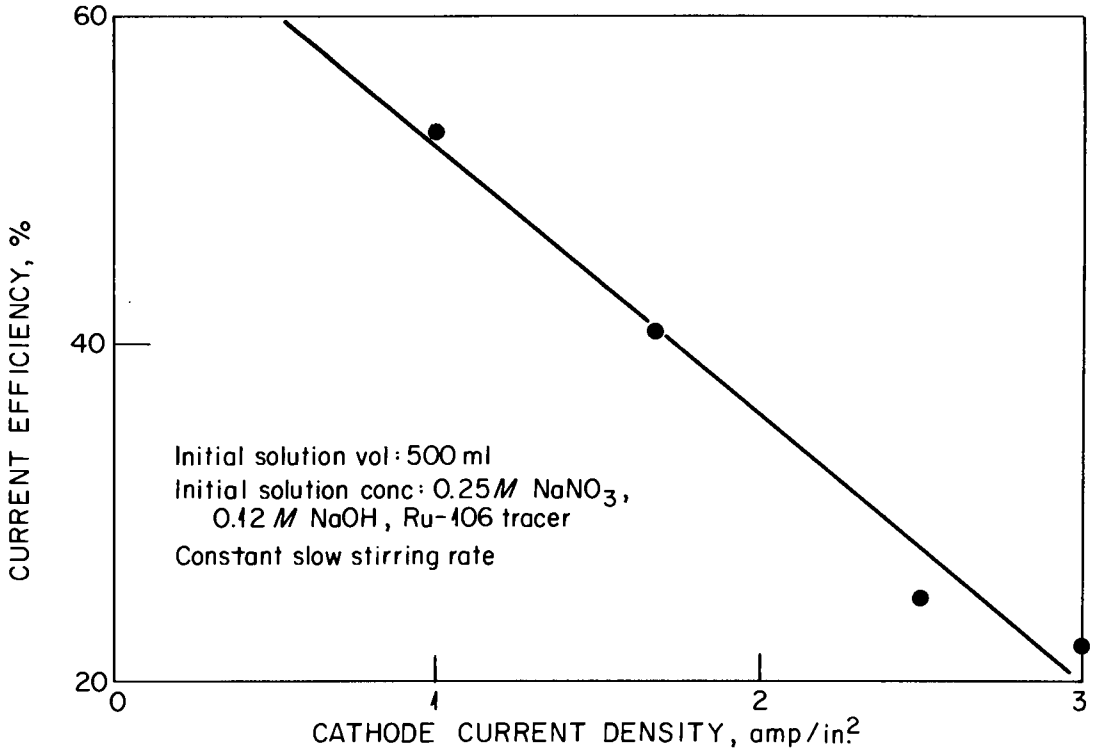

Fig. 3.1. Effect of current density on nitrate reduction current efficiency.

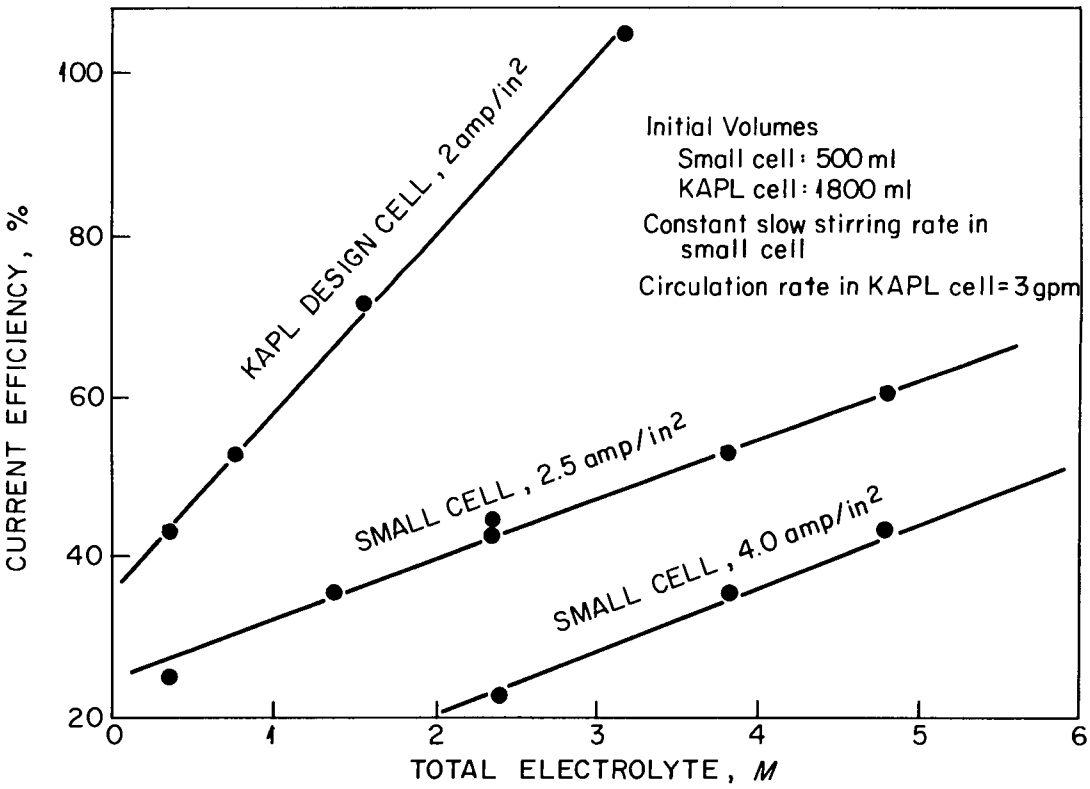

Fig. 3.2. Effect of varying solution total salt concentration and current density on nitrate reduction current efficiency.

Table 3.6. Effect of Stirring on Nitrate Reduction Current Efficiency

Initial solution volumes: 500 ml
Anode and cathode areas: 3 in.^{2} each
Current: 12 amp
Anode and cathode current densities: 4 amp/in.^{2}

Stirring Rate	Initial Solution Concentration, M			Potential, volts	Temp. °C	Nitrate Reduction Current Efficiency, %
	$NaNO_3$	NaOH	Total			
Slow	1.71	0.70	2.41	7.0	75	23
Slow	2.75	1.08	3.83	5.9	92	36
Slow	3.43	1.40	4.83	6.2	89	44
Fast	0.85	0.45	1.30	5.2	81	22
Fast	1.87	0.49	2.36	5.5	81	36
Fast	2.74	0.94	3.68	6.4	81	52

Table 3.7. Demonstration of Maximum Decontamination from Ruthenium

Initial solution volumes: 500 ml
Anode and cathode areas: 3 in.^{2} each
Constant slow stirring rate

Potential volts	Current amp	Anode and Cathode Current Density, amp/in.^{2}	Temp. °C	Initial Solution Concentration, M			Ruthenium Plating			Nitrate Reduction Current Efficiency, %
				$NaNO_3$	NaOH	Total	k, hr^{-1}	$T_{1/2}$, hr	D.F.	
3.0	0.4	0.13	60	0.44	0.11	0.55	0.203	3.4	>250 (25hr)	62
6.2	12.0	4.0	89	3.43	1.40	4.83	0.357	2.0	>210 (15hr)	44

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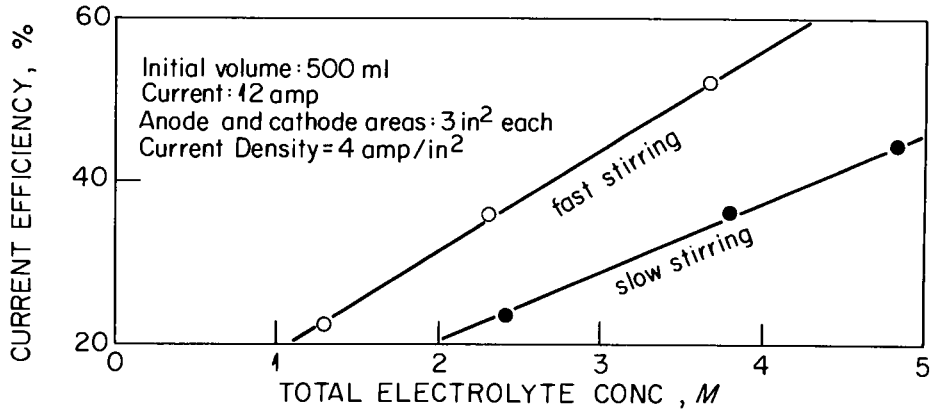

Fig. 3.3. Effect of solution slurry rate on nitrate reduction current efficiency.

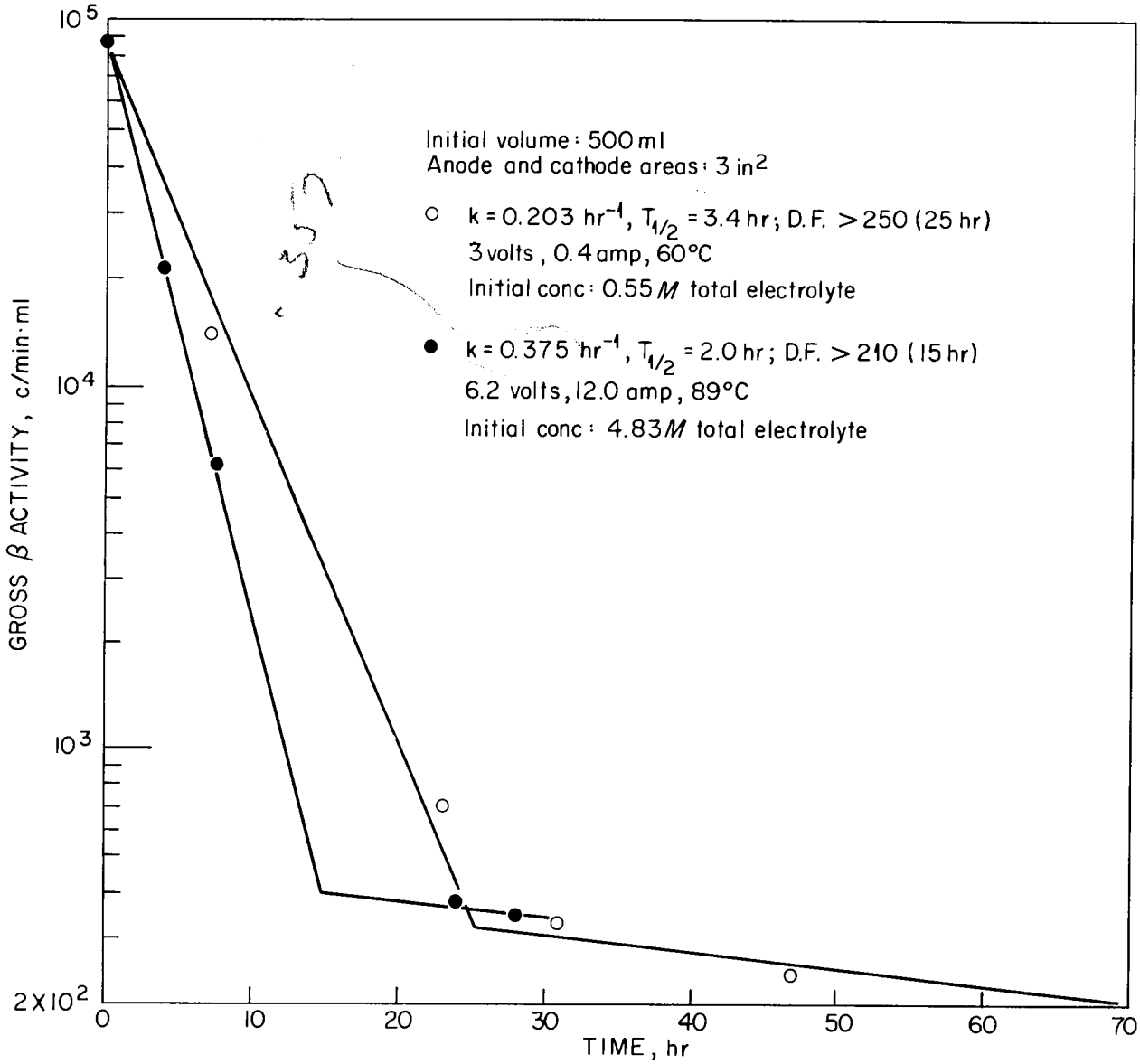

Fig. 3.4. Demonstration of maximum decontamination factors by ruthenium plating.

from a solution 4.83 M in total electrolyte, at a potential of 6.2 volts, current density of 4 amp/in.^{2}, and a temperature of 89°C, the ruthenium half-life was 2.0 hr. As in the above case a break occurred in the plot, at about the same level of activity but after about 15 hr. This represented a decontamination factor of >210. Whether or not the remaining activity was due to ruthenium or to a contaminant of the Ru-106 tracer was undertermined. Further investigation is needed.

3.7 Anode Corrosion During Electrolysis

An anodic dissolution test of type 347 stainless steel in 1.8 M $NaNO_3$--0.6 M NaOH indicated no weight loss in 24 hr at a current density of 2 amp/in.^{2} when either sulfate or chloride was added in a concentration of 1000 ppm.

4.0 SODIUM AND NITRATE MIGRATION STUDIES

Electrolysis of synthetic alkaline nitrate was investigated in an acid base membrane cell since the sodium hydroxide and nitric acid regenerated can be reused. An Ionics design and a simpler design membrane cell were compared for the following reason: Although the Ionics design cell is highly engineered and expected to be more efficient than the simple design cell, it is also more complicated in construction (see Appendix). Remote operation and maintenance are thus expected to be more difficult. Therefore, if performance characteristics of the two cells are not too different, the simply constructed cell would be preferred. However, it was found that >5 kwh of power was required to transfer 1 lb of nitrate from ORNL type waste in either case. This is compared to 4.7 kwh of power required to reduce nitrate to ammonia in the nitrate reduction cell at a solution total electrolyte concentration of 3.0 M. This, along with the lesser adaptibility of the membrane cells for remote control, has led to the recommendation that the nitrate reduction cell be used in the process.

Conditions of cell operation were chosen arbitrarily for simulated ORNL waste. Ruthenium was not added to the waste since the process assumes prior removal in a ruthenium plating cell. Three stages of a simple membrane cell were equivalent in performance to two stages of an Ionics design cell. The sodium was reduced from 0.6 to 0.03 M while nitrate was reduced from 0.3 to 0.01 M in each system. Current efficiencies were about the same in the two cell types and were independent of current density but dependent on concentrations of electrolyte in the feed, anode, and cathode compartments. The power consumption in the Ionics cell was about 60-70% of that required in the simpler cell for the same performance. With a solution 0.6 M Na^+--0.03 M NO_3^- at a current density of 1 amp/in.^{2} the power consumption in the Ionics cell was 5.1 kwh/lb of nitrate transferred in comparison to 8.4 kwh/lb in the open cell. Current efficiencies were about 60% at anolyte and catholyte concentrations of about 1.1 M HNO_3 and 1.5 M NaOH, respectively.

4.1 Investigation of the Simple Design Acid-Base Membrane Cell

In the simple design acid-base membrane cell, three stages were required to reduce a solution 0.57 M Na^+--0.30 M NO_3^- to 0.06 M Na^+--0.01 M NO_3^- with a

corresponding volume reduction from 9000 to 5500 ml because of evaporation and water decomposition. Current efficiencies, based on sodium transfer, and power required to transfer 1 lb of nitrate increased from 62 to 68% and from 8.4 to 24.4 kwh, respectively, depending on the stage under investigation.

Under an applied potential of 15.3 volts and a current density of 1 amp/in.^{2}, 9000 ml of feed was reduced in volume to 8200 ml. As a result of migration, sodium was reduced from 0.57 to 0.38 M and nitrate from 0.30 to 0.19 M. Based on sodium transfer, the cell operated at a current efficiency of 62%. The power required to transfer 1 lb of nitrate and 0.65 lb of sodium from the waste was 8.4 kwh. When the effluent was passed back through the cell under a potential of 20.8 volts and a current density of 0.5 amp/in.^{2}, the volume was reduced to 6500 ml. Sodium and nitrate concentrations were reduced to 0.17 and 0.06 M, respectively. The current efficiency was 63% and the power requirement was 10.9 kwh for the transfer of 1 lb of nitrate and 0.64 lb of sodium. In a third pass through the cell the volume was reduced to 5500 ml and the concentrations to 0.06 M Na^+ and 0.01 M NO_3^-. The current efficiency was 68% with a power consumption of 24.4 kwh per lb of nitrate and 0.88 lb of sodium transferred, (Table 4.1).

Flow rates of feed to the feed compartment and water to the anode and cathode compartments was 31, 3.6, and 4.1 ml/min, respectively. Electrode surface areas were 20 in.^{2}.

4.2 Investigation of the Ionics Design Acid-Base Membrane Cell

In the Ionics design acid-base membrane cell the current efficiency was dependent on the solution concentration but essentially independent of current density. On electrolysis of a 0.6 M Na^+--0.3 M NO_3^- solution at current densities of 1.0 to 1.5 amp/in.^{2} current efficiencies were 56 to 59%, based on sodium transfer. The current efficiency was 61% on electrolysis of a 0.35 M Na^+--0.16 M NO_3^- solution at 1 amp/in.^{2} At current densities of 0.5-0.75 amp/in.^{2} and from solutions about 0.25 M Na^+--0.1 M NO_3^- the current efficiencies were 65 to 68% (Table 4.2).

Power requirements varied with the applied potential necessary to obtain a particular current density in a solution of a given concentration. From a solution 0.6 M Na^+--0.3 M NO_3^-, the power consumed in the migration of 1 lb of nitrate increased from 5.1 to 7.6 kwh as the current density was increased from 1.0 to 1.5 amp/in.^{2} and the potential from 9 to 14 volts. The effluent concentration decreased from 0.35 M Na^+--0.16 M NO_3^- to about 0.25 M Na^+--0.10 M NO_3^-. At 1 amp/in.^{2} the power consumption increased from 5.1 to 11.3 kwh/lb of nitrate transferred as the feed concentration was decreased from 0.58 M Na^+--0.30 M NO_3^- to 0.35 M Na^+--0.16 M NO_3^- with the effluent concentration decreasing from 0.35 M Na^+--0.16 M NO_3^- to 0.11 M Na^+--0.03 M NO_3^-, respectively. A potential increase of from 9 to 17 volts was required to obtain the desired current density.

Flow rates of feed to the feed receptacle and water to the anode and cathode receptacles were 28, 4.2, and 5.2 ml/min, respectively. Corresponding circulation rates between the cell compartments and receptacles were 195, 210 and 200 ml/min. Electrode surface areas were 20 in.^{2} each. As with the simpler cell, the effluent of a first run was used as feed for the second.

Table 4.1. Migration of Sodium and Nitrate from Alkaline Solution in a Simple Design Acid-Base Membrane Cell

Input flow rates: water to the anode and cathode compartments and feed to the feed compartment, 3.6 ml/min, 4.1 ml/min, and 36 ml/min, respectively
Electrode areas: 20 in.^{2} each
Current efficiencies based on sodium transfer

Potential, volts	Current Density, amp/in.^{2}	Feed			Effluent			H^+ Conc. in Anolyte, M	OH^- Conc. in Catholyte, M	Current Efficiency, %	Power, kwh/lb NO_3^-	Na^+ Transferred, lb/lb NO_3^-
		Vol, ml	Na^+, M	NO_3^-, M	Vol, ml	Na^+, M	NO_3^-, M					
15.3	1.00	9000	0.57	0.30	8200	0.38	0.19	1.22	1.40	62	8.4	0.65
20.8	0.50	8200	0.38	0.19	6500	0.17	0.06	0.72	1.39	63	10.9	0.63
28.5	0.50	6500	0.17	0.06	5500	0.06	0.01	0.27	0.73	68	24.4	0.87

Table 4.2. Migration of Sodium and Nitrate Ions from Alkaline Solutions in an Ionics Design Acid-Base Membrane Cell

Input flow rates: water to the anode and cathode receptacles and feed to the feed compartment, 4.2 ml/min, 5/2 ml/min, and 28 ml/min, respectively
Electrode areas: 20 in.^{2} each
Current efficiencies based on sodium transfer

Potential, volts	Current Density, amp/in.^{2}	Feed			Effluent			H^+ Conc. in Anolyte, M	OH^- Conc. in Catholyte, M	Current Efficiency, %	Power, kwh/lb NO_3^-	Na^+ Transferred, lb/lb NO_3^-
		Vol, ml	Na^+, M	NO_3^-, M	Vol, ml	Na^+, M	NO_3^-, M					
13.1	1.50	8200	0.60	0.29	7300	0.26	0.10	1.09	1.61	57	7.6	0.63
11.3	0.50	7300	0.26	0.10	6600	0.10	0.04	0.41	0.72	67	8.1	0.91
9.0	1.00	9000	0.58	0.30	8500	0.35	0.16	1.00	1.22	56	5.1	0.60
17.0	1.00	8500	0.35	0.16	7500	0.11	0.03	0.60	1.14	61	11.3	0.77
14.1	1.50	9000	0.59	0.30	7900	0.25	0.09	1.08	1.65	59	7.5	0.59
18.0	0.75	7900	0.25	0.09	7100	0.06	0.01	0.47	1.00	65	16.5	1.15
11.3	1.25	9000	0.56	0.26	8400	0.28	0.11	1.19	1.52	58	6.9	0.66
18.2	0.75	8400	0.28	0.11	6350	0.08	0.02	0.54	0.96	68	12.6	0.90

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6.0 APPENDIX

6.1 Experimental Cell Designs

Small Cell. The small cell was nothing more than a 2.25 in.-dia pyrex cylinder 8 in. high, capable of holding 500 ml of solution. Heating tape and cooling coils were wrapped around the cylinder alternately for temperature control. Stirring was maintained relatively constant by a magnetic stirrer. Potential and current were adjusted by varying the distance between the two electrodes. The electrodes were prepared as follows: Two stainless steel strips, 1 by 8 in. were cleaned in nitric acid and acetone. To one of the strips a sheet of bright platinum was silver-soldered, and both were then coated with polyethylene. Three square inches of polyethylene was stripped from the platinum surface, completing the anode. A 3 in.^{2} strip of polyethylene was then removed from the other electrode in the corresponding location. Plating the exposed stainless steel surface with ruthenium completed the cathode.

KAPL Design Cell. The nitrate reduction cell was a miniture model of the one designed by KAPL,[8] built to contain 1800 ml of solution (Fig. 6.1). Several type 347 stainless steel anodes and Amsco iron cathodes were spaced alternately, 0.25 in. apart, with a total cathode area of 142 in.^{2} The solution was stirred by rapid circulation with an external centrifugal pump.

Simple Design Acid-Base Membrane Cell. The simple design acid-base membrane cell was constructed of polyvinyl chloride and consisted of three 1-in.-wide compartments, each with a capacity of 380 ml (Fig. 6.2). The anode and cathode compartments were separated from the feed compartment by an anionic and cationic membrane, respectively. Platinum and stainless steel, each with a surface area of 20 in.^{2}, were used as the anode and cathode, respectively. During electrolysis, water was pumped into the anode and cathode compartments and feed to the feed compartments. Nitrate migrated to the anode compartment, where it overflowed as nitric acid, and sodium migrated to the cathode compartment, where it overflowed as sodium hydroxide. The depleted feed overflowed from the feed compartment as waste.

Ionics Design Acid-Base Membrane Cell. Though the Ionics cell operated under the same principle as the simpler design cell, it was highly engineered and more complex, (Fig. 6.3). The compartments were 0.25 in. wide and consist of several spacers through which the solutions are pumped in a tortuous path. Each compartment had an external receptacle from which the solutions were rapidly circulated through the cell by a centrifugal pump. Water and feed were added to the appropriate receptacle and the overflow was collected as product or waste. The anode, feed and cathode compartments and their

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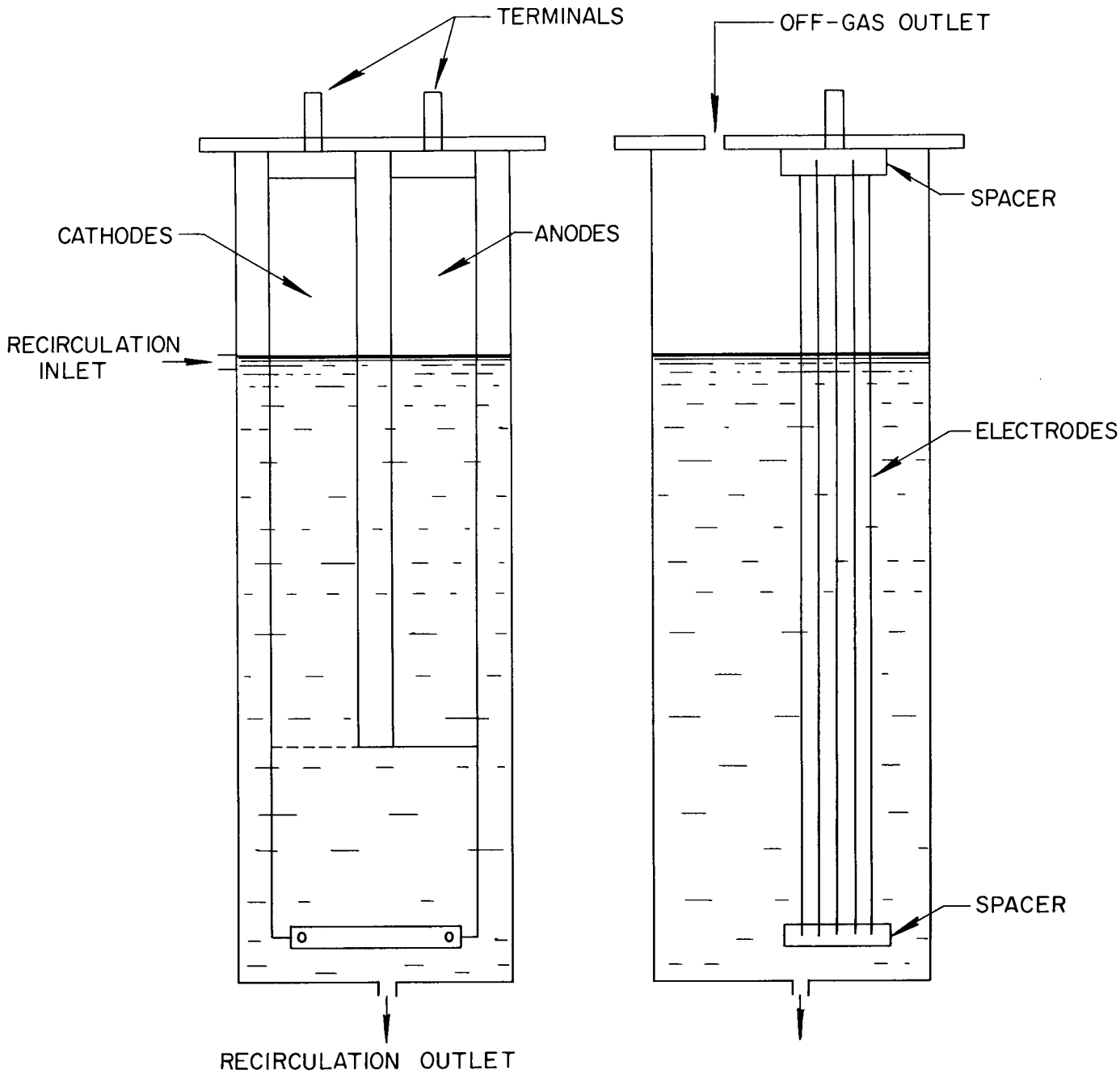

Fig. 6.1. Nitrate reduction cell, KAPL design. Cell diameter, 3 in.

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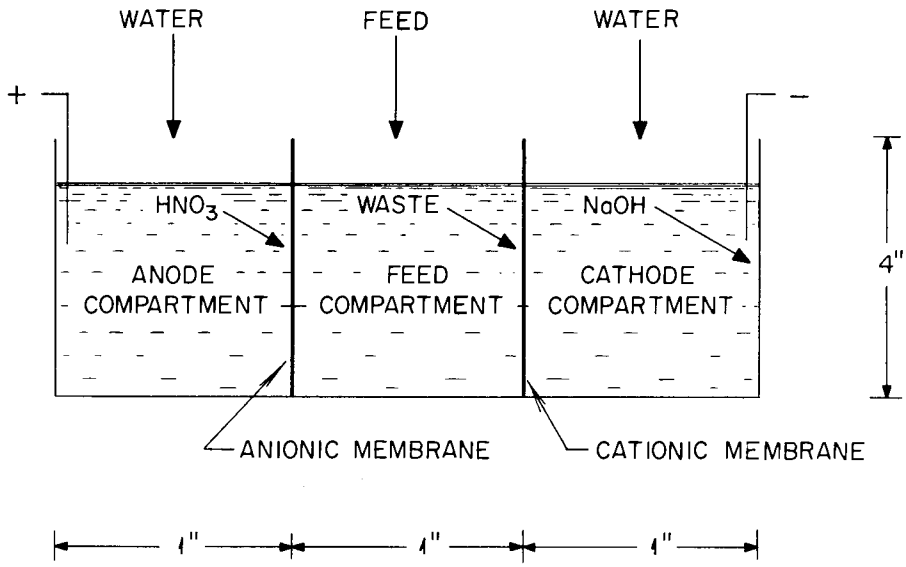

Fig. 6.2. Simple-design acid-base membrane cell. Cell length, 5 in.

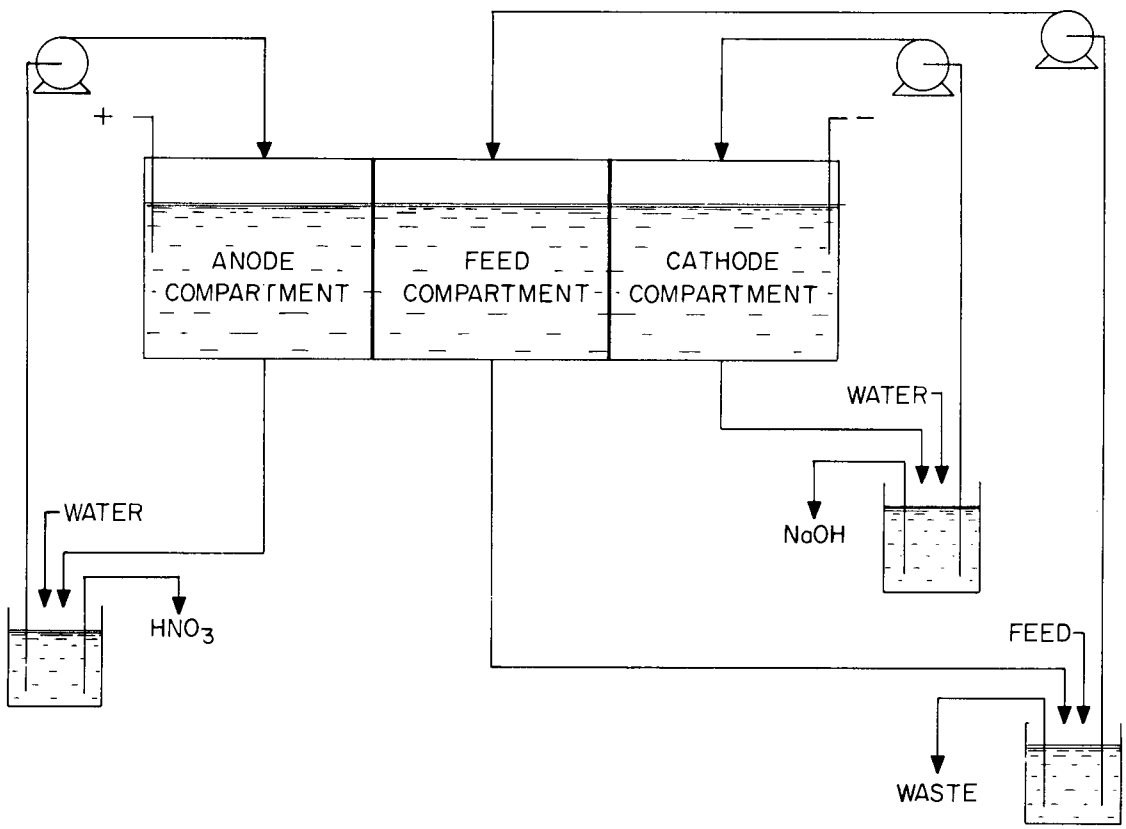

Fig. 6.3. Ionics design acid-base membrane cell. Compartment width, 1/4 in.

corresponding receptacle contained a total of 660, 660 and 650 ml of solution, respectively.

6.2 Experimental Run Data

Detailed experimental run data obtained in the investigation of the four electrolytic cells are included in the following tables. Ruthenium plating studies were performed in the small cell (Table 6.1), while nitrate reductions studies were made in the small cell and the KAPL design cell, (Tables 6.1 and 6.2). Migration of sodium and nitrate was studied in the simple design and an Ionics design acid-base membrane cell (Tables 6.3 and 6.4).

Table 6.1. Small Cell Run Data

Except where otherwise indicated, anode and cathode surface areas were each 3 in.^{2} and stirring was at a slow constant rate.

Date	Sample No.	Time, hr	Potential, volts	Current, amp	Temp, °C	Solution Volume, ml	Gross β c/ml·min	NO_3^-, mg/ml	OH^-, M	NO_2^-, mg/ml
1/8/58	Feed	---	---	---	---	500	37,600	15.9	0.12	---
	1	0.5	5.0	3.0	45.5	500	27,800	13.3	---	---
	2	1.0	5.0	3.0	46.0	490	29,600	13.3	---	---
	3	1.5	5.0	3.0	45.5	480	28,900	13.4	---	---
	4	2.0	5.0	3.0	46.0	470	28,600	12.0	---	---
	5	2.5	5.0	3.0	46.0	460	26,700	12.7	---	---
	6	3.0	5.0	3.0	46.0	450	23,400	11.5	---	---
	7	3.5	5.0	3.0	46.0	440	21,800	10.6	---	---
	8	4.0	5.0	3.0	46.0	430	20,600	11.1	---	---
	9	4.5	5.0	3.0	45.5	420	17,400	10.3	---	---
	10	5.0	5.0	3.0	46.0	410	16,300	9.75	---	---
	11	5.5	5.0	3.0	45.0	400	14,100	9.50	---	---
	12	6.0	5.0	3.0	48.0	390	12,600	9.49	---	---
	13	6.5	5.0	3.0	46.0	380	11,500	8.94	---	---
	14	7.0	5.0	3.0	46.0	370	10,500	8.39	0.16	---
1/9/58	Feed	---	---	---	---	500	22,000	14.9	0.12	---
	1	0.5	5.0	3.0	79.0	500	18,700	14.3	---	---
	2	1.0	5.0	3.0	79.5	490	15,000	14.2	---	---
	3	1.5	5.0	3.0	79.0	480	12,300	14.1	---	---
	4	2.0	5.0	3.0	79.5	470	10,200	12.7	---	---
	5	2.5	5.0	3.0	80.0	460	7,630	11.1	---	---
	6	3.0	5.0	3.0	80.0	450	5,350	11.7	---	---
	7	3.5	5.0	3.0	80.0	440	4,320	10.8	---	---
	8	4.0	5.0	3.0	80.0	430	3,290	10.4	---	---
	9	4.5	5.0	3.0	80.0	420	2,420	10.9	---	---
	10	5.0	5.0	3.0	80.0	410	2,010	9.46	---	---
	11	5.5	5.0	3.0	79.0	400	1,520	9.27	0.17	---
1/13/58	Feed	---	---	---	---	500	22,000	14.9	0.12	---
	1	0.5	5.0	3.0	57.0	500	17,000	14.0	---	---
	2	1.0	5.0	3.0	60.0	490	14,500	13.9	---	---
	3	1.5	5.0	3.0	60.0	480	12,000	12.7	---	---
	4	2.0	5.0	3.0	60.0	470	11,000	11.9	---	---

Table 6.1. (continued)

Date	Sample No.	Time, hr	Potential, volts	Current, amp	Temp, °C	Solution Volume, ml	Gross β, c/ml·min	NO_3^-, mg/ml	OH^-, M	NO_2^-, mg/ml
	5	2.5	5.0	3.0	60.0	460	9,380	11.4	---	---
	6	3.0	5.0	3.0	60.0	450	7,800	10.6	---	---
	7	3.5	5.0	3.0	60.0	440	6,900	10.3	---	---
	8	4.0	5.0	3.0	58.5	430	6,300	9.89	---	---
	9	4.5	5.0	3.0	61.5	420	5,300	9.56	---	---
	10	5.0	5.0	3.0	60.5	410	4,230	9.13	---	---
	11	5.5	5.0	3.0	60.5	400	3,900	8.47	---	---
	12	6.0	5.0	3.0	60.0	390	3,000	8.09	---	---
	13	6.5	5.0	3.0	60.0	380	2,900	8.24	0.16	---
1/15/58	Feed	Feed	---	---	---	500	13,700	15.2	0.12	---
	1	0.25	7.0	3.0	79.0	500	12,700	15.1	---	---
	2	0.5	7.0	3.0	80.0	490	11,100	14.7	---	---
	3	1.0	7.0	3.0	80.0	480	8,800	14.0	---	---
	4	1.5	7.0	3.0	80.0	470	7,000	13.4	---	---
	5	2.0	7.0	3.0	80.0	460	6,090	12.7	---	---
	6	2.5	7.0	3.0	80.0	450	4,330	12.4	---	---
	7	3.0	7.0	3.0	80.0	440	3,250	11.6	---	---
	8	3.5	7.0	3.0	80.0	430	2,610	10.9	---	---
	9	4.0	7.0	3.0	80.0	420	1,810	10.7	---	---
	10	4.5	7.0	3.0	80.0	410	1,580	9.69	---	---
	11	5.0	7.0	3.0	80.0	400	1,320	9.13	---	---
	12	5.5	7.0	3.0	80.0	390	1,080	8.56	0.18	---
1/16/58	Feed	---	---	---	---	500	13,700	15.2	0.11	---
	1	0.5	4.0	3.0	80.0	500	12,800	14.5	---	---
	2	1.0	4.0	3.0	80.0	490	12,600	14.5	---	---
	3	1.5	4.0	3.0	80.0	480	8,560	13.2	---	---
	4	2.0	4.0	3.0	80.0	470	6,070	12.5	---	---
	5	2.5	4.0	3.0	80.0	460	5,310	11.4	---	---
	6	3.0	4.0	3.0	80.0	450	4,300	11.4	---	---
	7	3.75	4.0	3.0	80.0	440	2,980	10.5	---	---
	8	4.5	4.0	3.0	80.0	430	2,190	9.65	---	---
	9	5.0	4.0	3.0	80.0	420	1,600	8.75	---	---
	10	5.5	4.0	3.0	80.0	410	1,240	9.46	---	---

Table 6.1. (continued)

Date	Sample No.	Time, hr	Potential, volts	Current, amp	Temp, °C	Solution Volume, ml	Gross β, c/ml·min	NO_3^-, mg/ml	OH^-, M	NO_2^-, mg/ml
	11	6.0	4.0	3.0	80.0	400	1,000	8.75	---	---
	12	6.5	4.0	3.0	80.0	390	706	8.56	0.15	---
1/20/58	Feed	---	---	---	---	500	13,000	15.2	0.11	---
	1	0.25	7.0	7.5	80.0	500	8,550	14.4	---	---
	2	0.5	7.0	7.5	80.0	490	7,950	14.2	---	---
	3	0.75	7.0	7.5	80.0	480	8,240	13.7	---	---
	4	1.0	7.0	7.5	80.0	470	6,010	12.9	---	---
	5	1.5	7.0	7.5	80.0	460	5,300	12.8	---	---
	6	2.0	7.0	7.5	80.0	450	3,470	11.8	---	---
	7	2.5	7.0	7.5	80.0	440	3,670	11.5	---	---
	8	3.0	7.0	7.5	80.0	430	2,460	11.0	---	---
	9	3.75	7.0	7.5	80.0	420	1,240	10.2	---	---
	10	4.5	7.0	7.5	80.0	410	668	9.20	---	---
	11	5.0	7.0	7.5	80.0	400	620	8.54	---	---
	12	5.5	7.0	7.5	80.0	390	504	8.30	---	---
	13	6.0	7.0	7.5	80.0	380	420	7.69	0.19	---
1/21/58	Feed	---	---	---	---	500	33,000	15.2	0.11	---
	1	0.5	7.0	5.0	80.0	500	27,000	13.5	---	---
	2	1.0	7.0	5.0	80.0	490	19,200	12.2	---	---
	3	1.5	7.0	5.0	80.0	480	18,100	11.8	---	---
	4	2.0	7.0	5.0	80.0	470	13,000	11.9	---	---
	5	2.5	7.0	5.0	80.0	460	9,120	11.0	---	---
	6	3.0	7.0	5.0	80.0	450	6,240	10.6	---	---
	7	3.5	7.0	5.0	80.0	440	5,300	10.6	---	---
	8	4.0	7.0	5.0	80.0	430	4,830	9.46	---	---
	9	4.5	7.0	5.0	80.0	420	3,170	9.13	0.15	---
1/24/58	Cathode area reduced to 1 in.^{2} in this run.									
	Feed	---	---	---	---	500	40,000	14.9	0.11	---
	1	0.5	7.0	3.0	80.0	500	25,400	14.6	---	---
	2	1.0	7.0	3.0	80.0	490	25,600	14.4	---	---
	3	1.5	7.0	3.0	80.0	480	19,000	13.8	---	---
	4	2.0	7.0	3.0	80.0	470	13,900	13.2	---	---

Table 6.1. (continued)

Date	Sample No.	Time, hr	Potential, volts	Current, amp	Temp, °C	Solution Volume, ml	Gross β, c/ml·min	NO_3^-, mg/ml	OH^-, M	NO_2^-, mg/ml
	5	2.5	7.0	3.0	80.0	460	13,300	11.6	---	---
	6	3.0	7.0	3.0	80.0	450	11,300	11.6	---	---
	7	3.75	7.0	3.0	80.0	440	8,100	12.9	---	---
	8	4.5	7.0	3.0	80.0	430	5,970	12.5	---	---
	9	5.5	7.0	3.0	80.0	420	4,470	11.8	---	---
	10	6.5	7.0	3.0	80.0	410	2,500	11.9	---	---
	11	7.0	7.0	3.0	80.0	400	2,050	11.5	0.12	---
1/25/58	Anode area reduced to 1 in.^{2} in this run.									
	Feed	---	---	---	---	500	27,000	14.9	0.11	---
	1	0.5	7.0	3.0	80.0	500	22,500	15.0	---	---
	2	1.0	7.0	3.0	80.0	490	20,600	14.4	---	---
	3	1.5	7.0	3.0	80.0	480	14,700	13.2	---	---
	4	2.0	7.0	3.0	80.0	470	10,600	12.7	---	---
	5	2.5	7.0	3.0	80.0	460	7,170	12.1	---	---
	6	3.0	7.0	3.0	80.0	450	6,430	11.7	---	---
	7	3.5	7.0	3.0	80.0	440	5,680	11.4	---	---
	8	4.0	7.0	3.0	80.0	430	4,480	10.5	---	---
	9	4.5	7.0	3.0	80.0	420	3,730	9.93	---	---
	10	5.0	7.0	3.0	80.0	410	3,080	9.27	---	---
	11	5.5	7.0	3.0	80.0	400	2,550	8.61	---	---
	12	6.0	7.0	3.0	80.0	390	1,950	8.04	0.16	---
2/6/58	Feed	---	---	---	---	500	68,100	56.5	0.28	---
	1	0.5	7.0	7.5	74.5	500	59,200	56.0	---	---
	2	1.0	7.0	7.5	80.0	490	44,300	57.3	---	---
	3	1.75	7.0	7.5	80.0	480	31,500	54.4	---	---
	4	2.5	7.0	7.5	80.0	470	21,500	46.8	---	---
	5	3.25	7.0	7.5	80.0	460	15,200	46.5	---	---
	6	4.0	7.0	7.5	80.0	450	8,490	44.9	---	---
	7	4.75	7.0	7.5	80.0	440	5,770	44.2	---	---
	8	5.5	7.0	7.5	80.0	430	3,720	41.1	---	---
	9	6.25	7.0	7.5	80.0	420	3,020	39.9	---	---
	10	7.0	7.0	7.5	80.0	410	2,020	40.6	0.36	---

Table 6.1. (continued)

Date	Sample No.	Time, hr	Potential, volts	Current, amp	Temp, °C	Solution Volume, ml	Gross β, c/ml·min	NO_3^-, mg/ml	OH^-, M	NO_2^-, mg/ml
2/11/58	Feed	---	---	---	---	500	38,500	43.2	0.20	---
	1	0.75	5.0	3.0	74.0	500	33,100	40.5	---	---
	2	1.5	5.0	3.0	80.0	490	24,400	39.8	---	---
	3	2.25	5.0	3.0	79.0	480	17,200	38.1	---	---
	4	3.0	5.0	3.0	81.0	470	12,700	39.6	---	---
	5	3.75	5.0	3.0	80.0	460	9,680	36.6	---	---
	6	4.5	5.0	3.0	80.0	450	7,130	35.4	---	---
	7	5.5	5.0	3.0	80.0	440	4,760	35.9	---	---
	8	6.5	5.0	3.0	80.0	430	3,780	35.1	---	---
	9	7.5	5.0	3.0	80.0	420	2,330	33.0	0.29	---
2/14/58	Feed	---	---	---	---	500	---	112.	0.62	---
	1	2.0	5.6	7.5	80.0	500	---	108.	---	---
	2	4.0	5.6	7.5	80.0	495	---	106.	---	---
	3	7.0	5.6	7.5	80.0	490	---	98.9	0.72	5.75
2/18/58	Feed	---	---	---	---	485	---	98.9	0.72	5.75
	1	7.0	7.0	12.0	75.0	485	---	89.2	0.83	4.90
2/20/58	Feed	---	---	---	---	500	---	85.0	0.91	---
	1	6.0	5.8	7.5	80.0	500	---	74.5	0.97	4.80
2/24/58	Feed	---	---	---	---	500	86,100	213.	1.40	---
	1	4.0	5.9	12.0	88.0	500	21,200	196.	1.42	11.4
	2	8.0	6.1	12.0	89.0	495	6,030	152.	1.70	9.74
	3	24.0	6.1	12.0	89.0	490	395	144.	1.94	6.34
	4	28.0	6.1	12.0	89.0	485	355	105.	2.54	3.94
2/26/58	Feed	---	---	---	---	500	---	170.	1.08	0.131
	1	4.0	5.9	12.0	91.0	500	---	165.	1.20	8.02
	2	7.5	5.9	12.0	92.0	495	---	147.	1.20	8.69
	3	24.0	5.9	12.0	92.0	490	---	107.	1.32	5.29
2/27/58	Feed	---	---	---	---	500	---	170.	1.08	0.131
	1	7.0	5.2	7.5	72.0	500	---	153.	1.14	7.19
	2	23.0	5.2	7.5	72.0	495	---	107.	1.40	3.32

Table 6.1. (continued)

Date	Sample No.	Time, hr	Potential, volts	Current, amp	Temp, °C	Solution Volume, ml	Gross β, c/ml·min	NO_3^-, mg/ml	OH^-, M	NO_2^-, mg/ml
3/6/58	Feed	---	---	---	---	500	---	221.	1.23	---
	1	20.0	4.5	7.5	65.0	500	---	168.	1.85	7.20
3/11/58	Fast stirring rate									
	Feed	---	---	---	---	500	---	170.	0.94	---
	1	18.0	5.2	12.0	81.0	500	---	94.9	1.42	4.40
3/11/58	Fast stirring rate									
	Feed	---	---	---	---	500	---	116.	0.49	---
	1	18.0	5.5	12.0	81.0	500	---	73.6	0.80	4.64
3/13/58	Fast stirring rate									
	Feed	---	---	---	---	500	---	52.9	0.45	---
	1	18.0	6.4	12.0	87.0	500	---	23.8	1.85	2.24
4/7/58	Feed	---	---	---	---	500	87,200	26.5	0.11	---
	1	7.0	3.0	0.4	60.0	500	14,000	23.7	---	---
	2	23.0	3.0	0.4	60.0	495	710	20.9	---	---
	3	31.0	3.0	0.4	60.0	490	336	20.2	---	---
	4	47.0	3.0	0.4	60.0	485	244	17.7	---	---
	5	71.0	3.0	0.4	60.0	480	206	16.0	0.18	---

Table 6.2. Nitrate Reduction in the KAPL Design Cell

Cathode surface area: 142 in.^{2}
Applied current: 284 amp for a current density of 2 amp/in.^{2}
Solution volumes: 1800 ml
Final analyses performed after makeup of water losses due to evaporation and electrolytic decomposition; power consumption based on the nitrate reduced to ammonia and does not include the quantity reduced to nitrate

Run No.	Length of Run, min	Potential, volts	Concentration, M					Current Efficiency, %	Power Consumption, kwh/lb
			Initial		Final				
			NO_3^-	OH^-	NO_3^-	NO_2^-	OH^-		
1	25	4.4	0.28	0.10	0.08	0.09	0.34	43	19.0
2	35	3.4	0.54	0.24	0.08	0.31	0.64	53	14.4
3	60	3.0	1.13	0.44	0.19	0.56	1.33	71	9.0
4	120	2.9	2.38	0.78	0.32	0.64	2.40	105	4.7

Table 6.3. Run Data for the Simple Design Acid-Base Membrane Cell

Input flow rates: water to the anode compartment at 3.6 ml/min, feed to the feed compartment at 31 ml/min, and water to the cathode compartment at 4.1 ml/min

Platinum anode and stainless steel cathode: surface areas of 20 in.^{2} each

Date	Sample No.	Time, hr	Potential, volts	Current, amp	Compartment Temperatures, °C			Effluent Volumes, ml			Concentrations			
					Anode	Feed	Cathode	Anolyte	Waste	Catholyte	H^+ in Anolyte, M	Waste Na^+, mg/ml	Waste NO_3^-, mg/ml	OH^- in Catholyte, M
9/27/57	Feed, 9000 ml, was 13.2 mg/ml Na^+, 18.9 mg/ml NO_3^-, and 8.1 mg/ml $CO_3^=$; anode and cathode compartments charged with 380 ml of 0.99 M HNO_3 and 380 ml of 0.76 M NaOH, respectively													
	1	0.5	16.2	20	---	75	---	147	876	127	1.02	9.25	12.5	1.06
	2	1.0	15.7	20	---	79	---	118	861	139	1.10	8.62	11.3	1.20
	3	1.5	15.4	20	---	80	---	100	875	136	1.17	8.75	11.4	1.30
	4	2.0	15.3	20	---	80	---	99	881	137	1.22	8.62	11.4	1.34
	5	2.5	15.3	20	---	80	---	100	885	136	1.27	8.30	11.3	1.40
	6	3.0	15.3	20	---	80	---	98	876	137	1.26	8.62	11.4	1.46
	7	3.5	15.3	20	---	80	---	97	880	137	1.30	9.25	11.3	1.49
	8	4.0	15.3	20	---	80	---	99	879	137	1.30	8.70	11.1	1.49
	9	4.5	15.3	20	---	80	---	98	879	137	1.30	8.30	11.3	1.49
		Volume remaining in compartments at end of run						430	320	220	---	---	---	---
	10	Total volume and composite analysis						1386	8212	1442	1.22	8.62	11.7	1.40
10/4/57	Feed, 8200, was 9.00 mg/ml Na^+, 11.7 mg/ml of NO_3^-, and 6.9 mg/ml $CO_3^=$; anode and cathode compartments charged with 380 ml of 0.42 M HNO_3 and 380 ml of 0.50 M NaOH, respectively													
	1	0.5	22.7	10	91	95	82	124	718	169	0.60	3.95	3.67	0.76
	2	1.0	22.0	10	92	98	85	138	748	163	0.68	4.00	2.44	1.04
	3	1.5	21.5	10	92	98	84	130	738	161	0.72	3.95	2.51	1.22
	4	2.0	21.3	10	92	98	84	130	743	160	0.75	3.95	2.46	1.37
	5	2.5	21.1	10	92	97	85	130	746	166	0.76	4.15	3.71	1.44
	6	3.0	20.8	10	92	97	84	130	743	167	0.77	4.15	3.10	1.51
	7	3.5	20.8	10	92	97	84	130	751	166	0.77	4.15	4.00	1.56
	8	4.2	20.8	10	92	97	84	172	1002	204	0.77	4.15	3.83	1.58
		Volume remaining in compartments at end of run						296	330	436	---	---	---	---
	9	Total volume and composite analysis						1380	6519	1792	0.72	4.00	3.26	1.39
10/16/57	Feed, 6500 ml, was 4.15 mg/ml Na^+, 3.55 mg/ml NO_3^-, and 3.71 mg/ml $CO_3^=$; anode and cathode compartments charged with 380 ml of 0.15 M HNO_3 and 380 ml of 0.29 M NaOH, respectively													
	1	0.75	28.3	10	79	80	68	260	1238	202	0.25	1.28	0.87	0.53
	2	1.5	29.8	10	85	90	76	235	1216	191	0.28	1.10	0.74	0.68
	3	2.25	29.1	10	85	90	77	228	1221	192	0.30	1.10	0.87	0.78
	4	3.2	28.5	10	85	90	78	302	1469	249	0.30	1.20	0.80	0.84
		Volume remaining in compartments at end of run						294	330	410	---	---	---	---
	5	Total volume and composite analysis						1309	5474	1234	0.27	1.28	0.84	0.73

Table 6.4. Run Data for the Ionics Design Acid-Base Membrane Cell

Input flow rates: water to the anode compartment at 4.2 ml/min, feed to the feed compartment at 28 ml/min, and water to the cathode compartment at 5.2 ml/min

Circulation rates between the cell compartment and the compartment receptacles: 210 ml/min, 195 ml/min, and 200 ml/min, respectively

Platinum anode and stainless steel cathode: surface areas of 20 in.^{2}

Date	Sample No.	Time, hr	Potential, volts	Current, amp	Compartment Temperatures, °C			Effluent Volumes, ml			Concentrations			
					Anode	Feed	Cathode	Anolyte	Waste	Catholyte	H^+ in Anolyte, M	Waste Na^+, mg/ml	Waste NO_3^-, mg/ml	OH^- in Catholyte, M
10/22/57	Feed, 8200 ml, was 13.3 mg/ml Na^+, 17.8 mg/ml NO_3^- and 8.78 mg/ml $CO_3^=$; anode and cathode compartments charged with 660 ml of 1.00 M HNO_3 and 650 ml of 0.81 M NaOH, respectively													
	1	1.0	13.0	30	62	61	59	320	1585	403	1.12	5.60	4.61	1.28
	2	2.0	13.1	30	74	70	66	372	1492	352	1.17	5.42	4.50	1.58
	3	3.0	13.1	30	75	70	66	410	1427	348	1.12	5.60	4.61	1.77
	4	4.0	13.1	30	75	70	66	422	1425	342	1.05	5.82	4.61	1.84
	5	4.5	13.1	30	75	70	66	211	710	171	1.02	5.85	4.61	1.83
		Volume remaining in compartments at end of run						610	640	605	---	---	---	---
	6	Total volume and composite analysis						2345	7279	2221	1.09	6.00	5.50	1.61
10/24/57	Feed, 7200 ml, 5.95 mg/ml Na^+, 5.91 mg/ml NO_3^-, and 4.87 mg/ml $CO_3^=$; anode and cathode compartments charged with 660 ml of 0.54 M HNO_3, and 650 ml of 0.55 M NaOH, respectively													
	1	1.0	11.3	10	38	36	36	343	1629	334	0.47	3.18	2.25	0.65
	2	2.0	11.3	10	39	37	36	314	1590	310	0.42	3.05	2.13	0.71
	3	3.0	11.3	10	39	38	37	320	1584	307	0.40	3.05	2.20	0.76
	4	3.67	11.3	10	39	38	37	211	1041	204	0.38	3.05	2.15	0.78
		Volume remaining in compartments at end of run						660	640	650	---	---	---	---
	5	Total volume and composite analysis						1848	6634	1815	0.41	3.28	2.44	0.72
11/5/57	Feed, 9000 ml, was 13.42 mg/ml Na^+, 18.4 mg/ml NO_3^-, and 8.40 mg/ml $CO_3^=$; anode and cathode compartments charged with 660 ml of 1.02 M HNO_3 and 650 ml of 0.84 M NaOH, respectively													
	1	1	9.3	20	44	43	42	292	1634	356	0.97	8.12	10.2	1.03
	2	2	9.1	20	46	45	45	270	1617	327	1.00	7.95	9.18	1.20
	3	3	9.0	20	47	46	45	273	1620	326	1.00	7.88	9.22	1.28
	4	4	9.0	20	47	46	45	273	1616	326	1.00	7.95	9.08	1.32
	5	4.9	9.0	20	47	46	45	238	1385	285	1.00	7.95	9.08	1.34
		Volume in compartments at end of run						625	630	615	---	---	---	---
	6	Total volume and composite analysis						1971	8502	2235	1.00	8.20	9.70	1.22

Table 6.4. (continued)

Date	Sample No.	Time, hr	Potential, volts	Current, amp	Compartment Temperatures, °C: Anode	Feed	Cathode	Effluent Volumes, ml: Anolyte	Waste	Catholyte	Concentrations: H^+ in Anolyte, M	Waste: Na^+, mg/ml	NO_3^-, mg/ml	OH^- in Catholyte, M
11/8/57	Feed, 8400 ml, was 8.18 mg/ml Na^+, 9.46 mg/ml NO_3^-, and 5.70 mg/ml $CO_3^=$; anode and cathode compartments charged with 660 ml of 0.57 M HNO_3, and 650 ml of 0.62 M NaOH, respectively													
	1	1	17.5	20	62	59	56	392	1508	370	0.62	2.51	2.13	0.88
	2	2	17.0	20	66	64	60	402	1485	349	0.61	2.08	1.66	1.09
	3	3	17.0	20	65	64	60	402	1484	342	0.59	2.18	1.78	1.19
	4	4.67	17.0	20	66	64	60	651	2378	553	0.60	2.28	1.85	1.25
		Volume in compartments at end of run						615	620	595	---	---	---	---
	5	Total volume and composite analysis						2462	7475	2209	0.60	2.62	2.13	1.14
11/6/57	Feed, 9000 ml was 13.5 mg/ml Na^+, 18.3 mg/ml NO_3^-, and 8.68 mg/ml $CO_3^=$; anode and cathode compartments charged with 660 ml of 1.02 M HNO_3 and 650 ml of 0.84 M NaOH, respectively													
	1	1	12.0	30	58	58	56	367	1575	406	1.14	5.74	4.97	1.23
	2	2	14.3	30	65	63	62	315	1546	365	1.22	5.38	3.93	1.54
	3	3	14.2	30	75	69	67	421	1450	362	1.10	5.32	4.57	1.66
	4	4	14.1	30	75	71	67	437	1414	342	1.04	5.57	4.45	1.77
	5	5	14.1	30	75	71	67	407	1323	327	0.97	5.54	4.45	1.80
		Volume in compartments at end of run						613	605	595	---	---	---	---
	6	Total volume and composite analysis						2560	7913	2397	1.08	5.64	5.12	1.65
11/8/58	Feed, 7800 ml was 5.79 mg/ml Na^+, 5.85 mg/ml NO_3^-, and 4.80 mg/ml $CO_3^=$; anode and cathode compartments charged with 660 ml of 0.57 M HNO_3, and 650 ml of 0.62 M NaOH, respectively													
	1	1	18.0	15	57	54	52	302	1574	284	0.56	1.31	0.92	0.90
	2	2	18.3	15	58	59	55	339	1549	307	0.59	1.11	0.74	1.01
	3	3	18.0	15	61	62	57	336	1543	308	0.45	1.00	0.62	1.05
	4	4.2	18.0	15	61	62	57	410	1849	374	0.43	1.01	0.78	1.10
		Volume in compartments at end of run						618	620	597	---	---	---	---
	5	Total volume and composite analysis						2005	7135	1870	0.47	1.43	0.98	1.00
11/16/57	Feed, 9000 ml, was 12.96 mg/ml Na^+, 16.3 mg/ml NO_3^-, and 9.00 mg/ml $CO_3^=$; anode and cathode compartments charged with 660 ml of 1.01 M HNO_3 and 650 ml of 0.94 M NaOH, respectively													
	1	1	11.5	25	52	52	52	278	1534	347	1.06	6.00	6.01	1.22
	2	2	11.3	25	57	56	56	266	1547	329	---	---	---	---

Table 6.4. (continued)

Date	Sample No.	Time, hr	Potential, volts	Current, amp	Compartment Temperatures, °C: Anode	Feed	Cathode	Effluent Volumes, ml: Anolyte	Waste	Catholyte	Concentrations: H^+ in Anolyte, M	Waste: Na^+, mg/ml	NO_3^-, mg/ml	OH^- in Catholyte, M
	3	3	11.3	25	56	55	55	265	1550	327	1.20	5.80	5.53	1.56
	4	4	11.3	25	56	55	55	264	1554	325	1.23	5.74	5.49	1.61
	5	5.1	11.3	25	56	55	55	279	1628	340	1.25	5.93	5.77	1.61
		Volume in compartments at end of run						620	620	600	---	---	---	---
	6	Total volume and composite analysis						1972	8433	2268	1.19	6.28	6.10	1.52
11/13/57	Feed, 6900 ml, was 6.50 mg/ml Na^+, 6.70 mg/ml NO_3^-, and 5.28 mg/ml $CO_3^=$; anode and cathode compartments were charged with 660 ml of 0.57 M HNO_3 and 650 ml of 0.63 M NaOH, respectively													
	1	1	18.3	15	54	53	49	352	1562	333	0.57	1.90	1.48	0.82
	2	2	18.2	15	58	59	53	355	1530	309	0.55	1.46	0.99	0.96
	3	3	18.2	15	58	59	55	352	1515	307	0.52	1.42	1.13	1.05
	4	3.15	18.2	15	58	59	55	259	1117	224	0.51	1.56	1.20	1.08
		Volume in compartments at end of run						620	630	605	---	---	---	---
	5	Total volume and composite analysis						1938	6354	1778	0.54	1.80	1.44	0.96

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