Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

Low Effluent Processing in the Pulp and Paper Industry: Electrodialysis for Continuous Selective Chloride Removal

Peter H. Pfromm[a]
[a] INSTITUTE OF PAPER SCIENCE AND TECHNOLOGY ATLANTA, GEORGIA, USA

To cite this Article Pfromm, Peter H.(1997) 'Low Effluent Processing in the Pulp and Paper Industry: Electrodialysis for Continuous Selective Chloride Removal', Separation Science and Technology, 32: 18, 2913 — 2926
To link to this Article: DOI: 10.1080/01496399708000787
URL: http://dx.doi.org/10.1080/01496399708000787

SEPARATION SCIENCE AND TECHNOLOGY, **32**(18), pp. 2913–2926, 1997

Low Effluent Processing in the Pulp and Paper Industry: Electrodialysis for Continuous Selective Chloride Removal

PETER H. PFROMM
INSTITUTE OF PAPER SCIENCE AND TECHNOLOGY
ATLANTA, GEORGIA 30318-5794, USA

ABSTRACT

Pollution prevention is currently a major focus of the United States pulp and paper industry. Significant process changes are inevitable to implement low effluent processing. The kraft pulping process is prevalent for the production of wood pulp. About 50 million tons of wood pulp are produced annually in the United States alone using the kraft process. Water consumption is currently roughly between 30 and 200 m^3 of water per ton of air dry bleached kraft pulp. In-process recycling of water is now being implemented by many mills to reduce the use of increasingly scarce water resources and to reduce the need for wastewater treatment. Mass balance considerations and industrial experience show that nonprocess elements, which are detrimental to the kraft process, such as chloride and potassium, will quickly build up once water use is significantly reduced. High concentrations of chloride and potassium can cause corrosion and lead to more frequent mill shutdowns due to fouling of heat exchanger surfaces in the kraft recovery furnace. Electrodialysis with monovalent selective anion and cation exchange membranes was explored here to selectively remove chlorine as sodium and potassium chloride from a feed stream with very high ionic strength. Experiments with model solutions and extended tests with the actual pulp mill materials were performed. Very good selectivities and current efficiencies were observed for chloride over sulfate. The outstanding performance of the process with actual mill materials containing organic and inorganic contamination shows great promise for rapid transfer to the pilot scale. This work is an example of the usefulness of membrane separations as a kidney in low effluent industrial processing.

Key Words. Electrodialysis; Monovalent selective membranes; Pulp and paper; Kraft process; Pollution prevention; Capital and operating costs

INTRODUCTION AND BACKGROUND

The theory and practice of the kraft pulping process are described in detail elsewhere (1). Wood is exposed to an aqueous solution of sodium sulfide and sodium hydroxide at elevated temperature and pressure in a batch or continuous reactor. Thereby the lignin is solubilized and the cellulose fibers are liberated. The solution of lignin and spent inorganic chemicals is then separated from the cellulose fibers. This solution is concentrated by evaporation and used as fuel in the kraft recovery furnace. In this furnace the lignin is burned to produce steam for the mill, and the inorganic chemicals are recovered as a molten salt mixture of sodium carbonate and sodium sulfide. This salt mixture is then dissolved in water and reprocessed to recover and reuse the active pulping chemicals.

One of the challenges for the pulp and paper industry is low effluent processing to minimize environmental impact and water use. A serious impediment to low effluent processing is the enrichment of certain elements, termed nonprocess elements (NPEs), in the process fluids when recycling of water is implemented. The term NPE loosely describes materials which are not needed in the process, but which are introduced through raw materials and replacement chemicals. The NPEs chlorine and potassium are attracting much attention due to their negative impact on the kraft pulping cycle. The good solubility of chloride and potassium ions in alkaline solutions is the cause of their accumulation.

Corrosion increases with the chloride concentration in kraft pulping fluids (2). Chloride and potassium also significantly increase shutdowns of kraft recovery furnaces by fouling of heat exchanger surfaces with sticky deposits (3, 4). The kraft recovery furnace is crucial to pulp mill operation. It produces most of the steam and energy used in a pulp mill by burning the unwanted organic wood components. Shutdowns of the recovery furnace cause costly pulp mill outages.

Chloride contributes to the inorganic deadload in the kraft pulping process. This can reduce the actual firing capacity of a recovery furnace for organic material if the furnace permit is written for overall solids fired. This can limit mill production. Even fractions of a percent of capacity decrease are economically very significant in a commodity industry such as pulp and paper.

Input of chloride into the pulping process can be minimized, but not eliminated, due to its presence in wood (5, 6). Chloride is highly soluble under the conditions encountered in kraft pulping and bleaching. If low effluent operation is pursued, tightening of the liquor cycles will lead to increasing concentrations of the soluble NPEs. A highly specific purge is therefore needed for chloride (7, 8).

The introduction of bleach plant effluents into the kraft recovery cycle is widely regarded as a way to achieve low effluent bleached kraft pulp production. The basic idea is to burn unwanted organics in the recovery furnace instead of discharging them with water. However, inorganic matter is recycled together with the organic material. This further emphasizes the need for a specific purge of soluble inorganics (9, 10). This is especially true if chlorine is present in some form in the bleaching chemicals (11).

A significant number of processes for removal of chloride from the kraft pulping process are described in the literature and in patents. The good solubility of chloride in kraft pulping liquors makes it difficult to purge without loss of large amounts of valuable chemicals that are also present. Purging of chloride as hydrochloric acid with the kraft recovery furnace flue gas has been considered, but does not appear to be a desirable long-term option because this would constitute only a shift of the emission problem. An economical chloride purge could be achieved by a highly selective removal process, or by discarding part or all of a chloride-rich process stream.

Capital investment, operating cost, maintenance, ease of operation, environmental impact, and the regulatory situation will all impact the choice of an optimum solution. The best choice will certainly vary from mill to mill.

An advantageous point for a selective chloride purge is the electrostatic precipitator that removes dust from the flue gas of the kraft recovery furnace. Several weight % of the total fired solids in the furnace are recovered as electrostatic precipitator dust (ESP dust) from the flue gas. This dust is then recycled to the process. It has been found that chloride and potassium are enriched in the ESP dust compared with their relative concentrations in the main process stream (black liquor) (4). This makes ESP dust a good candidate for a chloride purge process.

Differential solubility in water is clearly the selective chloride removal mechanism that has attracted the most interest in the past (8, 12). Excellent selectivity is inherent in crystallization processes, but a subsequent liquid/solid separation is necessary. This will cause some carryover of chloride-containing liquid. The chloride purge stream, if dissolved ESP dust is treated, will contain organic materials. These organics stem from incompletely combusted organics in the fuel (black liquor). The capital investment and maintenance for a crystallization/filtration process are not insignificant. Crystallizer performance, handling of two-phase systems, and startup/shutdown procedures may be other concerns.

The selective electrodialytical removal of chloride from dissolved ESP dust with chloride selective membranes is reported here. Similar technology involving a leaching step and subsequent electrodialysis, or a multistep pretreatment process for metal ion removal has been reported (13, 14). The work reported here differs in that no leaching or pretreatment of ESP dust was needed for successful long-term chloride separation experiments with actual mill materials. A schematic diagram of the process is shown in Fig. 1. The electrodialysis diluate (chloride-depleted sulfate solution) is recycled to the black liquor, where the water that was added for dust dissolution can be evaporated in the existing black liquor evaporators. The concentrate (aqueous chloride purge stream) can either be discharged directly, since it contains essentially no organics, or it can be routed to wastewater treatment. The chloride could also potentially be used in chlorine dioxide preparation for the bleach plant in bleached pulp production.

It is important to note that the goal is not to reduce the chloride concentration in the ESP dust to near zero, but to continuously remove the chloride input to the system to maintain steady state. This is a very advantageous situation where a membrane process serves as a kidney rather than as an ultimate polishing process. The result of the work presented here is a simple process for retrofitting existing mills. Estimates of operating costs and capital investment costs are given below.

Selective electrodialysis of chloride/sulfate solutions has been investigated elsewhere at concentrations far lower than those reported here (15).

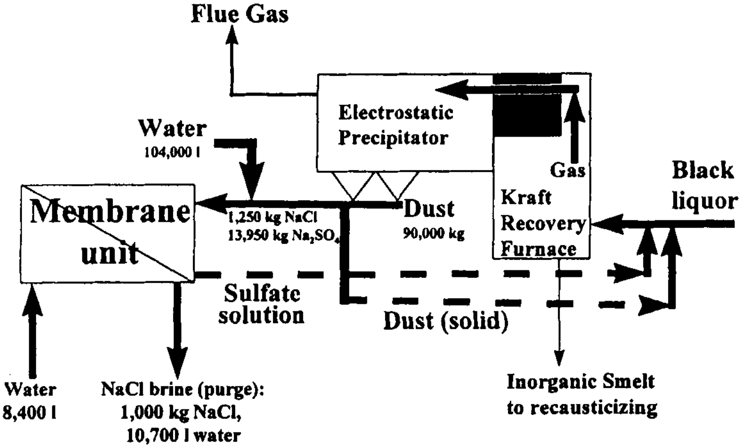

FIG. 1 Schematic for selective chloride purge from dissolved ESP dust in a kraft pulp mill. Mass balance basis: 1000 metric tons of air dry kraft pulp per day produced. Assumptions: NaCl input to the kraft process is 1000 kg per day (this is to be removed to maintain mass balance); 8.2 wt% of NaCl in the ESP dust (balance mainly Na_2SO_4); only NaCl is removed by electrodialysis.

In electrodialysis, the selectivity at high ionic strengths can usually be expected to decline. The results shown here give an example of good performance of commercial monovalent selective electrodialysis membranes, even at high ionic strengths.

EXPERIMENTAL

Materials

Mill Materials

In the experiments reported here, ESP dust from a kraft mill in the Southeastern United States was used. The dust is a white powder (micron-size particles, free flowing). It dissolves easily in water, forming a brownish solution; the colored organics settle overnight, leaving a clear supernatant. The ability to settle under gravity shows the rather large particle size of the organic material. No settling was allowed for any of the electrodialysis experiments. The composition of the dust is reported in Table 1. The organic carbon is either a product of incomplete combustion or direct carryover of lignin from the black liquor fuel fed to the kraft recovery furnace.

For the experiments with mill materials, a solution with 143 g ESP dust per liter of mill water was made and used as the diluate.

Components such as calcium, metals, and transition metals were not analyzed here. Both the organic and inorganic composition of ESP dust will vary from mill to mill and with time. Inorganic mill inputs vary due to geography (water supply, wood supply), seasonal changes of the wood, and impurities from replacement chemicals. Organic components in the ESP dust may vary with the firing strategy of the recovery furnace. Mill process water was used to dissolve the ESP dust for the chloride removal experiments. The main anions and cations in the mill process water were calcium (31 ppm), carbonate (93 ppm), and sulfate (28 ppm).

TABLE 1
Main Components of Dissolved Electrostatic Precipitator Dust from a Southeastern Kraft Pulp Mill (in wt%)[a]

Cl^-	CO_3^{2-}	SO_4^{2-}	Na^+	K^+	Organic carbon
3.46	2.35	57.15	25.09	10.74	0.10

[a] Mismatch of electrochemical equivalents anions/cations: 0.03%. Mass balance, total wt% accounted for: 98.89%.

Model Solutions

A representative composition for the model diluate (simulated ESP dust) was chosen based on the literature and on samples from several mills. The solution concentration was chosen not to exceed any solubility limits. Table 2 shows the initial diluate, concentrate, and electrode rinse compositions. The compositions for the electrode rinse solution and the initial concentrate solution were identical for all experiments, regardless of the diluate. The diluate composition was either as reported in Table 2 or it was actual ESP dust from the mill dissolved in mill process water.

Membranes

The membranes were donated for this work by Tokuyama Soda (Tokuyama America Inc., 1875 South Grant Street, San Mateo, CA). The membranes were NEOSEPTA CMS (monovalent selective cation-exchange membrane) and NEOSEPTA ACS (monovalent selective anion-exchange membrane). Two standard cation-exchange membranes (NEOSEPTA CM-1) were used for the ends of the stack. The bulk of ACS and CMS membranes consists of a strongly acidic (cation-exchange) and strongly basic (anion-exchange) resin, respectively. Both membranes contain a reinforcing fabric and are highly selective for monovalent ions, as measured in seawater and reported in the manufacturer's literature.

Inorganic Salts

The inorganic salts used here were technical grade for sodium sulfate and analytical grade for sodium chloride, sodium carbonate, and potassium sulfate.

TABLE 2

Initial Composition of Model Diluate (simulated ESP dust), Concentrate, and Electrode Rinse Solutions (all in deionized water) (in g/L)

	SO_4^{2-}	Cl^-	CO_3^{2-}	K^+	Na^+
Diluate (simulated ESP dust)	81.39	7.75	5.61	8.89	43.05
Concentrate	4.02	—	—	—	1.93
Electrode rinse	4.02	—	—	—	1.93

Water

Deionized water was used for all solutions. An exception was the diluate solution for experiments with actual mill ESP dust. The ESP dust was dissolved in mill process water (see Mill Materials above).

Electrodialysis System and Experimental Details

A laboratory electrodialysis system manufactured by Stantech, Inc. (Hamburg, Germany) was used. This system has an active cross-sectional membrane area of 100 cm^2 (10 cm by 10 cm). An electrodialysis unit consists of a parallel arrangement of alternating diluate and concentrate cells, separated by anion and cation membranes. The setup is similar to a conventional filter press. A polymeric netting is placed inside of the frames forming the cells to minimize boundary layer effects.

Four diluate and five concentrate cells were used for the experiments. The effective cell thickness (distance from membrane to membrane) was 0.25 cm. The superficial average flow velocities were 4.4 cm/s in the diluate cells and 4.9 cm/s in the concentrate cell. The electrodes were rinsed in series from anode to cathode with a flow of 80 L/h. The stack configuration and the goal of the separation process are shown in Fig. 2. The outermost cation-exchange membranes of the stack were standard membranes. All other anion- and cation-exchange membranes were monovalent ion selective.

A repeat unit is indicated in Fig. 2. Up to several hundred of these repeat units are used in commercial systems. The goal of the process investigated here is to selectively remove chloride as sodium and potassium chloride while retaining the sulfate, which is recycled to the kraft recovery furnace.

The experiments were run in batch mode with samples of 140 mL drawn at time intervals. A total electrical current of 1 A was passed through the stack, which corresponds to a current density of 10 mA/cm^2. Limiting current density experiments showed that this is far below the critical current density where boundary layer effects start to become important.

In batch experiments for electrodialysis, the concentrate circuit must be started with some electrical conductivity so that the electrical resistance of the stack is initially not too high. High electrical resistance would otherwise cause effects such as water splitting at the ion-depleted membrane boundary layers. In our runs, sodium sulfate was used to supply this initial conductivity. In a full-scale unit, the concentrate conductivity would be supplied by a small recycle stream from the concentrate outlet (purge) to the concentrate inlet of the stack.

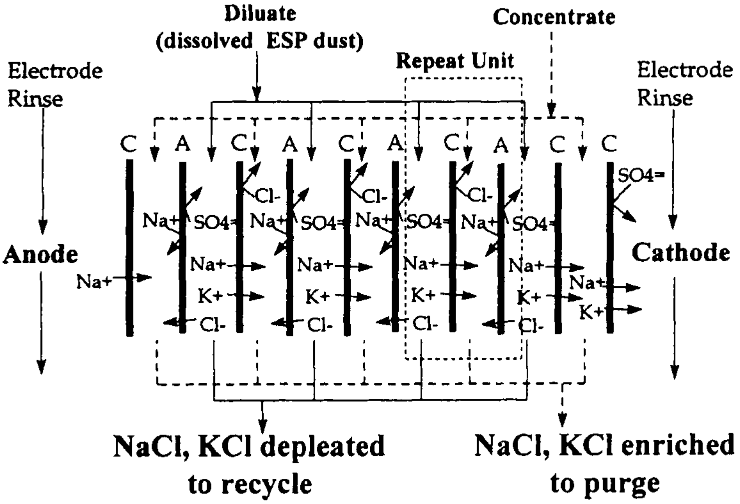

FIG. 2 Laboratory stack setup for chloride removal from dissolved ESP dust. (A, C: monovalent selective anion- and cation-exchange membranes, respectively.)

The pH of the solutions was measured with pH combination electrodes. The initial pH of the diluate solutions was high, generally above pH 10 for all experiments. The concentrate pH was near neutral, and rose during the experiments to about pH 10.

As indicated individually for each run reported here, 4 to 5.86 L diluate (actual ESP dust dissolved in mill process water, Table 1; or model ESP dust solution, Table 2), 4 to 5.86 L concentrate (see Table 2), and 4 L electrode solution (see Table 2) were used. The solution temperatures rose slightly during each experiment from room temperature (20°C) to about 27°C at maximum, mainly through heat transfer from the centrifugal pumps.

Analytical

The cations sodium and potassium were analyzed by atomic absorption spectroscopy. The anions carbonate, sulfate, and chloride where analyzed by ion chromatography.

RESULTS AND DISCUSSION

Model Solutions: Chloride Selectivity

Figure 3 shows the results from electrodialysis of a model solution to simulate ESP dust (initial composition, see Table 1). Figure 3 shows the electrochemical equivalents of chloride and sulfate transported from the diluate to the concentrate. Also shown is the theoretical maximum of transported ions as calculated by integrating the total electrical current that passed through the unit multiplied by the number of diluate cells.

The sulfate and carbonate concentrations remain constant within the accuracy of the analysis. The chloride concentration in the concentrate increases rapidly, which shows the selective transport of chloride. At the end point of this run, 65.9% of the chloride had been transferred from the diluate to the concentrate. The overall average current efficiency for chloride removal, when comparing the electrochemical equivalents of chloride transported with the total electrical current, was 73%.

It is clear that even with the highly concentrated feed solution containing a very large excess of divalent sulfate ions over chloride ions, the membranes are still very selective for monovalent anions. This encouraging result led us to initiate tests with the actual materials from a pulp mill.

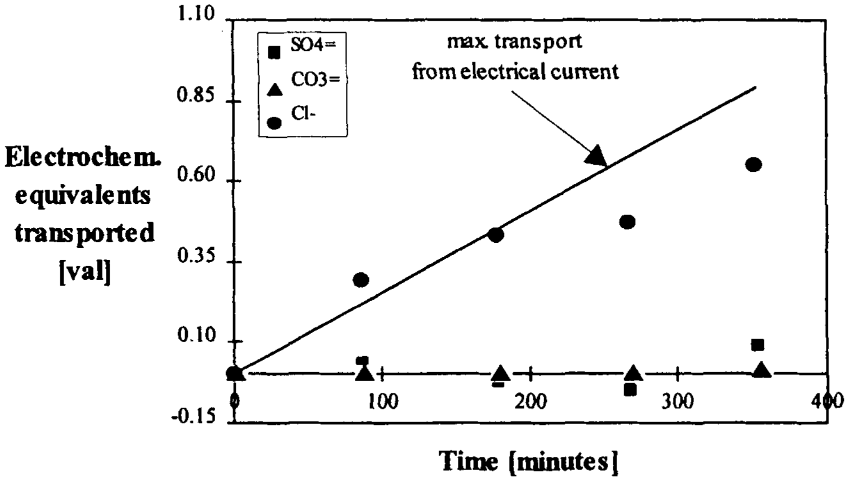

FIG. 3 Ion transport by batch electrodialysis of model ESP dust solution (20.8–26.2°C; 10 mA/cm^2; diluate, concentrate: 4 L each; diluate/concentrate start concentrations: Table 2).

Long-Term Tests with Mill Materials

Based on the encouraging selectivity for chloride found with the model solutions, long-term tests were initiated with mill materials. Figure 4 shows results from the first test with mill materials. Then a number of standard separation runs with mill materials were performed with the same set of membranes (these runs are not shown). Also shown in Fig. 4 are data for an experiment after a combined 86 hours of testing of this same set of membranes with the mill materials.

When comparing the initial run with the run after 86 hours of experiments with mill materials, no significant deterioration was detected. No special chemical or physical cleaning procedures were performed on the membranes during this extended series of tests. For storage between batch experiments, the stack was disassembled and the membranes sprayed with 0.5 M sodium chloride solution from a laboratory wash bottle. The membranes were then stored in 0.5 M sodium chloride solution. This was done four times during the time of performing the 86 hours of runs with actual materials. The loose brown deposits found on the diluate side of the membranes washed off quite easily, indicating that a simple backflush will likely suffice for in-place cleaning of a full-scale application.

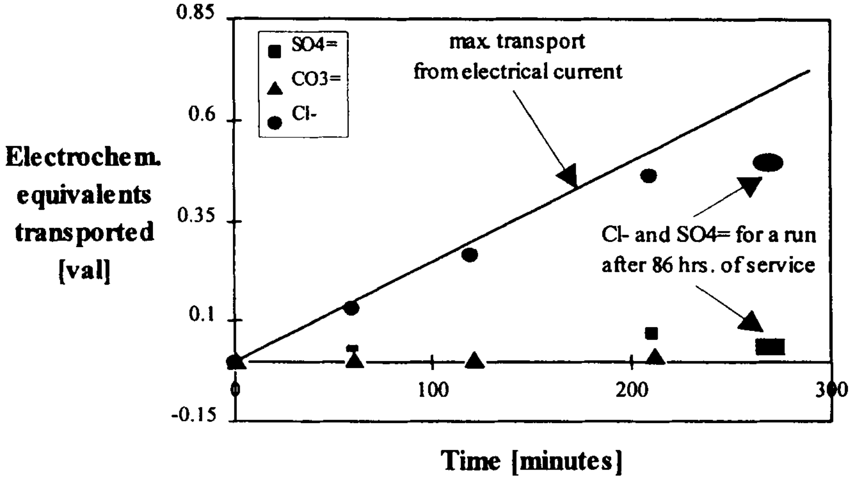

FIG. 4 Ion transport by batch electrodialysis of actual ESP dust dissolved in mill water. Dechlorination performance of the same set of membranes before and after 86 hours of service with actual mill ESP dust solution (20.2–24.6°C; 10 mA/cm^2; 5.86 L diluate, 5.86 L concentrate first run; 4.86 L diluate, 4.86 L concentrate final run after 86 hours; diluate start concentrations: Table 1; concentrate start concentrations: Table 2).

Figure 5 shows the stack voltage as measured between the anode and cathode as a function of batch run time for the two runs shown in Fig. 4. It is clear that the electrical resistance is not increased by extended runs with the actual mill materials. Therefore, fouling appears not to be a serious problem in this process. Dissolved anionic organic molecules, such as sulfonated lignin, could potentially foul the membranes. The high ionic strength of the feed solution may advantageously contribute to precipitation of any potentially soluble organics in the ESP dust.

The initial decrease of the stack voltage is due to improved conductivity of the concentrate by the chlorides that are introduced from the diluate. The final increase of the stack voltage is due to depletion of chloride ions in the boundary layers at the diluate side of the monovalent selective anion-exchange membranes. Experiments with model solutions with pure chemicals show a similar behavior.

The result of this extended series of experiments shows that a potentially viable process has been demonstrated. No pretreatment of any kind was applied to the mill ESP dust solution. Plugging of the flow channels in the system, fouling of the membranes, and precipitation of inorganics could be potential problems. However, no negative impact was observed.

Process Economics

A base case bleached kraft pulp mill (BKP mill) with a production of 1000 air dry tons of pulp per day (ADTP) was assumed. The energy needed

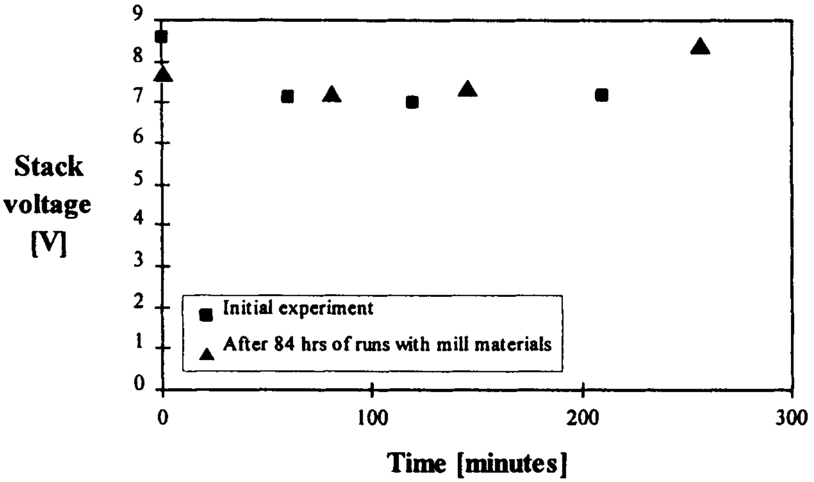

FIG. 5 Overall electrical voltage applied at current density 10 mA/cm^2. Values for the initial run with mill materials and after a total of 84 hours of experiments with mill materials.

to continuously remove the chloride input to this base case mill by electrodialysis of dissolved ESP dust is estimated in the following section and then compared to an energy demand estimate using data from the literature.

Assumptions for the Base Case Mill, Calculation of Energy Demand

The source for the total chloride input into the liquor system of the base case mill is published data (6). The total chloride input was estimated at 420 kg/day from wood, makeup chemicals, and water. No chloride input from the bleach plant was assumed (no bleach plant effluent recycle to the kraft chemical recovery process). Steady state was assumed (chloride input equals chloride purge by electrodialysis). Purge of chloride with the pulp produced was neglected.

Assumptions for the electrodialysis unit were 80% current efficiency for chloride (80% of the electrical current applied transports chloride), 1 m^2 active cross-sectional membrane area, and 150 concentrate/diluate cell pair units (see Fig. 2). The voltage drop per cell pair was assumed to be 2 V (from Fig. 5, about 8 V for 4 cell pairs). This is conservative, because the electrode voltage losses in the rather short laboratory stack have a significant impact; this will not be the case in a full-scale unit. The energy for pumping and mixing of the ESP dust with water is not taken into account.

Using Faraday's constant F (96,500 coulomb/mol of a univalent salt) and the chloride input assumed above, an electrical current of 13,214 A results to hold a steady-state chloride concentration in the pulping process. Because multiple use is made of the current passing between the anode and cathode in an electrodialysis stack (150 cell pairs), this value drops to 88 A passing between the anode and cathode. With a current efficiency of 80% for chloride removal, 110 A is the current estimated for the ED unit. This corresponds to 33 kW or 792 kW·h/day. With a cost of $0.026/kW·h (assumption of an advantageous utility contract for a pulp mill), the energy cost for the ED unit is $20.6/day.

Comparison with an Estimate of the Energy Demand from Published Data

The electrical power needed for electrodialytical chloride removal for a 1000-ADTP bleached pulp mill can be roughly estimated to be 751 kW·h/day, based on data for similar salt removal levels for water desalination published elsewhere (16). The chloride concentration in dissolved ESP dust is quite similar to seawater, and the sulfate and carbonate are assumed here not to influence the separation process.

The result of 751 kW·h/day from published data compares very well to the estimate of 792 kW·h/day shown above. Because no deterioration in membrane performance was detected in the laboratory experiments, a membrane lifetime of several years should be achieved.

The energy demand for separation would increase essentially proportional with additional chloride input from bleach plant effluent recycle. The membrane area also increases with increased load. In operation, the chloride removal level is easily adjusted through the current passing through the unit. Startup/shutdown of the chloride removal process is very simple.

Capital Investment

A quotation from a manufacturer of electrodialysis equipment was obtained. The specifications for the base case mill were used. A full-scale electrodialysis unit for the above process was quoted at $210,000 (electrodialysis unit only, no dissolving equipment or process modifications included). The energy demand for the unit was quoted to be 528 kW·h/day by the equipment vendor. This, again, compares well to the estimates above.

CONCLUSIONS

Chloride removal from the kraft pulping process under low effluent conditions was the goal of this work. Electrodialysis with monovalent selective membranes was used successfully to separate chloride from a feed solution with a very high concentration of sulfate. The process was shown to operate without significant problems using an actual feed solution containing organic and inorganic contaminants. Electrical current efficiencies above 70% were found. The energy demand of the electrodialysis for a full-scale application was estimated at 1.9 kW·h/kg of chloride removed.

The work reported here is an example of membrane technology as a kidney for low effluent processing in a major manufacturing industry. The performance of the electrodialysis process with realistic feed solutions is very encouraging. Pilot tests at a kraft pulp mill in the Southeastern United States are underway.

ACKNOWLEDGMENTS

The support of this work by the Member Companies of the Institute of Paper Science and Technology is gratefully acknowledged. The donation of membranes by Tokuyama Soda Inc. is also gratefully acknowledged.

REFERENCES

1. T. M. Grace, B. Leopold, and E. W. Malcolm (technical editors), *Volume 5: Alkaline Pulping*, in M. J. Kocurek and F. Stevens (Eds.), *Pulp and Paper Manufacture*, Joint Textbook Committee of the Paper Industry of the United States and Canada, Atlanta, GA, 1991.
2. P. E. Ahlers, H. Norrstrom, and B. Warnqvist, *Chlorides in the Kraft Recovery System. Part II: Process and Equipment Aspects on a Closed Bleached Kraft Mill*, Paper Presented at the TAPPI/CPPA International Pulp Bleaching Conference, May 1976, Chicago, IL.
3. D. W. Reeve, H. N. Tran, and D. Barham, *Pulp Pap. Can.*, *82*(9), 105 (1981).
4. H. N. Tran, *TAPPI J.*, *69*(11), 102 (1986).
5. O. Keitaanniemi and N.-E. Virkola, *Ibid.*, *65*(7), 89 (1982).
6. NCASI (National Council of the Paper Industry for Air and Stream Improvement, Inc.), *A Study of Kraft Recovery Furnace Hydrochloric Acid Emissions* (NCASI Technical Bulletin No. 674), 1994.
7. P. O. Karjalainen, J. E. Lofkrantz, and R. D. R. D. Christie, *Pulp Pap. Can.*, *73*(12), 95 (1972).
8. W. A. Moy, P. Joyce, and G. E. Styan, *Ibid.*, *75*(4), 88 (1974).
9. W. H. Rapson, *Ibid.*, p. T635 (December 1967).
10. D. W. Reeve, *Ibid.*, *77*(8), 35 (1976).
11. K. L. Holman, G. P. Golike, and K. R. Carlson, *Process Simulation of Bleach Plant Effluent Concentrate Recycle to Recovery with Leaching of Chloride from Salt Cake*, Poster Preprint at the 1994 International Pulp Bleaching Conference.
12. R. H. Wright, *Pulp Pap. Mag. Can.*, *57*(12), 171 (November 1956).
13. S. Maruko, T. Iwadare, Y. Kobuchi, K. Motomura, and Y. Matsunaga, "Method of Dechlorination," Japanese Patent 22,051-1980.
14. M. Paleologou, R. M. Berry, R. Thompson and J. T. Wearing, US Patent 5,567,293 (1996).
15. G. Saracco, M. C. Zanetti, and M. Onofrio, *Ind. Eng. Chem. Res.*, *32*, 657–662 (1993).
16. US DOE Report, *Separation Systems—A Research and Development Needs Assessment, Vol. II* (DOE/ER/30133-H1, Contract DE-AC01-88ER30133) April 1990.

Received by editor February 25, 1997
Revision received May 1997