# Permeation of Molecules Through Different Polymeric Membranes

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**ABSTRACT:** Permeation of molecules through the membranes plays an important role in immobilized cell systems. Permeation of molecules like glucose and bovine albumin serum was studied through chitosan, polyvinyl alcohol, and polyvinyl acetate membranes using a flow cell made of two detachable compartments. Permeation of molecules through chitosan, polyvinyl alcohol, and polyvinyl acetate membranes increases with decrease in hydrophobic characteristics of the membrane. Permeation of molecules also show its dependency on the molecular weight of the solute. Distribution coefficient of glucose and bovine serum albumin in these polymeric membranes measured by equilibration technique indicates that permeation of molecules through these membranes follows pore type mechanism. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 74: 3469–3472, 1999

**Key words:** diffusion coefficient; distribution coefficient; polymeric membranes; cell immobilization

## INTRODUCTION

Immobilized cell systems comprise a wide variety of natural and man made supporting matrices like alginate, carageenan, agar, hydrogels like polyacrylamide, etc.[1–3] Due to the presence of support material, there is no convective flow inside the system and cells receive nutrients only by diffusive mechanisms.[4] As the cells proliferate, the total nutrient consumption increases and the diffusive limitations start effecting the efficacy of the immobilized cell systems. To improve the performance of immobilized cell systems, it is important to study the permeation of the molecules through the supporting material. In the present study we have tried to study the permeation of molecules through some polymeric membranes in order to know their ability to be used as immobilization matrix.

## MATERIALS

Chitin was obtained from Central Institute of Fisheries Technology—Cochin, India. Polyvinyl alcohol (PVA) was obtained from BDH, UK, and polyvinyl acetate (PVAC) was supplied by Sigma, USA. Glucose and bovine serum albumin (BSA) of BDH, India, and Sigma, USA, were used as received.

## METHODS

Chitin was deacetylated to chitosan[5] using sodium hydroxide solution (50% w/w) at 110°C for 2 h to obtain polymer of high molecular weight and swellability. Polyvinyl alcohol films were prepared and crosslinked with gluteraldehyde.[6] Films with different crosslink density were used

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**Table I Effect of the Amount of Crosslinking Agent on Swelling**

| Sample | Amount of Gluteraldehyde (% v/v) | Degree of Swelling |
|---|---|---|
| 1 | 2 | 0.98 |
| 2 | 3 | 0.89 |
| 3 | 4 | 0.86 |
| 4 | 5 | 0.81 |
| 5 | 6 | 0.77 |

to study permeation of molecules (Table I). Films of different crosslink density show a difference in the degree of swelling. Films of known weight were dipped in phosphate buffer (pH 7.4) for 24 h. They were then taken out wiped and weighed again. Degree of swelling was calculated using the following equation:

$$S \text{ (degree of swelling)} = W_2 - W_1/W_1 \quad (1)$$

where $W_1$ is the weight of dry sample and $W_2$ is the weight of the wet sample.

Polyvinyl acetate films were made by preparing 5% (w/v) solution of PVAC in acetone.

The diffusion (permeation) measurements were conducted using a continuous flow cell made of two detachable glass compartments.[7] The membrane was placed in between the two compartments and two sides were clamped together. One compartment was filled with phosphate buffer (0.1*M,* pH 7.4) and other with a solution of solute (BSA/glucose) in buffer. Aliquots of the buffer were taken out after a given period of time (1–8, and 12 h). The concentration of the solute(s) in the solution was determined spectrophotometrically and the solute permeability was calculated using[8]:

$$P = [-d \ln\{(((1 + V_1/V_2)C_1)/C_0) - V_1/V_2\}]/(A(1/V_1 - 1/V_2)t) \quad (2)$$

Where $V_1$, $V_2$, $A$, $d$, $C_0$, and $C_t$ were the volumes of the concentrate and dilute compartments, membrane area (2.55 $cm^2$) thickness, and concentration of the concentrate compartments at times 0 and $t$, respectively.

Distribution coefficient of the membranes was determined by placing a disk (18 mm in diameter) in the solute solution for a period of 48 h at 37°C. The membrane was removed quickly and blotted on the outer surface to remove excess solution and placed in a known volume of phosphate buffer (100 mL) at 37°C. After 48 h of equilibration the concentration of the solute was determined in the buffer ($C_2$) spectrophotometrically. The membrane was again placed in the solute solution ($C_1$) and the amount of solute sorbed was calculated[9]:

$$K_d = C_2V/V_P(C_1 - C_2) \quad (3)$$

where $V$ and $V_p$ refer to volume of the buffer and of swollen membrane and $K_d$ is the experimentally determined distribution coefficient.

## RESULTS AND DISCUSSION

The molecular weight of the chitosan obtained decreases continuously with an increase in reaction time (Fig. 1) from 44 to 19 $\times$ $10^4$ Daltons. This indicates that the molecular weight of chitosan is highly dependent upon the time of treatment. This further affects the properties like solubility, viscosity, etc., of the polymer.[10]

The swelling behavior of chitosan as a function of deacetylation time is shown in Figure 1. It shows that the degree of swelling of chitosan varies from 0.95 to 1.0 on increasing the reaction time from $\frac{1}{2}$ to 5 h. The amount of glucose perme-

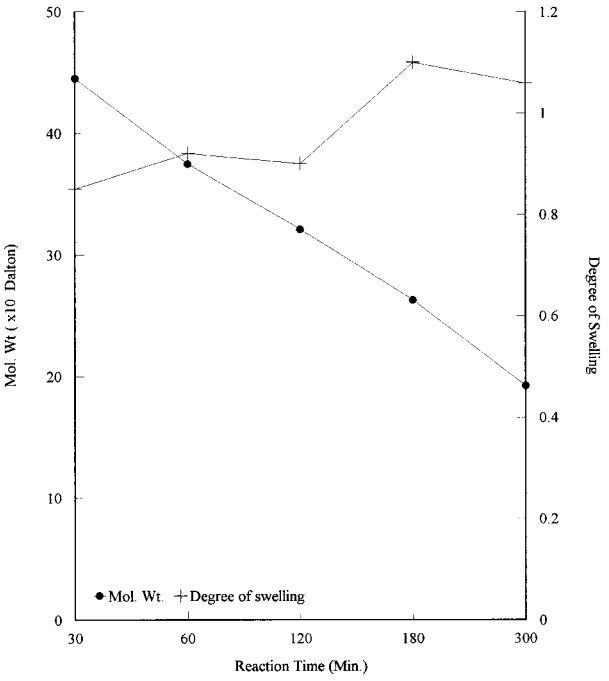


**Figure 1** Effect of reaction time on molecular weight and degree of swelling of chitosan.

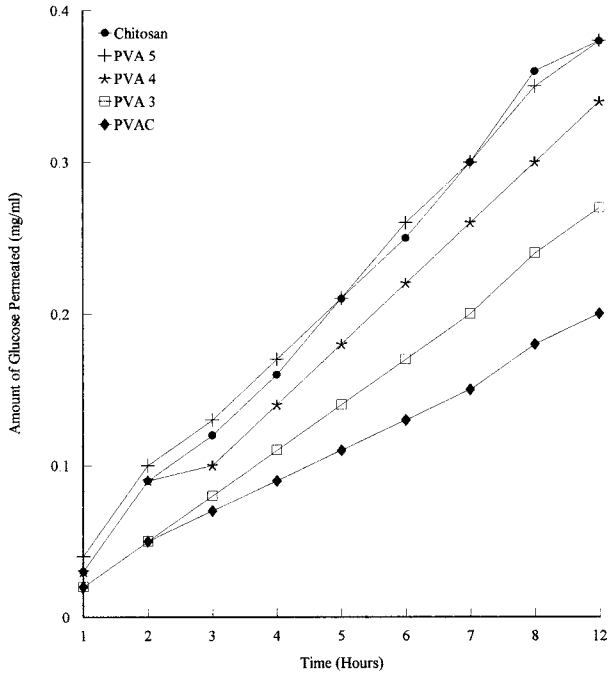


**Figure 2** Permeation of glucose through the polymeric membranes.

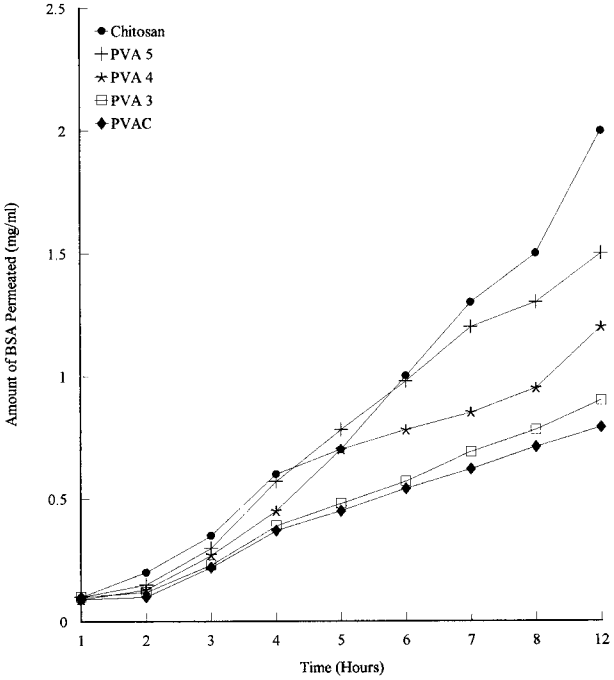


**Figure 3** Permeation of BSA through the polymeric membranes.

ated through these different films is shown in Figure 2, and the values of permeability and distribution coefficient are given in Table II. The permeability coefficient of glucose was found to be in the range of $10^{-7}$ $cm^2/s$.

An increase is observed in the permeability of glucose through crosslinked PVA films with increase in the swelling ability (Fig. 2). This is attributed to the decrease in the degree of the crosslinking. PVAC shows low permeability compared to PVA and chitosan films. This may be due to the hydrophobic nature of the membrane (Fig. 2).

The amount of BSA permeated through these films with respect to time is shown in Figure 3. The values of permeability and distribution coefficient are given in Table III. The amount of BSA permeated through membranes was found to be in the range of $10^{-8}$ $cm^2/s$, which is significantly lower than that for glucose, but the phenomenon was same as observed in the case of glucose. The lower value of permeability coefficient for BSA shows the molecular weight dependence of permeation through membranes.

The measurement of distribution coefficient of swollen membranes using equilibration technique supports the fact that permeation through the membrane follows pore-type mechanism.[11–13] The average values of distribution coefficient for glucose and BSA were 0.30 and 0.25, respectively, in chitosan films, 0.42 and 0.37 in PVA films, and 0.25 and 0.20 in PVAC films, indicating there was little if any preferential dissolution of solute in

**Table II Permeability of Glucose Through Polymeric Membranes**

| Sample | Degree of Swelling | $P \times 10^7$ ($cm^2/s$) | $K_d$ |
|---|---|---|---|
| Chitosan | 0.75 | 3.89 | 0.30 |
| PVA 5 | 0.77 | 4.73 | 0.40 |
| PVA 4 | 0.81 | 4.91 | 0.42 |
| PVA 3 | 0.86 | 5.09 | 0.44 |
| PVAC | 0.53 | 3.82 | 0.25 |

**Table III Permeability of BSA Through Polymeric Membranes**

| Sample | Degree of Swelling | $P \times 10^8$ ($cm^2/s$) | $K_d$ |
|---|---|---|---|
| Chitosan | 0.75 | 2.10 | 0.25 |
| PVA 5 | 0.77 | 2.30 | 0.35 |
| PVA 4 | 0.81 | 2.55 | 0.37 |
| PVA 3 | 0.86 | 2.95 | 0.38 |
| PVAC | 0.53 | 1.23 | 0.20 |

the polymer matrix; values less than unity have been interpreted as transport via a pore diffusion mechanism. It also indicates low interaction between solute and polymer interaction in the hydrogel. Thus it can be concluded that desired rate of permeation can be obtained from PVA polymeric films by varying the crosslink density. The chitosan and PVAC films should be further modified using different means like grafting, blending, etc. if the rate of solute permeation is to be altered from the rate found in this study.

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