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Low-Frequency Ultrasound to Improve Dead-End Ultrafiltration Performance

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ABSTRACT

Low-frequency ultrasound (20 kHz) has been used to facilitate dead-end ultrafiltration of polymer solutions. The influence of ultrasonic irradiation on permeate fluxes and retention ratios was investigated and compared with conventional stirred and unstirred operations. In addition, the effect of the ultrasonic field on the membrane permeability and on the polymer solution was examined. The efficiency of ultrasound in enhancing ultrafiltration performance was demonstrated. This improvement is due to a decrease of the boundary layer resistance against permeation flow, which may be attributed to disturbance induced within the cell and at the membrane surface by physical effects associated with ultrasonic waves propagation.

Key Words. Dead-end ultrafiltration; Ultrasound; Flux enhancement; Boundary layer; Resistances-in-series model

INTRODUCTION

Separative membrane technologies have advanced significantly in recent years. Ultrafiltration (UF) is one of the very promising techniques. However, in practical applications, as the solute accumulates at the membrane surface, a

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progressive flux decline with time is observed. This phenomenon, known as concentration polarization, has been studied in many works and several models have been developed to account for this flux reduction (1–6). Another important problem that can occur during UF is membrane fouling caused by solute adsorption or blocking of pores (7, 8). These phenomena remain major disadvantages of the UF process; they affect performance by altering membrane permeability and selectivity.

A growing interest exists about technical advances that may overcome such limitations. Among the different approaches that have been considered, the use of ultrasonic waves appeared as a potentially effective means. Ultrasonic waves are mechanical vibrations operating at frequencies above the human hearing threshold, in the range of 18–100 MHz. As a result of ultrasound (US) propagation, a number of physical and mechanical phenomena, such as heating, radiation pressure, acoustic streaming, formation and subsequent collapse of cavitation bubbles, are generated in the medium (9). The benefits that ultrasonically induced effects can provide to a large variety of processes have been recognized for many years. Classical industrial applications include cleaning, sterilization, plastic welding, emulsification, and degassing (10). The use of US to improve separation technologies is a new field of application that is gaining importance; very promising results have been reported. Indeed, several-fold increases in diffusion rates were recorded as ultrasonic energy was supplied to dialysis operations (11–15). US was also found to efficiently enhance liquid flow through a wide variety of porous media (16–19) and microfiltration membranes (20–22).

Recently, it was shown that a similar increase in UF productivity can be brought about by ultrasonic irradiation. However, few detailed studies have been performed (23–26). Ahner et al. (23) used thin ceramic piezoelectric disks to intermittently transmit ultrasonic energy to polymeric UF membranes. The feasibility tests of the piezoelectric drivers were carried out on aqueous solutions of polyethylene glycol and dextran. By application of moderate power (40 W), over short periods of time (20–90 sec), enhancements as high as a factor of 8 were recorded. Kokugan et al. (24) also examined the effect of US on UF properties by irradiating about one-eighth of the permeation surface of a ceramic membrane in an ultrasonic cleaning bath. It was confirmed that ultrasonic irradiation can help to increase permeate flux whether a gel or a boundary layer was formed at the membrane surface. However, ultrasonic radiation was ineffective in removing fouling that occurred inside the pores. Sabri et al. (25) also investigated acoustically assisted cross-flow UF during the processing of a paper mill effluent. When the ultrasonic field was applied (40 kHz/40 W), higher flux values were achieved at low cross-flow velocities. Chai et al. (26) studied the UF process of various molecular weight

dextran solutions on a flat sheet polyacrylonitrile membrane immersed in an ultrasonic cleaner (45 kHz). The ultrasonic irradiation was found to significantly increase permeate fluxes, particularly because the phenomenon of concentration polarization was important. The enhancement was attributed to the increase in bulk mass transfer due to vibrations of the membrane caused by US.

Although the beneficial use of ultrasonic energy in filtration processes seems quite obvious according to these studies, the application of the ultrasonic field still has to be optimized. Moreover, the mechanisms involved in the reported enhancement are not yet fully understood.

The present work aims to determine the potential of low-frequency US to improve dead-end UF performance, and also to propose an interpretation of the observed improvement. Results reported in this paper illustrate the effect of US on fluxes and retention ratios, as well as the influence of some parameters such as sonication mode and ultrasonic power. The permeation resistances affected by ultrasonic irradiation are determined and the results are compared to those obtained in a mechanically stirred cell.

EXPERIMENTAL METHODS

Materials

Dextran, with an average molecular weight of 260 kDa (Sigma Chemical Co., St. Louis, MO), was chosen for the feed solution because this polymer is generally used as a model solute in ultrafiltration studies. Solutions at $0.3 \text{ g}\cdot\text{L}^{-1}$ were prepared in demineralized and microfiltered water.

UF membranes (PTTK type, Millipore, Bedford, MA) with a 30 kDa molecular weight cut-off and an effective filtration area of 39.6 cm^2 were used in all experiments. As specified by the manufacturer, these membranes have an asymmetric structure composed of a thin skin layer of polysulfone on a polypropylene support.

Equipment

The schematic diagram of the experimental apparatus is given in Fig. 1. It is composed of a batch cell ultrafilter (model 8400, diameter 7.5 cm and total volume $V_0 = 350 \text{ mL}$, Amicon, Millipore, Bedford, MA) that can be fitted (Fig. 1A) with a mechanical stirrer, the speed (ω) of which was measured with a digital tachymeter and fixed at $36.6 \text{ Rad}\cdot\text{sec}^{-1}$.

For UF experiments with US (Fig. 1B), the cell is equipped with a specially designed cover that enables us to introduce the ultrasonic emitter. Low-frequency US was produced by a classical probe system (3 mm diameter) operating at 20 kHz and supplied by a generator, the output of which can be var-

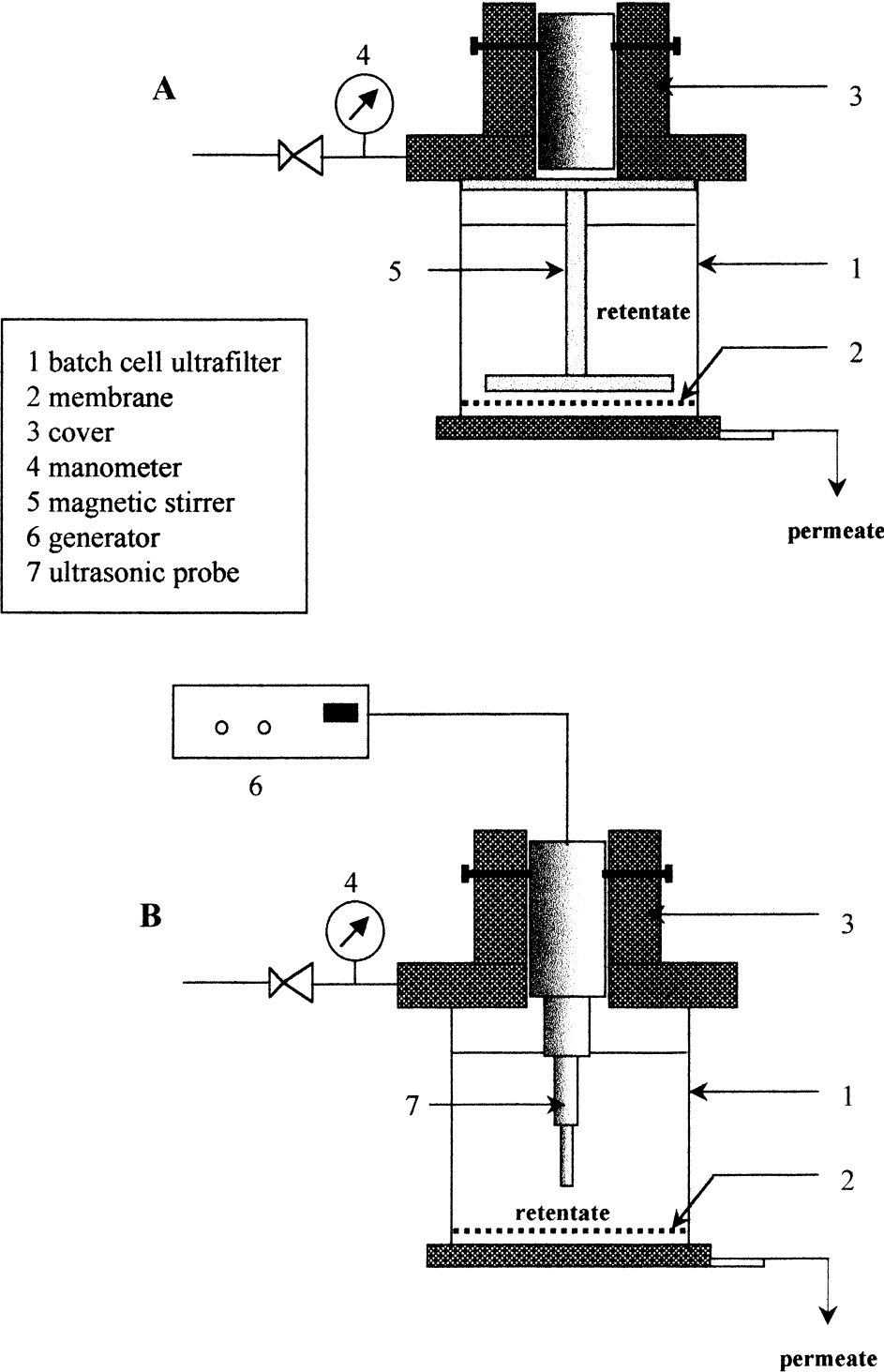


FIG. 1 Experimental setup: (A) for stirred UF, (B) for ultrasonically assisted UF.

ied from 20 to 100% of the maximal electrical power (40 W). The ultrasonic powers transmitted to the medium (P_{us}) were determined by calorimetric measurements (27). From this method, an approximate conversion yield of 50% was noted. For all experiments, the emitter was located at 14 mm from the membrane surface.

Procedure

The following procedure was employed to measure permeate fluxes under all experimental conditions:

1. The membrane permeability was determined by measuring the pure water flux (PWF) under different transmembrane pressures (ΔP),
2. The water was replaced by 350 mL of aqueous solution of dextran at 30°C. The cell was placed in a thermostated water bath in order to keep the operating temperature at $30 \pm 1^\circ\text{C}$ and then the cell was pressurized with nitrogen gas. The permeate flux (J_v) was obtained by weighing the permeate (m) collected at regular time intervals (Δt) and was calculated according to the following equation:

$$J_v = \frac{m}{\rho \cdot S \cdot \Delta t} \quad (1)$$

where ρ and S are the solution density and the membrane surface, respectively.

3. The solution of dextran was removed and the membrane was chemically cleaned (NaOH 0.1 M, HCL 0.1 M, ultrapure water) until the original PWF was restored.

The concentrations of the feed solution C_0 , of the retentate C_r , and of the permeate C_p , were determined by gel permeation chromatography (NaNO₃ 0.8 g·L⁻¹ solution as eluent Controller W600, Refractometer W410, Ultrahydrogel 1000 column, Waters, MilFord, MA). Total organic carbon determination was also used as a control analytical method (Dorhmann-Xertex DC 80, Martec, Sevres, France). The instantaneous observed retention ratio $R_{obs}(t)$ was then calculated, taking into account the variation of the initial solute concentration due to the batch mode of filtration:

$$R_{obs}(t) = \left(1 - \frac{C_p(t)}{C_b(t)} \right) \quad (2)$$

where the bulk concentration $C_b(t)$, was calculated as follows:

$$C_b(t) = \frac{C_0 V_0 - \sum C_p(t) V_p(t)}{V_0 - \sum V_p(t)} \quad (3)$$

EXPERIMENTAL RESULTS AND DISCUSSION

Preliminary Control Experiments

Prior to any filtration operation under sonication, control experiments were performed to investigate the effect of the ultrasonic field on the membrane and the polymer solution characteristics, respectively.

Effect of Ultrasound on Ultrafiltration Membrane

The influence of US on the membrane characteristics was investigated to ensure that no modification was induced by sonication. First, the PWF was measured as a function of the transmembrane pressure without US. A linear relationship between PWF and operating pressure was observed, as predicted by Darcy's law:

$$J_w = \frac{\Delta P}{\mu \cdot R_m} \quad (4)$$

where μ and R_m are the solution dynamic viscosity and the hydraulic membrane resistance, respectively. From the above equation, R_m was found to be equal to $1.1 \times 10^{12} \text{ m}^{-1}$. The PWF was then measured in the presence of the acoustic field and after the membrane was irradiated at the highest ultrasonic power for 2 hr.

Figure 2 shows that the PWF and thereby the hydraulic membrane resistance (R_m) remain unchanged in all cases. Therefore, note that ultrasonic waves do not enhance PWFs. Thus, it might be thought that the intrinsic per-

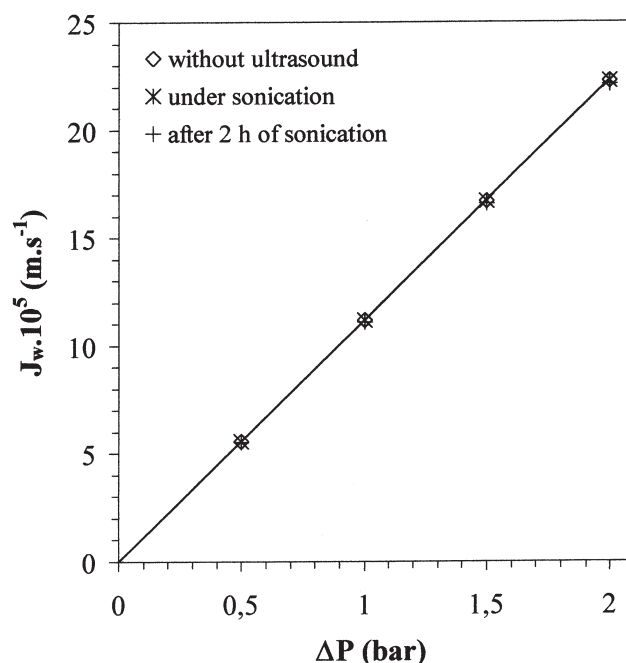


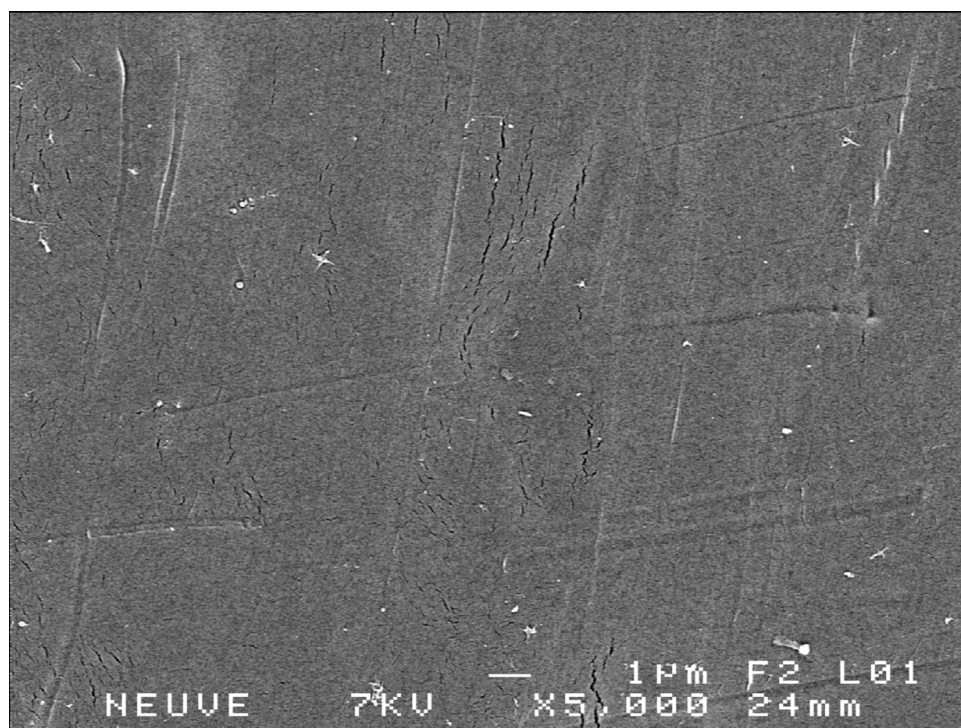
FIG. 2 Pure water flux (PWF) as a function of transmembrane pressure.

meation properties of the membrane are not modified by the presence of the low-frequency ultrasonic field and also are not altered by a long sonication time. Furthermore, scanning electron microscopy observations have confirmed this result; any structural difference between sonicated and fresh membranes could have been detected as illustrated by comparison of Photographs 1 and 2.

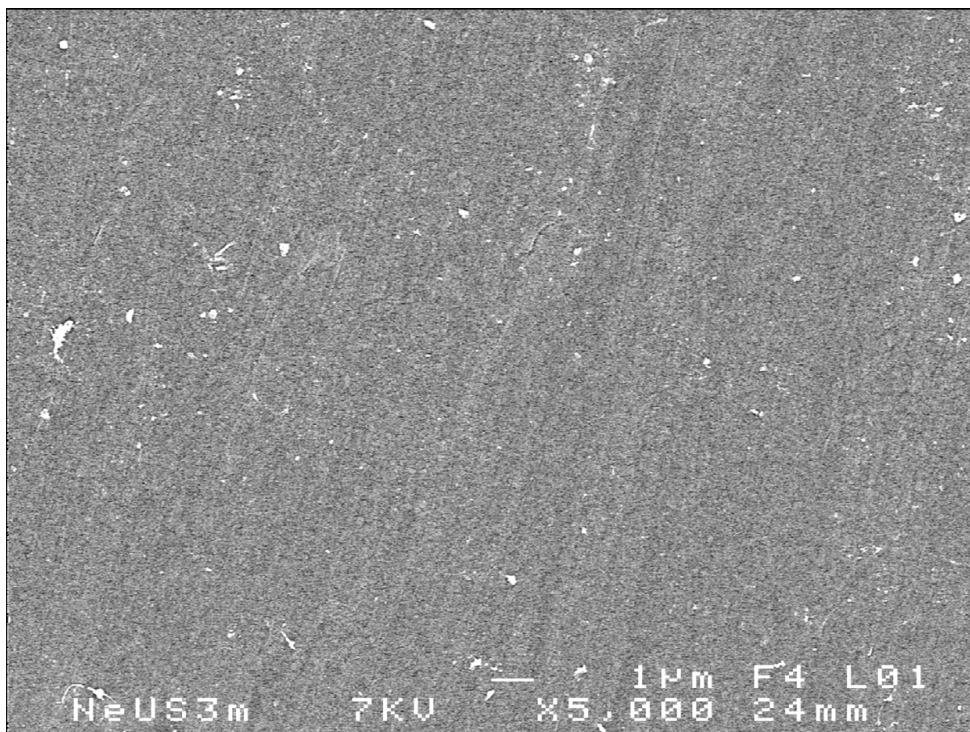
Effect of Ultrasound on Polymer Solution

As widely reported in literature, ultrasonic irradiation of polymer solutions results in a reduction of the solute molecular weight, i.e., depolymerizing effect. According to Eq. (4), such a phenomenon may induce a decrease in solution viscosity and thereby results in an enhancement of permeate fluxes. It was therefore of interest to investigate the effect of US on the aqueous solutions of dextran used in this study.

As previously mentioned (28), degradation experiments were performed in the UF cell and the influence of many parameters (such as sonication time, ultrasonic power, and volume and concentration of the polymer solution) were investigated. Despite the depolymerization effect on dextran molecules that was evidenced from this series of experiments, the polymer degradation was shown to be sufficiently slow that it could be neglected, owing to the time required for UF operation. Conversely, the polymer solution viscosity remained



PHOTOGRAPH 1. SEM observation of the surface of a fresh UF membrane (Millipore, PTTK type).



PHOTOGRAPH 2. SEM observation of the surface of an UF membrane (Millipore, PTTK type) after 2 h of exposure to ultrasound ($P_{us} = 18$ W).

unchanged even after sonication and was found to be that of the pure solvent (0.8×10^{-3} Pa·sec) because of the low polymer concentration.

Ultrafiltration Experiments

Effect of Ultrasound on Permeate Fluxes

Permeate fluxes versus filtration time for the three UF modes (with US, with a magnetic stirrer, and unstirred UF) are given in Fig. 3. In each case, an important reduction in flux compared to the PWF ($J_w = 5.2 \times 10^{-5}$ m·sec⁻¹) was observed at the beginning of the filtration run. This phenomenon is caused by the build-up of the polarized layer (4). Afterward, the permeate flux for unstirred UF continued to decrease sharply, whereas further decline was more gradual under mechanical stirring. In the presence of the acoustic field, the flux decline was considerably reduced compared to the unstirred UF, leading to a higher permeate flux, which remains constant throughout the UF experiment. To quantify the magnitude of the permeate flux improvement a theoretical “ultrasonic enhancement factor” (E_{us}) was defined as:

$$E_{us} = \frac{J_{v,ultrasound}/PWF_1}{J_{v,unstirred}/PWF_2} \quad (5)$$

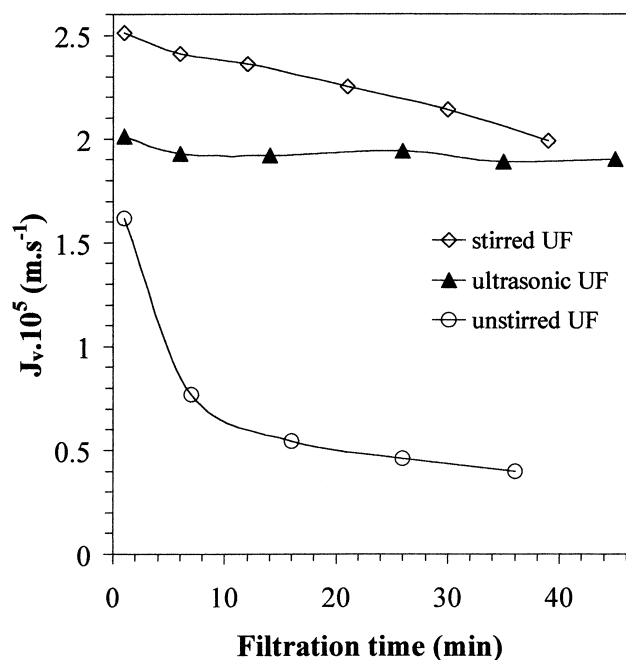


FIG. 3 Permeate flux J_v as a function of filtration time for the three operating configurations ($C_o = 0.3 \text{ g}\cdot\text{L}^{-1}$, $\Delta P = 0.5 \text{ bar}$, $P_{us} = 16 \text{ W}$).

where $J_{v,\text{ultrasound}}$ and $J_{v,\text{unstirred}}$ are the permeate fluxes achieved under ultrasonic and unstirred UF conditions. The corresponding values are those obtained for a given reference filtration time (26 min), at which one can consider that $J_{v,\text{unstirred}}$ has reached a “quasi-steady state.” PWF_1 and PWF_2 are the pure water fluxes measured before each experiment. In Eq. (5), they take into account possible modifications of the membrane permeability between different runs. Owing to the efficiency of the chemical procedure used for membrane cleaning, the initial PWF is systematically restored. Therefore, PWF_1 and PWF_2 are identical and E_{us} can then be calculated according to

$$E_{us} = \frac{J_{v,\text{ultrasound}}}{J_{v,\text{unstirred}}} \quad (6)$$

Effect of Ultrasound on Retention Ratios

From the same series of experiments, the observed retentions ($R_{\text{obs (26 min)}}$) were found to be 50.0, 89.0, and 95.1% for unstirred, ultrasonic, and stirred UF, respectively, indicating that the higher the permeate flux, the higher the solute retention. This trend can easily be explained for stirred UF because it is well known that the high shearing stress induced at the membrane interface by the mechanical stirring hinders the growth of the boundary layer (6, 29), thereby, both flux and retention ratio increase.

Because the permeate flux improvement achieved by means of US can be attributed neither to a modification of the membrane permeability nor to changes in the polymer solution characteristics, it might be supposed that similar mechanisms are involved in stirred and ultrasonically assisted UF.

Effect of the Sonication Mode

Further experiments were then conducted under continuous and pulsed sonication modes. As shown in Fig. 4, both modes effectively provided significant enhancement of permeate flux. When US is used intermittently, the permeate flux starts to increase as soon as ultrasonic energy is supplied to the system. Afterward, it reaches the same value as that under continuous application. Because sonication is suspended, J_v decreases and behaves as if under unstirred filtration. The same trend can be observed whenever the pulse is applied. Such a result tends to confirm the assumption that US may have a depolarizing effect. In fact, US seems effective both to prevent the establishment of part of the polarized layer when applied at the beginning of the filtration run and to disrupt a polarized layer previously formed when applied at a later time.

Our experimental results (see Fig. 4) indicate that the intermittent use of US appears less effective than continuous use. Although the permeate flux starts

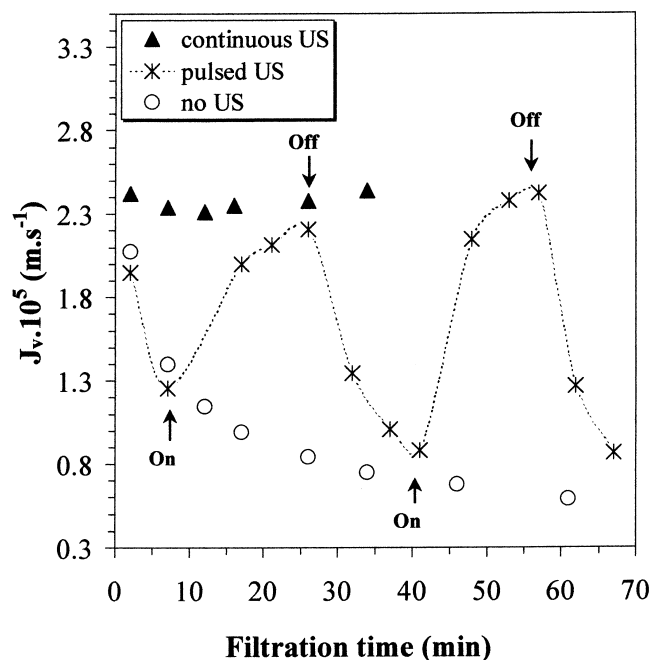


FIG. 4 Permeate flux J_v as a function of filtration time. Influence of the sonication mode ($C_o = 0.3 \text{ g}\cdot\text{L}^{-1}$, $\Delta P = 1 \text{ bar}$, $P_{us} = 16 \text{ W}$).

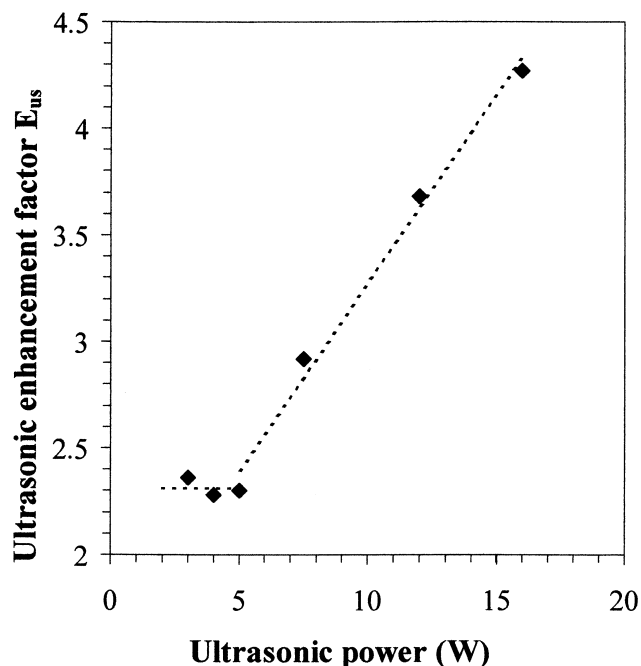


FIG. 5 Ultrasonic enhancement factor E_{us} as a function of ultrasonic power ($C_o = 0.3 \text{ g}\cdot\text{L}^{-1}$, $\Delta P = 0.5 \text{ bar}$).

to increase as soon as the pulse is applied, a recovery time of about 20 min is required to gain the same improvement as that observed under continuous sonication mode. Furthermore, the achieved flux enhancement is not maintained after the irradiation is suspended.

Effect of the Ultrasonic Power

As illustrated in Fig. 5 and Table 1, the efficiency of US in enhancing both UF permeate fluxes and retention rates strongly depends on the ultrasonic power. Below a power threshold of 5–6 W, the ultrasonic field provides the same enhancement E_{us} , and observed retention (R_{obs}) remains constant, whatever the ultrasonic power (P_{us}). Above this threshold, both flow and retention ratio increase quite linearly with the power in our experimental range.

TABLE 1
Variation of E_{us} (Dimensionless) and Observed Retention Rate (%) with Ultrasonic Power

	Ultrasonic power (W)					
	3	4	5	7.5	12	16
E_{us}	2.36	2.28	2.30	2.92	3.68	4.27
$R_{obs(26 \text{ min})}$	71.7	71.4	67.3	74.3	87.8	89.0

Influence of Ultrasound on Permeation Resistances

Experimental Determination of Permeation Resistances

Additional UF experiments were performed at different ultrasonic powers. The resistances-in-series model, which was shown to be suitable to predict the UF rate of macromolecular solutes such as dextran (30), was employed to express our experimental results. Such a model allows one to distinguish among those resistances affected by the sonication. According to this theory, the solvent flux is predicted to vary as follows:

$$J_v = \frac{\Delta P}{\mu \cdot R_{\text{tot}}} \quad (7)$$

where R_{tot} is the global resistance against solvent permeation. Under our experimental conditions, R_{tot} is the sum of the resistances induced by the membrane R_m , the boundary layer R_{bl} , and the fouling layer R_f :

$$R_{\text{tot}} = R_m + R_{\text{bl}} + R_f \quad (8)$$

This latest resistance, R_f , can be mainly attributed to pores blocked by dextran molecules. Thus, it can be considered as a resistance against permeation flow, which can only be suppressed after a cleaning procedure, whereas that induced by the boundary layer can be easily removed by washing away the cell.

To determine the value of each resistance, the experimental procedure described in the procedure section was modified as follows:

1. The PWF is measured to obtain R_m ;
2. dextran solution (350 mL) was ultrafiltered. Using Eq. (7), the permeate flux measured after a given filtration time leads to R_{tot} ;
3. The solution of dextran is removed from the cell, and the fluid boundary layer is washed away and replaced by water;
4. The PWF is measured once again immediately after the UF run, providing $(R_m + R_f)$;
5. The membrane is chemically cleaned until the original R_m value is restored before the next run.

According to this procedure, R_{tot} , R_m , and R_f can be obtained individually. The value of the resistance induced by the boundary layer R_{bl} can then be deduced from Eq. (8).

Influence of the Ultrasonic Power on Permeation Resistances

The contribution of each resistance against permeation flow is given in Table 2. From these experiments, the resistance induced by the boundary

TABLE 2
Contribution of Each Resistance (%) Against Permeation Flow

	Ultrasonic power (W)				
	5	7.5	10	12	16
R_m/R_{tot}	19.2	22.8	24.0	26.4	37.0
R_f/R_{tot}	9.6	7.0	10.6	10.2	12.8
R_{bl}/R_{tot}	71.2	70.2	65.4	63.4	50.2

layer, R_{bl} , was shown to be the major resistance against permeation flow as commonly observed with dextran. In comparison, the resistances induced by the membrane R_m and the fouling layer R_f were found to be minor.

As shown in Fig. 6, increasing the ultrasonic power led to a decrease of R_{tot} and hence to higher permeate fluxes. As expected, the hydraulic resistance of the membrane, R_m , was insensitive to US. The resistance induced by the fouling layer, R_f , was also unaffected by sonication and remained constant whatever the applied ultrasonic power. This demonstrates that US was not effective in removing fouling that occurred inside the pores, and that increasing ultrasonic power did not allow the foulant layer to be reduced. Therefore, flux enhancement was achieved as a consequence of a decrease of the boundary layer resistance R_{bl} .

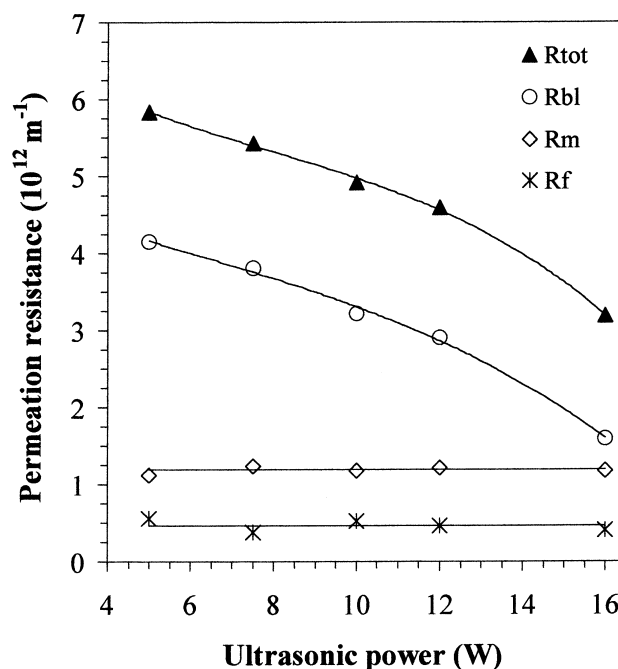


FIG. 6 Permeation resistances values as a function of ultrasonic power ($C_o = 0.3 \text{ g} \cdot \text{L}^{-1}$, $\Delta P = 0.5 \text{ bar}$).

Evolution of Permeation Resistances with Filtration Time

To illustrate the decrease of the flux during stirred UF (see Fig. 3), a series of experiments was performed during different filtration times (10, 20, and 30 min) to investigate the evolution of permeation resistances.

Deduced experimental permeation resistances are reported in Table 3. Note that the total resistance against solvent permeation (R_{tot} value) gradually increased with filtration time. The resistance of the boundary layer (R_{bl}) remained quite constant, whereas the fouling resistance (R_f) increased throughout the experiment. Such observations are consistent with the corresponding permeate flux profile described, previously. Under US, it can be assumed that R_{tot} is constant because the permeate flux reaches a quasi-steady-state. It might thereby be thought that the resistance of the boundary layer (R_{bl}) as well as the fouling resistance (R_f) are not modified. As a result, it can be considered that ultrasonic waves prevent a progressive plugging with time. However, they do not prevent initial fouling because the value of the fouling resistance ($R_f = 0.41 \times 10^{12} \text{ m}^{-1}$) is identical to that obtained after 10 min of filtration under stirred condition ($R_f = 0.50 \times 10^{12} \text{ m}^{-1}$).

On the other hand, data in Table 3 illustrate that after 30 min of filtration, an identical resistance is offered against solvent permeation in both configurations of stirred and ultrasonically assisted UF. Even though the boundary layer resistance (R_{bl}) value is two times higher in the presence of US, that induced by fouling (R_f) is lower. Owing to the ability of US to avoid progressive plugging and owing to the evolution of resistances with filtration time when the cell is stirred, a higher permeate flux might be expected for a longer filtration time in the presence of the ultrasonic field.

Cleaning Efficiency of Ultrasound

As reported by Deqian (31), US is one of the most effective cleaning technologies for reverse osmosis and UF membranes. To investigate this effect, a fouled membrane was subjected to US and the PWF was determined after 1 hr of sonication. At the same time, a second membrane fouled under identical op-

TABLE 3
Evolution of Each Permeation Resistance (10^{12} m^{-1}) with Filtration Time During Stirred UF and Ultrasonically Assisted UF

	Filtration time	R_{tot}	R_{m}	R_f	R_{bl}
Stirred UF	10 min	2.56	1.22	0.50	0.84
	20 min	2.79	1.22	0.58	0.99
	30 min	2.91	1.22	0.86	0.83
UF under ultrasound	30 min	3.20	1.19	0.41	1.60

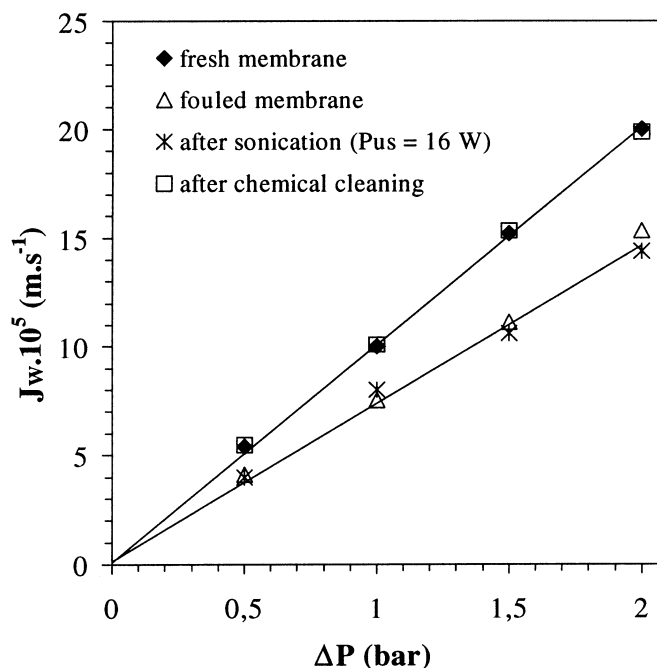


FIG. 7 PWF as a function of transmembrane pressure. Comparison between ultrasonic and chemical cleaning of a fouled UF membrane.

erating conditions was chemically cleaned. Figure 7 shows that US does not effectively clean the used membrane. Hence, the PWF cannot be restored after sonication, although restoration is possible by the classical chemical cleaning procedure. This means that, under our operating conditions, the cleaning action of ultrasound acts at the membrane surface but not inside the pores.

DISCUSSION

This study showed that US is an effective means of improving dead-end UF performance. Hence, comparison between unstirred and acoustically assisted separation operations illustrates the enhancement provided by the presence of the acoustic field: higher permeate fluxes and higher retention ratios. This improvement cannot be attributed to either change of the membrane permeability (which could result in higher permeate flux but worse selectivity) or to change of the solution characteristics. Therefore, the effect of US has to be explained in terms of induced hydrodynamics. As suggested by comparison with classical mechanically stirred UF, analogy can be made between mechanisms involved in these two configurations (32). Indeed, ultrasonic waves provide a vigorous mixing within the whole volume of the UF cell. At a macroscopic scale, strong convective currents, known as acoustic streaming, are at the origin of a stirring of the medium (33, 34). This effect can be easily visualized

using small suspended particles in water. In this way, it can be qualitatively demonstrated that the higher the ultrasonic power, the more intense the acoustic streaming, and thereby the more intense the induced stirring (35). Such an experimental observation is confirmed by theoretical velocity profiles of these convective currents predicted by Eckart's model (36), and may explain why an increase of ultrasonic power results in a higher ultrasonic enhancement factor.

At a microscopic scale, the physical effect associated with cavitation bubbles contributes also to generate micromixing in a liquid (37). Moreover, it is well known that the violent collapse of cavitation bubbles near a solid surface leads to formation of liquid microjets (38). This effect is equivalent to high-pressure jetting and is the main reason that US is widely used for cleaning. Such a phenomenon, even if it remains localized in the vicinity of the tip of the horn, has to be taken into account because it could have significant mechanical effects when occurring at the membrane surface. In fact, these different phenomena induced by US propagation may be thought to generate rigorous stirring within the US cell. From a chemical engineering viewpoint, this apparatus can be regarded as a completely stirred tank reactor (35). Owing to this effect, applying US during UF operations results, in terms of permeation resistances, in a many-fold decrease of the boundary layer resistance as demonstrated in this work. Such an observation is consistent with some previous works (22, 24).

Otherwise, it was shown that US can be used to minimize the resistance due to the foulant layer even if it is not able to prevent the initial fouling which takes place at the beginning of the filtration. Because the ultrasonic waves can cross the UF membrane but are reflected at the solid bottom of the cell, one can consider that the membrane is then subjected to vibrations. Thus, fouling can be avoided because the oscillations of the membrane may keep dextran molecules from entering the pores. It was demonstrated that the effect of US is limited because it cannot be used as an efficient method to clean fouled membranes and remove plugged materials.

CONCLUSION

Low-frequency US was applied to facilitate dead-end UF and was found to provide significant improvement in flow and retention ratios compared to unstirred operations. Comparison with mechanically stirred UF showed that the observed enhancement has to be related to hydrodynamics generated by ultrasonic waves propagation. However, the ultrasonic field appears to be a promising technology to facilitate separation membrane processes. The scale-up of such a device to a laboratory-scale tangential filtration system will be the subject of future work. The ultrasonic field will be optimized to provide more

effective results without causing damage to the membrane surface. It is expected that the use of US in conjunction with low cross-flow velocities will offer the potential advantage of reduced pump costs.

NOMENCLATURE

C	solute concentration ($\text{g}\cdot\text{L}^{-1}$)
E_{us}	ultrasonic enhancement factor (dimensionless)
J_{v}	permeate flux ($\text{m}\cdot\text{sec}^{-1}$)
J_{w}	pure water flux ($\text{m}\cdot\text{sec}^{-1}$)
P_{us}	ultrasonic power (Watt)
R	hydraulic resistance (m^{-1})
R_{obs}	observed retention ratio (%)
S	membrane surface (m^2)
V	volume (L)

Greek Letters

ΔP	driven pressure (Pa)
μ	dynamic viscosity ($\text{Pa}\cdot\text{sec}$)
ρ	solution density ($\text{kg}\cdot\text{m}^{-3}$)

Subscripts

b	bulk
bl	boundary layer
f	fouling
m	membrane
p	permeate
o	feed
r	retentate

REFERENCES

1. W. F. Blatt, A. Dravid, A. S. Michaels, and L. Nelsen, *Solute Polarisation and Cake Formation in Membrane Ultrafiltration: Causes, Consequences and Control techniques* (J. Flinn, Ed.) Plenum Press, New York, 1970.
2. M. C. Porter, "Concentration Polarisation with Membrane Ultrafiltration," *Ind. Chem. Prod. Res. Develop.*, **11**, 234 (1972).
3. G. Jonsson, "Boundary Layer Phenomena during Ultrafiltration of Dextran and Whey Protein," *Desalination*, **51**, 6 (1984).
4. M. W. Chudacek and A. G. Fane, "The Dynamics of Polarisation in Unstirred and Stirred Ultrafiltration," *J. Membr. Sci.*, **21**, 145 (1984).
5. G. B. Van den Berg and C. A. Smolders, "The Boundary Layer Resistance Model for Unstirred Ultrafiltration: a New Approach," *J. Membr. Sci.*, **40**, 149 (1989).

6. S. Bhattacharjee, A. Sharma, and P. K. Bhattacharya, "A Unified Model for Flux Prediction during Batch Cell Ultrafiltration," *J. Membr. Sci.*, **111**, 243 (1996).
7. E. Matthiasson, "The Role of Macromolecular Adsorption in Fouling of Ultrafiltration Membranes," *J. Membr. Sci.*, **16**, 23 (1983).
8. M. S. Le and J. A. Howell, "Alternative Model for Ultrafiltration," *Chem. Eng. Res. Des.*, **62**, 373 (1984).
9. I. E. El'piner, *Ultrasound: Physical, Chemical and Biological Effects*, Consultants Bureau, New York, 1964.
10. D. Ensminger, *Ultrasonics: Fundamentals, Technology, Applications*, 2nd ed., Marcel Dekker, New York, 1988.
11. S. D. Howkins, "Diffusion Rates and Effect of Ultrasound," *Ultrasonics*, **4**, 129 (1969).
12. I. Lenart and D. Ausländer, "The Effect of Ultrasound on Diffusion through Membranes," *Ibid.*, **18**, 216 (1980).
13. S. K. Dutta and A. P. Sinha, "Studies on Diffusion in the Presence of Ultrasonics," *Ind. Chem. Eng.*, **23**, 19 (1981).
14. J. D. Floros and H. Liang, "Acoustically Assisted Diffusion through Membranes and Biomaterials," *Food Technol.*, **48**, 79 (1994).
15. H. Li, E. Ohdaira, and M. Ide, "Effect of Ultrasound on Driving Force of Diffusion Dialysis," *Jpn. J. Appl. Phys.*, **36**, 3138 (1997).
16. H. V. Fairbanks, "Use of Ultrasound to Increase Filtration Rates," Ultrasonic International Conference Proceedings, 1973, p. 11.
17. A. Semmelink, "Ultrasonically Enhanced Liquid Filtering," Ultrasonic International Conference Proceedings, 1973, p. 7.
18. H. S. Muralidhara, N. Senapati, D. Ensminger, and S. P. Chauhan, "Electro-acoustic Separation Process for Fine Particle Suspensions," *Filtr. Sep.*, **23**, 351 (1986).
19. S. Mariana, "Experimental Researches Concerning the Productivity Increase with Solid-liquid Suspension Filtration in an Ultrasonic Field," *Stud. Univ. Babes-Boyai Chem.*, **35**, 31 (1990).
20. R. J. Wakeman and E. S. Tarleton, "An Experimental Study of Electroacoustic Cross-flow Microfiltration," *Trans. IChem. E.*, **69**, 386 (1991).
21. E. S. Tarleton and R. J. Wakeman, "Electro-acoustic Crossflow Microfiltration," *Filtr. Sep.*, 425 (1992).
22. Y. Matsumoto, T. Miwa, S. I. Nakao, and S. Kimura, "Improvement of Membrane Permeation Performance by Ultrasonic Microfiltration," *J. Chem. Eng. Jpn.*, **29**, 561 (1996).
23. N. Ahner, D. Gottschlich, and S. Narang, "Piezoelectrically Assisted Ultrafiltration," *Sep. Sci. Technol.*, **28**, 895 (1993).
24. T. Kokugan, S. Fujiwara, and M. Shimizu, "Ultrasonic Effect on Ultrafiltration Properties of Ceramic Membrane," *Membrane*, **20**, 213 (1995).
25. N. Sabri, P. Pirkonen, and H. Sekki, "Ultrasonically Enhanced Membrane Filtration of Paper Mill Effluent," Proceedings of 1st Conference on Applications of Power Ultrasound in Physical and Chemical Processing, Toulouse, France, 1997, p. 99.
26. X. Chai, T. Kobayashi, and N. Fujii, "Ultrasound Effect on Cross-flow Filtration of Polyacrylonitrile Ultrafiltration Membranes," *J. Membr. Sci.*, **148**, 129 (1998).
27. T. J. Mason, *Practical Sonochemistry. User's Guide to Application in Chemistry and Chemical Engineering*, Ellis Horwood, 1991.
28. S. Marsteau, A. Simon, N. Gondrexon, S. Taha, and G. Dorange, "Investigations on Ultrasonic Degradation of Dextran in Aqueous Solutions," European Commission Publication, EUR 18024 EN, (J. L. Luche, C. Balny, S. Benefice, J. M. Denis, and C. Petrier, Eds.), 1997, p. 87.
29. C. Bhattacharjee and P. K. Bhattacharya, "Prediction of Limiting Flux in Ultrafiltration of Kraft Black Liquor," *J. Membr. Sci.*, **72**, 137 (1992).

30. J. G. Wijmans, S. Nakao, J. W. A. van den Berg, F. R. Troelstra, and C. A. Smolders, "Hydrodynamic Resistance of Concentration Polarization Boundary Layers in Ultrafiltration," *J. Membr. Sci.*, **22**, 117 (1985).
31. R. Deqian, "Cleaning and Regeneration of Membranes," *Desalination*, **62**, 363 (1987).
32. A. Simon, L. Penpenic, N. Gondrexon, S. Taha, and G. Dorange, "A Comparative Study between Classical Stirred and Ultrasonically Assisted Dead-end Ultrafiltration," *Ultrason. Sonochem.*, in press.
33. R. M. G. Boucher, "Sonochemistry at Low and High Ultrasonic Frequencies," *Br. Chem. Eng. J.*, **15**(3), 148 (1970).
34. M. L. Cadwell and H. S. Fogler, "Ultrasonic Gas Absorption and Acoustic Streaming Observations," *Chem. Eng. Progr. Symp. Series*, **67**(109), 124 (1971).
35. N. Gondrexon, V. Renaudin, C. Petrier, M. Clement, P. Boldo, Y. Gonthier, and A. Bernis, "Experimental Study of the Hydrodynamic Behaviour of a High Frequency Ultrasonic Reactor," *Ultrason. Sonochem.*, **5**, 1 (1998).
36. C. Eckart, "Vortices and Streams by Sound Waves," *Phys. Rev.*, **73**, 68 (1948).
37. H. Monnier, A. M. Wilhelm, and H. Delmas, "Influence of Ultrasound on Mixing on the Molecular Scale for Water and Viscous Liquids," *Ultrason. Sonochem.*, **6**, 67 (1999).
38. K. S. Suslick, *Ultrasound—Its Chemical, Physical and Biological Effects*, VCH Publishers, New York, 1998, p. 110.

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