Cyanide Degradation by Direct and Indirect Electrochemical Oxidation in Electro-active Support Electrolyte Aqueous Solutions

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Abstract. The cyanide degradation was performed in an electrochemical reactor (with anode of reticulated vitreous carbon and cathodes of graphite) with three compartments separated by a polymeric membrane. The experiments were made under different conditions; supporting electrolytic media (NaOH, NaCl, $NaNO_3$ and Na_2SO_4), applied potentials (3.0, 4.0, 5.0 and 6.0 V) and volumetric flows of 130, 240, 480, 1058 and 1610 mL/min with recirculation. The best conditions for the cyanide degradation are: NaCl (0.1 M) as supporting electrolyte, applied cell potential of 5.0 V and volumetric flow of 480 mL/min. Under these conditions almost 100% of cyanide degradation was achieved.

Keywords: cyanide, electrochemical reactor, electrolyte, reticulated vitreous carbon, polymeric membrane

Resumen. La degradación de cianuro se realizó en un reactor electroquímico (con ánodo de carbono vítreo reticulado y cátodos de grafito), con tres compartimentos separados por una membrana polimérica. Los experimentos se realizaron en condiciones diferentes; medios electrolíticos (NaOH, NaCl, Na_2SO_4 y $NaNO_3$), potenciales aplicados (3.0, 4.0, 5.0 y 6.0 V) y el flujo volumétrico de 130, 240, 480, 1058 y 1610 ml / min con recirculación. Las mejores condiciones para la degradación del cianuro son: electrolito de soporte NaCl (0.1 M), potencial de celda aplicado 5.0 V y un flujo volumétrico de 480 mL / min. Bajo estas condiciones se logró una degradación de cianuro cercano al 100%.

Palabras clave: cianuro, reactor electroquímico, electrolito, carbón vítreo reticulado, membrana polimérica.

Introduction

The cyanide as sodium cyanide and hydrocyanic acid is widely used as reactant in the industry. The main sources of wastes containing cyanide are the electrolytic coating industry and the mining industry. Also the synthetic fiber industry generates a considerable quantity of wastes containing cyanide.

There are many process reported for cyanide wastes treatment, the most used is the chlorination. The cyanide can be partially oxidized to cyanate (CNO^-) or completely oxidized to carbon dioxide (CO_2) and nitrogen (N_2). Cyanide destruction by chlorination is conduced with direct addition of sodium hypochlorite or chlorine gas plus sodium hydroxide into the wastes. In the second process, chlorine reacts with sodium hydroxide forming sodium hypochlorite. The cost of the second treatment is half of the treatment with sodium hypochlorite but the use of chlorine gas is too risky and the cost of equipment is also expensive. The oxidation of cyanide with sodium hypochlorite produces cyanate which is less toxic than cyanide [1], but if is necessary that cyanate can be oxidized to carbon dioxide and nitrogen by additional chlorination or onto carbon dioxide and ammonia by acidic hydrolysis.

Effective alternatives to typical chemical oxidation treatments are the electrochemical methods. These methods have been studied by many authors since 1970´s. Many of these investigations have been aimed to the destruction of free cyanide and on elucidation of reaction mechanism. It has been demonstrated that direct oxidation is slow on platinum [2] and graphite [3], exhibiting low current efficiencies. The best re-

sults on direct oxidation have been reported using nickel anodes [4], electrodes coated with PbO_2 [5]; reticulated electrodes [6, 7] and anodes of fluidized or packed bed [8-13].

Regarding to electrochemical methods for cyanide degradation, Yasuda, Ohkawa and Yasukawa [10] reported the treatment of diluted wastes containing cyanide by using a cell with an electrode of fluidized bed of graphite particles. They investigated the effect of bed height and cell current on the direct oxidation of cyanide. They found that increasing the height of the bed, time and energy consumption can be reduced, yielding a degradation of 99 %. The energy consumption increases almost proportionally with the cell current.

The cyanide oxidation with tetrahydroxoargentate (III) ion in strongly alkaline media was reported by Sun and Kirschenbaum [14]. They reported some kinetic parameters like reaction order, the K_{obs} and the variation of cyanide concentration against electrolysis time. Tissot and Frangniere [5] reported the use of an electrode of reticulated vitreous carbon coated with lead dioxide (RVC-PbO_2) for the cyanide oxidation. They concluded that this electrode is suitable for the cyanide oxidation when is present at low concentration in wastes, reporting an excellent stability of electrode after 120 hours of use. Unappreciable change in electrode weight was observed and only 0.1 ppm of Pb, coming from PbO_2, was found in solution after 120 h.

When the cyanide is complexed with some metals like iron, copper or zinc many problems appear because of high stability of these compounds. Several studies where the decomposition rate of cyanide is improved by the presence of copper in solu-

tion have been reported [8, 15], stating that a layer of copper oxide is formed on the anode surface at long electrolysis time, this oxide layer apparently helps in the oxidation of cyanide. Hine *et al* [4] considered that this copper layer with a blue dark color is composed of cupric oxyhydroxide. Hwang *et al* [16] analyzed the layer formed on the anode during the alkaline electrolysis of cupric cyanide (pH > 12) and they reported that was a mixture of cupric and cuprous oxides. Wels and Johnson [17] experimentally confirmed that this layer of copper has some catalytic proprieties on the cyanide electro-oxidation.

The copper catalyzed the cyanide destruction with a porous electrode of RVC was reported by Hofseth and Chapman [6]. They reached a reduction of cyanide concentration from 100 ppm to 1 ppm, indicating that a strict pH control is necessary to keep high current efficiencies.

Zhou and Chin [18] reported the cyanide destruction and simultaneous recovering of copper using an electrochemical cell with cathode of platinum drum and anode of graphite packed bed. Cyanide destruction took part directly and indirectly by adding NaCl. The total concentration of cyanide was reduced from 580 ppm to less than 10 ppm with an energy consumption of 80 to 340 kWh/kg of cyanide destroyed. The current efficiency was improved by addition of NaCl. The authors concluded that this method is more expensive than conventional alkaline chlorination.

In this study the cyanide degradation by electrochemical oxidation from diluted wastewater was realized using an electrochemical reactor, consisting of a reticulated vitreous carbon anode and two graphite cathodes. Effect of the cell applied potential, the support electrolyte, NaCl concentration and cyanide concentration on the electrooxidation of cyanide was studied.

Results and Discussion

For clarity of presenting the results of this work, the effects of parameters like applied cell potential, the composition of the electrolyte, the concentration of NaCl and the volumetric flow of the anodic solution on the cyanide degradation; are going to be presented and discussed separately for each parameter.

1. Effect of the cell voltage on the electrochemical degradation of cyanide

As shown in Figure 1 (a), cyanide degradation depends on the cell voltage; when the cell voltage is increased from 3.0 V to 5.0 V, the degradation rate of cyanide increases, however the degradation rate is slowed down when the applied potential is 6.0 V. This may be due to the fact that the applied cell potential was too high; this leaded to the oxidation of water at the anode surface. It was observed that oxygen gas was produced, the gas was adsorbed at the anode surface; this would reduce the availability of anodic surface for the direct electro-oxidations of chloride and cyanide. As later experimental work demonstrated the formation of chlorine greatly increased the rate of cyanide
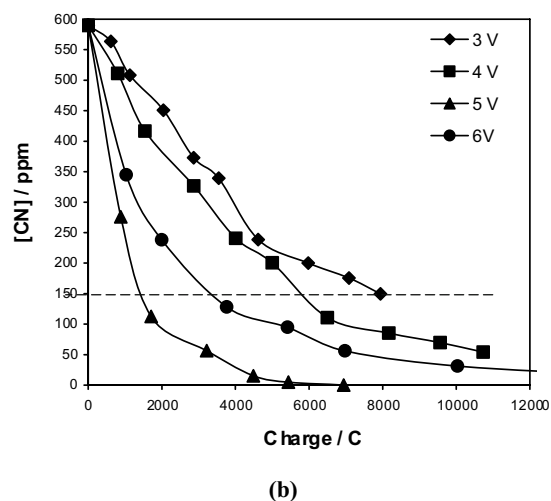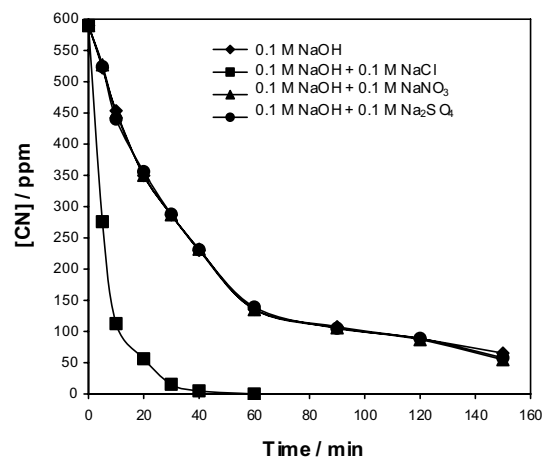

(b)

Fig. 1. Effect of applied cell potential on the cyanide degradation. (a) Cyanide concentration vs time and (b) Cyanide concentration vs charge consumption during electrolysis. Volumetric flow 480 mL/min with recirculation.

degradation, this is the result of indirect oxidation of cyanide by chlorine in the anodic solution.

Results for the cyanide concentration versus charge consumption are presented in Figure 1 (b). The graphic shows a horizontal dotted line at which a 75% of cyanide is degraded. At an applied potential of 5.0 V the charge consumption is 1500 Coulombs to achieve 75% cyanide degradation while at 6.0 V the charge consumption was 3500 C. In the cases of 3.0 V and 4.0 V, the charge consumptions were 8000 C and 6000 C, respectively. Based on this experimental result we decided to carry out the rest of the experiments at applied cell potential of 5.0 volts.

2. Effect of electrolyte type on the electrochemical degradation of cyanide

Figure 2 (a) shows the results of cyanide concentration versus reaction time in various electrolyte systems. With NaCl present in the anodic solution, the velocity of cyanide degradation was relatively fast in comparison with the rest electrolyte systems used for this group of experiments. The degradation rate of cyanide was almost the same as the degradation of cyanide in the presence of NaOH only, when sodium sulfate and sodium nitrate were used. This finding was unexpected because we considered that, when sodium sulfate was used in the anodic solution, the cyanide degradation rate could be fast too, because at this applied cell potential (5.0 volts) persulfate ions are generated by the oxidation of sulfate at the anode surface [19-23]. The presence of electro-generated persulfate in our experiments was confirmed by treating a sample of anodic solution, obtained during the test run, with potassium iodide and a few drops of a 10% w/v starch solution, a blue color appeared; this is the indication of the presence of the oxidizing agent in the anodic solution. However the presence of persulfate did not speed up the cyanide degradation, but the NaCl did. Further evidence for
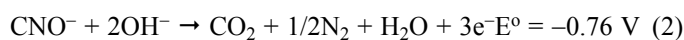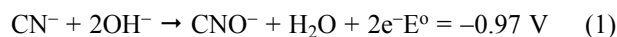
(a)
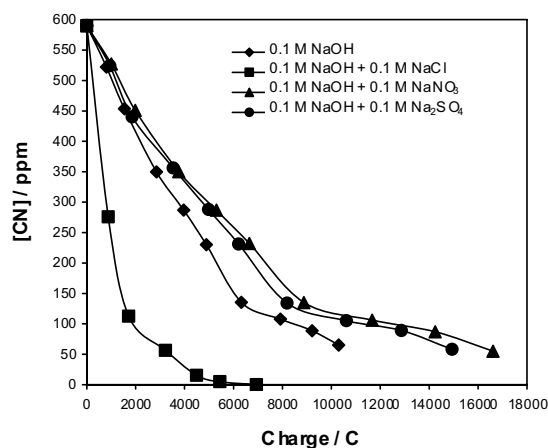
(b)

Fig. 2. Effect of supporting electrolyte on cyanide degradation. (a) Cyanide concentration vs time and (b) Cyanide concentration vs charge consumption during electrolysis. Volumetric flow 480 mL/min with recirculation and applied potential of 5.0 V.
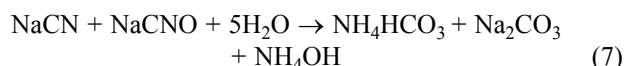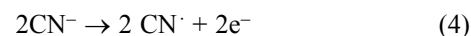
the advantage having NaCl in anodic solution is given in the plot of cyanide concentration in anodic solution versus charge consumption as shown in Figure 2 (b). This Figure reveals that cyanide degradation with NaCl as electrolyte has the lowest charge consumption and in the cases of $NaNO_3$ and Na_2SO_4 the charge consumption is almost the same as for the anodic solution containing NaOH only. Apparently in our work in the absence of NaCl, the cyanide degradation might take place by direct electro-oxidation according the following reactions:

$$CN^- + 2OH^- \rightarrow CNO^- + H_2O + 2e^- E^o = -0.97 \text{ V} \quad (1)$$

$$CNO^- + 2OH^- \rightarrow CO_2 + 1/2N_2 + H_2O + 3e^- E^o = -0.76 \text{ V} \quad (2)$$

Here cyanide is oxidized to cyanate (reaction (1)) and then the cyanate is then oxidized to produce carbon dioxide, nitrogen and water (reaction (2)). Also OH^- ions can be oxidized to give molecular oxygen (reaction (3)).

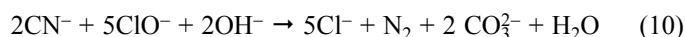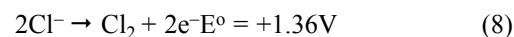$$4OH^- \rightarrow O_2 + 2H_2O + 4e^- E^o = +0.40 \text{ V} \quad (3)$$
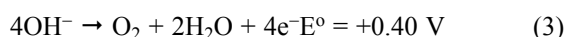
E^o is the standard oxidation potential [24] versus SHE (standard hydrogen electrode at 25 °C). The said reactions shown above were reported by Tissoy and Frangniere [5], Hofseth and Chapman [6], Yasuda and Ohkawa [10] and Brockris [25]. Also Petterson [1] reported the reactions for the direct electrochemical oxidation of cyanide as follows:

$$2CN^- \rightarrow 2 \ CN^{\cdot} + 2e^- \quad (4)$$

$$2CN^{\cdot} \rightarrow (CN)_2 \quad (5)$$

$$(CN)_2 + 2 \ NaOH \rightarrow NaCN + NaCNO + H_2O \quad (6)$$

$$NaCN + NaCNO + 5H_2O \rightarrow NH_4HCO_3 + Na_2CO_3 + NH_4OH \quad (7)$$

Reactions (4) to (6) correspond to the chemical reaction of converting cyanide to cyanate, once the cyanate is formed, the reaction (7) goes very fast [25].

The high degradation rate of cyanide with NaCl as support electrolyte is due to the fact that Cl⁻ ions are oxidized directly on anode surface to produce molecular chlorine (reaction (8)) and subsequently it reacts in the basic solution with OH⁻ ions to generate ClO⁻ ion (reaction (9)) which is an effective oxidizing agent widely used to cyanide degradation [1, 26]. Then the hypochlorite reacts with cyanide in alkaline media to yield gas nitrogen, water and chloride and carbonate ions (reaction (10)). The presence of the oxidizing agent was confirmed by the same way as described for persulfate ions.

$$2Cl^- \rightarrow Cl_2 + 2e^- E^o = +1.36V \quad (8)$$

$$Cl_2 + 2OH^- \rightarrow Cl^- + ClO^- + H_2O \quad (9)$$

$$2CN^- + 5ClO^- + 2OH^- \rightarrow 5Cl^- + N_2 + 2 \ CO_3^{2-} + H_2O \quad (10)$$

The advantage of having ClO⁻ in the anodic solution rather than $S_2O_8^{2-}$ may come from its contribution to form [O•] from the dissociation of ClO⁻ ; this free radical is very reactive, it combines with CN⁻ to form CNO⁻, which as shown in reaction (7), reacts very fast with cyanide. This may be the reason for the degradation of cyanide in the electrolyte system contains NaCl.

3. Effect of NaCl concentration on electrochemical degradation of cyanide

As shown in Figure 3 (a), the presence of chloride in the anodic solution greatly increased the rate of cyanide degradation. In the absence of NaCl in the anodic solution (only NaOH 0.10 M), degradation of 75% cyanide concentration required 60 min and the charge consumption was 6000 C (Figure 3(b)). Addition of NaCl as low as 0.02 M to the anodic solution, resulted in the fact that the time required for 75% cyanide degradation was reduced to 48 min and the charge consumption was 5800 C. When NaCl concentration was increased to 0.05 M, the time
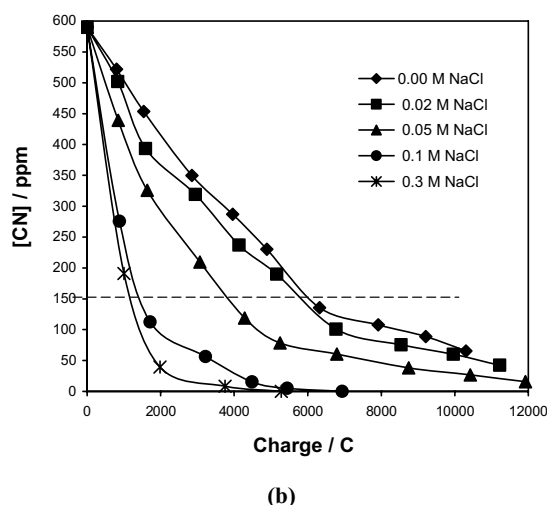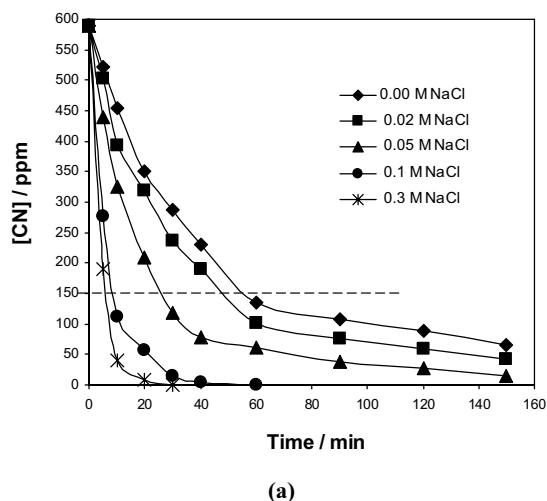
(a)

(b)

Fig. 3. Effect of NaCl concentration on cyanide degradation. (a) Cyanide concentration vs time and (b) Cyanide concentration vs charge consumption during electrolysis. Volumetric flow 480 mL/min with recirculation and applied potential of 5.0 V.

required for the same degradation of cyanide was shortened to 25 min and the charge consumption was 3750 C . Increasing the concentration of NaCl to 0.10 M, it tooks only 10 min to achieve 75% degradation of cyanide with a charge consumption of 1300 C, further increase of NaCl concentration to 0.30 M, resulted in 5 min time to degrade 75% of cyanide from the anodic solution with a charge consumption of only 1000 C.

4. Effect of anodic volumetric flow on the electrochemical degradation of cyanide

As shown in Figure 4, increasing the volumetric flow from 130 mL/min to 480 mL/min, the degradation of cyanide was increased. Further increase of volumetric flows (1058 mL/min and 1610 mL/min), resulted in less cyanide degradation than that at a flow rate of 480 mL/min, at 20 min of reaction time. This may due to the fact that for the first three volumetric flows, the velocity of the solution coming into the anode compartment was adequate to enter into RVC pores and to remove oxygen

bubbles formed on its surface, improving the area available for the electrode surface reaction. However, in cases of 1058 mL/min and 1610 mL/min flows a turbulent pattern may form, affecting the solution passage through the RVC pores and not removing all the oxygen gas formed, thus resulting in a reduced area of anode surface for electro-oxidation of cyanide. At 40 min of reaction time, the effect of volumetric flow of the anodic solution on the removal of cyanide and on the energy consumption is shown in Figure 5. The cyanide degradation was ~100, 98 and 86% at anodic volumetric flows of 480, 1058 and 1610 mL/min, respectively. The energy was observed to be the lowest when the volumetric flow was 480 mL/min (5400 C) and highest at a volumetric flow of 1610 mL/min (7000 C).

Conclusions

Our experimental results clearly indicate that the type of electro-active supporting electrolyte plays an important role in the cyanide degradation; among the three electrolytes used in this
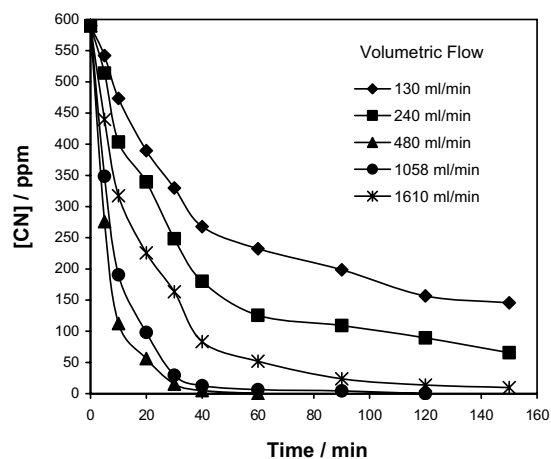

Fig. 4. Cyanide concentration vs time and volumetric flow with recirculation. Applied potential 5.0 V.
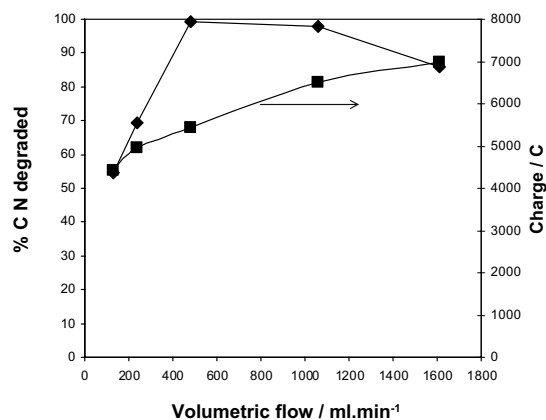

Fig. 5. Percentage of cyanide degradation and charge consumption vs anodic volumetric flow with recirculation. Applied potential of 5.0 V. Reaction time of 40 minutes.

work (NaCl, NaNO$_3$ and Na$_2$SO$_4$), the sodium chloride yielded the highest percentage of cyanide degradation. It is evident that NaCl is oxidized to produce hypochlorite which is a strong oxidant agent; it oxidized cyanide present in the anodic solution. We believe that part of cyanide is degraded by direct oxidation on anode surface and the rest of the cyanide is degraded indirectly in solution by chemical reaction with the oxidizing agent generated from NaCl. The indirect electro-oxidation of cyanide may involve the free radical reaction of [O•], generated by dissociation of ClO$^-$ and its reaction with CN$^-$ to form CNO$^-$.

The sodium chloride concentration and the applied cell potential are the other two very important parameters that affect the rate of electrochemical oxidation of cyanide. We achieved higher degradation rates for cyanide by increasing the sodium chloride concentration to 0.3 M and by applying a cell potential of 5.0 V.

Our experimental results also reveal that the presence of persulfate in the anodic solution did not affect the rate of cyanide degradation appreciably. This suggests that when sodium sulfate and also sodium nitrate and sodium hydroxide are used, the cyanide degradation takes place only by direct electro-oxidation on anode surface. This lead us to conclude that direct electrochemical oxidation is slower than indirect electrochemical oxidation for cyanide.

Experimental

Reagent grade sodium cyanide, sodium hydroxide, sodium chloride, sodium nitrate and sodium sulfate were purchased from Aldrich. All solutions were prepared with deionized water obtained from a SYBON/Barnstead model 02610 water purification system. The pH of the solutions was kept between 11 and 13 by adding 0.1 M NaOH to avoid the release of HCN (pK$_a$ 9.2).

The experiments were performed in a three compartments electrochemical cell as shown in Figure 6. The cell was fabricated with five acrylic blocks of 100 mm long × 75 mm wide × 75 mm thickness, three blocks were machined to form the channel for the electrolytes flows; one of them was for anodic compartment and on its centre a reticulated vitreous carbon anode of 25 mm long × 25 mm wide × 21 mm thickness was placed. For the electrical contacts a graphite rod was joined to the anode with a conductive epoxy resin (provided by Epo-Tech). In each cathodic compartment a graphite plate of 25 mm long × 25 mm wide × 10 mm thickness was placed.

To separate the anodic compartment from the cathodic compartments a microporous polysulfone membrane was used in between of them. The polysulfone membrane was fabricated in our laboratory by the phase inversion method [27]; the membrane casting solution prepared with 20% p/v polysulfone and 80% p/v N-metyl-2-pyrrolidone (NMP) as solvent. The features of this membrane were: 6.5 × 10^{-3} inches of total thickness, 11.76 cm^3/min of volumetric cross-flow and a A-value of 332.87 × 10^{-5} g/(cm^2 · s · atm) at an applied cross-flow
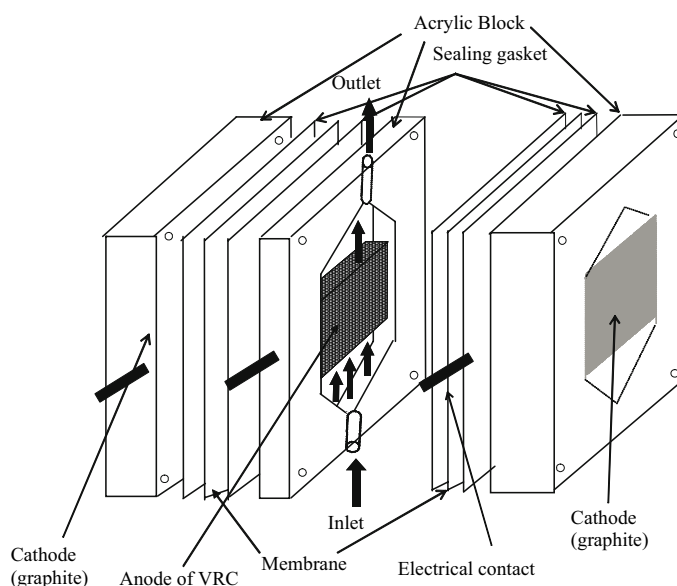

Fig. 6. Diagram of the electrochemical reactor used for the experiments of cyanide degradation.

filtration pressure of 40 psig, at 25 °C and using distilled water as the feed.

All experiments were conducted at room temperature, which fluctuated from 25-28 ºC and the applied electrical potential was kept constant using a model 6038 A Hewlett-Packard power supply. The current was monitored at range of 0-10 ampere.

Samples of 5 mL of anodic solution were taken from the electrochemical reactor at each time interval and the cyanide concentration was measured with a CN$^-$ ion selective electrode from Orion Company. Details for experiments to study the effect of changing parameters like applied cell potential, composition of the electrolyte, concentration of NaCl and volumetric flow, on the cyanide degradation are given below.

One of the most important parameters in electrolysis is the cell voltage. The applied potentials were in a range of 3.0V to 6.0V and it was increased stepwise as indicated in each experiment. The conditions for this series of experiments were: anodic compartment was fed with 1000 mL of solution containing 0.023 M NaCN, 0.100 M NaOH and 0.100 M NaCl, the solution was recirculated at 480 mL/min, the cathodic compartment was fed with a similar solution at a recirculating volumetric flow of 10 mL/min.

Some of the experiments were carried out with three different electrolytes; NaCl, NaNO$_3$ and NaSO$_4$. The anodic solution contained 0.023 M NaCN and 0.100 M NaOH plus the respective electro-active electrolyte at 0.1 M in concentration. The applied cell potential was always at 5.0 V. The solution was fed through the anodic compartment at recirculating volumetric flow of 480 mL/min and in the cathodic compartment was at 10 mL/min.

Several experiments were carried out with NaCl concentration ranged from 0.0 to 0.3 M. The conditions for these experiments were: in the anodic compartment, solution con-

tained NaCN 0.023 M, NaOH 0.100 M and NaCl (at different concentrations) and the cathodic compartment contained NaCl at same concentration of the anodic solution. The applied potential for the cell was kept constant at 5.0 V and the solution was recirculated at 480 mL/min in the anodic compartment and at 10 mL/min in the cathodic compartment.

Some experiments were carried at different volumetric flow through the anode compartment. The volumetric flows used for these experiments were: 130, 240, 480, 1058 and 1610 mL/min. The conditions for all experiments were: the solution for anodic compartment contained NaCN 0.023 M, NaOH 0.100 M and NaCl 0.100 M. The solution for cathodic compartment contained similar solution with recirculation at 10 mL/min. The applied potential was 5.0 V.

Acknowledgements

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