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## **The Effect of Temperature on the Flux from a Stirred Ultrafiltration Cell**

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### **Abstract**

The effect of viscosity and temperature on the flux from a stirred ultrafiltration cell was studied. Fluxes of water, sucrose solutions, and sucrose solutions containing bovine serum (nonpermeating species) were measured at various temperatures. The effect of sucrose concentration and temperature on flux was explained by determining the effect of these two variables on the bulk viscosity and the diffusivity of the nonpermeating species, and then correlating flux by the equation  $Sh = A(Re)^a(Sc)^{1/3}$ . The flux at various temperatures could also be adequately estimated from changes in viscosity alone.

### **INTRODUCTION**

Flux through ultrafiltration membranes depends on temperature (1, 2). This dependence has usually been described by demonstrating that flux is a linear function of temperature (3, 4) or that flux and temperature can be related by an Arrhenius equation (3, 5, 6). Such descriptions fail to account for the fact that flux at constant temperature depends on other functions, such as the Reynolds and Schmidt numbers, which are themselves dependent upon temperature. This report presents data to show that the

effect of temperature on flux through ultrafiltration membranes can be described through equations relating flux to mixing, and to the physical properties of the solution being ultrafiltered.

## EXPERIMENTAL DESIGN

A 60-ml, stirred ultrafiltration cell fitted with an Abcor HFA-300 membrane with an area of  $13.5 \text{ cm}^2$  was used in this study. The ultrafiltration experiments were carried out in a continuous mode with feed solution supplied from a reservoir under pressure. The ultrafiltration cell temperature was controlled by a thermostatted water bath.

In experiments with bovine serum solutions, the ultrafiltration cell was initially completely filled with the protein solution, and either water or sucrose solution was fed from the reservoir to the ultrafiltration cell. Back diffusion of protein from the ultrafiltration cell into the feed line was negligible. Protein concentration inside the ultrafiltration cell was essentially constant throughout the experiment. All solutions were Millipore-filtered to remove extraneous matter that could affect the flux through the membrane. To achieve reproducible results, it was necessary to condition a new membrane by passing a large volume of water at  $50^\circ\text{C}$  through it.

An Ostwald viscometer was used to determine the kinematic viscosity of protein and protein-sucrose solutions. Viscosities of water and sucrose solutions at different temperatures were obtained from the literature (7, 8). Lyophilized bovine serum was supplied by Miles Labs, Inc. Kankakee, Illinois.

## RESULTS AND DISCUSSION

### Pressure-Dependent Systems

Solutions permeable to ultrafiltration membranes are normally characterized by linear flux-pressure relationships. Figure 1 illustrates this effect for water and for sucrose solutions at low pressures at temperatures from 5 to  $60^\circ\text{C}$ . The deviation from linearity of the flux-pressure relationship for sucrose solutions was unexpected, and was found to be less at higher stirring speeds. No rejection of sucrose, as measured by a refractometer, was found.

Figure 2 shows that the effect of temperature on the flux of water and sucrose at low pressures through the ultrafiltration membrane can be

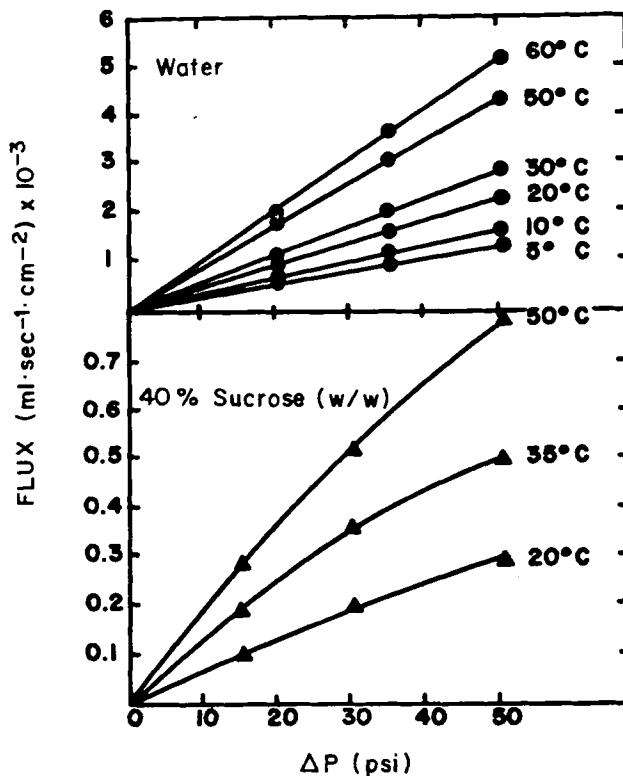


FIG. 1. The effect of pressure and temperature on the flux of water and sucrose through an ultrafiltration membrane.

described by

$$J = A(\Delta P/\mu) \quad (1) \quad (5)$$

Furthermore, Fig. 2 demonstrates that this relationship also holds for sucrose solutions permeating under high pressure, and the constant ( $A$ ) was found to be a function of stirrer speed.

### Pressure-Independent Systems (Polarized Region)

The typical flux-pressure curve for the ultrafiltration of macromolecular solutions is characterized by two well-defined regions. At low pressures there is a pressure-dependent region in which flux is a function of pressure, essentially independent of mixing. At higher fluxes (or higher pressures) the flux becomes independent of pressure across the membrane, and dependent on mixing. This phenomenon is known as concentration polarization, and most ultrafiltration units operate in the polarized region. Flux-pressure curves for the ultrafiltration of bovine serum solutions under various conditions are given in Fig. 3.

It is well established (1, 5) that flux in the polarized region is a function of mixing, temperature, and the nature and concentration of the solution being ultrafiltered. As mentioned previously, the effect of temperature on flux in the polarized region is usually described by demonstrating either that the flux is a linear function of temperature or that flux and temperature can be related by an Arrhenius equation (3-6). These two types of relationships would appear to fit the same data over the narrow range of temperature usually studied (5 to 60°C). Such plots for the data in Fig. 3 are given in Figs. 4 and 5. These figures also demonstrate temperature-related changes in viscosity.

The viscosity-temperature curves roughly parallel the flux-temperature curves. A plot of viscosity against flux reveals that flux is inversely proportional to viscosity (Fig. 6). Thus expressions describing temperature-related changes in flux apparently reflect temperature-related changes in viscosity. Forbes (9) also found that variation in the flux of a silica sol with temperature could be accounted for by the change in the solution's viscosity with temperature.

It has been suggested (1, 5) that the mechanism of ultrafiltration is such that the following equation holds:

$$Sh = A'(Re)^\alpha(Sc)^{1/3} \quad (2)$$

If this is true, the effect of temperature on flux should be calculable from

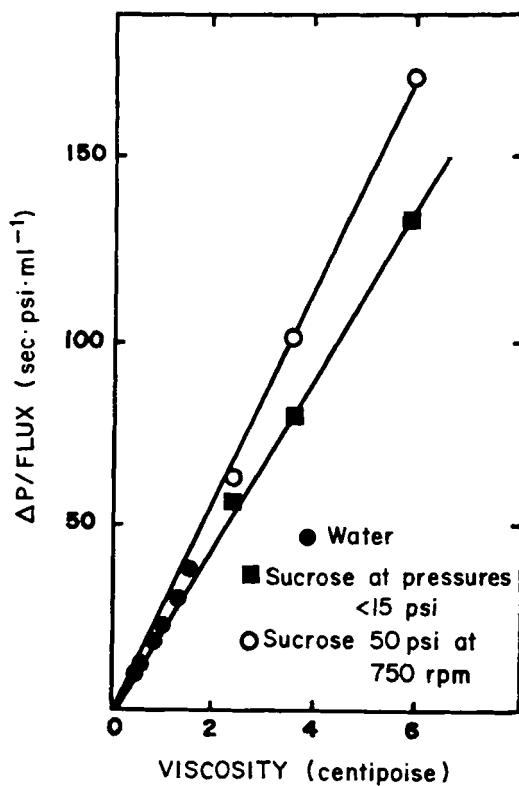


FIG. 2. The relationship between the ultrafiltration flux and the viscosity of water and sucrose solutions at various temperatures.

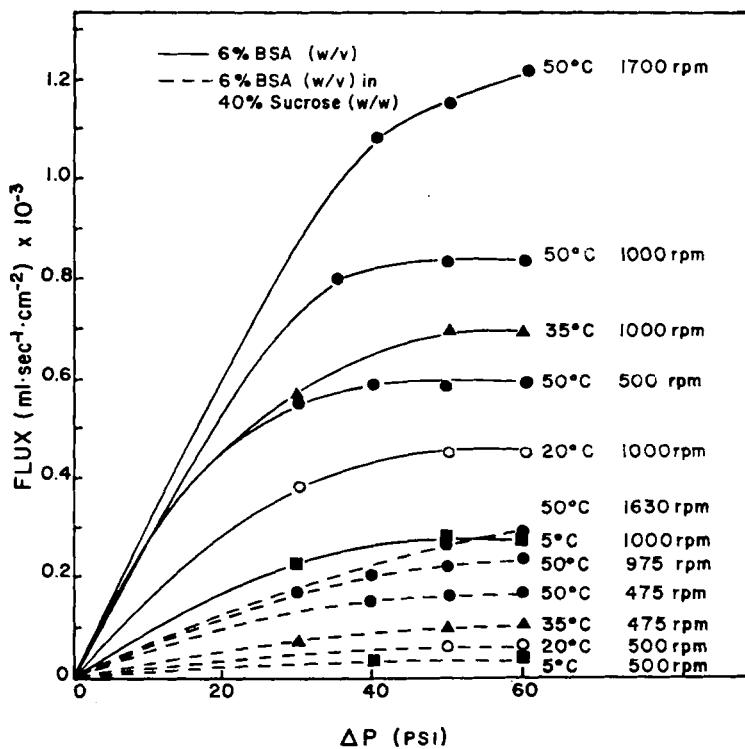


FIG. 3. The effect of pressure and temperature on the rate of ultrafiltration of a 6% bovine serum solution and a 6% bovine serum solution containing 40% (w/w) sucrose.

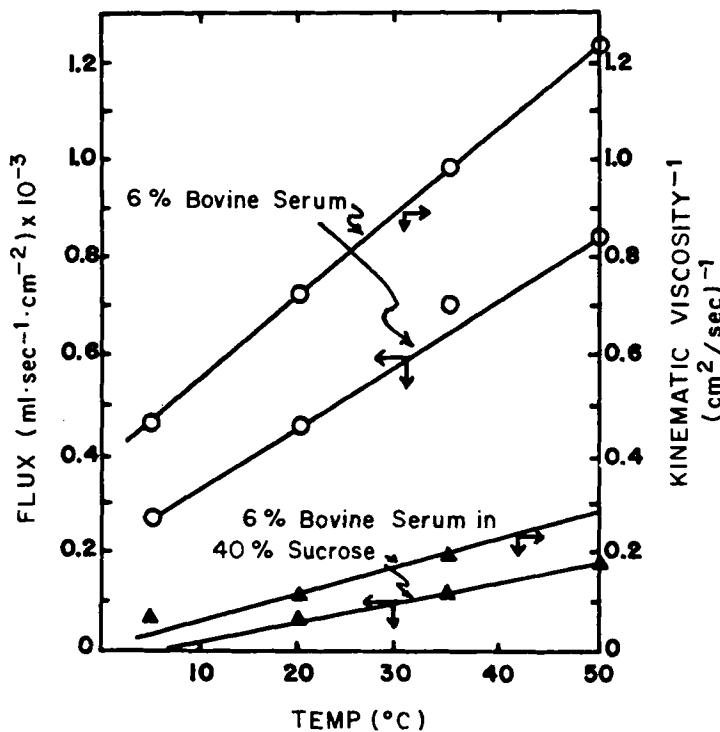


FIG. 4. The relationship between the ultrafiltration flux, temperature, and viscosity for bovine serum and bovine serum plus sucrose solutions.

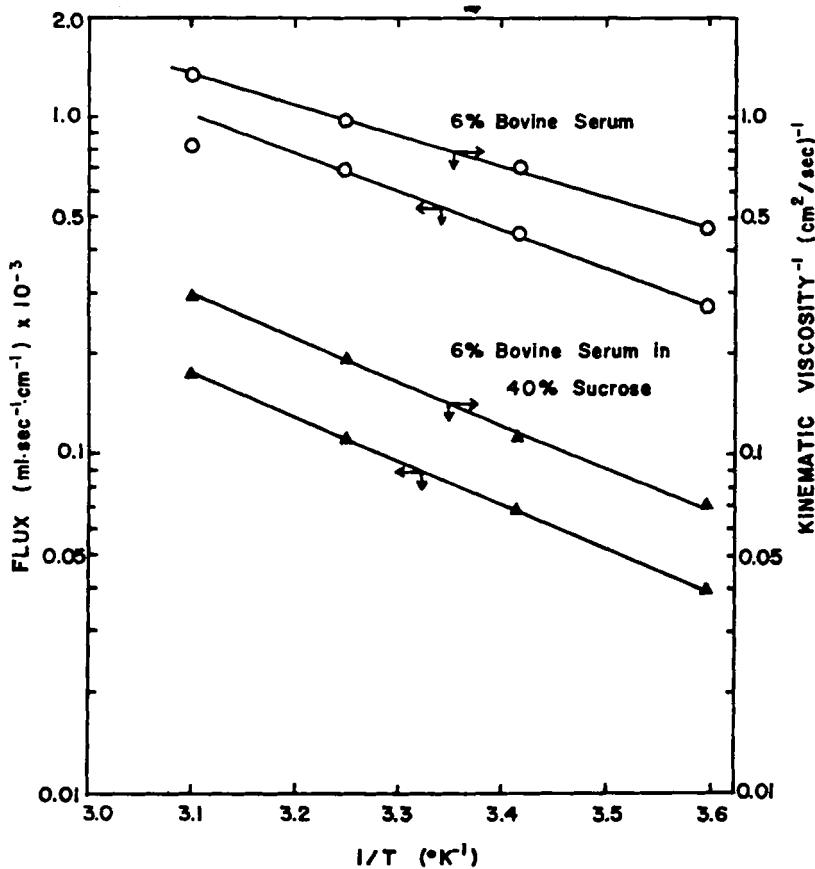


FIG. 5. Arrhenius plots for the ultrafiltration flux and viscosity of bovine serum and bovine serum plus sucrose solutions.

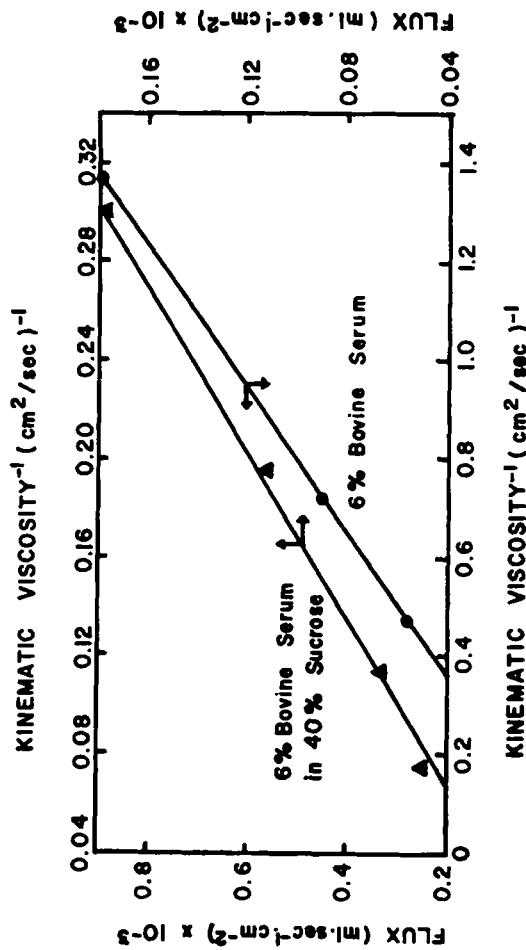


FIG. 6. The relationship between the ultrafiltration flux and viscosity for bovine serum and bovine serum plus sucrose solutions.

its effect on the viscosity and diffusivity of the solution being ultrafiltered.

In stirred ultrafiltration cells the exponent ( $\alpha$ ) is related to the magnitude of the Reynolds number and to the geometry of the ultrafiltration unit (1). In this instance  $\alpha$  was found to be 0.47 for bovine serum-sucrose solutions ( $6,000 < Re < 20,000$ ), and 0.59 for bovine serum solutions ( $27,000 < Re < 93,000$ ) (Fig. 7). In experiments to test the validity of the application of Eq. (2) to flux-temperature data, the concentration of the non-permeating species (bovine serum) was kept constant. Hence the flux through the membrane became an indirect measurement of the mass transfer coefficient  $k$ :

$$J = k \ln(Cg/C_B) \quad (3) (1)$$

Furthermore, the nature of the permeating species was varied by adding

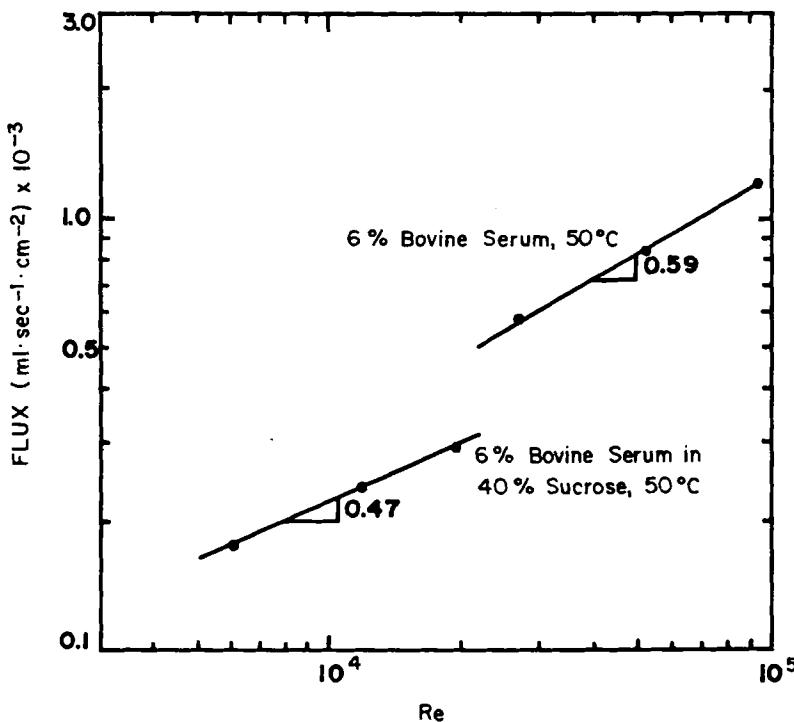


FIG. 7. The relationship between the ultrafiltration flux and Reynolds number for a bovine serum and bovine serum plus sucrose solution. In this instance the flux is proportional to  $(Sh) \cdot (Sc)^{-1/3}$ .

sucrose to the protein solution, which allowed the viscosity and diffusivity to vary while the protein concentration was held constant. Using an average value (10) of  $6 \times 10^{-7}$  cm<sup>2</sup>/sec for the diffusivity of the bovine serum at 20°C, and the Stokes-Einstein equation (Eq. 4), the diffusivity of the protein at different viscosities and temperatures was calculated:

$$D = (k'T)/(6\pi\mu r_p) \quad (4)$$

Then, when the temperature, viscosity, diffusivity, and rpm were known, data for the polarized region in Fig. 3 (points taken at 60 psi) were fitted to Eq. (2) (Fig. 8). This figure shows that the effect of temperature on flux can be described in terms of the accepted mass transfer correlation for ultrafiltration (1, 5).

Furthermore, with concentration, solution density, and stirring speed constant, Eqs. 2 and 3 can be combined and reduced to

$$\ln J = \ln(A'') + 0.67 \ln(T) + 0.81 \ln(1/\mu) \quad (\alpha = 0.47) \quad (5)$$

or

$$\ln J = \ln(A'') + 0.67 \ln(T) + 0.93 \ln(1/\mu) \quad (\alpha = 0.59) \quad (6)$$

In both these equations, variations in the term 0.67 ln(T) over the range of absolute temperature 278 to 333° (5 to 50°C) are small, while the effect of temperature on viscosity is large. Hence the correlation of viscosity with flux (Fig. 6) is explained.

## SYMBOLS

$A, A', A'', a$	constants
$C_g$	gel concentration (constant in this case)
$C_B$	concentration of nonpermeating species (constant in this case)
$D$	diffusivity of nonpermeating species
$J$	flux through membrane
$k$	mass transfer coefficient
$k'$	Boltzman constant
$\Delta P$	transmembrane pressure
$r$	radius of ultrafiltration cell
$r_p$	radius of nonpermeating species
$T$	absolute temperature
Sh	Sherwood number = $kr/D$

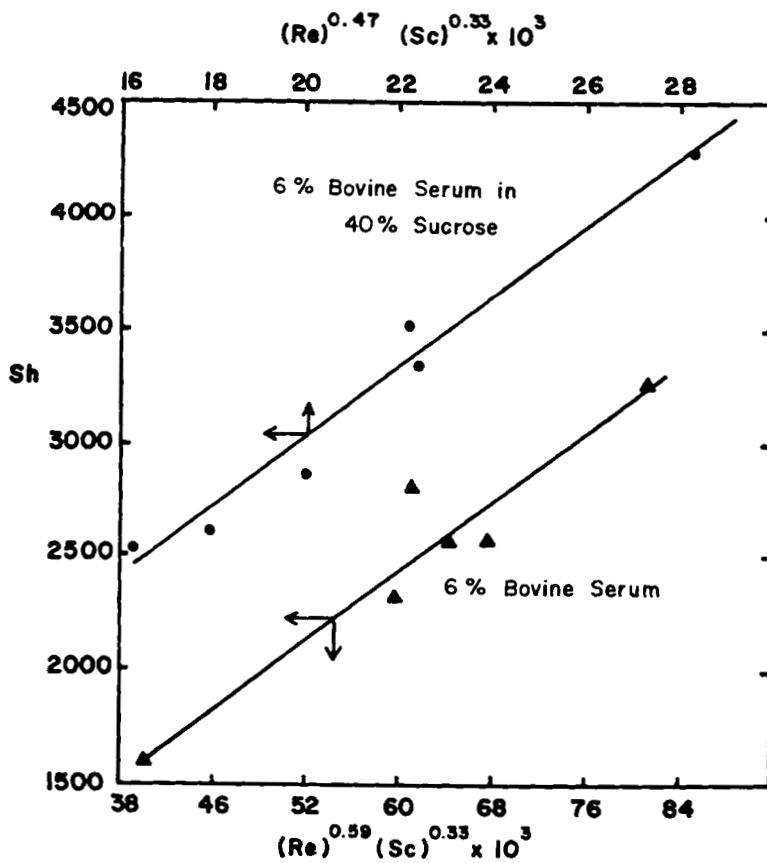


FIG. 8. Correlation of Sherwood with Reynolds and Schmidt numbers for the ultrafiltration of bovine serum and bovine serum plus sucrose solutions.

Re      Reynolds number =  $wr^2/\eta$   
Sc      Schmidt number =  $\mu/\rho D$

$\eta$       kinematic viscosity  
 $\mu$       viscosity  
 $w$       stirrer speed  
 $\rho$       density

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