

Hydrogen Anode for Nitrate Waste Destruction (U)

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Abstract

The use of a hydrogen gas-fed anode and an acid anolyte in an electrochemical cell used to destroy nitrate was demonstrated. A mixed $\text{Na}_2\text{SO}_4/\text{H}_2\text{SO}_4$ anolyte was shown to favor the nitrate cell performance, and the generation of a higher hydroxide ion concentration in the catholyte. The suggested scheme is an apparent method of sodium sulfate disposal and a possible means through which ammonia (to ammonium sulfate, fertilizer) and hydrogen gas could be recycled through the anode side of the reactor. This could result in a substantial savings in the operation of a nitrate destruction cell.

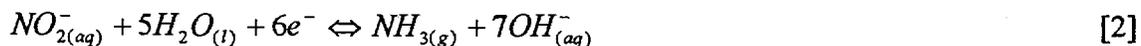
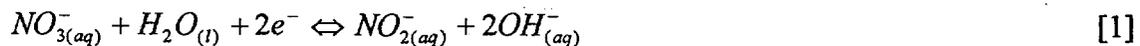
Introduction

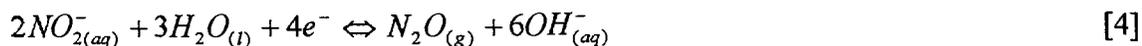
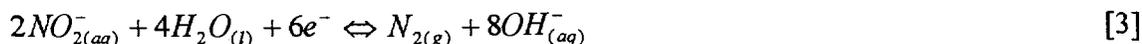
Large quantities of radioactive and hazardous wastes have been generated from nuclear materials production during the past fifty years. Processes are under evaluation to separate the high level radioactive species from the waste and store them permanently in the form of durable solids. The schemes proposed will separate the high level radioactive components, cesium-137 and strontium-90, into a small volume for incorporation into a glass wasteform. The remaining low-level radioactive waste contain species such as nitrites and nitrates that are capable of contaminating ground water. Electrochemical destruction of the nitrate and nitrite before permanent storage has been proposed^{1, 2}. Not only will the electrochemical processing destroy these species, the volume of the waste could also be reduced.

Li *et al.*³ studied the electrochemical reduction of nitrate at nickel, zinc, lead, and iron cathodes in alkaline solutions. They found that nitrite was an intermediate in the electrochemical reduction of nitrate and lead electrode appears to be the best cathode for the reduction process. Hobbs *et al.*² reached the same conclusion with the effective treatment of both simulated and real wastes in a parallel-plate cell. Unfortunately, long term use of lead electrode is not environmentally acceptable. Using concentrated NaOH solutions with platinum and nickel electrodes, Li *et al.*⁴ found that the generation of ammonia or nitrogen is highly dependent on the applied current density. At lower current densities (less than 100 mA/cm²), nitrogen was the major gaseous product, while at higher current densities (460 mA/cm²) ammonia was the major product. Nickel appears to be the best choice cathode material for the nitrate reduction process.

Nitrate/Nitrite Cell Reactions

In an alkaline solution at a lead or nickel electrode, the important cathodic reactions involving nitrogenous species are believed to be¹:





The main reaction at the anode in a separated cell is the evolution of oxygen as shown in equation [6]. For an undivided cell, the nitrite species are re-oxidized at the anode.



The overall cell reaction shows that for every mole of nitrate or nitrite destroyed, one mole of hydroxide ion is produced.

Because ammonia gas is considered hazardous, the reduction of nitrate/nitrite to nitrogen gas is highly preferred. Unfortunately, operating current required to achieve an economic reduction of nitrate (with respect to processing time), results in the generation of ammonia. Also, the oxygen generated at the anode, (eqn. [6]), could combine with ammonia, in the absence of a separator, to form an explosive mixture. Anodic evolution of oxygen gas consumes a high amount of energy. It is of interest, therefore, to reduce the energy that will be required to operate a nitrate cell at an economically useful current density. With a hydrogen anode, the high energy consumption associated with oxygen evolution can be avoided and the hydrogen generated at the cathode recycled to the anode for use.

Hydrogen anode can be used in an anolyte of sodium hydroxide (NaOH), sulfuric acid (H₂SO₄) or a mixture of Na₂SO₄/H₂SO₄. An anolyte of sulfuric acid or a mixture of sodium sulfate and sulfuric acid can be beneficial to a nitrate cell equipped with a hydrogen consuming gas anode, because the ammonia produced on the cathode can be recycled through the sulfuric acid generated in the anolyte to produce ammonium sulfate (a fertilizer).

There is also an interest in the recovery of sodium hydroxide from sodium sulfate. Sodium sulfate (Na₂SO₄) is a by-product of sulfuric acid neutralization of alkaline process waste

water. One method of recovery of NaOH from sulfate waste, involves an electrochemical splitting of sodium sulfate in a membrane cell. In the splitting process, Na⁺ ions from the Na₂SO₄ anolyte migrate into the cathode to combine with OH⁻ that is formed from H₂O reduction. Oxygen is generated on the anode. For the nitrate destruction process, use of acid sodium sulfate anolyte is attractive because of the possible formation of a more concentrated alkaline solution in the catholyte. High alkaline concentration does not favor hydrogen gas generation - an inefficient use of current in the nitrate cell. The formation of NaOH is favored by the migration of sodium ions from the anolyte. The alternative use of pure sulfuric acid as anolyte promotes the formation of a very dilute NaOH on the catholyte side, since the migrating protons from the anode side will mostly be used in the neutralization of the hydroxide ions formed in the catholyte. This favors hydrogen gas generation. Thus, in the proposed use of an acid sodium sulfate anolyte for the nitrate reduction process, the current efficiency for the formation of sodium hydroxide in the catholyte will depend on the ratio of protons [H⁺] to the sodium ion [Na⁺] in the anolyte. Because the effective transport number of Na⁺ ions and H⁺ ions in Nafion is about 2:1⁵, Na⁺ ions will be transported into the catholyte from the anolyte more favorably.

Although hydrogen consuming anode can be employed in an alkaline environment, the commercially available acid based hydrogen gas diffusion electrodes show superior performance over those used in alkaline solutions. This is a significant reason in this work for the evaluation of both acid and alkaline based hydrogen anodes. Alkaline based hydrogen anode is justified on the basis of cost since the catholyte (alkaline) product could, after purification, be recycled to the anode side and thus eliminate the use of an all fresh NaOH anolyte. On the other hand, use of sulfuric acid or sodium sulfate/sulfuric acid mixture as an anolyte could involve an added cost for the nitrate processing. However, since sodium sulfate is a readily available waste, and its dumping into the environment is under strict control, it could be readily obtained at a minimal cost.

The purpose of this paper is to report the preliminary results of the evaluation of a nitrate cell using an hydrogen consuming anode in both alkaline and acid media. We intend to

demonstrate the feasibility of an acid based anolyte in the nitrate destruction cell. The use of a mixture of sodium sulfate and sulfuric acid (mixed anolyte) in the nitrate cell has not been demonstrated before.

Experimental Apparatus and Procedures

A bench-scale multi-purpose continuous flow electrochemical reactor capable of being configured either in a parallel plate electrode mode or gas diffusion electrode mode was used. The reactor was manufactured by Electrocell Co. (MPCell, Akersberga, Sweden). The electrode-to-separator gap (ion exchange NAFION[®] membrane) and the cross-section area are 0.7 cm and 7.0 cm², respectively. Each electrode has a macroscopic active surface area of 100 cm². In our present work, a monopole nickel electrode 3.0 mm thick (Alrich Chemical Co., Inc., Milwaukee, Wisconsin) was used as the cathode in all the runs. Run as a parallel plate cell, nickel electrode was used as the anode. In the gas diffusion mode, the anode consisted of an hydrogen consuming anode supplied by E-Tek Inc. (Cleveland, OH) for the acid based anolyte. An Alupower (New Jersey) supplied gas diffusion electrode was used in the alkaline anolyte. It is believed that platinum is the catalyst for both electrodes.

A schematic of the equipment used is presented in Fig. 1. The flow circuit includes recycled anolyte and catholyte loops. A metered flow of the catholyte was recycled through the cathode from a 5.0 liter tank. The reactor was not cooled. A water cooled condenser was mounted at the top of each tank to condense vapor carried over by the disengaged gaseous products from the reactor. Gilmont flow meters, with manually controlled valves were used in measuring the anolyte and catholyte flows. The catholyte flow rate was kept at a constant rate of 3.8 liters/min. A dc power supply with a maximum power output of 1.8 kVA and capable of either voltage or current controls up to 60.0 volts or 30 amperes was used. All parts in contact with the electrolytes were made of polypropylene. Laboratory grade chemicals were used in deionized water. Hydrogen was of greater than 99.9% purity. In the diffusion anodes, the hydrogen supply into the anode was maintained at a constant flow rate of 30 cm³/min. at an overpressure of 375 cm H₂O.

The major independent variables were the superficial operating current densities (100 - 250 mA/cm²) and anolyte type (H₂SO₄, Na₂SO₄, NaOH). The concentration of pure sulfuric acid anolyte was between 1.5 - 1.6 M. For a mixed anolyte, H₂SO₄ and Na₂SO₄ concentration was 0.95 M and 1.6 M, respectively. For a pure NaOH anolyte, the anolyte concentration varied between 1.6 - 6.1 M. In all the experiments in which sodium sulfate was used as an anolyte, changes in the anolyte Na⁺ ion concentration with time were followed. The catholyte and anolyte were charged with known concentrations of their respective components (NaOH, NaNO₂ and NaNO₃ for the catholyte and H₂SO₄, or H₂SO₄ + Na₂SO₄ or NaOH for the anolyte). The concentrations of the nitrate and nitrite were determined by ion chromatographic method while atomic absorption spectroscopy was used for the Na⁺ ions. The OH⁻ and H⁺ were determined by titration. The cell voltage and applied current was measured by a voltmeter and an ammeter, respectively.

Results and Discussion

The nitrate and nitrite species are considered non-benign in this process. The desired objective in the electroprocessing of nitrate/nitrite solution is to totally reduce the nitrates and nitrites to nitrogen with a minimal formation of ammonia. Thus, a cell's performance could be expressed in terms of the fraction of unreacted nitrate/nitrite species. Equations [1] - [5] show that the reduction of these non-benign species is a consecutive reaction process. In a membrane process, as this, water is transported across the membrane by electrosmotic and concentration differences. This results in variable volume processing. To account for this effect, use is made of the concept of 'fraction of unreacted species per unit volume change', β_{pu} (see Appendix). The concept was used as a criteria to account for the performance of a membrane cell in which electrosmotic water transport and consecutive species reaction occur. This criteria is useful when either the initial or final volume of the electrolyte in the recirculation tank is not known. In this work, the composition and identity of the evolved gases on the cathode were not measured or determined.

For the proposed hydrogen gas consuming anode, oxygen evolution, reaction [6], will be replaced by hydrogen oxidation:



Reaction [7] occurs both in pure sulfuric acid or mixed anolyte of sodium sulfate and sulfuric acid. However, in a mixed anolyte, Na^+ and H^+ ions will be available for transport across the membrane as current carriers. The overall reaction for a Na_2SO_4 anolyte is therefore represented by:



This in effect translates to the splitting of sodium sulfate into sulfuric acid (anode side) and sodium hydroxide (cathode side). Irrespective of the anolyte used, reactions [1] - [5] will take place on the cathode.

Figure 2 shows the fraction of unreacted non-benign (nitrate and nitrite) species per unit volume change as a function of run time in NaOH and acid based hydrogen consuming anodes. The current density is the superficial current density based on the macroscopic surface area of the electrode. In the figure, the overall combined concentrations of both nitrate and nitrite decreased at all times. This was not the case when the individual species are plotted as a function of time. Whereas the nitrate species concentration steadily decreased with time of run, the concentration of the nitrite rises and falls. The increase in nitrite ion suggests that initially, the high concentration of nitrate favors its conversion to nitrite. As the nitrite level increased, the subsequent conversion of nitrite to either ammonia or nitrogen is favored. The result in Figure 2 suggests that hydrogen anode performance in acid anolyte is slightly better than that in alkaline. This advantage in acid is very negligible. The main impetus for recommending the use of an acid based anolyte is for the better control of hydrogen oxidation in acid media with commercially available gas diffusion electrodes.

The interaction between the concentration of the acid anolyte and the alkaline catholyte and its effect on cell performance is given in Figure 3. With a hydrogen gas consuming

anode, hydrogen is oxidized on the anode and the H_3O^+ formed in the process is transported across the membrane to the catholyte side, where it interacts with OH^- ions to form water. Since there is competition between the reduction of water and the non-benign species, the fraction of unreacted non-benign species will be related to the $[\text{H}^+]/[\text{OH}^-]$ ratio where the H^+ and OH^- ions are measured in the anolyte and catholyte recirculating tanks, respectively. In Figure 3, β_{pu} is plotted against the $[\text{H}^+]/[\text{OH}^-]$ ratio. From the figure, it is observed that the ratio decreases with the current density suggesting that the production of H^+ was not keeping up with the production of OH^- . The $[\text{H}^+]/[\text{OH}^-]$ ratio is important because the $[\text{H}^+]$ generation rate in the anolyte and its migration into the catholyte for subsequent neutralization of hydroxide ions is controlled by the applied current density. At higher current densities, the high generation rate is offset by the high migration rate into the catholyte which leads to low hydroxide ion concentration. On the other hand, at 150 mA/cm^2 the rate of H^+ ion generation and migration is low. Similarly, the hydroxide ion generation rate is also low. Also, a plot of the nitrate/nitrite concentration against the $[\text{H}^+]/[\text{OH}^-]$ shows that the nitrate level increases with an increase in the $[\text{H}^+]/[\text{OH}^-]$ ratio.

A comparison is made of the effects of sodium sulfate in the anolyte in Figures 4-6. As a result of the hydrogen ion migration from the anolyte to the catholyte, the use of sulfuric acid as an anolyte limits the concentration level of the OH^- ions generated in the cathode. When sulfuric acid is used as the anolyte, protons are the main current carrier across the membrane. The transported protons into the catholyte neutralizes the OH^- ions. On the other hand, in a $\text{Na}_2\text{SO}_4/\text{H}_2\text{SO}_4$ anolyte, protons and Na^+ ions compete as current carriers. Figure 4 compares the use of pure sulfuric acid and a mixture of sulfuric acid and sodium sulfate. The x-axis in Figure 4 represents the total energy required to reduce unit mass of combined nitrate and nitrite at 50% conversion. Relatively, the presence of sodium sulfate seems to favor the reduction process. The reason for this is not quite clear, except that with proton transport occurring in pure acid, a cyclical process of OH^- generation and neutralization could be occurring in the catholyte. This inefficient use of applied current means less destruction of non-benign species. This is not the case when sodium ion is part of the current carrier.

The effect of sodium sulfate on catholyte side hydroxide ion is shown in Figure 5. With pure sulfuric acid, the hydroxide ion level in the catholyte is limited to under 2.0 M after 10 hours of run. However, with the use of sodium sulfate anolyte, the hydroxide ion concentration in the catholyte rises faster than that obtained with a sulfuric acid anolyte. The merit of sodium sulfate lies in the higher transport number of Na^+ ions over H^+ in Nafion[®] membrane. The concentration of hydroxide ion in a cell utilizing $\text{H}_2\text{SO}_4/\text{Na}_2\text{SO}_4$ anolyte is dependent on the ratio of $[\text{H}^+]/[\text{Na}^+]$ in the anolyte. This is shown in Figure 6. The curve approaches an optimum $[\text{H}^+]/[\text{Na}^+]$ ratio (~ 2.0) above which the concentration of NaOH seems to level off. If this maximum value is exceeded, the hydroxide ion concentration starts to decrease. This is because the anolyte Na^+ ion has decreased to a level that H^+ transport into the cathode side favors water formation and cyclical OH^- generation and neutralization. Thus, in a practical application, sodium sulfate make-up has to be continuously added in the anolyte to maintain a $[\text{H}^+]/[\text{Na}^+]$ ratio equal to or less than the optimal value. Since the hydroxide ion competes with the nitrite and nitrate ions on the cathode for reaction, the anolyte side $[\text{H}^+]/[\text{Na}^+]$ ratio at which the catholyte hydroxide concentration levels off may be used as a means of controlling the availability, and hence, the reactivity of hydroxide ions on the cathode.

The performance of a parallel plate (nickel anode) electrode is compared to that of a hydrogen gas consuming anode at high current density in Figure 7, while the cell voltage for these two systems are compared in Figure 8. As pointed out earlier, at high current density, both high proton migration and hydroxide ion neutralization leads to a cyclical process involving inefficient hydrogen gas generation on the cathode. Thus, the performance of the parallel-plate cell appears better than that obtained with a gas diffusion anode.

The variation of cell voltage with time and current density is shown in Figure 8. Relative to the results obtained in the absence of a gas diffusion electrode, a substantial voltage drop of about 0.4 - 1.8 V at 250 mA/cm^2 is observed. This translates to a substantial

energy savings for the use of an hydrogen gas consuming anode. The energy savings here may not translate into any substantial cost advantages when the costs of H₂ gas and hydrogen electrode are factored in. The high energy consumption of the oxygen generating step at the anode is due to the high overvoltage associated with oxygen evolution. It is conceivable that the required hydrogen in the anode, could be recycled from that produced at the cathode in this process.

Conclusion

The feasibility of an acid based anolyte in the nitrate destruction cell has been demonstrated. Further experiments are required to investigate the interaction of the key parameters (current density, flow rates, Temperature, [H⁺], *etc.*) in the process. Splitting of acidic sodium sulfate is attractive anolyte process for waste minimization. A further incentive for this process is the substantial energy savings that will result with the use of a gas diffusion electrode and possible recycle and treatment of generated ammonia. The use of an acid anolyte is not dependent on the use of a gas diffusion electrode. Thus, both sodium sulfate splitting, and nitrate destruction could be carried out simultaneously with or without the use of a gas diffusion electrode.

Acknowledgments

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APPENDIX

The performance factor, "fraction of unreacted species/volume change", β_{pu} derived below, is a dimensionless quantity and measures the extent the sum total of unwanted species (pollutants) that are unreacted per unit volume change. It is a measure of the cell's "extent of performance for unreacted species" (β_{pu}) at the prevailing operating conditions and is a conservative performance criteria for a cell involving osmotic water transport, when the electrolyte tank volume change is not known. The β_{pu} value of a cell departs from the traditional fraction of unreacted species by the ratio of the initial electrolyte volume to the final volume after reaction (dilution ratio). A simple species balance at the start of a batch processing gives the total amount of species to be destroyed as:

$$F^{\circ} = \sum_{i=1}^n m_i c_i^{\circ} V^{\circ} \quad [A-I]$$

where, F° (gm) is the total mass of the species contained in an initial electrolyte volume, V° (cm^3) in the tank (anode or cathode side) of interest, m_i and c_i° are the formula weight and initial concentration of species i , respectively. At the end of the batch processing time, t , the total electrolyte tank volume of interest changes. The change in this volume could either be an increase (water received) or a decrease (water loss). A species material balance yields,

$$F_t = \sum_{i=1}^n m_i c_i V_t \quad [A-II]$$

where V_t is the electrolyte volume at time, t . Thus, the fractional change, γ (in the amount of unreacted species) is obtained by dividing equation [A-II] by equation [A-I] as,

$$\gamma = \frac{\left(\sum_{i=1}^n m_i c_i\right) V_t}{\left(\sum_{i=1}^n m_i c_i^{\circ}\right) V^{\circ}} = \frac{F_t}{F^{\circ}} \quad [\text{A-III}]$$

If we define β_{pu} as follows,

$$\beta_{pu} = \frac{\left(\sum_{i=1}^n m_i c_i\right)}{\left(\sum_{i=1}^n m_i c_i^{\circ}\right)} \quad [\text{A-IV}]$$

then, equation [A-III] could be re-written as

$$\gamma = \beta_{pu} \frac{V_t}{V^{\circ}} \quad [\text{A-V}]$$

But, the species extent of reaction, ε is defined as

$$\varepsilon = 1 - \gamma \quad [\text{A-VI}]$$

On substituting for γ in [A-VI] using [A-V] and solving for β_{pu} , we obtain

$$\beta_{pu} = (1 - \varepsilon) \frac{V^{\circ}}{V_t} \quad [\text{A-VII}]$$

Thus, β_{pu} will be equal to the traditional "fraction of unreacted species", $(1-\varepsilon)$ when there is no volume change or dilution of initial electrolyte tank volume. Conceptually, β_{pu} is proportional to $(1-\varepsilon)$ where the constant of proportionality is the extent of dilution resulting from water transport. This performance criteria should not be confused with $(1-\varepsilon)$.

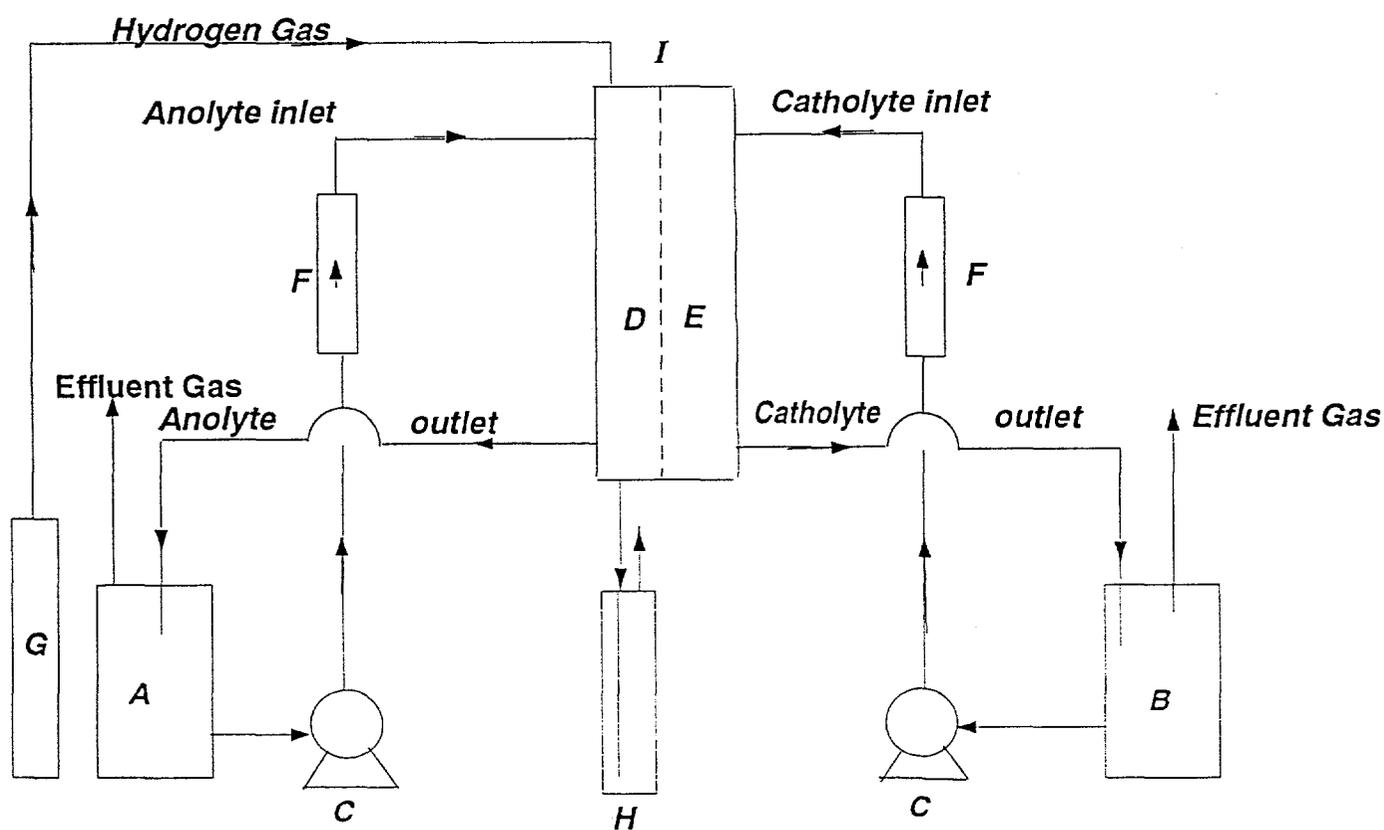


Fig. 1: Schematic of the Nitrate Cell flow-system. A: Anolyte reservoir, B: Catholyte reservoir, C: Pump, D: Anode chamber of MP-Cell, E: Cathode chamber of MP-Cell, F: Flow meter, G: Hydrogen gas tank, H: Water column for hydrogen overpressure, I: MP-Cell.

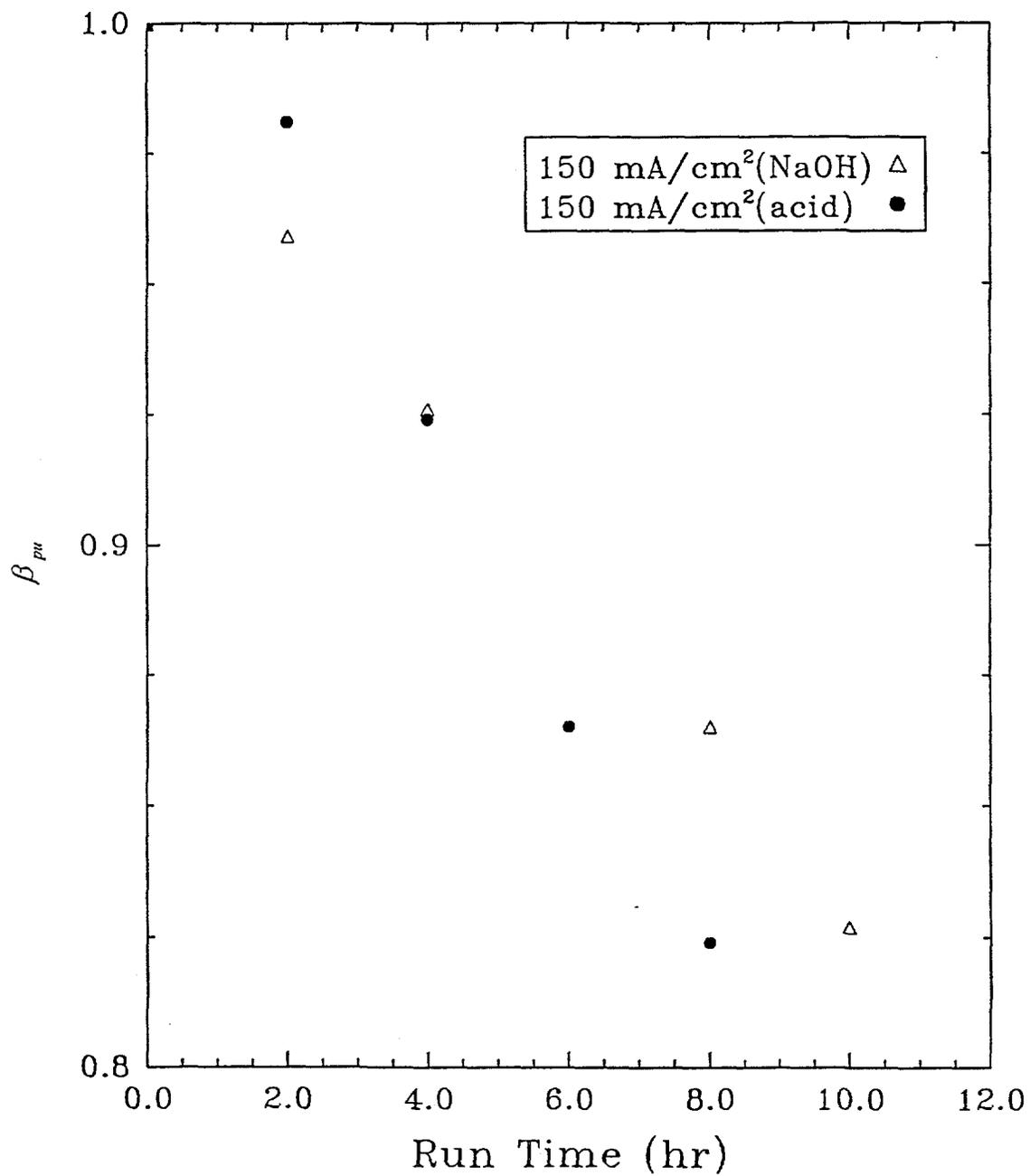


Fig. 2: Performance of hydrogen gas consuming anodes in NaOH and H₂SO₄ anolytes

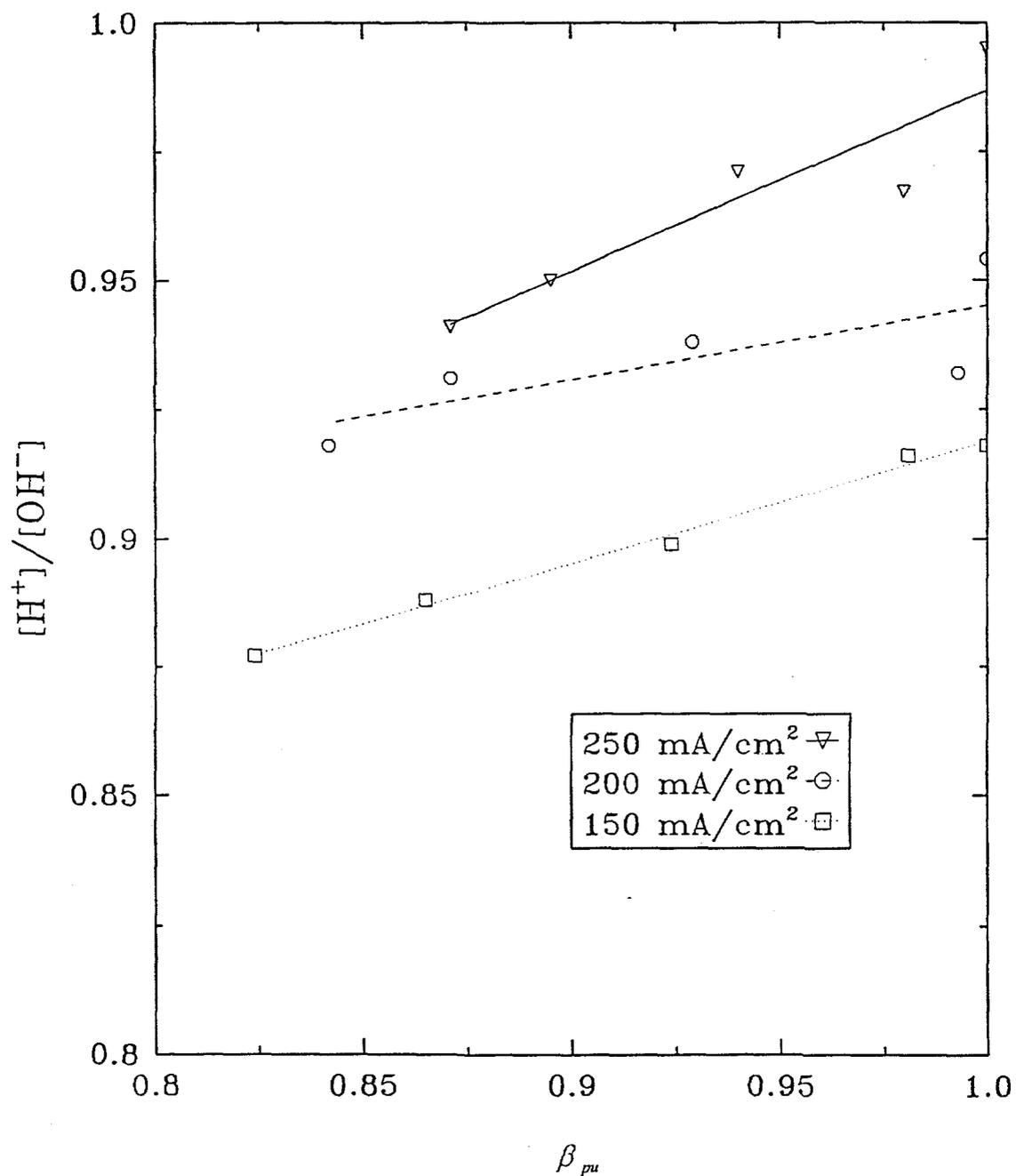


Fig. 3: Ratio of anolyte side $[H^+]$ concentration to catholyte side $[OH^-]$ concentration versus fraction of unreacted nitrate and nitrite species per unit volume change

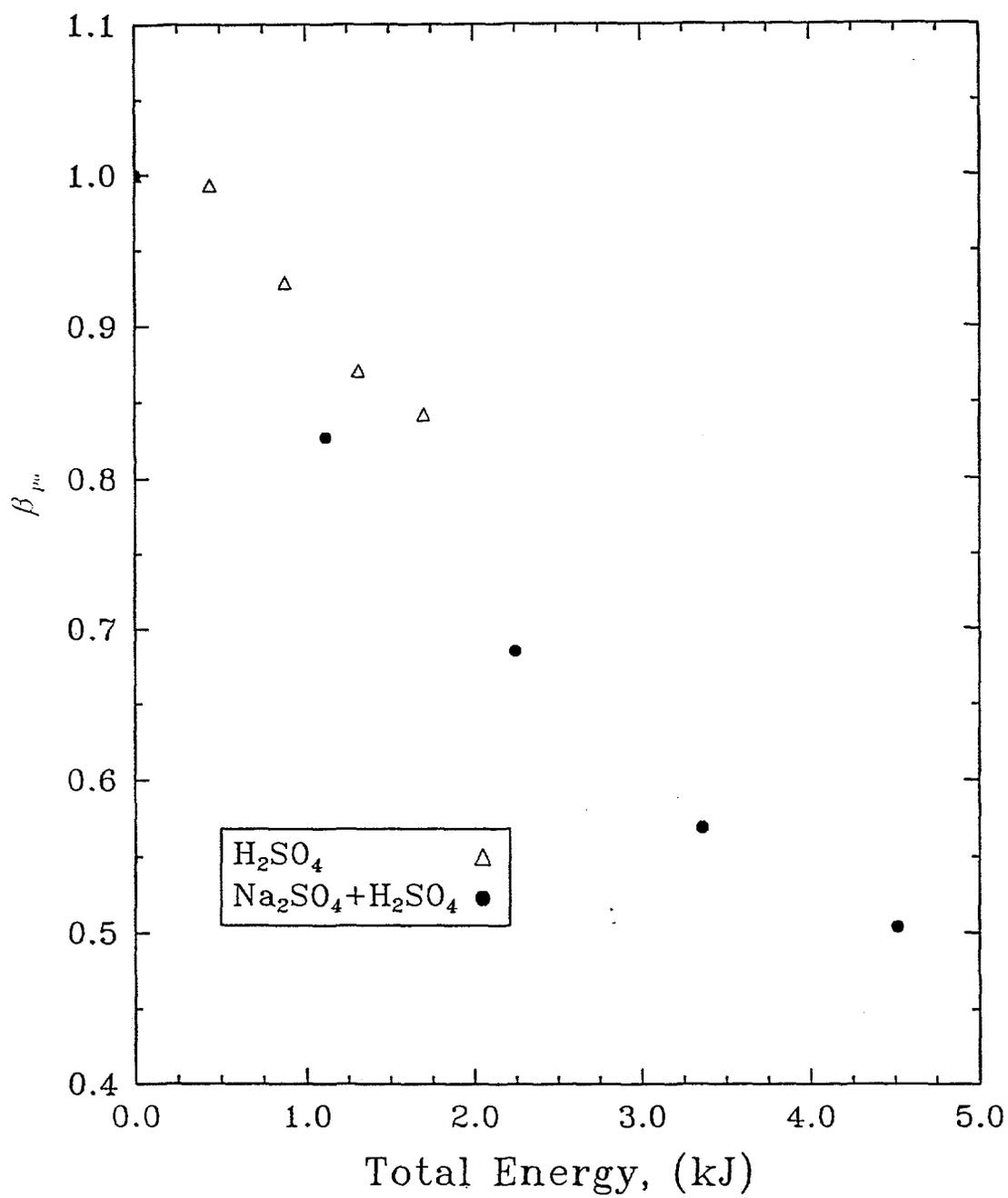


Fig. 4: Change in the fraction of unreacted benign species, β_{pu} , with and without Na_2SO_4 at 200 mA/cm²

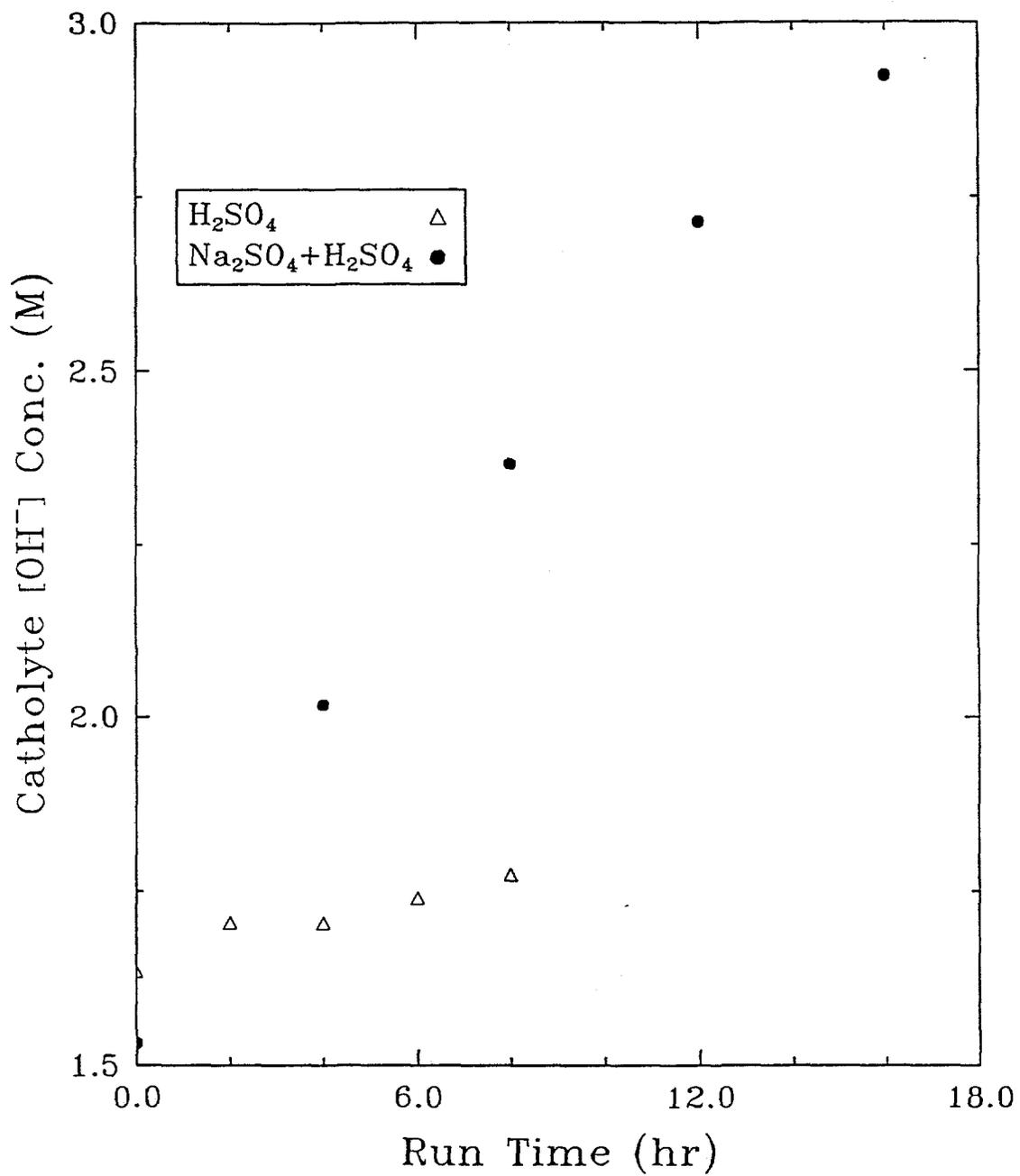


Fig. 5: Catholyte OH⁻ concentration with and without Na₂SO₄ anolyte

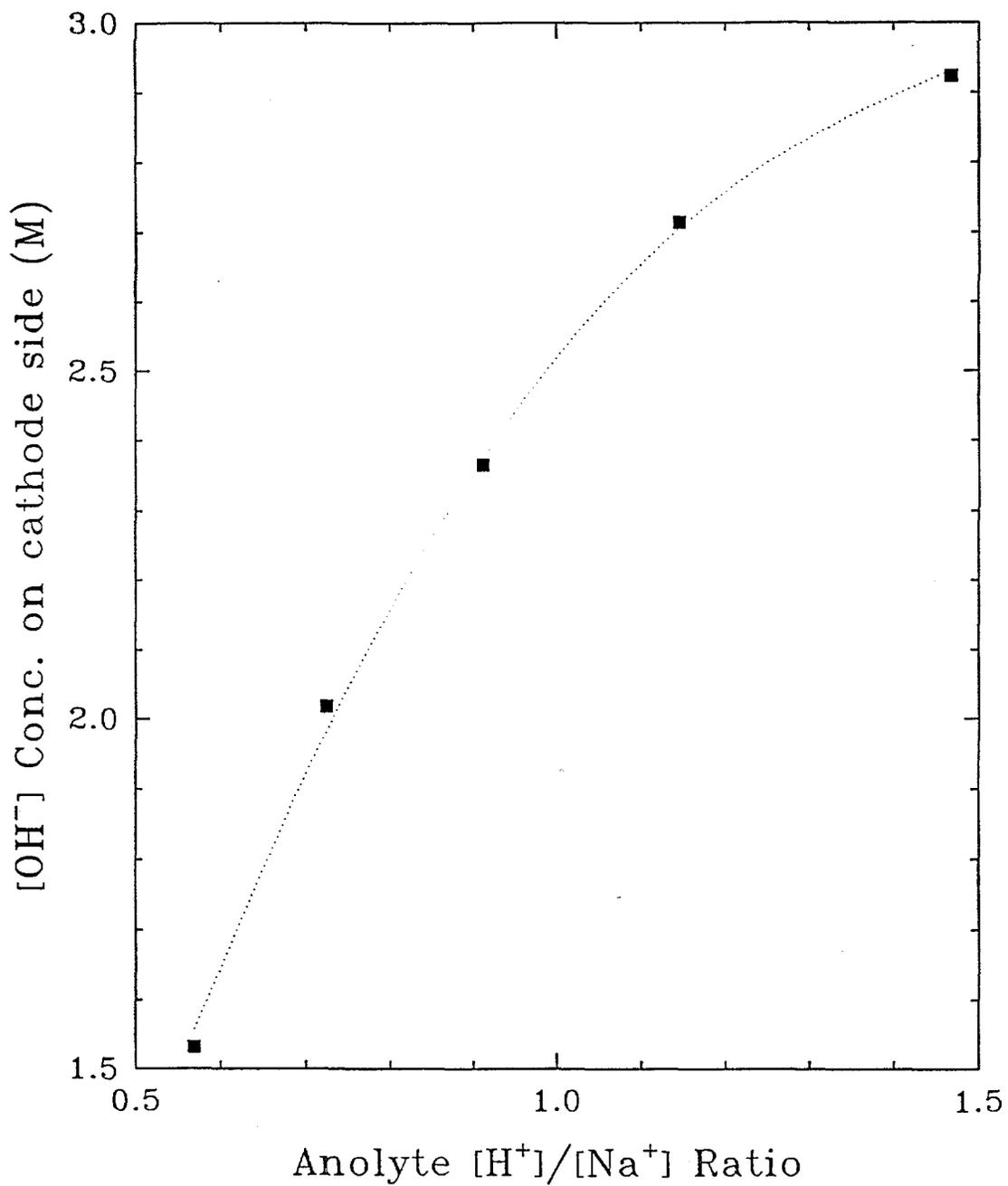


Fig. 6: Catholyte OH⁻ dependence on anolyte [H⁺]/[Na⁺] ratio at 200 mA/cm²ee

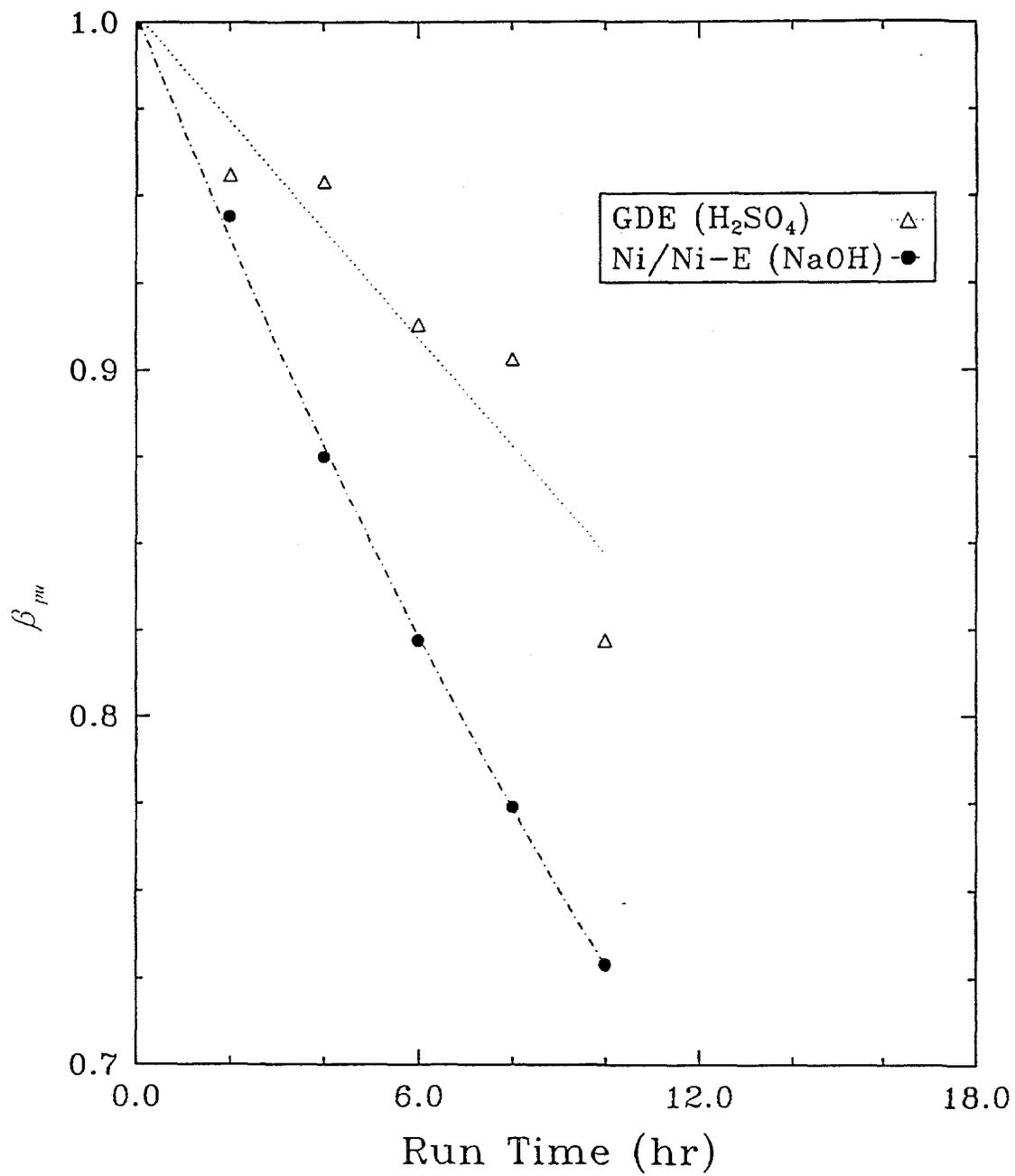


Fig. 7: Cell performance in NaOH anolyte (no GDE) vs Acid anolyte with GDE at 250 mA/cm²

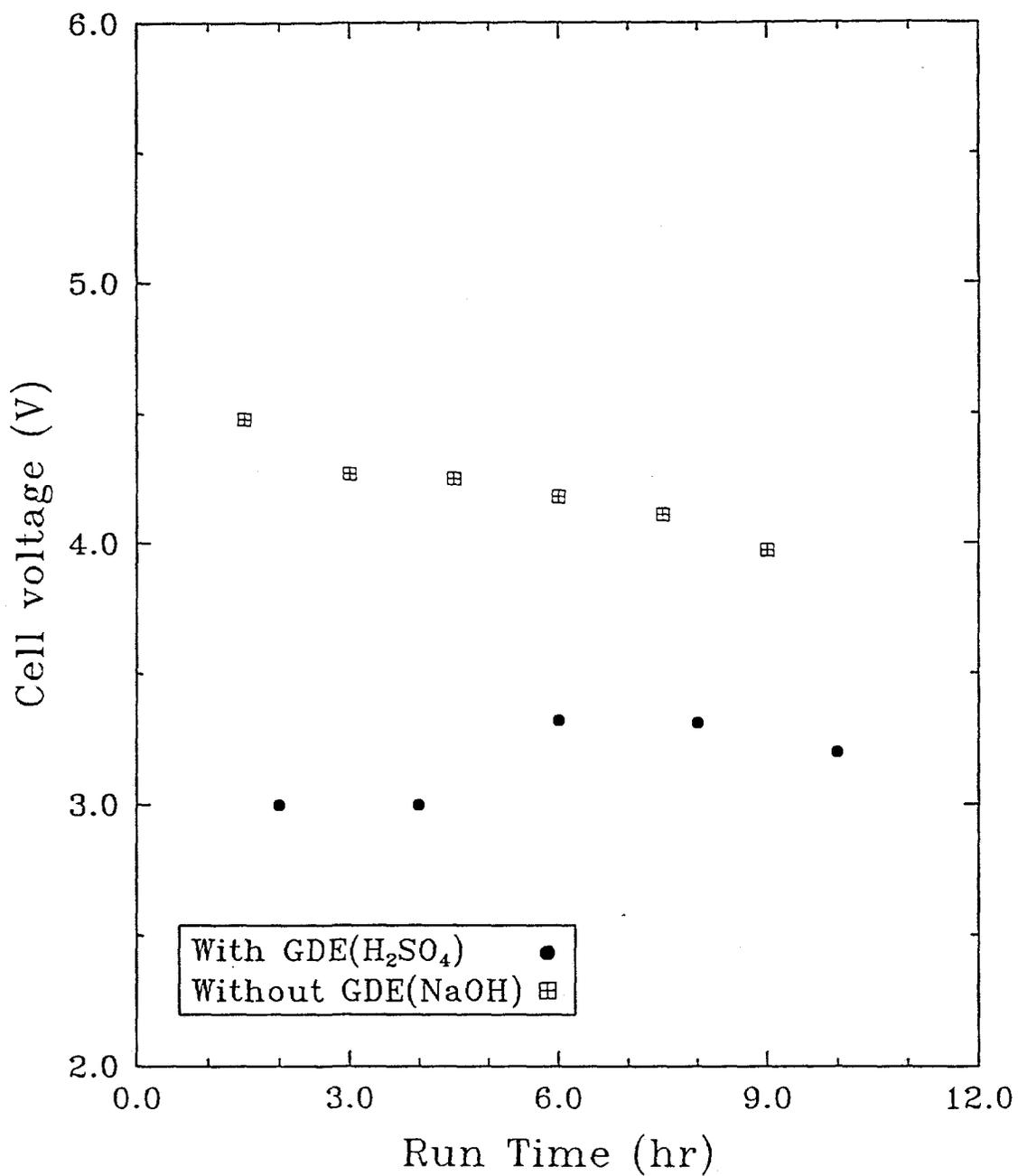


Fig. 8: Variation of cell voltage w/o gas electrode at $250\text{mA}/\text{cm}^2$