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The Removal of Fatty Acids from Edible Oil. Removal of the Dispersed Phase of a Water-in-Oil Dispersion by a Hydrophilic Membrane

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Abstract

Fatty acids can be extracted from an oil phase by forming a dispersed phase of saponified fatty acids/water/isopropanol in oil. This dispersion can be separated in the two phases by two membranes of opposite polarity in series. In this study the separation of the water phase from the dispersion by a hydrophilic membrane and the mechanisms underlying the flux characteristics are investigated. The permeation flux through a PAN ultrafiltration membrane is optimized with respect to the fatty acid/water/isopropanol ratio. It appears that a 1:6.5:3 (v/v) ratio gives the highest flux [95 L/(m^2·h·bar)]. The dispersion at these conditions consists of a continuous oil phase as well as a continuous water phase between 20 and 65% water phase hold up. The flux/pressure curve shows a linear increase of the flux with pressure at low pressures (determined by the membrane resistance), followed by a maximum flux value for the case where the volume of the water phase present in the inflow is limiting. It is not possible to remove the water phase with membranes below a water phase hold up of 20%. At this hold up value the transition between a bicontinuous and a discrete dispersion also occurs.

INTRODUCTION

Fatty acids have to be removed from oils for different purposes. In refining procedures of edible oils, the free fatty acids (FFA) have to be removed as a contaminant (*1*, *2*) since too high levels of FFA will result in rancidity of the oil (*3*). In the enzymatic production of fatty acids from triglycerides, the reaction rate strongly decreases with an increasing fraction of fatty acids in the oil phase (*4*). To maintain a sufficiently high reaction rate, the fatty acids should be removed continuously.

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The classical method for the removal of fatty acids from an oil is so-called caustic refining. Soaps are formed by adding alkali to the oil, and the soapstock formed is consequently separated from the oil by high-speed separators (*5*). The most important problem occurring in this procedure is the inclusion of triglycerides in the soapstock. This amount will usually equal the amount of fatty acids saponified (*6*). This included oil is difficult to recover and is therefore considered to be a loss. Caustic refining of oils with a high fatty acid content will evidently introduce considerable losses of triglycerides. In enzymatic oil splitting, the soapstock phase contains the fatty acids, which are the products. As a recovery process for these fatty acids in the enzymatic oil splitting (*4*), caustic refining will result in a very poor product, containing about 50% fatty acids.

The crude oil losses inherent to caustic refining can be avoided by the use of other refining procedures such as distillation or steam distillation (*7*). The high temperatures needed for these processes may especially affect highly unsaturated fatty acids, either present as free fatty acids or in triglycerides. A membrane separation process might be a mild alternative for these processes. For the application of membranes for fat/fatty acid separation, three configurations can be envisaged. First, direct filtration with a retention based on molecular size differences can be used. This will be difficult because of the small differences in molecular weights. Second, an extraction mode can be applied. However, the extractants found in the literature (mostly alcohols) are not very specific (*8*, *9*) and will also introduce losses of crude oil. The third mode to apply membranes is the formation of a dispersion that can be separated in the two phases by a hydrophilic and a hydrophobic membrane in series. The two-membrane system for the separation of this dispersion is shown schematically in Fig. 1 (*10*). To separate one phase from a two-phase mixture, the membrane used has to fulfill the requirement that it is preferentially wetted by this phase. The phase that does not wet the membrane (exhibits a contact angle on the surface larger than 90°) can be retained in the case that the Laplace pressure is higher than the applied transmembrane pressure.

The separation of dispersions will largely depend on the type of dispersion to be separated. The most simple types of dispersions are emulsions of a dispersed aqueous phase in a continuous organic phase (a W/O emulsion) and of a dispersed organic phase in a continuous aqueous phase (an O/W emulsion). In the absence of a stabilizing agent (surfactant) or in the presence of sufficiently large droplets (in the order of 1 mm), these dispersions are not very stable and their separation into two phases is relatively easy. Other, more complicated, types of dispersions may also occur. Examples are dual emulsions and microemulsions. In dual emulsions the continuous phase is also present as small droplets in the dispersed phase

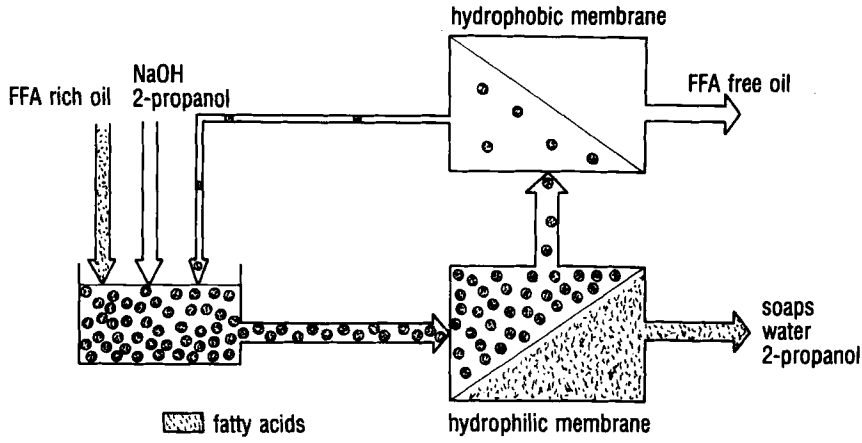

FIG. 1. Two membrane system for the removal of fatty acids from an oil.

(*11*, *12*). Microemulsions can be formed in the presence of a surfactant and a cosurfactant (usually an alcohol). They are thermodynamically stable water-in-oil dispersions with very small droplet sizes [of the order of 20 nm (*13*)]. Upon increasing the fraction of the dispersed phase in such a microemulsion system, a transition into a bicontinuous system can be observed (*14*). Several types of bilayer and liquid crystal structures have been found in this type of dispersed systems (*15*). However, no literature is available on the filtration characteristics of the more complicated types of dispersions.

In this study the soapstock obtained by the addition of alkali to a fatty-acid-rich oil is solubilized by an alcohol under the formation of a dispersion: soaps solubilized in water and alcohol form the dispersed water phase. The objectives of this study are the selection of a suitable membrane for the removal of the dispersed water phase from this dispersion and the establishment of the mechanisms underlying the permeation characteristics of the water phase.

MATERIALS

Dispersions are prepared by adding alkali to oleic acid containing soybean oil to saponify the oleic acid. Subsequently, isopropanol is added to solubilize the soaps, and water is added to lower the viscosity of the water/soap/alcohol phase. The soybean oil used is of edible quality (obtained from Rhenus Inc., The Netherlands). In this two-phase system the solubility of isopropanol in the oil phase is determined to be below 0.5%. To avoid the formation of insoluble calcium and manganese soaps, deminer-

alized water is used. All chemicals used were purchased from Merck (FRG) and are reagent grade. The dispersion is stored in a stoppered bottle to avoid the evaporation of isopropanol and is continuously stirred by means of a magnetic stirrer.

A qualitative analysis for fatty acids, its soaps, and triglycerides is made by means of TLC. The stationary phase is 0.2 mm Silicagel 60 F 254 purchased from Merck (FRG) and the mobile phase petroleum ether 40–60, diethyl ether, and acetic acid [80:20:1 (v/v/v)]. After developing, the sheets are colored with iodine vapor.

Several hydrophilic membranes have been tested for their capability to separate this dispersion. They are summarized in Table 1.

Flux measurements of the flat-sheet membrane are carried out in a New Brunswick Scientific Megaflow TM 100 test module. The membrane surface in this module is 64.5 cm^2 and the channel height is 0.4 mm. All membranes are rinsed thoroughly with demineralized water to remove preservative liquids before use.

The viscosity of the water phase has been determined by using an Ostwald viscometer. After phase separation, the interfacial tension between the two phases is measured by using a spinning drop tensiometer (*16*, *17*). A small drop of the low-density phase is brought into a rotating tube containing the high-density phase. The drop will deform along the axis of the tube. In case the length of the droplet is more than 4 times its height, the Vonnegut approach gives the following relation:

$$\gamma = \Delta\rho\omega^2 r_d^3/4 \tag{1}$$

in which γ is the interfacial tension between the two phases, $\Delta\rho$ is the density difference between the two phases, ω is the angular velocity, and r_d is the half height of the droplet.

To establish the transition from a continuous into a discrete oil phase, a thin layer of the pure oil phase (containing Sudan III to color it intensely red) is placed on top of a nonstirred dispersion. In the case of a continuous

TABLE 1
Hydrophilic Membranes Tested

Membrane	Type[a]	Pore size/cut off	Manufacturer
Cellulose	HF	6,000	ENKA
Cellulose acetate	HF	200,000	ENKA
Polyacrylonitrile (PAN)	FS	30,000	Rhone-Poulenc
Polyamide	HF	0.2 μm	ENKA

[a]HF = hollow fiber, FS = flat sheet.

oil phase, the red-colored oil moves from the top downward by diffusion and probably convection, whereas in the case of a discrete oil phase no movement of the colored oil downward occurs. The transition from a continuous into a discrete water phase is established by conductivity measurements. Conductivity measurements have been performed by using a 400 Hz ac current to avoid electrophoresis. All measurements have been carried out at 20°C, unless stated otherwise.

RESULTS AND DISCUSSION

Membrane Selection

The membrane selection experiments have been carried out with a dispersion containing 54% soybean oil, 10% sodium oleate, 14% water, and 22% isopropanol (v/v). The water phase contains the soaps, water, and 2-propanol, while the organic phase contains oil and only traces of 2-propanol. It is not possible to detect any soaps in the organic phase. The membranes given in Table 1 have been tested for their capability to separate this dispersion. The results are summarized in Table 2.

For all membranes, the permeate is examined for the presence of soaps and triglycerides by means of TLC. In the case of a cellulose and PAN membrane, no triglycerides could be detected in the permeate, thus indicating that the separation of the fatty acids from the triglycerides is complete. From Table 2 it can also be concluded that the pore size of the membrane influences the separation characteristics: the membranes with the large pore sizes could not retain the oil phase. However, the cellulose acetate and polyamide are expected to be slightly more hydrophobic compared to cellulose and PAN (*18*). This also might be part of the reason for the permeation of both phases. It is evident from Table 2 that the PAN membrane gives the best flux and a complete separation. Therefore, the PAN membrane was used for the flux optimization experiments.

TABLE 2
Fluxes Determined with the Standard Dispersion

Membrane	Flux [L/(m²·h·bar)]
Celulose	1
Cellulose acetate	—[a]
PAN	30
Polyamide	—[a]

[a]Both phases permeate.

Flux Optimization in the PAN Membrane

The clean water flux of the PAN membrane is found to vary from 300 to 700 L/(m^2·h·bar) depending on the sheet of material used. To allow a comparison of the results obtained with different sheets of the membrane material, the flux is standardized to a virtual clean water flux of 500 L/(m^2·h·bar), the average of the measured clean water flux of the sheets used. This correction is allowed, since in our system permeation of the water phase is determined entirely by the resistance of the membrane. This is shown in Fig. 2 in which the measured (noncorrected) flux is plotted versus the inverse of the viscosity of the water phase with different compositions. This appears to be a straight line through the origin. The clean water flux of this particular sheet is 580 L/(m^2·h·bar), which falls within the range of clean water fluxes measured. This means that although a dispersion is present at the retentate side of the membrane, the membrane is completely wetted by the water phase and the resistance against permeation is completely determined by only the hydrodynamic resistance of the membrane: there is no additional resistance at the retentate side.

To optimize the flux of the PAN membrane, the fatty acid to water and 2-propanol ratio is varied. The experiments are carried out at 1 bar transmembrane pressure. Varying the isopropanol content of the dispersion at a fixed water content results in a permeation flux and fatty acid flux as shown in Fig. 3. It can be seen that although the permeation flux increases with an increase in isopropanol content (due to a decrease in viscosity of the water phase), the fatty acid flux (this is the permeation flux times the fatty acid concentration) has an optimum value at an isopropanol to fatty

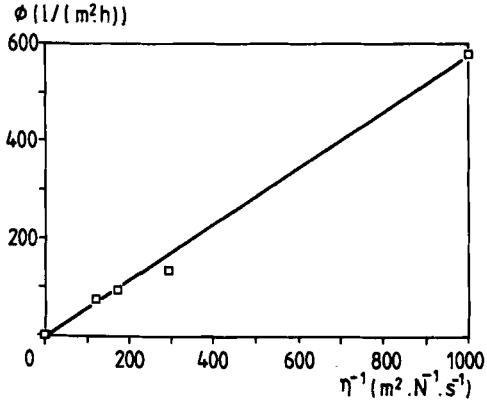

FIG. 2. Permeation flux through the PAN membrane versus the inverse of the viscosity of the water phase at 1 bar transmembrane pressure.

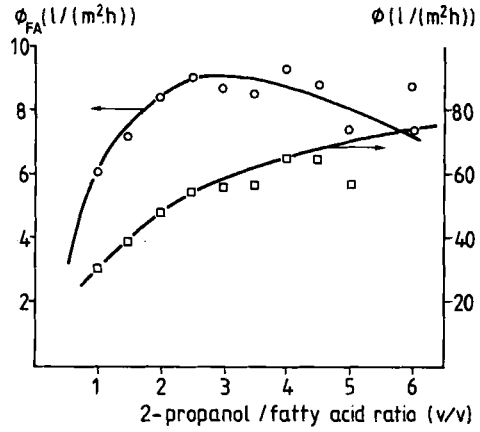

FIG. 3. Permeation flux and fatty acid flux through a PAN membrane at different isopropanol/fatty acid ratios (P = 1 bar).

acid ratio of 3:1. At this optimum isopropanol content the water content is varied and the same types of curves are obtained (Fig. 4). The optimum composition of the water phase with respect to the fatty acid flux appears to be a fatty acid, water, isopropanol ratio of 1:6.5:3. At this optimum composition of the water phase a permeation flux of 95 L/(m²·h·bar) can be achieved, resulting in a fatty acid flux of 15 L/(m²·h·bar). All experiments described further are carried out with this composition of the water phase.

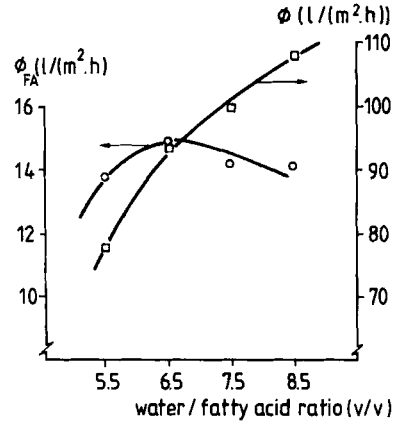

FIG. 4. Permeation flux and fatty acid flux through a PAN membrane at different water/fatty acid ratios at the optimum isopropanol content (P = 1 bar).

Although not performed here because it is not relevant for our purposes, the relation between flux and viscosity and therewith with the composition can easily be brought in an optimization model with the fatty acid flux as the parameter to be optimized.

Long-Term Performance of the PAN Membrane

In a series of consecutive batch experiments the performance of the PAN membrane has been investigated. Every new batch is started without cleaning the membrane. In these experiments the permeate is recirculated to the feed vessel. From Fig. 5 it can be seen that the flux gradually decreases from an initial flux of 105 L/(m^2·h·bar) to around 30 L/(m^2·h·bar) after 560 h. Every new batch initially gives a higher flux; however, after 1 day the flux decrease continues according to the pattern of the batch before. Rinsing the membrane with isopropanol for 3 h restores the flux to 53 L/(m^2·h·bar), a value comparable with the value after 60 h. Rinsing with nitric acid (0.1%), however, has no effect, indicating that the flux decay is probably due to clogging of the membrane with nondissolved soap molecules.

Characterization of the System

To reveal the nature of the dispersion, several experiments have been performed. It appeared to be impossible to estimate the particle size by microscopy. This indicates that a more complicated system than a water-in-oil dispersion is formed. This might be due to low interfacial tension. From spinning drop measurements at different angular velocities it follows that γ equals 0.27 ± 0.01 mN/m. This value is sufficiently low to form a

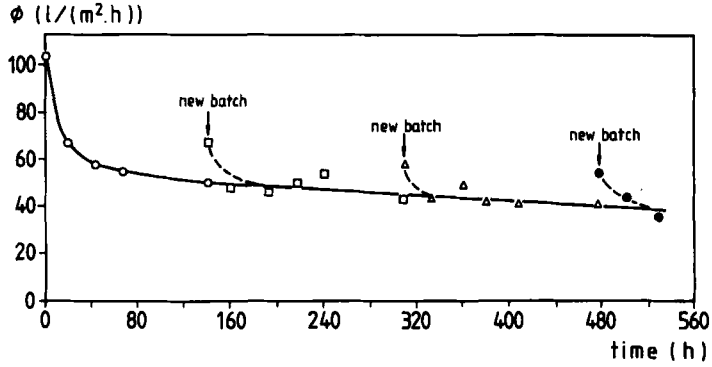

FIG. 5. Long-term performance of a PAN membrane in the separation of a two-phase system (P = 1 bar).

microemulsion. However, laser light-scattering experiments indicate that in the water phase as well as in the oil phase, no microemulsion is formed.

By performing conductivity measurements it can be seen (Fig. 6) that the conductivity of the system increases almost two decades stepwise at a water-phase content of around 20%. It also shows that no other stepwise increase in conductivity takes place, which indicates that water is present as a continuous phase above 20% water phase in the dispersion. The experiments with colored oil show a similar abrupt change around 35% oil phase. It can therefore be concluded that the oil phase is present as a continuous phase above 35% oil phase.

From these experiments it can be concluded that the system behaves similarly to the concentrated microemulsions used by Lagües (*19*), and it forms a bicontinuous system at a water-phase content between 20 and 65%. However, it has to be noted that bicontinuous in this context is not the same as the expression bicontinuous in the classical way: the dispersion is not transparent, and it definitely is not the result of a highly concentrated microemulsion (*20*, *21*). Lowering the fraction of the water phase below 20% will result in a transition from bicontinuous into a discrete emulsion containing water spheres in oil. According to Kirkpatrick (*22*), this transition is expected to take place between 15 and 29% dispersed phase in the system, based on a percolation theory approach. Above this threshold value, the conductivity is expected to increase according to a power law with an exponent of around 1.6 (*22*). However, from a log-log plot of the results given in Fig. 6, a maximum exponent of 0.8 follows, showing that the conductivity of the system increases less with an increase of the water-phase content than expected from the percolation theory.

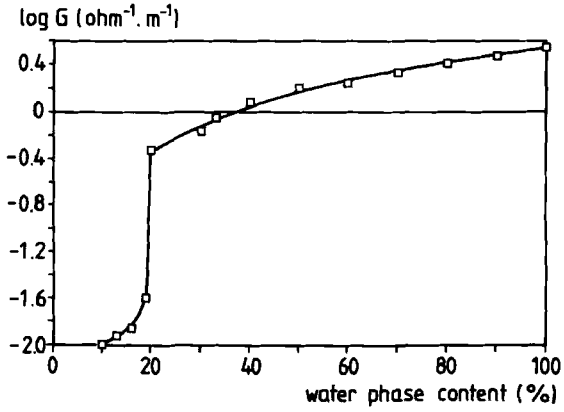

FIG. 6. Conductivity of the dispersion at different water phase contents.

It can be concluded that three regions can be distinguished for our system: between 0 and 20% water phase a dispersion of water droplets in oil is formed, between 20 and 65% water phase the water phase as well as the oil phase is present as a continuous phase, although it is not yet clear which type of bicontinuous system is present. Above 65% water phase a dispersion of oil droplets in water is formed.

Flux Versus Dispersed Fraction

In a batch experiment the water-phase content will decrease upon filtration. For a dispersion containing 45% water phase, the flux can be plotted versus the water-phase content, giving Fig. 7 as a typical result. The maximum flux found in this plot equals the flux in case the water phase only is applied as a feed solution on the same sheet of membrane, which is in agreement with the fact that the water phase is present as a continuous phase and is wetting the membrane completely. It is also found that the same curves are obtained in the case of sodium, potassium, and lithium soaps, and an increase in the temperature only influences the maximum flux, which is merely due to a decrease in viscosity of the water phase. In Fig. 7 a steep decrease of the flux can be observed below 20% water phase in the system, and the flux finally becomes zero at 18%. The same phenomenon has been observed in the case of the cellulose membrane, thus indicating that this stepwise flux decrease is a property of the dispersion rather than an effect caused by the membrane.

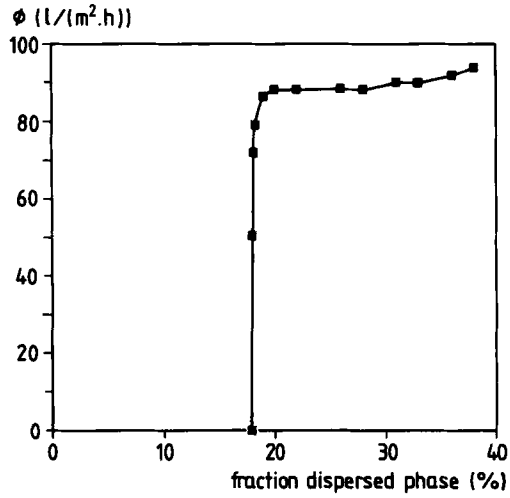

FIG. 7. The permeation flux through a PAN membrane varying with the fraction dispersed phase in the system.

This transition point coincides with the transition from a continuous water phase into a water phase consisting of discrete spheres in oil. Apparently the dispersed droplets cannot coalesce with the same phase present in the membrane. The size of the droplets is many times smaller than the height of the flow channel, and dispersed droplets will be lifted away from the surface because of the tubular pinch effect (*23*, *24*). This will result in an extremely low flux.

By removing the oil phase from this poor emulsion it is possible to obtain a dispersion with more than 20% water phase again. Consequently, the flux then can be restored to the value it has above 20% water phase. This can be done in the two-membrane system as proposed for the separation of this dispersion (*10*).

The Effect of Pressure and Flow Conditions on Filtration

For a filtration in which no concentration polarization occurs, the flux will be entirely determined by the membrane resistance. It has been shown above that the flux decreases linearly with an increase in viscosity. When at a fixed feed flow velocity the pressure is increased, the flux will be limited by the membrane resistance, and it will increase linearly with the transmembrane pressure:

$$\phi = \phi_{cw} A P \eta_w / \eta \tag{2a}$$

where ϕ is the permeation flux, and ϕ_{cw} is the clean water permeation flux at 10^5 N/m^2 transmembrane pressure and 1 m^2 membrane area, P is the transmembrane pressure, A is the membrane surface, η is the viscosity of the permeate, and η_w is the viscosity of water. A mass balance then yields

$$\phi = Q(f_{in} - f_{out})/A \tag{2b}$$

in which Q is the feed flow and f_{in} and f_{out} are the dispersed fractions in the feed flow and the flow leaving the system, respectively. From the experiments mentioned above it follows that f_{out} cannot be smaller than 0.2. This implies that the flux cannot increase above the value calculated from Eq. (2b) with $f_{out} = 0.2$:

$$\phi_{max} = Q(f_{in} - 0.2)/A \tag{2c}$$

In Fig. 8 it is shown that the predicted and measured permeation flux at conditions for which $f_{out} > 0.2$ (Eq. 2b) show good agreement at the condition of pressure-dependent permeation. From Fig. 9 it can be con-

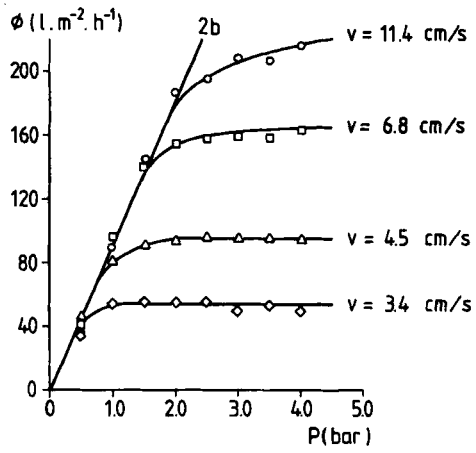

FIG. 8. Permeation flux through a PAN membrane at different feed flow velocities and transmembrane pressures.

cluded that the maximum permeation flux, i.e., for $f_{out} = 0.2$ (Eq. 2c), in this system is well described by the amount of water phase that is present in the inflow. Both figures confirm that the pressure-limited permeation at low transmembrane pressures (determined by the membrane resistance) and inflow limited permeation at high transmembrane pressure determine the flux/pressure curve.

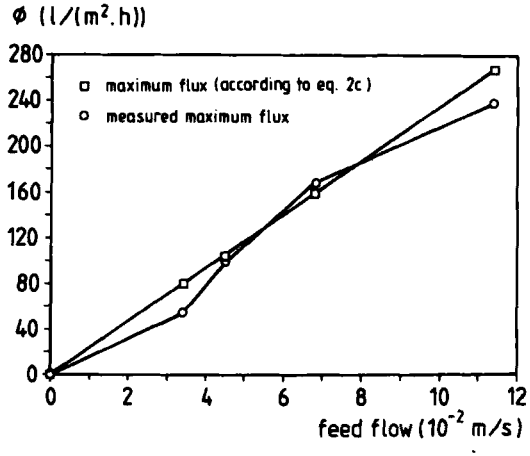

FIG. 9. Permeation flux through a PAN membrane at different feed flow velocities. The maximum flux is calculated according to Eq. (2c).

Prediction of the Membrane Performance

With the experimental data obtained it is possible to predict the performance of a membrane used in this separation. First, it has to be established whether a membrane is capable of retaining the oil phase or not. Subsequently, the performance can be determined as shown schematically in Fig. 10. A change in the composition of the water phase will result in a change in viscosity. By using the ratio of the viscosity of the water phase over the viscosity of pure water together with the clean water flux, the permeation rate can be calculated. A mass balance over the system gives the maximum flux that can be attained. Combining the concentration of the product in the water phase with the maximum flux finally results in the product flux.

CONCLUSIONS

From this work it is concluded that it is possible to separate fatty acids from an oil by forming a dispersion of saponified fatty acids/water/isopropanol in oil. It follows from conductivity and diffusion measurements that a bicontinuous system is formed between 20 and 65% water phase in the system. The filtration characteristics at a hydrophilic membrane have been investigated. The permeation flux through a PAN ultrafiltration membrane is optimized with respect to the fatty acid/water/isopropanol ratio. It follows that a 1:6.5:3 (v/v) ratio gives the highest flux [95 L/(m^2·h·bar)]. In these experiments the flux is limited by the amount of dispersion across the membrane. It appears that it is not possible to go below 20% water phase in the dispersion, which can be explained by a transition from a bicontinuous system into a discrete dispersion. This 20% value falls within

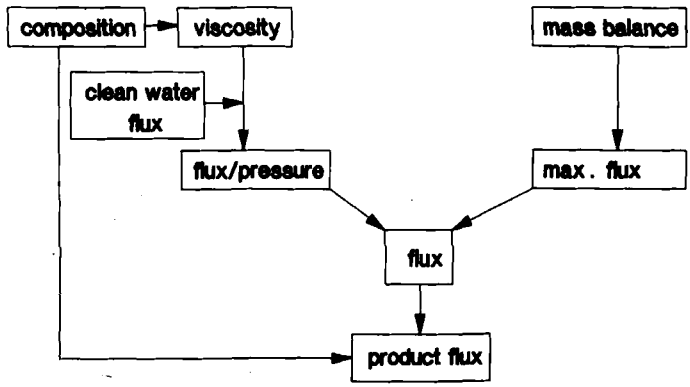

FIG. 10. Route for the prediction of the performance of a membrane for the removal of the water phase from the dispersion.

the range predicted by percolation theory for the transition between spheres and a bicontinuous system. However, by removing the oil phase by a hydrophobic membrane in series, it is possible to restore the water-phase content to a value above 20%.

SYMBOLS

A	membrane surface area (m^2)
f_{in}	dispersed fraction in feed (—)
f_{out}	dispersed fraction leaving the system (—)
G	conductivity ($ohm^{-1} \cdot m^{-1}$)
r_d	half droplet height (m)
P	transmembrane pressure (N/m^2)
Q	feed flux (m^3/h)
γ	interfacial tension (N/m)
ϕ	permeation flux ($m^3 \cdot m^{-2} \cdot h^{-1}$)
ϕ_{cw}	clean water flux ($m^3 \cdot m^{-2} \cdot h^{-1}$)
ϕ_{max}	maximum attainable permeation flux ($m^3 \cdot m^{-2} \cdot h^{-1}$)
η	viscosity (Ns/m^2)
η_w	viscosity of water (Ns/m^2)
Dp	density difference (kg/m^3)
ω	angular velocity (rad/s)

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