

Membrane Stability Testing

by

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Aiken, South Carolina 29808

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WSRC-OS-97-00007

Keywords: Caustic, Recovery,
Salt Solution,
Electrochemical,
Waste Management

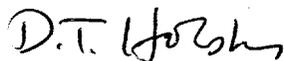
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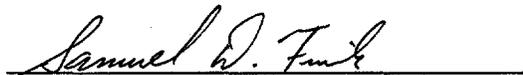
Membrane Stability Testing (U)

Attached is the final report submitted to the Westinghouse Savannah River Company by the Electrosynthesis Company, Inc. in accordance with terms specified under contract #AB93799N. This report details test conditions and findings from a 1000-hour stability test using a DuPont Nafion[®] Type 350 membrane in an electrochemical salt splitting process for the recovery of caustic from alkaline salt solution. The salt solution used in the test simulates the average flowsheet composition for the Savannah River Site Decontaminated Salt Solution which is produced in the In-Tank Precipitation process and disposed in the Saltstone low-level waste disposal facility. This work was funded by the Tank Focus Area Program of the Office of Science and Technology under Technical Task Plan SR16WT41, Subtask A, "Caustic Recycle."

Key findings of the testing included: (1) successfully recovered caustic from alkaline salt solutions over a 1079-hour period at high electrical efficiency, (2) no evidence of attack of the nickel cathode and platinized-titanium anode during the test, (3) nickel is not a suitable anode material for this application, (4) aluminum-containing solids can precipitate decreasing electrical efficiency, (5) periodic cleaning of the cell with dilute acid solution restores the electrical efficiency and (6) over the 1079-hour test period, the current efficiency dropped from >95% to approximately 90% indicating that the membrane had possibly suffered some degree of permanent damage.



D. T. Hobbs, Subcontract Technical Representative



Authorized Derivative Classifier



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Prepared for the Department of Energy Office of Technology Development, Office of Environmental Management under Contract DE-AC09-88SR18035

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“Membrane Stability Testing”

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EXECUTIVE SUMMARY

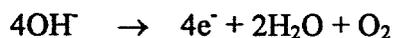
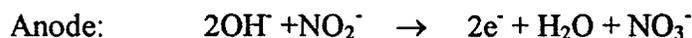
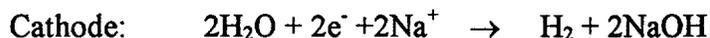
The Electrosynthesis Co., Inc. (ESC) has successfully demonstrated the operation of the ICI FM01 electrochemical flow cell over a 1000 hour period for the recovery of 13-14% caustic from a simulated SRS waste solution using a platinised titanium anode, nickel cathode, and a Nafion 350 membrane. At the end of the test period (1009 hrs) the anode showed no signs of deterioration in performance (based on operating potential) when compared to a new anode surface, and visual inspection revealed no obvious signs of wear. The cathode was very stable throughout the entire test period and visual inspection at the end revealed no obvious signs of wear. The membrane, however, has possibly suffered some degree of permanent damage. The damage manifests itself in the form of a slow but steady decline in caustic current efficiency, from a high of >95% down to approximately 90% after 1079 hours. It is believed that soluble Al and Si species present in the anolyte were transported into the membrane where they formed complex precipitates and possibly disrupted the membrane structure.

We have also shown that nickel is unsatisfactory as an anode material in this application. Over the period that it was tested (143 hrs), the anode suffered severe corrosion, losing a significant amount of mass and also contaminating the bulk anolyte solution with soluble nickel ions. It is believed that the high current density at the anode results in depletion of caustic in the boundary layer adjacent to its surface thereby facilitating attack by corrosive species present in the anolyte such as sulfate.

INTRODUCTION

The Electrosynthesis Co., Inc. (ESC) was contracted by the Westinghouse Savannah River Company under Subcontract # AB93799N to investigate the long term performance and durability of cell components (anode, membrane, cathode) in an electrochemical caustic recovery process using a simulated SRC liquid waste as anolyte solution. This report details the results of two long-term studies conducted using an ICI FM01 flow cell. This cell is designed and has previously been demonstrated to scale up directly into the commercial scale ICI FM21 cell. The first study used nickel electrodes, a Nafion 350 membrane and was run for 143 hours. The second study used a platinised titanium anode, nickel cathode, a new piece of Nafion 350 membrane and was run for 1155 hours.

The reactions occurring at the anode and cathode are as follows:



The reactions occurring at the anode may be any combination of the three listed. The cell voltage, electrode potentials, and membrane voltage drop were monitored continually as they provide an excellent indication of any deterioration in the cell components. The caustic current efficiency and water transport across the membrane were also measured throughout the run. The flow rate and purity of the anode gaseous products were measured as they provide an indication of the type of reaction occurring at the anode. On several occasions the anode and membrane were acid washed (usually due to increased cell voltage), either individually after opening up the cell or combined when the cell was washed *in situ*. Several of the washings were sent out to a commercial laboratory for

analysis. The cell components were also visually inspected at the end of the run for signs of deterioration.

EXPERIMENTAL

Process Description

The 1000-hour run was carried out using an ICI FM01 electrochemical flow cell (Appendix 1) equipped with PTFE spacers and gaskets. The anode was initially a flat plate nickel electrode. It was subsequently replaced by a platinised titanium flat plate anode. The cathode was a flat nickel plate, and the membrane was a DuPont Nafion 350 perfluorinated cation exchange membrane. This membrane contains sulfonate ion exchange groups with a higher molecular weight polymer on the cathode side to resist hydroxide back-migration. This membrane typically operates at about 85% current efficiency for the production of 15-20% NaOH in the chlor-alkali industry and is generally less sensitive to the influence of impurities¹. It was pretreated by soaking in 3.5M NaOH for several days. Turbulence promoters (PTFE) were placed on either side of the membrane for support and to prevent it from touching the electrodes. Power was supplied using a Sorenson # DCS 20-50 DC power supply.

The anolyte loop consisted of a 1 liter glass reservoir and a March MDK-MT3 circulating pump with a Kynar head. The flow rate was monitored using a Signet Scientific Co #3-2507.100-6V Kynar magnetic paddle wheel flow sensor coupled with an Omega #DPF75 flow controller. The temperature was monitored using an Omega #CN310 temperature controller coupled with a type "J" thermocouple mounted in a glass thermowell in the solution reservoir. On top of the reservoir was mounted a glass condenser to remove water vapor from the gases exiting the reservoir. The gas was vented to the atmosphere. Synthetic SRS solution was circulated continuously from the bulk (200 gallon) reservoir to the glass reservoir and back at approximately 53-73 ml/min. The anode potential was monitored continually using an Ag/AgCl reference

electrode mounted in a separate circulation loop and connected to a Luggin probe placed at the electrode.

The catholyte loop consisted of a 2 liter glass reservoir and a magnetically coupled gear pump (#H07144-91 motor, #H07001-40 SS head, Cole Parmer Instrument Co.) for circulation. The flow rate was monitored using a Signet Scientific Co #3-2507.100-6V Kynar magnetic paddle wheel flow sensor coupled with an Omega #DPF78A flow controller. The temperature was monitored using an Omega #CN310 temperature controller coupled with a type "J" thermocouple mounted in a glass thermowell in the solution reservoir. On top of the reservoir was mounted a glass condenser to remove water vapor from the gas leaving the reservoir. This gas was vented to the atmosphere. Deionized water was fed continually to the catholyte reservoir to maintain the caustic strength and the product caustic overflowed to a separate collection vessel. Solution samples were also taken frequently and analyzed for caustic strength. The cathode potential was monitored continually using an Ag/AgCl reference mounted in a separate circulation loop and connected to a Luggin probe placed at the electrode. A diagram of the test rig is shown in Appendix 2. Selected data (cell voltage, electrode potentials, membrane voltage drop, solution temperatures) were monitored and recorded continually using the DucksoupTM Version 1.23 data acquisition software coupled with a Keithly Metrabyte DAS1401 data acquisition board. All voltage signals coming into the board were first passed through a National Instruments #SCXI 1120 signal isolation amplifier which was connected to a #SCXI 1000 chassis (for power) through a #SCXI 1320 terminal block. This system filtered out AC "noise" in the signals caused by the close proximity to other electrical equipment. Other data (recirculation rates, volumes, concentrations etc were recorded on log sheets. All major electrical components of the system were connected to a Sola model # 510 uninterruptible power supply to ensure continued operation . A summary of actual operating conditions is listed below.

Anolyte flow rate : 0.7 gal / min

Catholyte flow rate : 0.7 gal / min

Anolyte temperature : Ambient
Catholyte temperature : Ambient
Current Density : 400 mA / cm²
Initial SRS solution caustic strength : 1.5M
Caustic concentration : 13-15%

Safety Interlocks

The safety interlock system was designed to shut down the test rig in the event an unsafe condition occurred. In addition the system could be shut down manually. The unsafe conditions are as follows:

Low anolyte flowrate (set at 0.3 gal / min)
Low catholyte flowrate (set at 0.3 gal / min)
Liquid spill on floor near bulk anolyte tank
Liquid spill in drip tray under setup

If an unsafe condition occurred then the shutdown was automatically initiated with the following result:

Power supply shutdown
Anolyte circulation pump shutdown
Catholyte circulating pump shutdown
Dilution water pump (for caustic produced) shutdown
SRS soln circulation pump (from bulk reservoir) shutdown

Sampling and Analysis

Caustic solution samples were taken daily, usually over a six-hour period and analyzed for strength using the standard acid/base titration method with phenolphthalein as

indicator. The caustic current efficiency was then calculated for that time period. The carbonate content of the caustic was also calculated from the titration using the bromocresol green indicator end point. The water transport across the membrane was calculated based on the volume of water fed to the caustic reservoir, and the volume, density, and concentration of caustic collected.

The caustic content of the anolyte along with the carbonate content was measured using the same procedure described above. The SRS solution was also analyzed periodically for anions (NO_2^- , NO_3^- , F^- , Cl^- , PO_4^{3-} , SO_4^{2-}) using a Dionex DX 500 Ion Chromatograph equipped with an IonPac AS4A-SC analytical column, AG4A-SC guard column, CD-20 conductivity detector, GP-40 gradient pump, and controlled using the Peaknet™ software system. The catholyte solution was also frequently analyzed for these anions. The anolyte off gas flow rate was measured using a simple burette/soap solution type bubble meter. It was qualitatively analyzed on a GowMac gas chromatograph equipped with a series 550 Thermal Conductivity detector and a Hewlett Packard #3396A integrator. Oxygen was detected on a Molecular Sieve 60/80 6 ft column using helium as carrier gas. The SRS solution was analyzed for silicon, aluminum, and chromium by a commercial laboratory (Galbraith Laboratories, Inc.).

Acid Washing

The anode and membrane were acid washed on several occasions throughout the run. For the first two times the cell was taken apart and the components were washed separately with 1.0M HCl. The third acid wash was performed *in situ* where the cell was first drained and then rinsed three times with deionized water. Then 1.0M HCl was added to the anolyte compartment and circulated for 15 minutes. The washing was then collected and stored. The last three acid washes were also performed *in situ* as before with the exception that 1.0M HNO_3 was used instead of HCl. Selected acid washing samples were sent out for analysis.

Anolyte Preparation

The SRS synthetic solution was prepared by dissolving calculated amounts of reagent grade chemicals to give 610 L of final solution as summarized in Table 1. The assumption was made that the caustic content of the anolyte should initially be enough to supply all the Na^+ needed for caustic production in the catholyte as well as to maintain sufficient alkalinity in the anolyte to avoid precipitation at the end of the run.

Table 1

| Element | Initial Concentration (Nickel Anode run) | Initial Concentration (Platinised Ti Anode run) | Final Concentration (Platinised Ti Anode Run) |
|--------------------|---|--|--|
| NO_2^- | 0.57 M | 0.59 M | 0.081 M |
| NO_3^- | 2.10 M | 2.30 M | 2.89 M |
| F^- | 0.017 M | 0.017M | |
| Cl^- | 0.024 M | 0.024 M | 0.022 M |
| PO_4^{3-} | 0.006 M | 0.0072 M | 0.006 M |
| SO_4^{2-} | 0.14 M | 0.15 M | 0.148 M |
| CO_3^{2-} | 0.17 M | 0.12 M | 0.098 M |
| OH^- | 1.55 M | 1.34 M | 0.69 M |
| Si^{4+} | 89.7 ppm | 99.6 ppm | 57.0 mg/l |
| Al^{3+} | 0.319 % | 0.325 % | 4.04 g/l |
| Cr^{6+} | 0.011 % | 0.013 % | 0.185 mg/l |
| Ni | <0.4 ppm | 4.6 ppm | |

RESULTS AND DISCUSSION

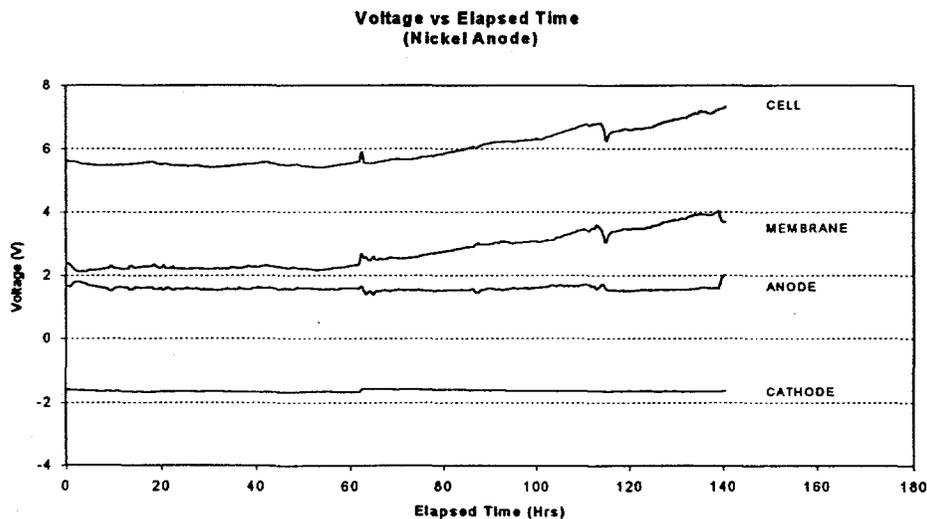
Nickel Anode/Cathode Configuration

The initial run used a cell configured with a Ni anode, Ni cathode and a Nafion 350 membrane.

Cell Potentials

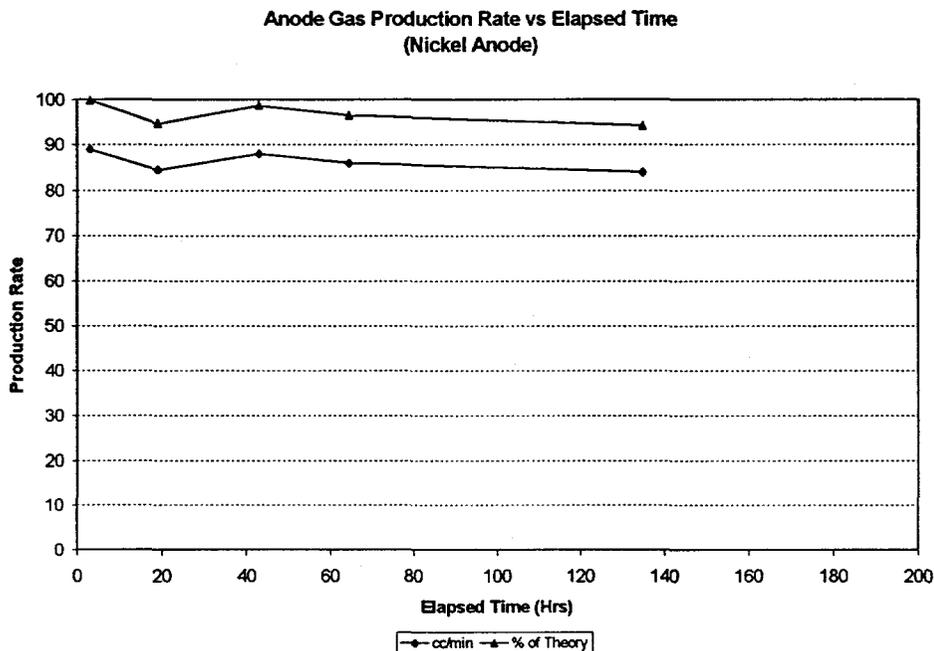
The overall cell voltage consists of several components; the decomposition voltage for the reaction, anode overpotential, cathode overpotential, membrane IR, and electrolyte and structural IR. The decomposition voltage is independent of the cell design or hardware used. The anode and cathode overpotentials represent the extra energy above the decomposition voltage needed for the reaction to proceed at a reasonable rate. The membrane, electrolyte and structural IR are resistive losses due to the passage of electricity through them and are affected by factors such as electrolyte concentration, temperature, interelectrode gap, presence of impurities etc. Figure 1 shows the voltage vs. elapsed time for the individual cell components as well as the overall cell voltage measured throughout the run.

Figure 1



The cell was operated for 143 hours with a Ni anode before it was shut down due high voltage and the presence of an increasing amount of solids in the anolyte causing difficulties with maintaining a consistent flow rate. The results show that the overall cell voltage stayed constant at around 5.5-5.6V for the first 60 hours before it began to rise, reaching a high of 7.3V before the run was terminated. The increase coincided with a similar increase in the membrane voltage drop. The anode potential throughout the run remained relatively constant at around 1.6 Volts vs. Ag/AgCl reference. Measurements of the anolyte off gas flow rate (Figure 2) and composition indicate that for the duration of the run, >95% of the current was consumed by the O₂ producing reactions and very little was used for the oxidation of NO₂⁻ to NO₃⁻. Frequent GC analysis of the off gas indicated the presence of O₂ only.

Figure 2



This is consistent with previous work done for Westinghouse² which has shown that nickel anodes are very inefficient for the oxidation of NO₂⁻ in SRS simulants. The

cathode potential was also very stable for the duration of the run, averaging -1.6V vs. Ag/AgCl reference.

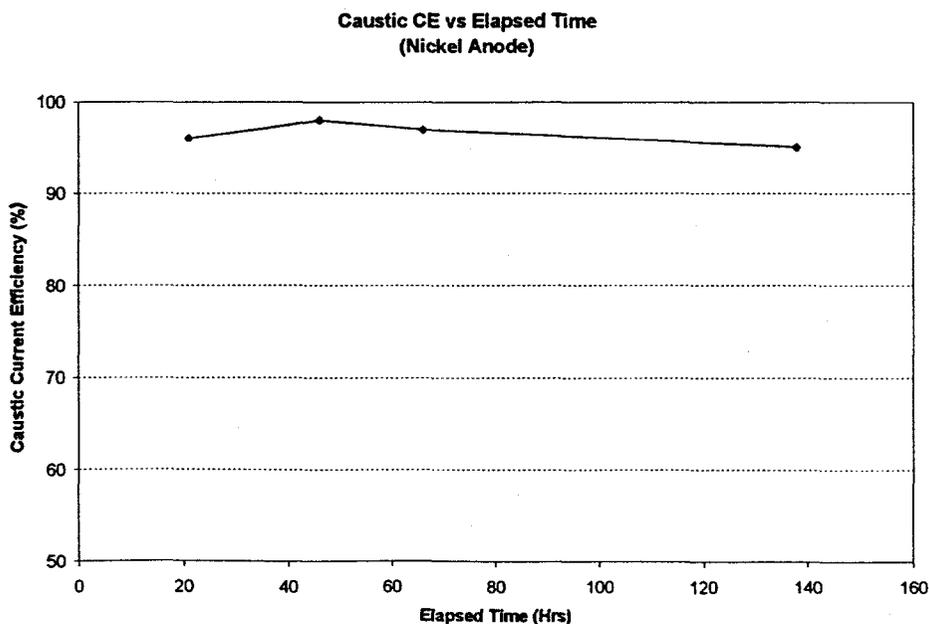
Cell Disassembly and Inspection

Examination of the cell components at the end of the run (see photographs Appendix 3) showed that the anode suffered severe corrosion, losing a significant amount of mass (approximately 7g). In addition, solution analyses by Galbraith Laboratories, Inc. (Appendix 4, sample B) indicate a significant increase in the nickel content of the anolyte. It is believed that the corrosion was caused by a depletion of hydroxide species in the boundary layer immediately adjacent to the anode (due to insufficient mixing at this very high current density) resulting in attack of the anode by corrosive ions such as SO_4^{2-} . The membrane appeared covered with a white precipitate on the anodic surface as well as within its structure.

Caustic Current Efficiency

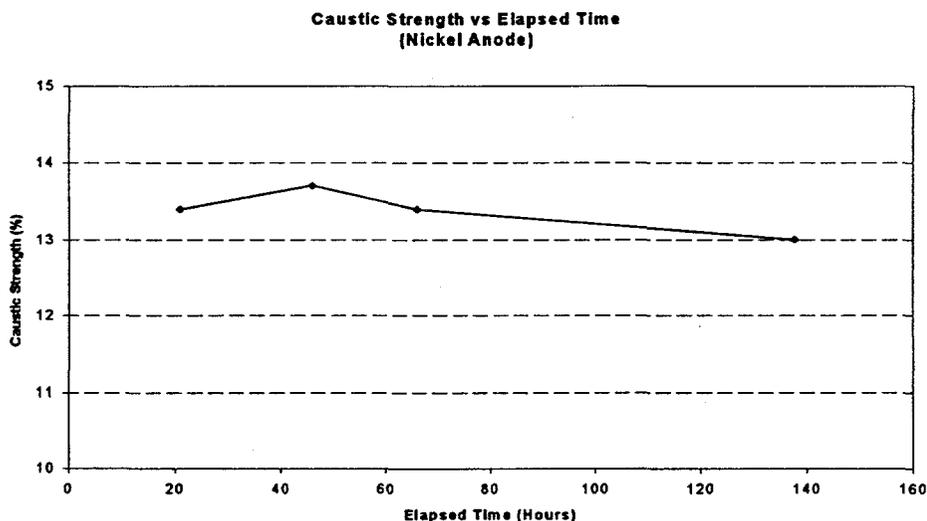
The current efficiency for caustic production is shown in Figure 3.

Figure 3



The current efficiency remained >95%, producing 13-14% caustic for the entire run (Figure 4). The current efficiency is significantly higher than is typically achieved for this type of membrane in chlor-alkali service (85%)³, and may be partly due to the high salt concentration in the anolyte which could tend to dehydrate the membrane and therefore reduce the backmigration of OH⁻ species from the catholyte compartment.

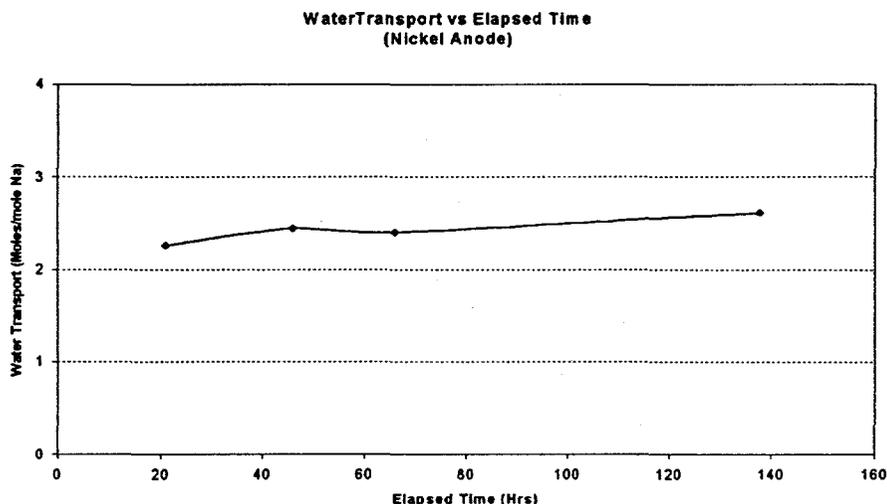
Figure 4



Water Transport

Water transport occurs in the cell from anolyte to catholyte because the sodium ion carries a solvation sphere with it through the membrane. Water transport is very sensitive to anolyte strength and usually decreases as anolyte strength increases⁴. The water transport measured for this experiment is shown in Figure 5.

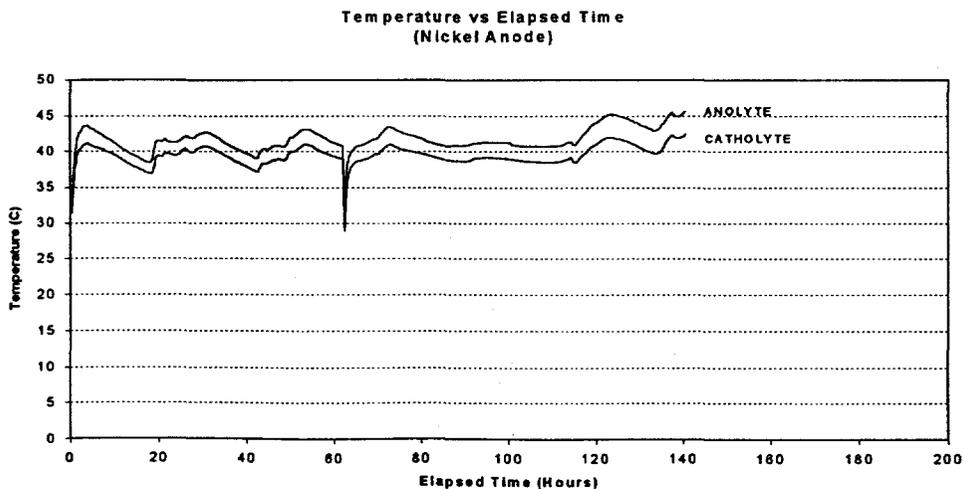
Figure 5



The water transport varied between 2.3 – 2.6 moles/mole Na^+ , and although there seems to be a slight increase over time, it is likely related to experimental error rather than changing anolyte strength.

The system was run at ambient temperature and thus was sensitive to fluctuations in the external laboratory temperature. These were small and thus had only a marginal impact on the cell operation. Figure 6 shows the temperature profile for the duration of the experiment. The operating temperature in the cell typically averaged 40^o C.

Figure 6



Platinised Titanium Anode/Nickel Cathode Configuration

Due to the premature failure of the Ni anode, a second long-term study was conducted with a platinised titanium anode. This run used the anolyte solution from the end of the first study. The composition of this solution was listed in Table 1 previously. During this study there were several events that took place to ensure continued operation of the experiment. These are listed in Table 2

Table 2

List of Events

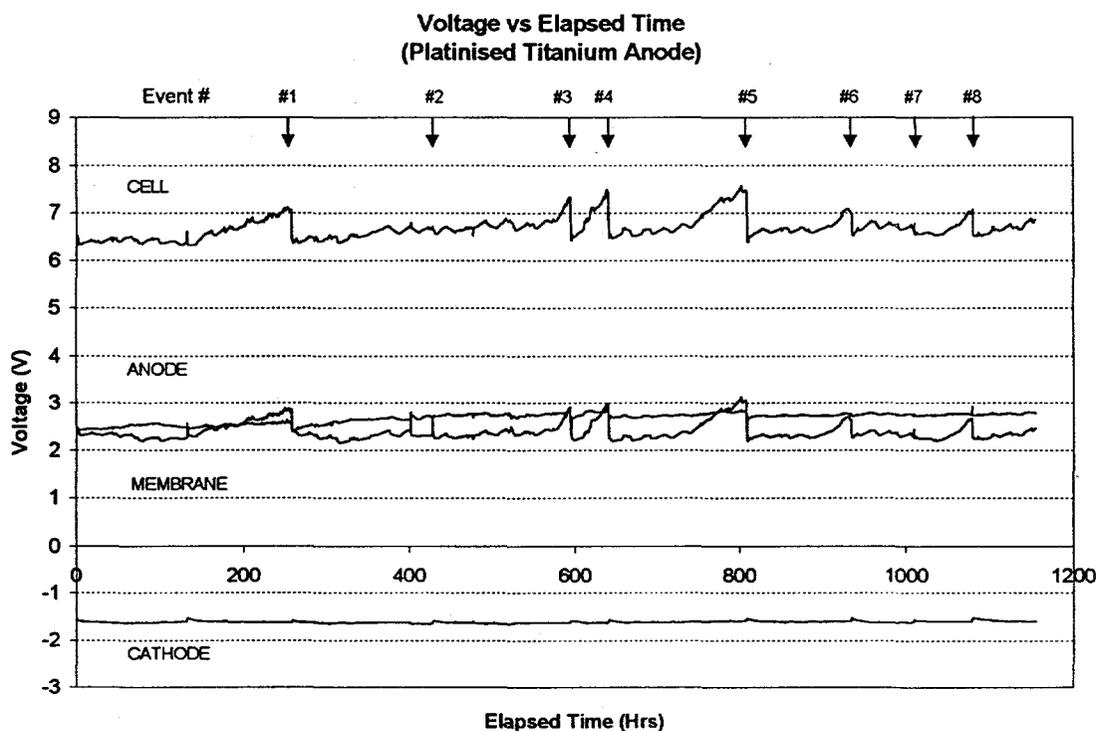
| Event # | Elapsed Time (Hours) | Description |
|---------|-------------------------|--|
| 1 | 257.5 | Cell opened, acid washed anode and membrane 1.0M HCl Membrane soaked in 4.0M NaOH over weekend Washings analyzed |
| 2 | 426 | Cell opened, acid washed anode and membrane 1.0M HCl Membrane soaked in 4.0M NaOH for 1 hour |
| 3 | 594 | <i>In situ</i> acid wash with 1.0M HCl |
| 4 | 640.25 | <i>In situ</i> acid wash with 1.0M HNO ₃ . Added NaOH soln to bulk anolyte |
| 5 | 808 | <i>In situ</i> acid wash with 1.0M HNO ₃ . Added NaOH pellets to bulk anolyte Washings analyzed |
| 6 | 934 | <i>In situ</i> acid wash with 1.0M HNO ₃ . Washings analyzed |
| 7 | 1009 | Cell opened, new platinised titanium anode inserted Added NaOH soln to bulk anolyte |
| 8 | 1079 | Cell opened, new Nafion 350 membrane inserted Added NaOH soln to bulk anolyte |

Cell Potentials

The individual and combined cell potentials measured during the experiment are presented in Figure 7. The cell was operated for a total of 1155 hours before the

experiment was ended. The cell voltage started at around 6.4V and remained stable for roughly 150 hrs before beginning to increase, reaching a high of 7.1V after 257 hours when the cell was shut down (event #1). The anode potential remained relatively constant during this time at 2.4-2.5V vs. Ag/AgCl reference. The cathode potential remained stable for the entire 1155 hours of the run at -1.6V vs. Ag/AgCl reference. The membrane voltage however, closely mirrored that of the cell, remaining relatively flat for 150 hours before rising significantly. Upon opening up the cell, the membrane was found to have a white precipitate on its surface as well as within its structure.

Figure 7



The anode was also coated with a white/yellow thin film (see photo Appendix 5). Analysis of the acid washings from the membrane and anode (Appendix 4, samples C, D) indicated elevated levels of Si and Al species as well as some Ca^{2+} . The cell was put

back together and restarted. Thereafter the cell was acid washed five more times (events #2 to #6) primarily due to increased membrane voltage, except for event #2 where the cell was acid washed to clear the plugged anode luggin probe which had been causing erroneous readings. Each acid washing appeared to return the membrane voltage drop back to its original value. Analysis of selected acid washings (from events #5 and #6) also indicated elevated levels of Al and Si species (Appendix 4, samples E, F).

It has been reported⁵ that silica species in the anolyte can be transported into the membrane where it may combine synergistically with Ca and Al species to form precipitates, which permanently damage the membrane structure. This movement is believed to be facilitated by the water transport⁶ occurring with the Na⁺ ions and ultimately results in a reduction of the membrane lifetime. Established maximums as low as 10 ppm⁷ as SiO₂ have been reported in chlor-alkali applications. (Note that these levels have been established for the bilayer membranes containing carboxylate polymer. It is known that these membrane structures are more sensitive to impurities than the 300 series membranes.)

The anode potential increased slowly but consistently from 2.4V to 2.7V vs. Ag/AgCl reference over the 1009 hours that it was used in the cell. This increase is likely due to the changing nature of the reactions occurring at the anode. The off gas produced at the anode provides an excellent indication of what reactions are occurring there. Measurements of the flow rate were taken throughout the run. The results are presented in Figure 8.

These results show that for the first 650 hours, 40% or less of the current is being consumed by the O₂ evolving reactions, with the rest being used to oxidize NO₂⁻ to NO₃⁻. Frequent GC analysis of the off gas indicated the presence of O₂ only. Analysis of the bulk anolyte (Figure 9) during the run showed a steady decrease in the NO₂⁻ content and a corresponding increase in the NO₃⁻ content. Thereafter the off gas production rate increases until the end of the run (1009 hours) when 68% of the current was consumed by the O₂ producing reaction. The almost instantaneous increases in off gas flow rates

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Figure 8

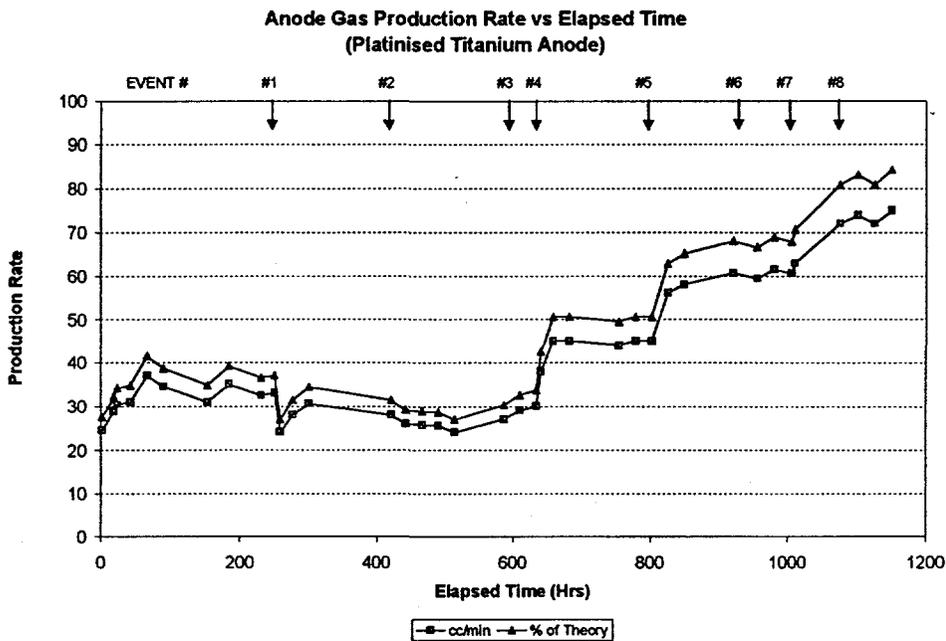
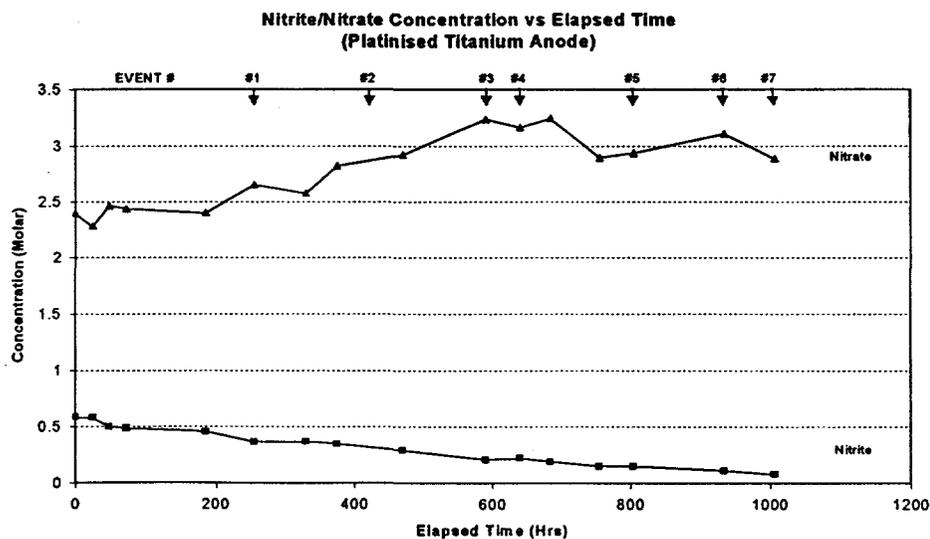
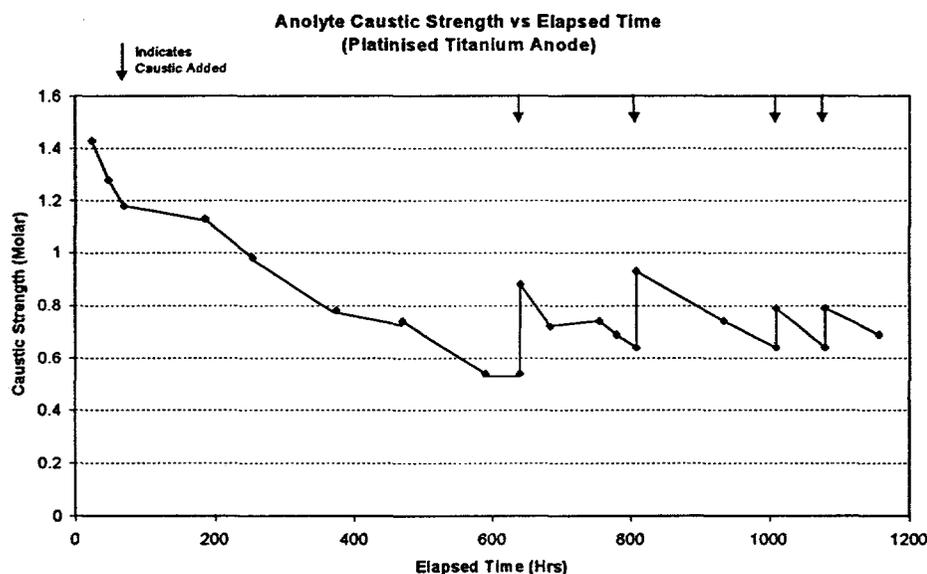


Figure 9



observed after events #4, #5, and #7 are likely due to the caustic added at those times, making conditions more favorable for oxygen evolution. A summary of the caustic additions to the bulk anolyte is shown in Figure 10.

Figure 10



It was necessary to add caustic to the bulk anolyte on several occasions to prevent precipitation from occurring. Precipitation was initially observed after approximately 594 hours when the caustic content dropped to 0.54N NaOH. This caused a rapid increase in cell voltage and necessitated an acid wash (Event #4) shortly after the previous one. Subsequently the caustic content was maintained above 0.6N NaOH. After 1009 hours (event #7) the anode was reversed so that the unused side was now exposed to the solution. The cell was run a further 146 hours with this anode surface to establish a new baseline with the changed anolyte composition. The potential profile for the new anode surface was almost identical to that of the used surface, indicating that there was no apparent damage to the anode over the 1009 hours that it was tested.

Current Efficiency

The caustic current efficiency measured during the run is shown in Figure 12 and the caustic strength produced is shown in Figure 11.

Figure 11

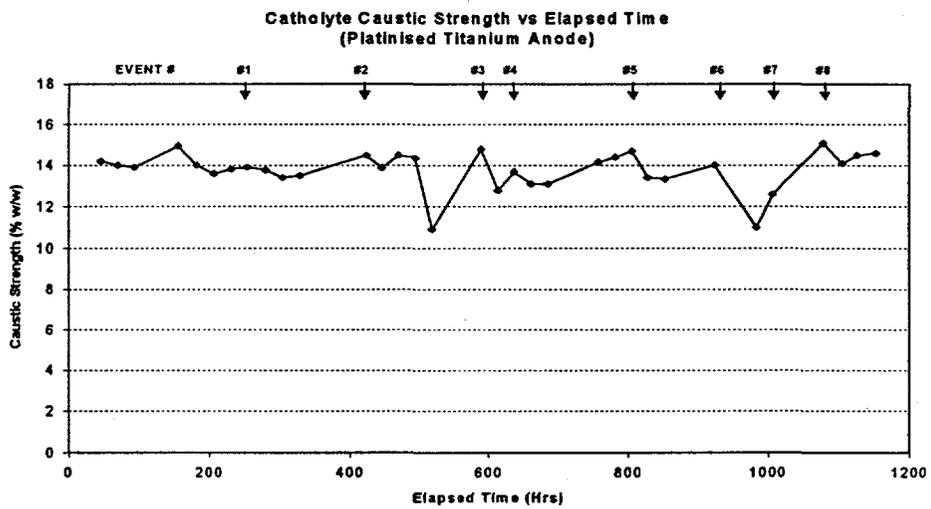


Figure 12

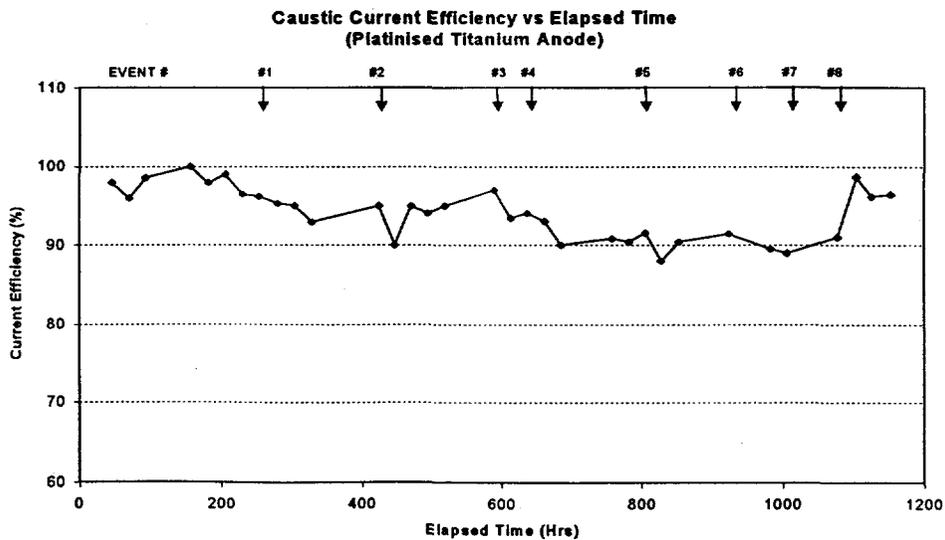
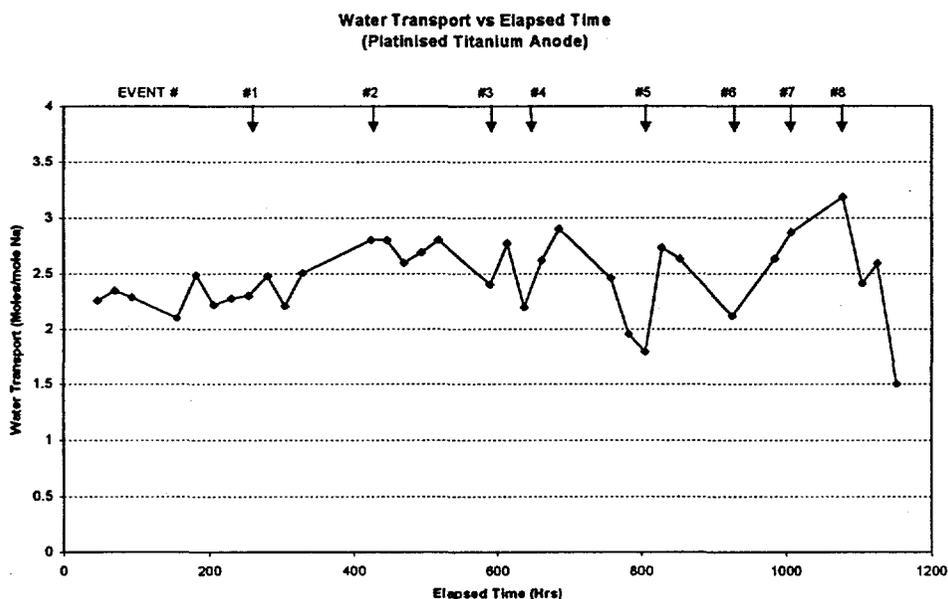


Figure 13



The results indicate that current efficiencies were quite high for roughly the first 200 hours, producing approximately 14% NaOH at over 95%. However, the efficiency declined slowly, finally averaging around 90%. After 1079 hours the Nafion 350 membrane was replaced to determine the effect on current efficiency (event #8). This resulted in an immediate and sustained increase in the current efficiencies back to >95%, as was measured initially. It is believed that the used membrane has suffered some degree of permanent damage, likely due to the precipitation of Si and Al species within the membrane referred to previously.

The water transport across the membrane is shown in Figure 13. For the majority of time the water transport ranged between 2-3 moles/mole Na^+ . This is similar to that observed previously. Several analyses by ion chromatography of the caustic produced over the 1000-hour run indicated that NO_3^- was the only detectable impurity present. The highest concentration obtained was 0.6 mM and the average was 0.4mM. These levels are

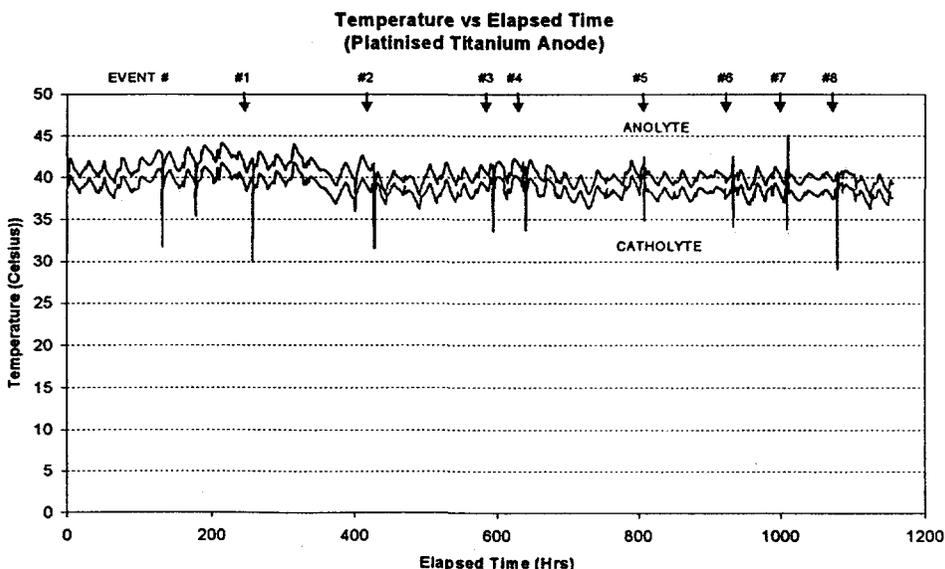
comparable to the Cl^- levels in caustic normally seen in the conventional chlor-alkali process⁸. It is possible to calculate a maximum diffusion coefficient of $3.6\text{E}^{-13} \text{ m}^2/\text{sec}$ from this data, which is quite low when compared to reported diffusion rates of other similar anions through perfluorosulfonic acid type membranes⁹.

The operating temperature of the system throughout the run was very similar to that observed earlier and averaged around 40°C (Figure 14).

Cell Disassembly and Inspection.

Examination of the cell components after they were removed from the cell (see photographs Appendix 6) showed that the anode appeared physically unchanged except for a slight surface discoloration. The cathode surface also appeared to have a dark discoloration, but this was easily removed by acid washing with dilute acid. There was no apparent physical damage to the cathode. The membrane appeared to have a white deposit both on its surface as well as within its structure. Acid washing of the membrane with dilute nitric acid was successful in removing all of the deposit from the surface and some from within the structure.

Figure 14



CONCLUSIONS AND RECOMMENDATIONS

We have successfully demonstrated that caustic can be recovered from a simulated SRS liquid waste using an ICI FM01 electrochemical cell equipped with a platinised titanium anode, Nafion 350 membrane, and nickel cathode. Over the period tested (1009 hrs), the cell consistently produced an average 13.8% NaOH solution of high purity. The performance (voltage profile) of the platinised titanium anode appeared unchanged after the run when compared to the performance of a new (unused) surface. The caustic current efficiency began at > 95% efficiency but dropped slowly down to 90% by the end of the run. Comparison of the used membrane performance with that of a new piece indicates that there has been some permanent damage to the membrane. This is likely due to the precipitation of Al and Si species which react synergistically to form complex precipitates within the membrane structure, thereby disrupting it. While it has been shown that the membrane performance can be partly restored by acid washing, it is not known how much further the current efficiency will drop at even longer operating times. No apparent damage to the cathode was noted.

We have also shown that nickel is an unacceptable anode material in this application. During the 143 hours that the cell was run with a nickel anode, severe degradation of its surface occurred, causing precipitation of nickel containing solids in the anolyte reservoir that necessitated stopping the run. The anolyte was also contaminated with soluble nickel species, which in high enough quantities are also known to affect membrane performance¹⁰. It is believed that the degradation is a result of the high current density in combination with the presence of corrosive species such as SO_4^{2-} in the anolyte, facilitating the attack on the anode surface. It is possible that the deterioration in membrane performance (due to Al/Si precipitates) could be reduced through the following approaches:

INFORMATION ONLY

- (a) Increase the amount of water in the membrane. This could be achieved by either diluting the catholyte or by using a lower equivalent weight polymer. The latter would likely affect the current efficiency of the process.
- (b) Reduce the current density. This should prolong the onset of precipitation in the membrane.
- (c) Increase the temperature. This may increase the solubility of the Si and Al species in the membrane.
- (d) Use a different membrane.

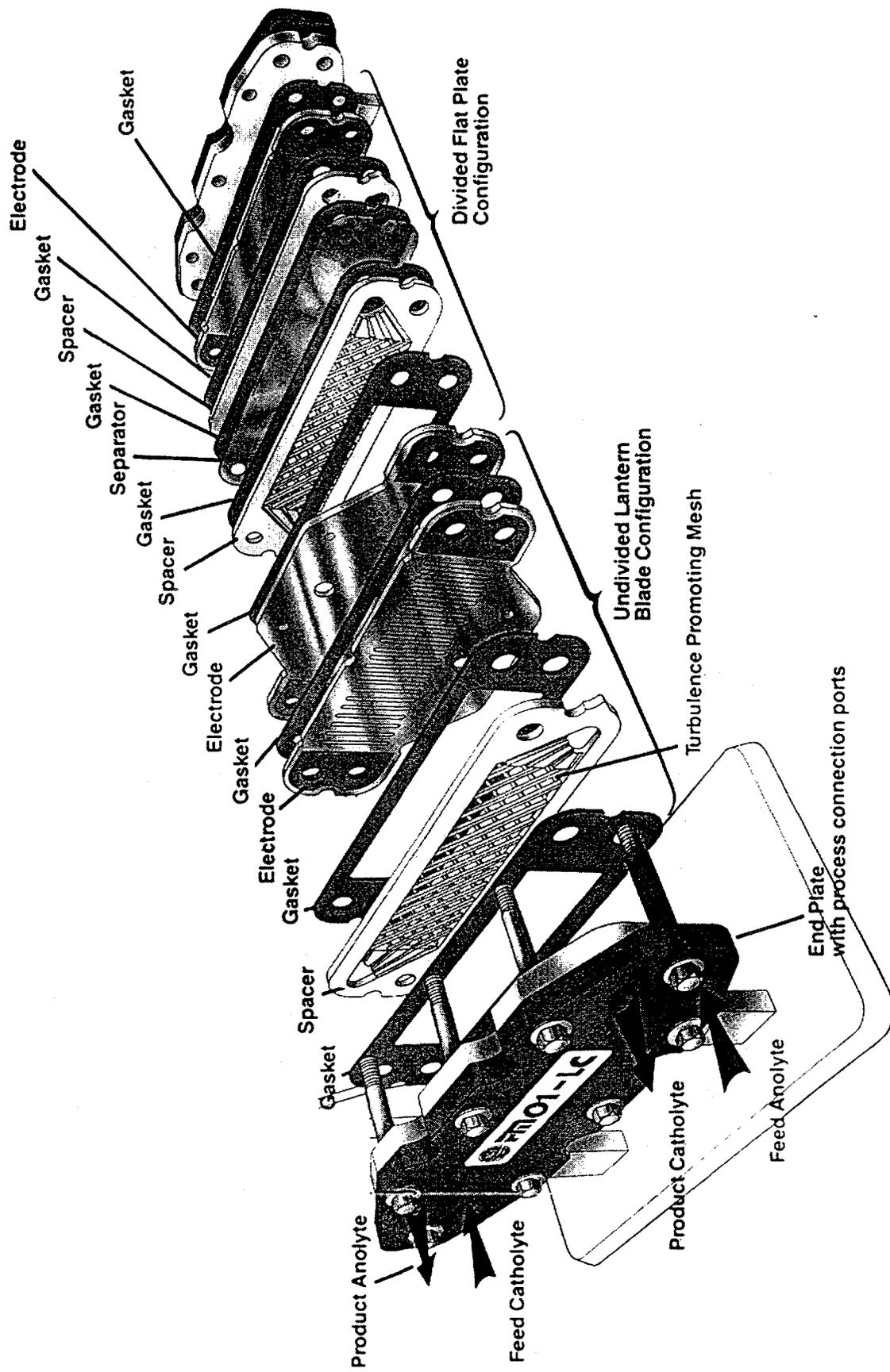
Clearly the tradeoff between membrane life, current efficiency and operating voltage will need to be assessed as a function of overall economics.

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Appendix 1

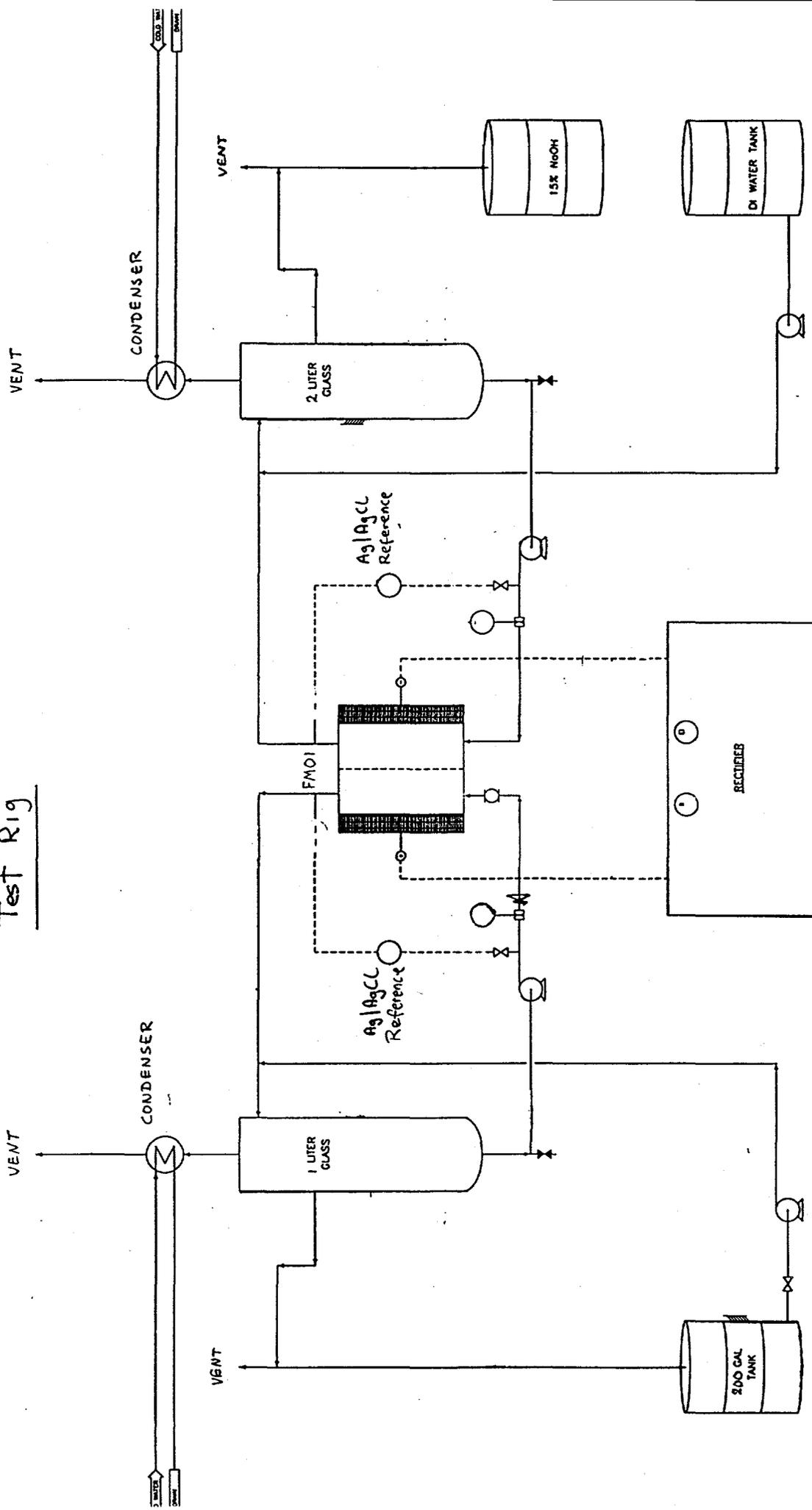
ICI FM01 Cell



Appendix 2

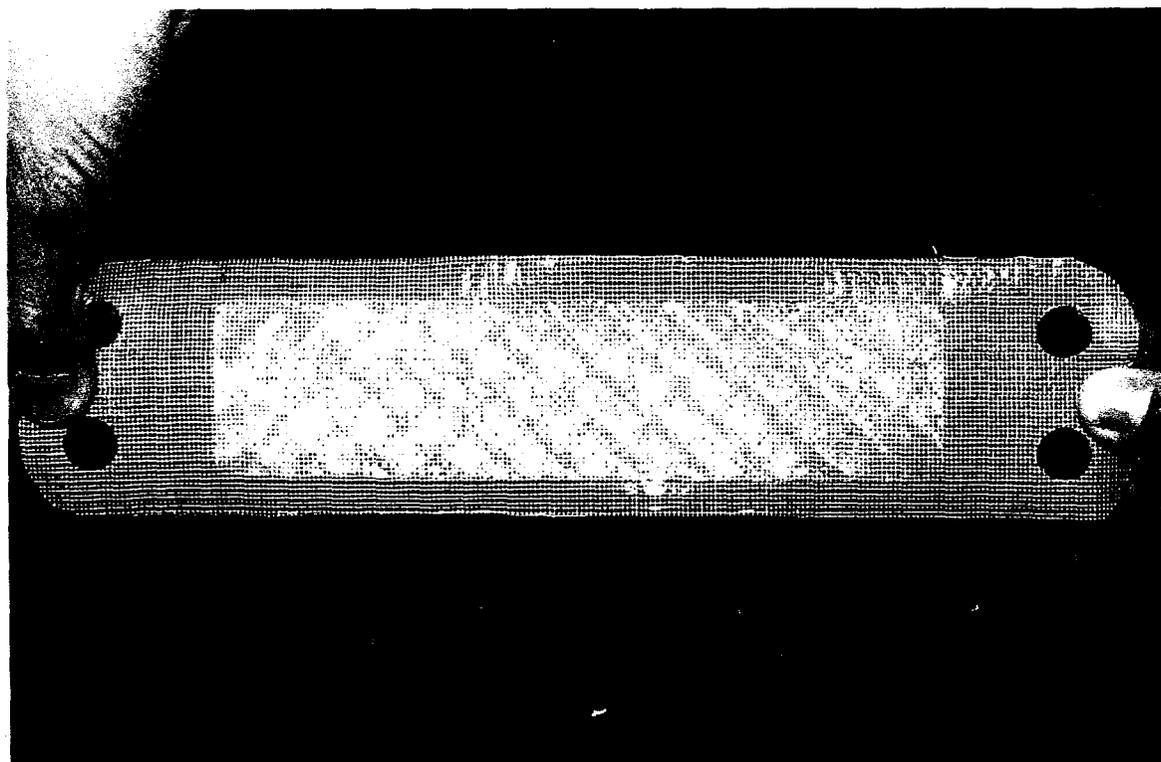
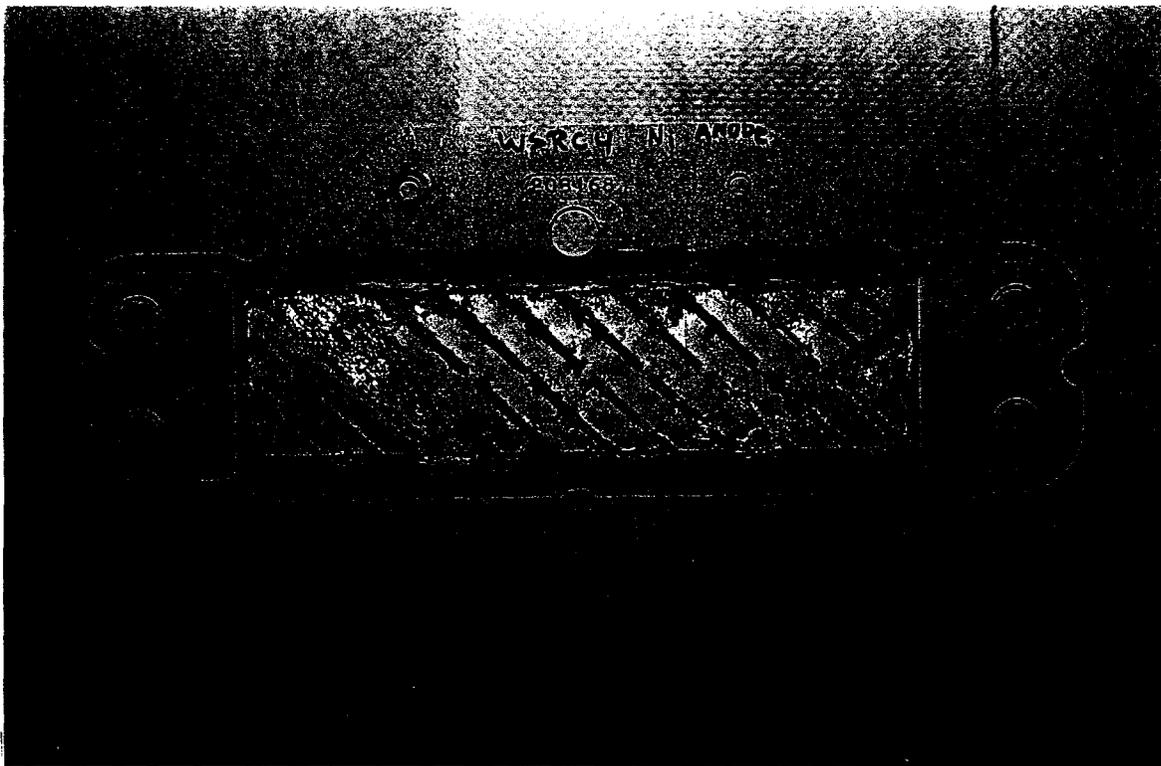
Schematic of Test Rig

Test Rig



Appendix 3

Photographs of Components from Nickel Anode/Cathode Run



Appendix 4

Galbraith Laboratories Analytical Results.

Summary Table

| Sample ID# | Description |
|------------|---|
| A | Initial Bulk Anolyte Soln (Ni Anode Run) |
| B | Initial Bulk Anolyte Soln (Platinised Ti Anode Run) |
| C | Membrane Acid Washing (255 Hrs Elapsed) |
| D | Electrode Acid Washing (255 Hrs Elapsed) |
| E | Acid Washing (808 Hrs Elapsed) |
| F | Acid Washing (934 Hrs Elapsed) |
| G | Bulk Anolyte (1009 Hrs Elapsed) |



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Dennis Chai
Electrosynthesis Co
72 Ward Rd
Lancaster NY 14086

Report Date: 07/14/97
Sample Received: 06/12/97
Purchase Order #: WSRC461197DC
Fax Number: 716-684-0511

| SAMPLE ID | LAB ID | ANALYSIS | RESULTS | |
|-----------|--------|----------|---------|-----|
| A | U-2076 | Aluminum | 0.319 | % |
| | | Silicon | 89.7 | ppm |
| | | Nickel | < 0.4 | ppm |
| | | Chromium | 0.011 | % |
| B | U-2077 | Aluminum | 0.325 | % |
| | | Silicon | 99.6 | ppm |
| | | Nickel | 4.6 | ppm |
| | | Chromium | 0.013 | % |

BDS:sc



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Dennis Chai
Electrosynthesis Co
72 Ward Rd
Lancaster NY 14086

Report Date: 07/28/97
Sample Received: 07/02/97
Purchase Order #: WSRC470197DC
Fax Number: 716-684-0511

| SAMPLE ID | LAB ID | ANALYSIS | RESULTS | |
|-----------|--------|----------|---------|------|
| C | U-4782 | Aluminum | 327 | mg/l |
| | | Silicon | 284 | mg/l |
| | | Calcium | 38 | mg/l |
| D | U-4783 | Aluminum | 78 | mg/l |
| | | Silicon | 68 | mg/l |
| | | Calcium | 6 | mg/l |

ICP:le



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LABORATORY REPORT

Dennis Chai
Electrosynthesis Co
72 Ward Rd
Lancaster NY 14086

Report Date: 08/07/97
Sample Received: 08/05/97
Purchase Order #: WSRC480497DC
Fax Number: 716-684-0511

| SAMPLE ID | LAB ID | ANALYSIS | RESULTS | |
|-----------|--------|----------|---------|------|
| E | U-8700 | Aluminum | 400 | mg/l |
| | | Silicon | 353 | mg/l |
| F | U-8701 | Aluminum | 459 | mg/l |
| | | Silicon | 368 | mg/l |
| G | U-8702 | Aluminum | 4040 | mg/l |
| | | Silicon | 57 | mg/l |
| | | Chromium | 185 | mg/l |

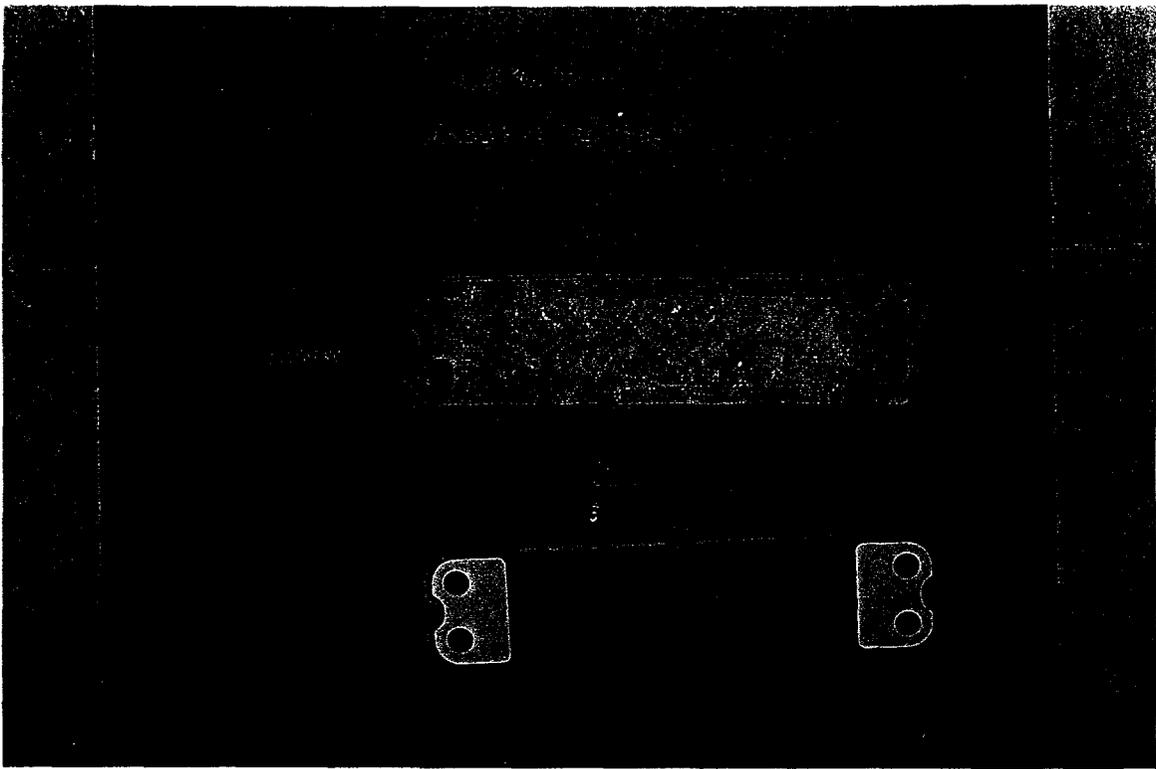
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Appendix 5

Photographs from Platinised Ti Anode/ Ni Cathode Run after 255 hours.



Appendix 6

Photographs from Platinised Ti Anode/Ni Cathode Run at end.

