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A New Centrifugal Ultrafiltration Device

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Abstract

A novel ultrafiltration device is described. It consists of a vertical dialysis cell in which the semipermeable membrane is supported by a metal or plastic porous, rigid sheet. The dialysis cell is mounted in a centrifuge swinging bucket, or rotor hole, with the membrane parallel to the centrifuge radius. Experiments are performed by filling one dialysis cell compartment with the liquid sample to be treated and by spinning it in a centrifuge. Effluent is collected from the other (initially empty) compartment of the dialysis cell. Membranes used in these cells have similar permeabilities but better retention than those used in unstirred ultrafiltration cells for small filtrate volumes.

INTRODUCTION

Ultrafiltration is now a well-established method for the concentration of macromolecular solutes in the laboratory and in industry. Characteristics of current ultrafiltration equipment and membranes have been reviewed (*1–4*).

Major extant problems in ultrafiltration are membrane concentration polarization (*5, 6*) and fouling (*7–9*), for which many remedies have been proposed: stirring (*5*), backwashing (*10*), concurrent use of electroosmosis (*11*) and convection (*12*), etc. These problems have prevented the use of ultrafiltration to concentrate highly viscous polymer solutions.

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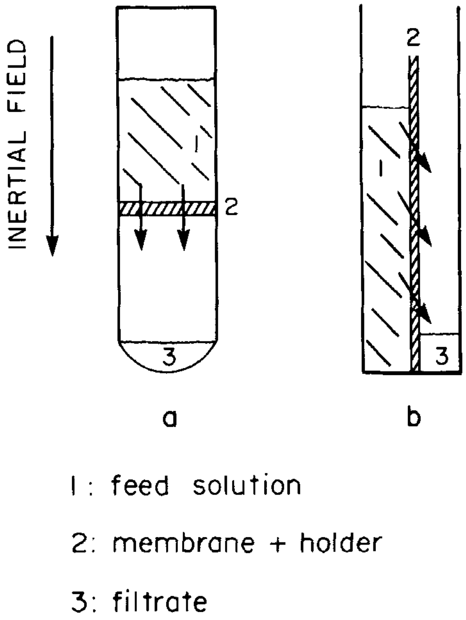

FIG. 1. Experimental arrangements for centrifugal ultrafiltration: (a) conventional, (b) this work. Arrows indicate liquid flow path.

It is possible to do ultrafiltration experiments by using the pressure generated by holding the liquid to be treated in a centrifugal field (centrifiltration) (*13*). This is very convenient for handling small volumes but leads to the accumulation of solute on top of the filter membrane and, concurrently, to a decrease of solvent flux. The accumulation of solute over the membrane can be eliminated by holding the membrane *parallel* to the centrifugal field and not *perpendicular* to it, because in the former case convective currents in the feed solution reservoir transfer the concentrated liquid from the membrane vicinity to the cell bottom (see Fig. 1). To verify this idea, ultrafiltration cells were built to fit in a centrifuges' swinging buckets. In these cells the membrane is parallel to the centrifugal field.

This report describes the use of vertical-membrane ultrafiltration cells for the concentration of polymer solutes.

EXPERIMENTAL

Dextran T 500 was obtained from Pharmacia Fine Chemicals (code no. 17-0320-01, lot G1-21917). Ovalbumin was prepared by the method of Kekwick and Cannan (*14*) and recrystallized six times. Poly(vinyl acetate)

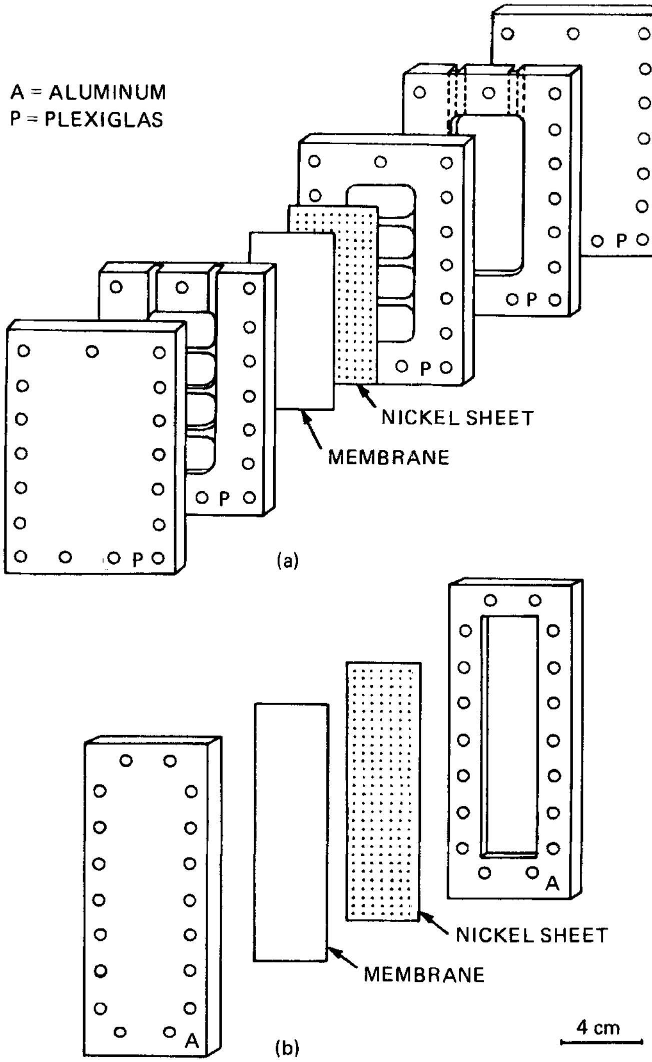

FIG. 2. Exploded view of centrifugal ultrafiltration cells used in experiments with (a) dextran and ovalbumin and (b) polystyrene and polyvinyl acetate solutions.

(PVA) was from Aldrich (Secondary Standard, cat 18, 250-8, lot 03). Polystyrene was from BDH ($\overline{M}_w = 100 \times 10^3$). Other reagents were of analytical grade. The ultrafiltration cells used in the centrifuge are depicted in Fig. 2. The cells were fitted with semipermeable membranes of cellulose acetate. These membranes were cast in this laboratory from acetic acid–acetone–aqueous solutions (*15*) and coagulated in water.

Cellulose acetate was from Carlo Erba (Italy), 53% acetyl content, and from May and Baker (England), 54-56% acetyl content.

For use in methanol solutions, the membranes were soaked in H_2O–methanol solutions of increasing concentration (20, 50, 80, and 100% v/v methanol) for 8-10 h in each solution. Ethanol-swollen membranes were transferred to toluene by an analogous procedure. The membrane divided the cell into two vertical compartments. A perforated nickel sheet (hole density: 1444 cm^{-2}) was used as the membrane support.

Prior to use, the assembled cells were filled with solvent and spun for ~30 min to check for leakage and to induce membrane compaction.

Experiments were performed by filling one cell compartment with solution and centrifuging it in a refrigerated swinging-bucket Sorvall RC-3B centrifuge. Ovalbumin solution concentration was determined by measuring A_{280} in a Micronal B 382 UV-spectrophotometer. Polystyrene and dextran concentrations were determined using a PAAR-DMA 60/602 densimeter and poly(vinyl acetate) concentrations by gravimetry.

Control runs were performed using unstirred ultrafilters. Aqueous solutions were run through a Millipore (10.2 cm^2) filter holder. For nonaqueous solutions an all-brass holder was built in this Department's workshop (membrane area: 12.7 cm^2).

RESULTS

Ultrafiltration of T500 Dextran

Centrifugation of 15.5 mL of a 0.15% (w/w) T500 dextran solution (2500 rpm, 4°C, 5 min) in the cell described in Fig. 2(a) gave 5.0 mL filtrate; solute retention was 99%. A similar experiment, run for 10 min, gave 8.3 mL filtrate and 99% retention.

In another run, under similar conditions (feed solution: 15.5 mL of 0.15% dextran, 2500 rpm, 4°C), the cell was loaded and centrifuged for 7 min, after which the filtrate was collected and the cell was centrifuged again. This was done four times, and after a total of 28 min centrifugation, 12.5 mL filtrate was collected. The concentrate (2.6 mL) contained 99% of the solute at a concentration of ~5-fold the initial concentration.

Other experiments were run using a higher initial dextran concentration (0.9%): after five cycles of centrifugation (7 min) and filtrate collection, 4 mL concentrate was obtained at 3.2 and 99% retention. These results were compared to those obtained in a nonstirred ultrafiltration cell (10.2 cm^2 filtration area) using membranes from the same lot as used in the centrifugation experiments: at 2 atm pressure difference, after 150

TABLE 1
Centrifugal Ultrafiltration of 15.8 mL of 0.1% (w/w) Ovalbumin Solution in 0.1 *M* NaCl at 4°C (r_B = 23.4 cm, r_T = 16.7 cm)[a]

Speed (rpm)	Time (min)	Filtrate volume (mL)	Concentrate volume (mL)	$\frac{c_{\text{final}}}{c_{\text{initial}}}$	Retention (%)
1500	5	5.3	10.3	1.3	98
1500	10	7.7	8.0	1.8	98
1500	15	8.7	7.0	2.0	98
1500	25	9.8	6.0	2.4	99
2000	5	6.5	9.3	1.6	98
2000	10	8.7	6.7	2.1	99
2500	5	7.7	7.7	1.8	99
2500	10	9.4	6.0	2.3	99
2500	21	10.1	5.3	2.5	98

[a] *r_B, r_T: distances between the centrifuge rotation axis and the solution bottom and meniscus, respectively.*

min, 4.2 mL filtrate was obtained with 99% retention. The performance of the centrifugation system is thus superior to the standard, unstirred cell when dealing with aqueous dextran.

Ovalbumin Ultrafiltration

15.8 mL ovalbumin solution (0.1% w/w in 0.1 *M* NaCl) was centrifuged (4°C, 2500 rpm) for a total of 21 min. During this time the centrifuge was stopped every 7 min for filtrate removal. As a result, 2.7 mL concentrate was obtained with 99% retention and $c_{\text{final}}/c_{\text{initial}} = 5.0$.

The results of some other runs are presented in Table 1. Using the data at 1500 rpm centrifugation speed, the membrane permeability was calculated ($L_p = 4.2 \times 10^{-10}$ $\text{cm}^2 \cdot \text{s/g}^1$) as the ratio between solvent flow and pressure head (*16*).

Standard ultrafiltration experiments gave a similar L_p (3.7×10^{-10} $\text{cm}^2 \cdot \text{s/g}^1$) but a slightly lower retention (91–97%).

Ultrafiltration of Poly(Vinyl Acetate) and Polystyrene

PVA dissolved in methanol and PS dissolved in toluene were also concentrated by centrifugal ultrafiltration. Table 2 gives the results of four runs for each solution. It was possible to concentrate them to ~1.5–1.7-fold (98% retention) in 30 min at 2000 rpm.

TABLE 2

Centrifugal Ultrafiltration of 16 mL PVA (in methanol) Solution, 10.1 mg/mL and 16 mL PS (in toluene) Solution, 4.9 mg/mL; 2000 rpm, 25°C

Solute	Centrifugation time (min)	Filtrate volume (mL)	Concentrate volume (mL)	$\frac{c_{\text{final}}}{c_{\text{initial}}}$	Retention (%)
PVA	5	2.8	12.6	1.2	98
	10	4.3	11.2	1.4	97
	20	6.8	8.6	1.7	98
	30	7.3	9.0	1.7	98
PS	10	2.8	13.0	1.1	87
	20	4.4	11.0	1.4	90
	30	5.1	10.4	1.5	96
	40	5.6	10.2	1.5	98

In other experiments, PVA/methanol solution was concentrated in three steps of 20 min each (2000 rpm) and PS/toluene solution in four steps of 25 min each (2000 rpm), the filtrate being removed at the end of each step. Feed solution volumes were 16 mL; after centrifugation a 3-4 fold concentration was obtained with 97-98% retention.

Solute retention in control experiments was lower than in centrifugal ultrafiltration (Table 3).

TABLE 3

Ultrafiltration of PVA (in methanol) Solution, 10.1 mg/mL, and PS (in toluene) Solution, 4.9 mg/mL; the Membranes Used Were from the Same Batch as Those Used in the Experiments in Table 2. Membrane Support: 12.7 cm^2 Brass Holder. $P = 2$ atm, $t = 25°C$

Solute	Filtration time (min)	Filtrate volume (mL)	Retention (%)
PVA	15	2	80
	52	4	94
PS	15	3	82
	32	5	92

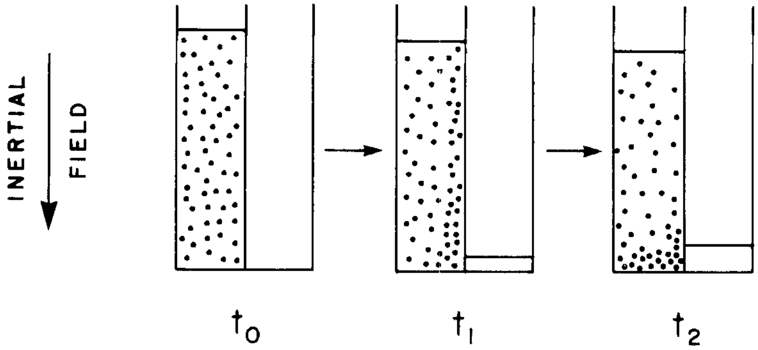

FIG. 3. A schematic description of accumulated solute removal from the membrane vicinity by convection. This prevents membrane impairment by solute gelation or caking.

DISCUSSION

The results described in this paper show that centrifugal ultrafiltration using cells in which the membrane is *parallel* to the inertial field is an effective technique for polymer concentration. Besides, it is a very convenient technique. Provided a low-speed centrifuge is available, it works without pressure cells, stirrers, recirculation devices, etc. It should be appreciated that this is an extremely gentle technique: there is hardly any chance of damaging polymer solutes by stirring and shearing of viscous solutions.

Another point is that useful pressures may be obtained very easily and safely: in the centrifugation experiments with dextran, at zero time (2500 rpm) the pressure at the liquid column bottom was 8.9 bar; at 5000 rpm it would go above 35 bar.

Given the current status of centrifuge construction and use, we believe that larger-scale centrifugal ultrafiltration may soon come into existence.

Last but not least, the retention of a given solute by a given membrane is better in centrifugation experiments than in standard ultrafiltration. This is not difficult to understand, considering that in the novel technique there is no accumulation of solute over the membrane to be pushed forward by a liquid stream. Concentrated solution adjacent to the membrane is convectively transferred to the cell bottom, as depicted in Fig. 3.

Recent, previous work from this laboratory showed that polymer solutes may be concentrated by centrifugation within dialysis cells (*17*) (osmocentrifugation). The present technique of centrifugal ultrafiltration appears to be as gentle and convenient as osmocentrifugation, but faster.

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