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

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N. Ahner[a]; D. Gottschlich[a]; S. Narang[a]; D. Roberts[a]; S. Sharma[a]; S. Ventura[a]
[a] SRI International, Menlo Park, CA

To cite this Article Ahner, N. , Gottschlich, D. , Narang, S. , Roberts, D. , Sharma, S. and Ventura, S.(1993) 'Piezoelectrically Assisted Ultrafiltration', Separation Science and Technology, 28: 1, 895 — 908
To link to this Article: DOI: 10.1080/01496399308019526
URL: http://dx.doi.org/10.1080/01496399308019526

SEPARATION SCIENCE AND TECHNOLOGY, 28(1-3), pp. 895-908 (1993)

PIEZOELECTRICALLY ASSISTED ULTRAFILTRATION

N. Ahner, D. Gottschlich, S. Narang, D. Roberts, S. Sharma, and S. Ventura
SRI International, 333 Ravenswood Ave., Menlo Park, CA 94025

ABSTRACT

We have demonstrated the feasibility of using piezoelectrically assisted ultrafiltration to reduce membrane fouling and enhance the flux through ultrafiltration membranes. A preliminary economic evaluation, accounting for the power consumption of the piezoelectric driver and the extent of permeate flow rate enhancement, has also shown that piezoelectrically assisted ultrafiltration is cost effective and economically competitive in comparison with traditional separation processes.

Piezoelectric transducers, such as a piezoelectric lead zirconate titanate (PZT) disc or a piezoelectric horn, driven by moderate power, significantly enhance the permeate flux on fouled membranes, presumably because they promote local turbulence. Several experiments were conducted on polysulfone and regenerated cellulose UF membranes fouled during filtration of model feed solutions. Solutions of poly(ethylene glycol) and of high-molecular weight dextran were used as models. We found that we could significantly increase the permeate flux by periodically driving the piezoelectric transducer, horn or PZT disc, by application of moderate power over short periods of time, from 20 to 90 seconds. Enhancements as high as a factor of 8 were recorded within a few seconds, and enhanced permeate fluxes were maintained over a prolonged period (up to 3 hours). The prolonged flux enhancement makes it feasible to drive the piezoelectric transducer intermittently, thereby reducing the power consumption of the piezoelectric driver.

As piezoelectric drivers of sonically assisted ultrafiltration, PZT disc transducers are preferred over the piezoelectric horn because of their small size and ease of adaptability to ultrafiltration test cells. The horn transmits sonic energy to the UF membrane through a titanium element driven by a separate piezoelectric transducer, but a piezoelectric ceramic disc transmits energy directly to the UF membrane. Moreover, because piezoelectric ceramic elements can be fabricated in several configurations, they are potentially feasible for piezoelectrically driven ultrafiltration spiral-wound membrane modules.

INTRODUCTION

Ultrafiltration (UF) is a process increasingly applied in liquid industrial waste treatment, food processing (fruit juices, cheese whey, milk, starch factory effluent), and pharmaceutical and medical industries.

Susceptibility of UF membranes to fouling, resulting in serious flux decline of the permeate, is a continuing problem adversely affecting the cost of using a membrane separation process as a unit operation. Commercial acceptance of ultrafiltration has been severely limited by the inability of membranes to perform at consistent productivity for an extended period of time.

The hydrophobic nature of UF membranes in conjunction with low surface porosity and non-uniform pore size distribution increases their vulnerability to fouling by pore blockage (1). The fouling phenomenon in general proceeds as follows:

1. Rapid initial decline in flux due to the buildup of concentration polarization, reducing the driving force for separation.

2. Adsorption of macromolecules such as proteins on the hydrophobic membrane material and competing plugging of pores.

3. Convective deposition of particles and compression of cake solids.

The rate of decrease of the flux of the permeate through the membrane, however, depends significantly on the particular composition of the liquid treated and on the interaction between the solute and the membrane.

Several approaches have been proposed to minimize fouling of UF membranes. Pretreatment of the membrane with hydrophilic surfactants (2,3) and polymers increases the initial flux and reduces the flux decline. This pretreatment of the membranes improves the homogeneity of the surface, and the increased hydrophilicity is thought to lower the degree of concentration polarization and reduces the possibility of protein adhesion to the original hydrophobic surface. A second approach involves prefiltration (4-8) of the solution with a larger pore size membrane to filter off the large particles. Pulsating flow has been used to create turbulence in the feed, thus postponing the onset of serious fouling. Backwashing is a common approach used to unplug the blocked pores and dislodge the cake from the membrane surface (9-12).

However, all these approaches have drawbacks. Pretreatment of the membrane or the feed provides only marginal and short-term improvement. Prefiltration also adds the cost of another unit operation. Backwashing implies interruption of operation to yield a batch process.

In view of the significant limitations of the fouling control options described above, it is highly desirable to develop a fouling control process applicable to

1. A wide variety of membrane materials
2. A wide variety of feed solutions
3. Many different configurations of separation modules
4. A continuous process, without involving an extra unit operation in series.

In this paper, we discuss a unique approach to solving the fouling problem by developing piezoelectric drivers as backings of membranes and utilizing the piezoelectric effect during the ultrafiltration process to produce strong local turbulence that will minimize concentration polarization and the rate of buildup of solutes and particulate matter on the membrane surface.

EXPERIMENTAL SECTION

To demonstrate the feasibility of using piezoelectrically assisted ultrafiltration to reduce fouling, we undertook two approaches. We used thin ceramic piezoelectric discs to intermittently transmit sonic energy to commercially available UF membranes, and, in another approach, we used a piezoelectric ultrasonic horn as driver . The effectiveness of the driver was evaluated by monitoring the permeate flux enhancement.

Commercial UF membranes with 10,000 and 100,000 molecular weight cut-off (MWCO) were used in this study. We studied polysulfone membranes mounted on a polypropylene support (Millipore type PTGC) and low-protein binding regenerated cellulose membranes bound to a polypropylene support (Millipore type PLGC). Stainless steel support filters with ten and twenty micron pores were used to provide structural support to the UF membranes, if required.

We tested aqueous solutions of polyethylene glycol with molecular weight of 10,000, a derivative of polyethylene glycol with molecular weight of 15,000-20,000, and dextran with molecular weight of 162,000. Before testing the effect of the piezoelectric driver on ultrafiltration, we allowed the UF membranes to foul to an approximately constant permeate flux. The piezoelectric driver was applied on the permeate side. The metalized ceramic transducer was carefully encapsulated by an insulating polymer coating to ensure proper electrical insulation. For this purpose we used room temperature vulcanized (RTV) rubber, polyvinylchloride (PVC), and epoxy polymers.

A cross-flow ultrafiltration test apparatus was assembled for long-term evaluation of the membrane fouling. The apparatus is designed to circulate feed solution through two test cells that hold ultrafiltration membranes (Figure 1) . This configuration allows the accurate and simultaneous evaluation of UF membranes driven and not-driven by a piezoelectric transducer, under the same operating conditions. The feed pump is a centrifugal five-stage pump with a maximum flow rate of 4 gallons per minute and a maximum pressure of 125 psig. The pressure in the system is adjusted through a back-pressure regulator. Our standard operating conditions are a pressure of 50 psig with a feed flow rate of about 1/2 gallon per minute. The feed stream is split so that the piezoelectric and non-piezoelectric cells can be tested side by side. The non piezoelectric-driven cell is identical to the piezoelectric-driven cell in all respects except that it does not have any power supplied to the piezoelectric driver. The residue streams from each of the cells is recombined before passing through a throttle valve. The throttle valve has the function to maintain the pressure in the system. Because equal lengths of tubing have been used to supply the feed to both cells, the feed streams to each cell are approximately similar. We measure the permeate flux through each membrane. Any difference in flux directly measures the effect of the piezoelectric vibration on the fouling of the UF membrane. Both the permeate stream and the combined residue stream can be sampled for analysis. To maintain constant feed conditions, the test apparatus is designed with total recycle. A coil with cooling water is inserted into the feed tank to maintain constant temperature in the system. The temperature of the feed is monitored with a thermocouple.

As a standard procedure, between different runs, the system is thoroughly cleaned to prevent contamination. A 2% solution of citric acid is run for one hour to remove traces of metal from the system. Deionized water is then run for fifteen minutes for rinsing the system. A 0.2% Micro detergent solution is run for one hour to remove proteins and amino acids. Several more deionized rinses follow. Care was taken to run the final two rinses with Milli-Q (18 megaohm) water. The entire cleaning procedure is always carried out with no membrane so as to ensure a thorough cleaning of both sides of the system.

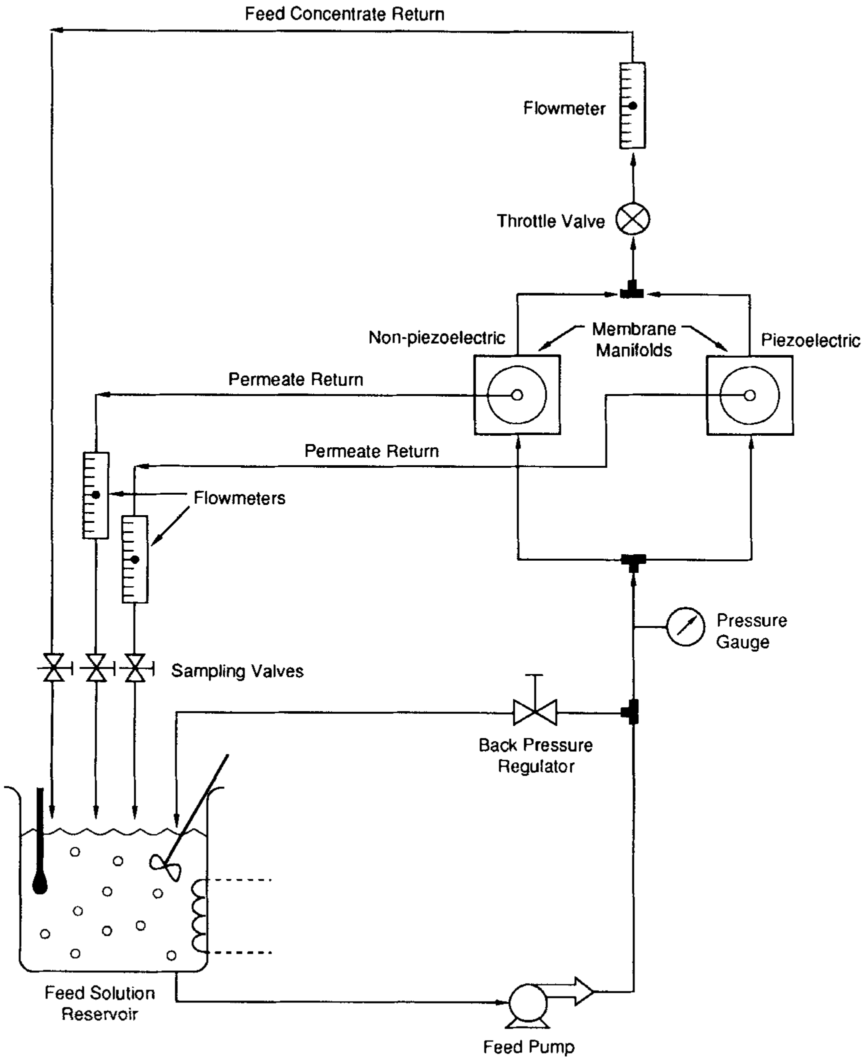

Figure 1. Cross-flow ultrafiltration test apparatus.

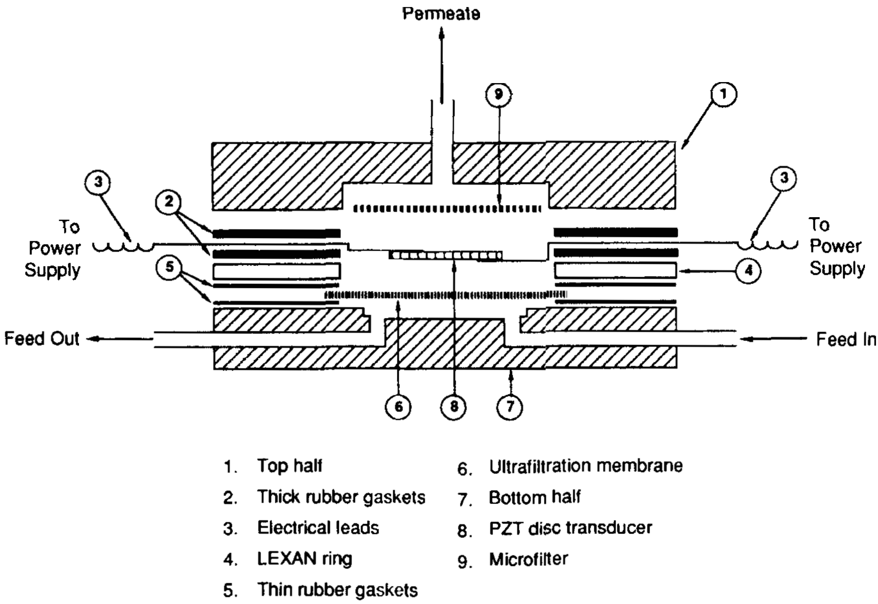

Figure 2. PZT disc-driven ultrafiltration test cell.

RESULTS AND DISCUSSION

Lead Zirconate Titanate (PZT) Piezoelectric Disc Assisted Ultrafiltration

Ring and disc-shaped PZT transducers were tested to transmit sonic power to UF membranes. The transducers were placed on the permeate side of the UF membrane, where they are not subjected to high pressure. Disc-shaped PZT transducers provided the best performances. Electrical wires were connected to a metalized PZT disc of 38 mm. diameter and of 2.5 mm thickness. The disc and the electrical leads were electrically insulated by encapsulation with a fluoroepoxy coating. The schematic representation of the PZT disc driven ultrafiltration test cell is illustrated in Figure 2. By application of a moderate power, such as 40 watts, mechanical strain is produced along orthogonal directions. Dimensional changes of thickness, length or width, depending on the resonance frequency, are responsible for the generation of local turbulence on the permeate side, which is transmitted to the UF membrane.

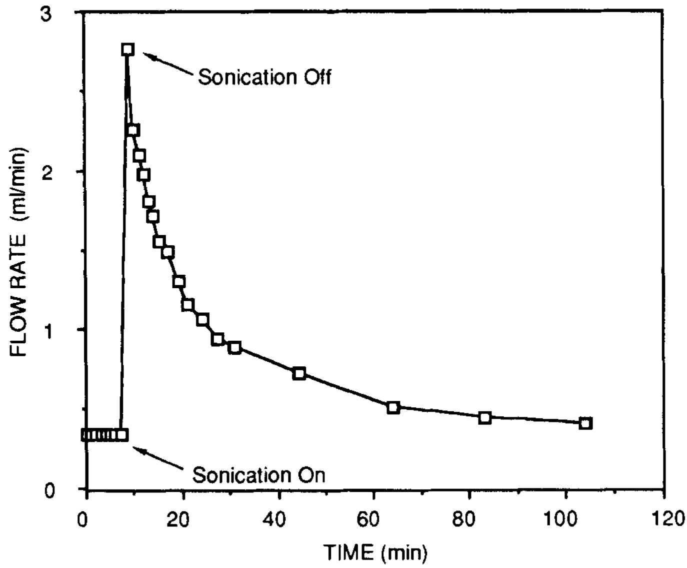

Figure 3. Ultrafiltration flow rate of a 0.45% dextran (MW 162,000) solution with a 100,000 MWCO polysulfone membrane piezoelectrically-enhanced by a PZT disc transducer.

The feasibility of PZT disc-driven ultrafiltration was tested in the following experiments. A 0.45 % solution of dextran (M_W 162,000 daltons) in water was filtered through a Millipore polysulfone UF membrane with a nominal MWCO of 100,000. The insulated PZT disc was inserted in the ultrafiltration test apparatus. The feed solution was filtered at 50 psig under cross-flow conditions for a few days to reach steady fouling conditions. The permeate flow rate was 0.34 mL/min. Forty watts of power was applied to the PZT transducer for 90 seconds by means of a suitable power supply. The permeate flow rate increased to 2.78 mL/min. We turned off the power, and after more than one hour and a half the permeate flow rate was still higher than the permeate flow rate before power was applied. The permeate flow rate changes are shown in Figure 3. Under these conditions, piezoelectric vibration increased the permeate flow rate as much as by a factor of 8.

In a similar test, we filtered a 0.58% dextran solution with a Millipore UF membrane with nominal MWCO of 100,000. At 50 psig a steady fouling condition

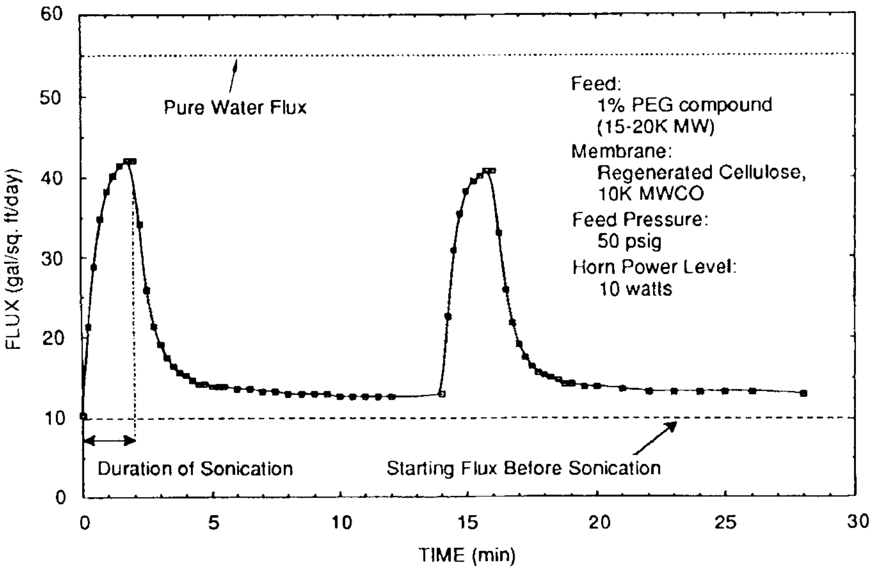

Figure 4. Effect of the piezoelectric horn on the ultrafiltration flux of 1.0% polyethylene glycol solution.

was reached with a permeate flow rate of 0.30 mL/min. Forty watts of power was applied to the PZT transducer for 90 seconds, and the permeate flow rate increased to 2.1 mL/min (a factor of 7). Permeate volumes were collected at constant intervals of time, and the cumulative volumes were plotted versus time. It took 3 hours for the system to return to the original permeate flow rate of 0.30 mL/min. It should be noted that no attempt has yet been made to optimize power consumption and performance of the piezoelectric ceramic disc driver.

The configuration of the PZT transducer is an important parameter, as shown by some experiments we did with a PZT ring-shaped transducer instead of the PZT disc. The ring had an internal diameter of 3/4 inch and an external diameter of 1 inch, and a thickness of 1/2 inch. The ring was metalized on the inner and outer surfaces and poled radially. We attached electrical leads to the two metalized surfaces and the ring was completely coated with a thin layer, of epoxy for electrical insulation. In air the PZT ring showed a minimum impedance at 40.394 KHz, corresponding to the radial resonance mode of vibration of the ring. At this frequency, in-phase dimensional changes of the inner and outer ring diameter take place. However, when the ring was immersed in water, this radial mode of displacement was severely attenuated, as shown by experimental measurements of impedance as a function of frequency. Nevertheless, a PZT ring-assisted ultrafiltration test was carried out by resonating the PZT ring in the frequency range of approximately 30 ± 10 KHz. The voltage applied to the ring was amplified by means of a amplifier. Under these conditions, even at maximum amplification, the PZT ring transducer assisted ultrafiltration showed an insignificant permeate flux increase of about 5% above the steady state flux.

Piezoelectric Horn-Assisted Ultrafiltration

We used a piezoelectric horn capable of transmitting up to 200 watts of power to a titanium tip at the frequency of 20 KHz. The horn was maintained in proximity to the UF membrane, on the permeate side, through a microporous stainless steel filter. The horn was adapted to constitute the top portion of the test cell. The permeate was therefore collected through an output channel drilled through the horn.

Feasibility tests of the piezoelectric horn assisted ultrafiltration were carried out on a simplified ultrafiltration apparatus where the whole feed solution was forced through the UF membrane without recirculation of the feed and of the permeate stream. Two experiments, with and without piezoelectric assistance, were conducted and compared. A 1% solution of poly(ethylene glycol) with average molecular weight of 10,000 daltons was used as feed through a 47 mm. 10,000 MWCO poly(sulfone) UF membrane, mounted on a polypropylene support. A microporous stainless steel support filter was used to give structural support to the UF membrane. The feed solution was pressurized at 80 psig.

The ultrafiltration test experiments, with and without power applied to the piezoelectric horn, were run independently on the same membrane.

A systematic study on the effect of sonication produced by the horn was conducted for the filtration of 1% polyethyleneglycol solution through a regenerated cellulose UF membrane (MWCO 10,000) and 0.1% dextran solution with a polysulfone UF membrane (100,000 MWCO). Several sonication pulses were applied during each run. Figure 4 shows the short-term variation of the permeate

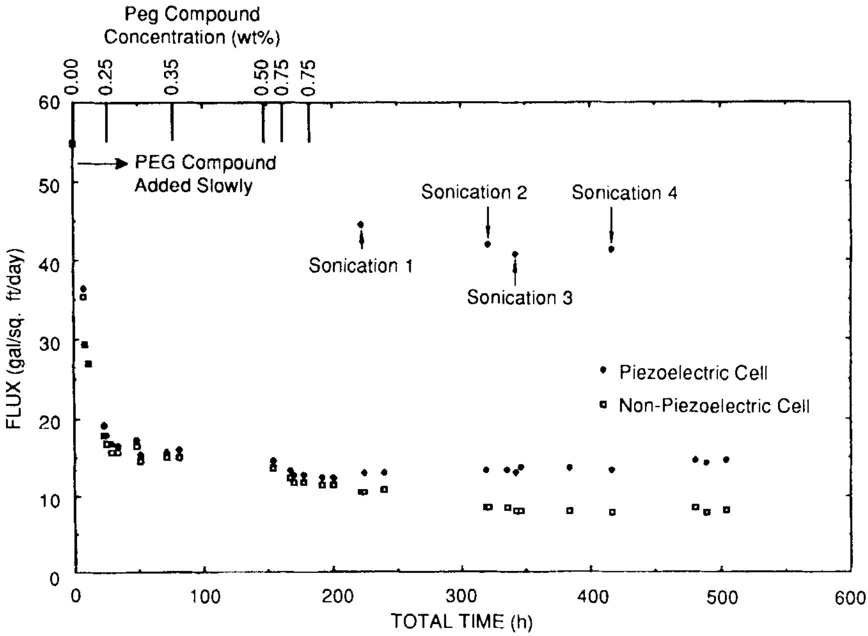

Figure 5. Long-term ultrafiltratrion performance on 1.0% polyethylene glycol solution. Piezoelectrically-assisted ultrafiltration was carried out by a piezoelectric horn.

flux with time for the polyethylene glycol experiments. The upper dotted line represents the flux of pure water through the membrane. Before the piezoelectric horn was driven to enhance ultrafiltration, the membrane was fouled over a period of a few days. The flux through the fouled membrane is indicated by the lower dotted line. The piezoelectric horn was then periodically driven for about two minutes by applying a formal power of 10 watts. The permeate flux was substantially enhanced by more than a factor of 4 when the horn was on. Figure 5 shows the long-term effect of sonication on long-term performance of the membrane. Initially the feed solution was progressively concentrated to quickly reach steady state fouling conditions. Then the flux was periodically enhanced by driving the piezoelectric horn with 10 watts power (2 minutes every 50 hours, on average). The figure compares the fluxes of the piezoelectrically-driven and non-piezoelectrically-driven cells. The permeate flux of the piezoelectrically-driven cell

TABLE 1.
COMPARISON OF ECONOMIC AND ENERGY ADVANTAGES OF PIEZOELECTRIC MEMBRANE TECHNOLOGY FOR SOLVENT RECOVERY IN DEASPHALTING OPERATIONS

	Process		
	Traditional	With Conventional Membrane	With Piezoelectric Membrane
Energy (Btu/lb DAO)*	1,370	450	463
Capital Cost ($1000)	4,330	8,410	6,190
Overall Processing Cost (Cents/lb DAO)	1.00	0.88	0.72

* DAO = Deasphalted oil.

is higher than that of the non-piezoelectrically driven cell at the end of 21 days. To examine in detail this effect, we are going to verify whether any pore size variation of the UF membrane takes place because of vibration.

The ultrafiltration of dextran (0.1%) with molecular weight of 162,000 through polysulfone 100,000 MWCO showed similar results. The permeate flux was enhanced by approximately a factor of 5 when the horn was piezoelectrically driven. The ultrafiltration of poly(ethyleneimine) (2.5%) , molecular weight 50,000-60,000, through polysulfone 10,000 MWCO also showed good results with a flux enhancement by a factor of 4.

Preliminary Economic Analysis

As an example of the economic potential and energy savings potential of this technology, we have considered the use of piezoelectrically-enhanced ultrafiltration (UF) membranes in the deasphalting of oil. In previous work, we examined the economics of traditional deasphalting processes and of a hybrid deasphalting process using conventional UF membranes (13). This previous work indicated that the capital cost of conventional UF membranes is so high that the economic advantage over traditional (nonmembrane) processing is minor, despite the substantial energy savings offered by the conventional UF membranes. Therefore, these energy savings will not be realized.

Piezoelectrically-assisted (PZ) membranes have the potential to allow these energy savings to be realized because: (a) much less surface area of membrane will be required because of the higher flux, reducing the capital cost (even though the capital cost of a unit area of PZ membrane will be greater); and (b) the additional energy required by the PZ membranes is insignificant. To estimate the economics of using the PZ UF membranes in solvent deasphalting, we assumed (a) the flux enhancement is a factor of 4 (average based on laboratory data), (b) the cost of a unit area of PZ membrane modules is twice that of conventional UF membrane modules (estimate; no technical basis at this time), and (c) the power necessary to drive the PZ membrane is 2.4 W/cm^2 of active membrane area (based on laboratory data).

With these parameters, the PZ membrane process for deasphalting oil is 28% less expensive than the traditional process and uses 34% of the energy of the traditional process (Table 1). In this table, we compute the overall processing cost by discounted cash flow with a 15% return on capital. The membrane life is one year. Other economic parameters are the same as those in Table VII-3 of Gottschlich and Roberts (13). Since our development is at an early stage, the economic analysis can serve only to assess the pay-off for further development work. With 283,000 barrel/day capacity for deasphalted oil (DAO; 1985 U. S. capacity), and a potential savings of 907 Btu/lb of DAO (Table 1), 6.8 trillion Btu/yr would be saved in the United States if 25% of the industry adopted the technology. History shows that economics of embryonic technologies tend to get worse as developments proceed. Nevertheless, we are encouraged by the potential for this technology in this application and in a host of ultrafiltration applications.

ACKNOWLEDGMENT

This work was supported by the DOE Office of Industrial Programs through the Idaho National Engineering Laboratory under Contract No. DE-FC07-89ID12906.

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