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

Pervaporation of Model Acetone-Butanol-Ethanol Fermentation Product Solutions Using Polytetrafluoroethylene Membranes

David L. Vrana[a]; Michael M. Meagher[b]; Robert W. Hutkins[a]; Bruce Duffield[c]
[a] Department of Food Science and Technology, University of Nebraska-Lincoln Lincoln, Nebraska [b] Department of Food Science and Technology and Department of Biological Systems Engineering, University of Nebraska-Lincoln Lincoln, Nebraska [c] Department of Biological Systems Engineering, University of Nebraska-Lincoln Lincoln, Nebraska

To cite this Article Vrana, David L. , Meagher, Michael M. , Hutkins, Robert W. and Duffield, Bruce(1993) 'Pervaporation of Model Acetone-Butanol-Ethanol Fermentation Product Solutions Using Polytetrafluoroethylene Membranes', Separation Science and Technology, 28: 13, 2167 — 2178
To link to this Article: DOI: 10.1080/01496399308016741
URL: http://dx.doi.org/10.1080/01496399308016741

SEPARATION SCIENCE AND TECHNOLOGY, **28**(13&14), pp. 2167–2178, 1993

Pervaporation of Model Acetone–Butanol–Ethanol Fermentation Product Solutions Using Polytetrafluoroethylene Membranes*

DAVID L. VRANA
DEPARTMENT OF FOOD SCIENCE AND TECHNOLOGY
UNIVERSITY OF NEBRASKA-LINCOLN
LINCOLN, NEBRASKA 68583-0919

MICHAEL M. MEAGHER†
DEPARTMENT OF FOOD SCIENCE AND TECHNOLOGY
AND
DEPARTMENT OF BIOLOGICAL SYSTEMS ENGINEERING
UNIVERSITY OF NEBRASKA-LINCOLN
LINCOLN, NEBRASKA 68583-0919

ROBERT W. HUTKINS
DEPARTMENT OF FOOD SCIENCE AND TECHNOLOGY
UNIVERSITY OF NEBRASKA-LINCOLN
LINCOLN, NEBRASKA 68583-0919

BRUCE DUFFIELD
DEPARTMENT OF BIOLOGICAL SYSTEMS ENGINEERING
UNIVERSITY OF NEBRASKA-LINCOLN
LINCOLN, NEBRASKA 68583-0919

ABSTRACT

A pervaporation apparatus was designed and tested in an effort to develop an integrated fermentation and product recovery process for acetone–butanol–ethanol (ABE) fermentation. A crossflow membrane module able to accommodate flat sheet hydrophobic membranes was used for the experiments. Permeate vapors were collected under vacuum and condensed in a dry ice/ethanol cold trap. The apparatus containing polytetrafluoroethylene membranes was tested using buta-

* Published as Paper 10120, Journal Series, Nebraska Agricultural Experiment Station, Lincoln, Nebraska 68583-0704.
† To whom correspondence should be addressed. FAX: (402) 472-1693. Telephone (402) 472-2342.

nol–water and model solutions of ABE products. Parameters such as product concentration, component effect, temperature, and permeate side pressure were examined.

INTRODUCTION

Production of acetone and butanol via industrial fermentation has been relatively inactive for more than 30 years. Favorable economic conditions and environmental concerns, however, have led to a renewed interest in industrial acetone–butanol–ethanol (ABE) fermentation (1–5). Alternative recovery processes, to use in lieu of or in conjunction with distillation, have the potential to reduce recovery energy requirements and costs associated with several industrial fermentations (4–7). This is especially true with fermentations such as the ABE fermentation which yields dilute quantities of butanol and acetone (3).

The toxic nature of butanol is one of the factors that limits the productivity and viability of the producing organism (2, 3, 8). Continuous removal of butanol may enhance fermentation performance (2, 4, 5). Alternative recovery processes, such as membrane separations, have also been shown to reduce butanol recovery costs and increase fermentation productivity (4, 9–11). One membrane process in particular, pervaporation, may have the greatest potential for industrial application because of cost, selectivity, and ease of use (4, 5). A recent review of butanol recovery integrated with fermentation processes indicated that of the five methods, stripping, adsorption, extraction, pervaporation, and perstraction (membrane assisted liquid–liquid extraction), the two with the greatest potential on a large-scale are liquid–liquid extraction and pervaporation (12).

Pervaporation, as described by Boddeker (13), denotes the transfer of matter from the liquid phase to the vapor phase through a nonporous polymeric membrane. One theoretical approach to describe this phenomena is the solution-diffusion model which consists of three steps: 1) sorption of the permeant from the feed liquid to the membrane, 2) diffusion of the permeant in the membrane, and 3) desorption of the permeant from the membrane as a vapor. A more recent transport model proposed by Okada and Matsuura describes pervaporation on the basis of a pore-flow model (14). The difference between the two models is that the pore-flow model denotes a boundary between the liquid and gas phase at some distance from the membrane surface in the pore of the membrane, while the solution-diffusion model does not incorporate a phase change. The pore-flow model incorporates the surface-flow model of Gilliland et al. (15). An adsorption layer of vapor molecules on the pore wall flows down the length of the pore without superimposition of any other gas flow. The

pore model loses validity when the pore size is so large that there is considerable contribution from Knudsen flow.

Microporous hydrophobic membranes have been investigated for pervaporation of alcohols derived from fermentation, though they do not adhere to the definition of Boddeker (13) as being nonporous. Microporous polytetrafluoroethylene (PTFE) (16, 17) and polypropylene (PP) membranes (18, 19) have been used for the pervaporation of ethanol. Friedl et al. (20) investigated the pervaporation of ABE from immobilized *Clostridium acetobutylicum* using a 0.2-μm PP hollow fiber module.

The objectives of this research were threefold: 1) to determine the flux and selectivity of PTFE microporous membranes for the pervaporation of model solutions, 2) to determine the effect of individual components on the flux and selectivity of butanol using model solutions, and 3) to evaluate a commercially available plate and frame crossflow filtration system for pervaporation.

MATERIALS AND METHODS

Configuration of the Pervaporation System

The pervaporation system consisted of an X-Flo crossflow membrane module (Bio-Recovery Inc., Hoboken, New Jersey). The module consisted of two stainless steel plates, 20 cm × 15.2 cm × 2.5 cm each, and a solvent-resistant membrane support plate, 20 cm × 15.2 cm × 1.1 cm, made of polyetheretherketone (PEEK) placed between the two stainless steel plates. The channel depth of the support plate was 0.16 cm. The membrane support plate was able to accommodate one flat sheet membrane on each side of the support plate for a total membrane area of 0.02 m^2. Compression bolts and two stainless pins held the membranes in place (Fig. 1).

A positive displacement rotary vane pump (PROCON Products, Murfreesboro, Tennessee) recirculated the feed solutions with a capacity of 15 L/min. A shell/tube heat exchanger connected to a Model 8000 IsoTherm recirculating water bath (Fisher Scientific, Pittsburgh, Pennsylvania) provided temperature control. Permeate vapor was drawn through $\frac{1}{8}$ in. PEEK tubing (Upchurch Scientific, Oak Harbor, Washington) by a Model 75 Precision vacuum pump (Precision Scientific, Chicago, Illinois) or a Model DD 100 Precision vacuum pump. The permeate passed through a series of two dry ice/ethanol cold traps. Glycerol filled gauges (Dresser Industries, Stratford, Connecticut) measured feed pressures and permeate side vacuum. Vacuum was regulated with a Model VR3600 vacuum regulator (Squire-Cogswell Company, Northbrook, Illinois). A diagram of the pervaporation system is shown in Fig. 2.

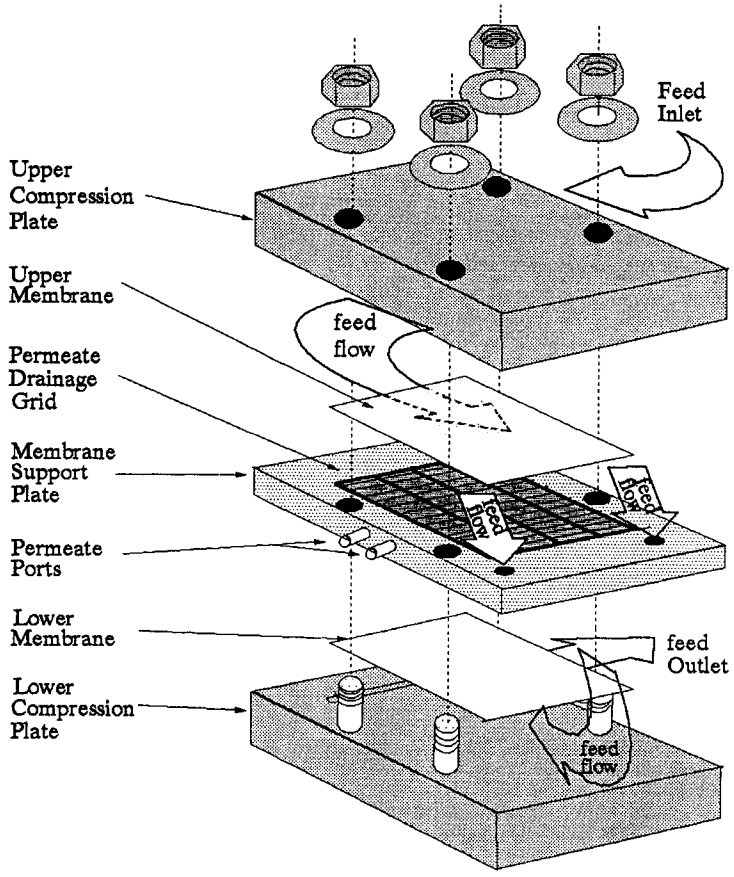

FIG. 1 Schematic of X-Flo membrane module.

Membranes

Polytetrafluoroethylene microporous flat sheet membranes were obtained from Bio-Recovery, Inc. The PTFE active layer was supported with polypropylene backing, and three pore sizes were tested: 0.10, 0.20, and 0.45 μm. All membranes were precut to fit the X-Flo membrane support plate.

Acetone–Butanol–Ethanol Solutions

The model acetone–butanol–ethanol (ABE) solution consisted of reagent-grade butanol, acetone, butyric acid, acetic acid, and ethanol (Al-

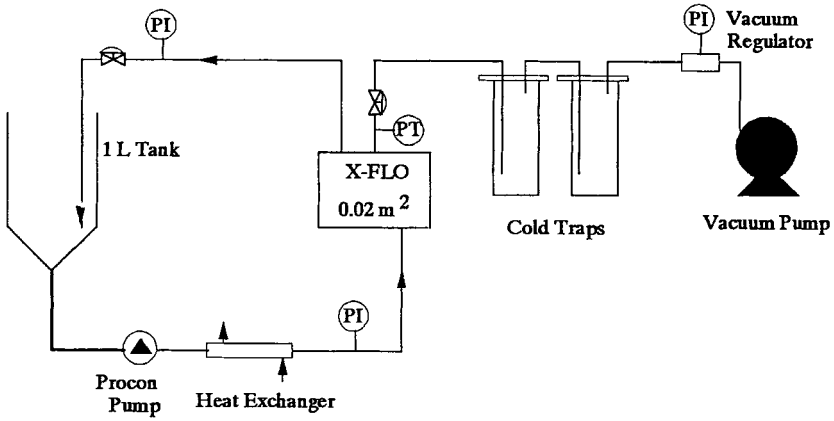

FIG. 2 Configuration of pervaporation system.

drich, Milwaukee, Wisconsin) at the following concentrations (v/v %), respectively: 1.25, 0.83, 0.18, 0.16 and 0.10%. The organic acids were added as free acids. These concentrations reflect ordinary ABE fermentation product proportions as reported by Soucaille (8). The solutions were adjusted to pH 5.3 with 10 N NaOH.

Pervaporation Procedure

Membranes were first seated in the membrane module with distilled water as recommended by Bio-Recovery, Inc. The inlet and outlet pressure was maintained at 10 psig, and the water was allowed to recirculate for 15 minutes. The distilled water was pumped out of the system. To determine if membranes were operating within set parameters, a standard flux ($L/m^2 \cdot h$) was performed with a 1.25% butanol solution for 0.5 hour. The standard operating parameters were: recirculation rate, 3 L/min; temperature, 39°C; and permeate vacuum, 100 mmHg. Experiments other than the standard flux lasted 2 hours. Permeate and retentate samples were taken every hour. After three experiments the standard flux was repeated to determine membrane performance as defined by the selectivity (α):

$$\alpha_{ij} = (Y_i/Y_j)/(Z_i/Z_j)$$

where i = solvent volume and j = total volume of the permeate (Y) and the retentate (Z). Selectivity is the ratio of solvent in the permeate over the ratio of solvent in the retentate (4). Flux was measured by dividing

the permeate volume by the membrane area × time, with the units of liters/(meter)^{2}hour.

Each parameter was repeated in triplicate, i.e., temperature, product concentration and composition, membrane material, flow rates, and permeate side pressure. Temperatures examined were from 30 to 55°C. This range was chosen because the optimum growth temperature of the ABE fermentation (37–39°C) falls approximately midway in the range (3).

The effect of membrane flux and selectivity as a function of the butanol concentration, 0.3 to 3.0% (v/v), was investigated. This range was chosen because a typical fermentation yield of butanol is about 1.25% (v/v) (1, 3, 8). The recirculation rate ranged from 2 to 8 L/min. The effect of each ABE component on the flux and selectivity of butanol was also examined.

Three different permeate side pressures, 250, 100, and 60 mmHg vacuum, were investigated. Pressures higher than 250 mmHg resulted in very low fluxes which were difficult to measure accurately.

Analytical Methods

Permeate and retentate sample concentrations were determined by gas chromatography (GC) using a Hewlett-Packard 5890 Series II (Avondale, Pennsylvania) with a flame ionization detector, a Hewlett-Packard 3396 A integrator, and a 2 m × 2 mm i.d. glass column packed with Chromosorb 101, 80/100 mesh size. GC conditions and column preparation were based on conditions described by Soucaille et al. (8). Oven temperature was 155°C (isothermic), and injector and detector temperatures were 200 and 350°C, respectively. Nitrogen carrier gas flow was 20 mL/min. Retentate samples were diluted twofold and permeate samples 10-fold with the internal standard, 50 mM isobutyric acid, prior to gas chromatography analysis.

RESULTS AND DISCUSSION

Membrane Comparisons

Results using three different pore size PTFE membranes, 0.1, 0.2, and 0.45 μm, are shown in Table 1. The data showed that 0.2 and 0.45 μm PTFE membranes had comparable selectivities. The flux rate was higher for the 0.45-μm membrane with the butanol/water solution, while lower with the model ABE solution. The 0.2-μm membrane experienced a higher flux rate with the ABE solution as compared to the 0.45-μm membrane. The 0.1-μm PTFE membrane was unable to prevent feed solutions from permeating through the membrane, and it wetted when a vacuum was applied to the permeate side. The fact that the 0.1-μm membrane wetted

TABLE 1
Comparison of Microporous PTFE Membrane

Pore size (μm)	Flux (LMH)		Selectivity	
	Model ABE	BuOH/H_2O	Model ABE	BuOH/H_2O
0.1	Wetted	ND[a]	ND	ND
0.2	0.98 (0.16)	0.17 (0.08)	9.5 (1.08)	8.5 (2.40)
0.45	0.41 (0.04)	0.54 (0.023)	10.4 (1.98)	10.4 (1.41)

[a] ND = not determined.

was unexpected. When the thicknesses of the PTFE membranes were measured, it was found that the 0.1-μm PTFE pore size was less thick (12 μm) compared with the 0.2 and 0.45 μm pore sizes which had thicknesses of 40 and 25 μm, respectively. This may be attributed to its inability to selectively permeate solvents. A thicker membrane should be more resistant to water permeation. Hickey and Slater showed that water flux increased as membrane thickness decreased, though selectivity stayed constant throughout the range of thicknesses examined (21). Nakao et al. stated that hydrophobicity is a very important property of a microporous membrane. If the membrane is not sufficiently hydrophobic, feed solution leakage will occur (17).

The 0.2-μm membrane was selected for the rest of the studies since it gave the highest selectivity for the model ABE solution. The 0.2-μm membrane should also be less resistant to plugging/fouling when dealing with whole broth systems.

Effect of Flow Rate and Permeate Side Pressure

Experiments were performed using recirculation rates ranging from 2.20 to 8.0 m/s and permeate side pressures of 250, 100, and 60 mmHg vacuum. There were no measurable differences in flux and selectivity for the recirculation rates examined (data not shown). Matsumura et al. (11) reported similar findings with butanol/isopropanol solutions in water, indicating that concentration polarization did not occur under their conditions.

As shown in Table 2 for a butanol/H_2O solution, a permeate side pressure higher than 100 mmHg caused the flux to drop dramatically. Butanol selectivity also dropped, but not as dramatically as the flux. A similar trend was seen by Hickey et al. (22) for an ethanol–water system over a pressure range from 1 to 40 mmHg when using a nonporous poly[1-(trimethylsilyl)-1-propyne] (PTMSP) membrane. The ethanol selectivity varied by approximately 20% while the flux decreased 10-fold as the pres-

TABLE 2
Effect of Permeate Side Pressure on Flux and Butanol Selectivity of a Butanol/H_2O Mixture

Temperature (°C)	Permeate pressure (mmHg)	Flux (LMH)	Selectivity
38	250	0.018	6.6
38	100	0.170	8.5
38	60	1.040	12.7

sure increased from 1 to 40 mmHg. Because the permeate side pressure provides the driving force for this process, a lower flux would be expected. Boddeker (13) stated that the effect of permeate pressure on pervaporation performance is dictated by the magnitude of the vapor pressures encountered and by the difference in the vapor pressure between them.

Effect of Butanol Concentration on Flux and Selectivity

The concentration of butanol in the ABE model solution and butanol/water solution was varied from 0.3 to 3% (v/v). The total flux of the model ABE solution gradually increased from 0.75 L/m^2·h at 0.3% to 1.01 L/m^2·h at 1.75% and then dropped to 0.58 L/m^2·h at 2.50 v/v % (Table 3). The selectivities of the ABE model solution started at 14.5 (0.3%), dropped to 9.5 (1.25%), and increased to 15 (1.75%) and remained relatively constant up to a butanol concentration of 3.0%. The total flux of the butanol/water mixture was consistently less than the ABE mixture and remained relatively constant at 0.15 L/m^2·h from 0.30 to 1.25% butanol. The flux increased to 0.48 L/m^2·h at 1.75%, maintaining this flux rate up to a butanol concentration of 3.0%. The selectivity of the PTFE membrane for the butanol/water system increased from 4.8 to 8.5 over a butanol concentra-

TABLE 3
Effect of Butanol Concentration on Flux and Butanol Selectivity

BuOH concentration (v/v %)	Flux (LMH)		Selectivity	
	Model ABE	BuOH/H_2O	Model ABE	BuOH/H_2O
0.30	0.75 (0.05)[a]	0.15 (0.02)	14.5 (1.78)	4.8 (0.96)
0.50	0.68 (0.11)	0.14 (0.07)	10.0 (1.32)	5.6 (0.93)
1.25	0.98 (0.16)	0.17 (0.08)	9.5 (1.08)	8.5 (2.35)
1.75	1.01 (0.06)	0.48 (0.07)	15.0 (0.25)	3.9 (0.35)
2.50	0.58 (0.13)	0.54 (0.13)	12.4 (1.67)	2.6 (0.20)
3.00	0.53 (0.04)	0.49 (0.09)	14.8 (2.40)	2.7 (0.82)

[a] Standard deviation.

tion range of 0.3 to 1.25% and then decreased to 3.9 at 1.75% butanol with no significant difference in selectivity (~2.6) between 2.5 and 3.0% butanol. Hickey et al. (22) reported a linear increase in both total flux and butanol flux for both *n*-butanol and *t*-butanol over a concentration range of 1 to 10% (w/w) with nonporous PTMSP. Nakao et al. (17), however, showed that ethanol flux with microporous PTFE membranes was independent of ethanol concentration.

Based on these results, it appeared that one or a number of components of the ABE model solution influenced the flux and selectivity of butanol.

Effect of Individual Components

In Table 4 the effects of individual fermentation product components on butanol flux and selectivity are shown. The butanol concentration remained constant (1.25%) as each of the ABE components was varied up to three times the standard concentration. The total flux remained relatively constant except when acetone was added. The total flux rate increased as the concentration of acetone increased. The selectivity of butanol in the presence of acetone appeared to reach a maximum at 0.83% (v/v) while the selectivity in the presence of acetic acid reached a minimum at 0.15% (v/v). Butyric acid appeared to have a negative effect on butanol selectivity as the concentration increases.

The effect of individual components on the flux of butanol may be attributed to flux coupling. The concept of flux coupling has been described by Mulder and Smolders (23) and Yeom and Haung (24). According to Mulder and Smolders, given a binary mixture, the flux is not only dependent on solubility and diffusivity in the membrane, but the flux of compo-

TABLE 4
Effect of Acetone, Acetic, and Butyric Acid on the Flux and Selectivity of Butanol[a] Using a 0.2-μm PTFE Membrane

Component	v/v %	Flux (LMH)	Selectivity
Acetone	0.27	1.63 (0.01)[b]	15.6 (0.28)
	0.83	1.80 (0.01)	24.7 (1.1)
	2.40	2.22 (0.12)	16.1 (5.44)
Acetic acid	0.05	1.84 (0.01)	25.0 (0.85)
	0.15	1.73 (0.06)	18.2 (2.82)
	0.45	1.73 (0.10)	25.2 (3.39)
Butyric acid	0.06	1.65 (0.01)	18.3 (0.78)
	0.18	1.72 (0.08)	17.9 (5.02)
	0.54	1.58 (0.07)	10.0 (1.13)

[a] Butanol concentration is 1.25% (v/v).
[b] Standard deviation.

nent i can be dependent on the diffusivity of component j. In cases of strong coupling, one component can be dragged along with the other. This could explain the increased selectivity of butanol in the presence of acetone and the decreased selectivity in the presence of butyric acid. Efforts are ongoing to determine if this is indeed occurring in this system.

Effect of Temperature

The model ABE solution showed a flux increase of approximately 10-fold, while the butanol/water solution showed a 60-fold increase over a temperature range from 30 to 55°C (Table 5). Figure 3 indicates an Arrhenius effect of temperature on the permeation of butanol through the membrane within the temperature range examined. It is interesting to note that the flux of the ABE solution was less temperature-sensitive than was the butanol/water system. Hickey et al. (22) reported an Arrhenius effect on the permeation rate for an ethanol/water system using nonporous PTSMP membranes.

Selectivities for the ABE system increased slightly with increasing temperature, while selectivities for butanol/water solutions were variable (Table 5). Butanol selectivities for the ABE model solutions were less variable, except at 50°C. Nguyen (25) stated that the relationship between temperature and membrane selectivity depends on the phenomenon that governs selectivity. If it is due to sorption, then selectivity will decrease with increasing temperature because the heat of sorption of the preferentially sorbed component would be negative while sorption of another component might be isothermal or endothermic. If selectivity is due to a difference in diffusion rates, then selectivity would increase with increasing

TABLE 5
Effect of Temperature on Flux and Butanol Selectivity[a]

	Flux (LMH)		Selectivity	
Temperature (°C)	Model ABE	$BuOH/H_2O$	Model ABE	$BuOH/H_2O$
30	0.19 (0.01)[b]	0.035 (0.03)	7.2 (0.70)	5.6 (2.35)
35	ND[c]	0.053 (0.01)	ND	3.7 (0.78)
40	0.98 (0.16)	0.170 (0.08)	9.5 (1.08)	8.5 (2.35)
45	1.23 (0.15)	0.680 (0.08)	8.1 (1.48)	12.9 (3.10)
50	1.79 (0.09)	0.805 (0.16)	13.7 (1.67)	9.9 (3.28)
55	ND	2.10 (0.07)	ND	5.2 (0.78)

[a] Permeate side pressure was 100 mmHg and 1.25% $BuOH/H_2O$ solution.
[b] Standard deviation.
[c] Not determined.

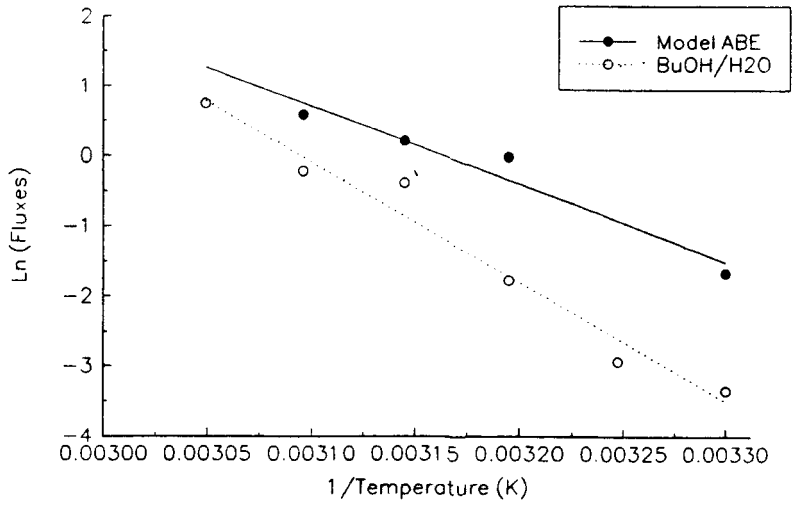

FIG. 3 Arrhenius plot of flux vs temperature for a 0.2-μm PTFE membrane on butanol/water and ABE model solutions.

temperature (25). Based on the data presented here, microporous PTFE membranes do not fall into either of these categories.

CONCLUSIONS

The recirculation rate of the pervaporation system did not affect butanol flux and selectivity at the rates examined in this study. Recirculation rate may be a factor when processing fermentation broths because of the potential fouling or polarization problems which have been reported in membrane systems that have been coupled to fermentation (17).

A tenfold difference in butanol concentration did not affect flux or selectivity in the model ABE solutions. Overall, the butanol/water system experienced lower selectivities as compared to the model ABE system. This indicates that the presence of the ABE mixture enhances the flux and selectivity of the PTFE membranes, suggesting flux coupling between the components. Nguyen (25) stated that a component that interacts strongly with the membrane polymer can allow components with less affinity to penetrate. Temperature had an obvious effect on the performance of the pervaporation system. By increasing the temperature, the flux increased by over 10-fold for model ABE solutions and 60-fold for butanol/water solutions. The increase followed an Arrhenius-type relationship.

It would appear that butyric acid has a negative effect on selectivity,

though at this time there is no explanation. Future research will focus on further understanding the interaction of butyric acid and butanol.

ACKNOWLEDGMENTS

This research was supported by the University of Nebraska Institute of Agriculture and Natural Resources, Industrial Agricultural Products Center, and the Nebraska Corn Development, Utilization, and Marketing Board.

REFERENCES

1. G. M. Awang, G. A. Jones, and W. M. Inlgedew, *CRC Crit. Rev. Microbiol.*, *15*, S33 (1988).
2. B. M. Ennis, N. A. Gutierrez, and I. S. Maddox, *Process Biochem.*, *21*, 131 (1986).
3. D. T. Jones and D. R. Woods, *Microbiol. Rev.*, *50*, 484 (1986).
4. S. A. Leeper, "Membrane Separations in the Production of Alcohol Fuels by Fermentation," in *Membrane Separations in Biotechnology* (W. C. McGregor, Ed.), Dekker, New York, 1986.
5. I. S. Maddox, *Biotech. Gen. Rev.*, *7*, 189 (1989).
6. M. Matsumura and H. Kataoka, *Biotech. Bioeng.*, *30*, 887 (1987).
7. M. Matsumura, H. Kataoka, M. Sueki, and K. Araki, *Bioprocess Eng.*, *3*, 93 (1988).
8. P. Soucaille, G. Joliff, A. Izard, and G. Goma, *Current Microbiol.*, *14*, 295 (1987).
9. W. J. Groot, M. C. H. den Reyer, T. Baart de la Faille, R. G. J. M. van der Lans, and K. Ch. A. M. Luyben, *Chem. Eng. J.*, *46*, B1–B10 (1991).
10. M. A. Larrayoz and L. Puigjaner, *Biotechnol. Bioeng.*, *30*, 692 (1987).
11. M. Matsumura, S. Takehara, and H. Kataoka, *Ibid.*, *39*, 148 (1992).
12. W. J. Groot, R. G. J. M. van der Lans, and K. Ch. A. M. Luyben, *Process Biochem.*, *27*, 61 (1992).
13. K. W. Boddeker, *J. Membr. Sci.*, *51*, 259 (1990).
14. T. Okada and T. Matsuura, *Ibid.*, *59*, 133 (1991).
15. E. R. Gilliland, R. F. Baddour, and J. L. Russel, *AIChE J.*, *4*, 90 (1958).
16. R. L. Calibo, M. Matsumura, J. Takahashi, and H. Kataoka, *J. Ferment. Technol.*, *65*, 665 (1987).
17. S. Nakao, F. Saitoh, T. Asakura, K. Toda, and S. Kimura, *J. Membr. Sci.*, *30*, 273 (1987).
18. K. Masumoto, H. Ohya, and M. Daigo, *Membrane*, *10*, 305 (1985).
19. K. Matsumoto and H. Ohya, in *Proceedings of the 3rd World Congress of Chemical Engineering*, 1986, p. 843.
20. A. Friedl, N. Qureshi, and I. S. Maddox, *Biotechnol. Bioeng.*, *38*, 518 (1991).
21. P. J. Hickey and S. Slater, *Sep. Purif. Methods*, *19*, 93 (1990).
22. P. J. Hickey, F. P. Juricic, and C. S. Slater, *Sep. Sci. Technol.*, *27*, 843 (1992).
23. M. H. V. Mulder and C. A. Smolders, *Ibid.*, *26*, 85 (1991).
24. C. K. Yeom and R. Y. M. Huang, *J. Membr. Sci.*, *67*, 39 (1992).
25. T. Q. Nguyen, *Ind. Membr. Proc.*, *82*, 1 (1986).

Received by editor October 8, 1992
Revised February 5, 1993