

인공신장을 위한 셀룰로즈 격막의 평가

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Evaluation of Cellulosic Membrane for Artificial Kidney

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요 약

전하나 흡착능이 거의 없는 셀룰로즈 격막의 물질전달에 관한 특성을 혈액투석기 설계를 위해서 조사하였다. 유니온 셀로판회사(한국)에서 제조한 셀로판 격막의 총괄격막 저항계수를 이중(二重) 폐쇄투석장치에 의해서 구했다. 혈류속도, dialyzate 속도 및 분자크기가 총괄저항계수에 미치는 영향도 아울러 살펴 보았다. Ultrafiltration 실험은 Amicon ultrafilter 를 사용하여 행하였고 용질 거부계수는 분자량 60에서 44,000까지의 범위에 걸쳐서 구했다. 분자반경(Stokes-Einstein 반경)이 격막의 저항계수를 추정하는데 있어서 분자량 보다 더 합당한 인자임을 알았다. 셀룰로즈 격막에 대한 관계식은 다음과 같다.

격막 1(22 μ 두께, 유니온 셀로판, 서울, 한국)

$$\log(R_0) = 2.224(r_0) + 0.745$$

격막 2(38 μ 두께)

$$\log(R_0) = 1.326 \log(r_0) + 0.945$$

R_0 ; 총괄격막저항계수(min/cm)

r_0 ; Stokes-Einstein 반경(\AA)

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격탁 2로 만든 혈액투석기를 신부전환자에게 적용시켰다고 했을 때 환자내의 노폐물(요소, 크레아티닌)의 동력학을 two compartment 모델을 사용하여 전산기로 simulation 하였다.

Abstract

The mass transfer characteristics of cellulosic membranes nearly devoid of any charge or adsorption property were investigated in vitro for the design of hemodialyzer. Overall membrane resistance(O.M.R.) of cellophane produced by Union Cellophane(Seoul, Korea) was determined by a dual closed-loop dialysis technique with which the effects of blood flow rate, dialyzate flow rate and molecular size on O.M.R. were examined. Ultrafiltration experiment was performed by making use of Amicon ultrafilter with which solute rejection was determined covering M. W. 60 to 44,000. Solute radius(Stokes-Einstein radius) is found to be more reasonable parameter in estimating membrane resistance than molecular weight. The correlations for cellulosic membranes have the following formula.

For membrane 1(22 μ thickness, Union Cellophane, Seoul, Korea)

$$\log(R_0) = 2.224 \log(r_0) + 0.745$$

For membrane 2(38 μ thickness)

$$\log(R_0) = 1.326 \log(r_0) + 0.945$$

where R_0 : overall membrane resistance(min/cm)

r_0 : Stokes-Einstein radius, (\AA).

The kinetics of metabolic wastes(urea, creatinine) of the uremic patient treated by hemodialyzer made of membrane 2(37 μm dry thickness, Union Cellophane, Seoul, Korea) was simulated with computer using a two compartment model.

1. Introduction

The artificial kidney(A.K.) is effective not only for sustaining life of a patient with renal failure for an indefinite period, but also for preserving life while awaiting kidney transplantation¹⁻³⁾.

The basic element of the A.K. is a membrane. For an A.K. device, cellulosic membranes such as cellophane and Cuprophane have been used since 1943 because of their good blood-compatibility. Cellulosic membranes are considered to be of microporous type and to act as size-selective sieve type barrier.

Throughout the world more than 60,000

patients are kept alive with hemodialysis. In Korea, it is estimated there are about 3,000 people with chronic uremia at this time⁴⁾. But only 1% of them are benefited by this special treatment. It is a cost that prevents A.K. therapy for becoming more widespread. About 8 million won per person per year is needed in Korea for hemodialysis. The most important portion of the cost for A.K. therapy is supplies cost. Hence membrane cartridge which accounts for the large portion of supplies cost should be produced in Korea so as to reduce the A.K. therapy cost. This will be a great help to 4,000 patients with renal failure or epidemic hemorrhagic fever.

The purpose of this study is to evaluate the

domestic cellophane membrane whether it is suitable for A.K. production in Korea.

2. Mathematical Development

In 1958 Kedem and Katchalsky⁵⁾ proposed an approximate pair of integrated flux equations for describing solute and solvent transport across membrane by using nonequilibrium thermodynamics.

$$J_v = k(\Delta p - \Delta\pi) \quad (1)$$

$$J_s = \omega RT \Delta C_s + J_v(1 - \sigma) \bar{C}_s \quad (2)$$

J_v, J_s : solvent and solute fluxes, respectively

k : hydraulic permeability

Δp : hydraulic pressure difference across membrane

$\Delta\pi$: osmotic pressure difference across membrane

ΔC_s : concentration difference across membrane

ω : solute permeability parameter

σ : reflection coefficient

The diffusive and convective contributions to J_s in Eq. (2) are given by the terms involving ω and σ , respectively.

A. Dialysis

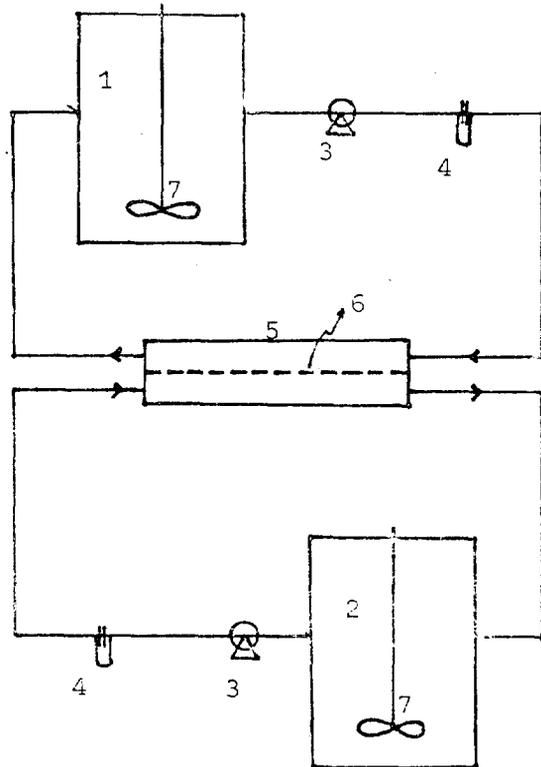
Fig. 1 shows the schematic diagram of a dual closed-loop dialysis. Mass balances are taken for each compartment.

$$\text{For blood compartment } -V_B \frac{dC_B}{dt} = J_s \cdot A \quad (3)$$

$$\text{For dialyzate compartment } V_D \frac{dC_D}{dt} = J_s \cdot A \quad (4)$$

$$t=0, C_D=C_{D0} \text{ and } C_B=C_{B0}$$

V_B and V_D are blood and dialyzate volume, respectively. C_B and C_D are blood and dialyzate concentration, respectively. A is mass transfer



experiment apparatus

- | | |
|----------------------|-------------------|
| 1. Blood bath | 2. Dialyzate bath |
| 3. Pump | 4. Manometer |
| 5. Permeability cell | 6. Membrane |
| 7. Agitator | |

Figure 1. Schematic diagram of diffusive permeability

area.

When the transmembrane pressure difference Δp is zero and the osmotic pressure difference $\Delta\pi$ is negligible, there is no volume flow (no convection). So the solute flux, J_s is:

$$J_s = \omega RT \Delta C_s \quad (5)$$

$$\text{or } J_s = P \Delta C_s \quad (6)$$

where P is the diffusive permeability.

If ΔC_s is concentration difference between two bulk phases, P is called "Overall Membrane Permeability" (P_0). Assuming a pseudo-steady state for the solute transport and con-

sidering the boundary layers and membrane to be resistive element in series, one finds:

$$\frac{1}{P_0} = \frac{1}{P_m} + \frac{1}{P_B} + \frac{1}{P_D} \quad (7)$$

The reciprocal of permeability is "resistance."

$$R_0 = R_m + R_B + R_D \quad (8)$$

R_0 is "Overall Membrane Resistance" (O.M.R.). R_m , R_B and R_D are membrane, blood part and dialyzate part resistance respectively. As no mass enters or leaves, mass is constant.

$$m = C_D V_D + C_B V_B = C_{D0} V_D + C_{B0} V_B \quad (9)$$

Integration of Eqs. (3) and (4) after substituting of Eqs. (6) and (9) gives:

$$\begin{aligned} \frac{1}{(V_D + V_B)} \ln \left\{ \frac{(V_D + V_B) C_B - m}{V_D (C_{B0} - C_{D0})} \right\} \\ = - \frac{P_0 \cdot A \cdot t}{V_D \cdot V_B} \end{aligned} \quad (10)$$

$$\begin{aligned} \frac{1}{(V_D + V_B)} \ln \left\{ \frac{m - (V_D + V_B) C_D}{V_B (C_{B0} - C_{D0})} \right\} \\ = - \frac{P_0 \cdot A \cdot t}{V_D \cdot V_B} \end{aligned} \quad (11)$$

Eqs. (10) and (11) can be used to find P_0 by measuring C_B or C_D at different time.

B. Ultrafiltration

The hydraulic permeability or ultrafiltration coefficient, k is defined as the following under the assumption of negligible osmotic pressure difference.

$$k = J_v / \Delta P \quad (12)$$

Fig. 2 shows the schematic diagram of ultrafiltration experiment. The volume and solute balance equations are given by:

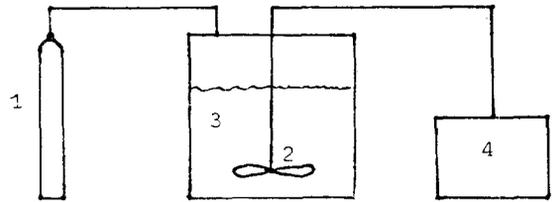
$$\frac{dV}{dt} = -A \cdot J_v \quad (13)$$

$$\frac{d(V \cdot C_s)}{dt} = -A \cdot J_s \quad (14)$$

③ $t=0, V=V_0$ and $C_s=C_{s0}$

where C_s is solute concentration in the ultrafiltration cell.

Although solute flux is provided by Kedem-Katchalsky equation, reflection coefficient σ



- 1. Nitrogen gas cylinder
- 2. Agitator
- 3. Ultrafiltration cell
- 4. R.P.M. controller
- 5. Mess cylinder

Figure 2. Schematic diagram for ultrafiltration test

is difficult to find experimentally. Hence another form of solute flux equation is required. Now we define distribution coefficient K_D as follows:

$$K_D = \frac{C_u}{C_s}$$

where C_u is solute concentration in the ultrafiltrate.

Solute flux equation is

$$J_s = J_v C_u = J_v K_D C_s \quad (15)$$

By substituting Eqs. (12) and (15) into Eqs. (13) and (14)

$$V = V_0 - k \cdot A \cdot \Delta P \cdot t \quad (16)$$

$$\ln \left(\frac{C_s}{C_{s0}} \right) = (1 - K_D) \ln(V_0/V) \quad (17)$$

Solute rejection, R_s is defined as:

$$R_s = 1 - K_D = \frac{\ln \left(\frac{C_s}{C_{s0}} \right)}{\ln(V_0/V)} \quad (18)$$

C. Solute Dimensions

Solute diffusion coefficients are taken from the literature⁶⁻⁸⁾ or estimated from the Wilke-Chang correlation.

$$D_s = 7.4 \times 10^{-8} (\phi M_v)^{\frac{1}{2}} \frac{T}{\mu_v \cdot V_s^{0.6}} \quad (19)$$

where D_s =diffusivity of solute

ϕ =association parameter of solvent

M_v =molecular weight of solvent

μ_v = viscosity of solvent

V_s = molal volume of the solute at its normal boiling point

Solute molecule dimensions can be obtained by the "Stokes-Einstein equation."

$$r_0 = \frac{k' \cdot T}{6 \pi \cdot D_s \cdot \mu_v} \quad (20)$$

where r_0 = Stokes-Einstein radius

k' = Boltzmann's constant

The Stokes-Einstein equation has been shown to be fairly good for describing the diffusion of large spherical molecules under conditions which the solvent appears to the diffusing species as a continuum⁹⁾. These data along with molecular weight are given in Table 1.

Table 1. Aqueous Diffusion Coefficients (T=20°C) and Characteristic Molecule Radii from Stokes-Einstein Equation

Component	M.W.	$D_s(\text{cm}^2/\text{sec}) \times 10^5$	$r_0(\text{Å})$
Urea	60	1.06	1.81
Creatinine	113	0.84	2.60
Glucose	180	0.60	3.50
Maltose	342	0.43	5.08
Naringine	580	0.302	7.24
P.E.G. 1000	1000	0.296	7.38
P.E.G. 2000	2000	0.195	11.20
P.E.G. 4000	3000	0.129	14.28
P.E.G. 6000	7500	0.089	24.68

3. Experimentals

The membranes used for this study are cellophane manufactured by Union Cellophane Co. (Korea). Membrane thickness measured by the micrometer is listed in Table 2.

The diffusive permeabilities are determined by a dual closed-loop dialysis technique. A schematic diagram of the apparatus is shown in Fig. 1. The channel heights of blood part and dialyzate part are 0.1 cm and 0.3 cm,

Table 2. Membrane Thickness Data

	Dry	Wet
Membrane 1	21.6 μ	38.0 μ
Membrane 2	38.1 μ	88.9 μ

Membrane thickness is measured by the micrometer.

(Model 549 Testing Machine, Inc. U.S.A.)

respectively. An aqueous solution is called "blood" and a distilled water is used as dialyzate. The transmembrane pressure difference should be as close to zero as possible so as to minimize convective transport. The volumes of blood and dialyzate are 5 l and 8 l, respectively. Mass transfer area is 16 cm².

The hydraulic permeabilities are determined by the Amicon Ultrafilter (Amicon Model 2,000). A schematic diagram of the apparatus is shown in Fig. 2. R.P.M. is large enough to eliminate "concentration polarization". Mass transfer area is 144 cm². The solutes used are urea, creatinine, glucose, maltose, naringine, polyethylene glycol 1,000, 2,000, 4,000 and 6,000 and albumin. The methods of analysis are provided in Ref. (10).

4. Results and Discussion

A. Effect of Dialyzate Flow Rate (D.F.R.) on Overall Membrane Resistance (O.M.R.)

Fig. 3 shows the effect of D.F.R. on O.M.R. for membrane 2 at 20°C. When D.F.R. is larger than 1.6 l/min, one can ensure the validity of assumption of negligibly small fluid-film resistance. For laminar range, the following result is obtained.

$$\text{O.M.R.} \propto (\text{D.F.R.})^{-0.31}$$

for D.F.R. < 1.6 l/min

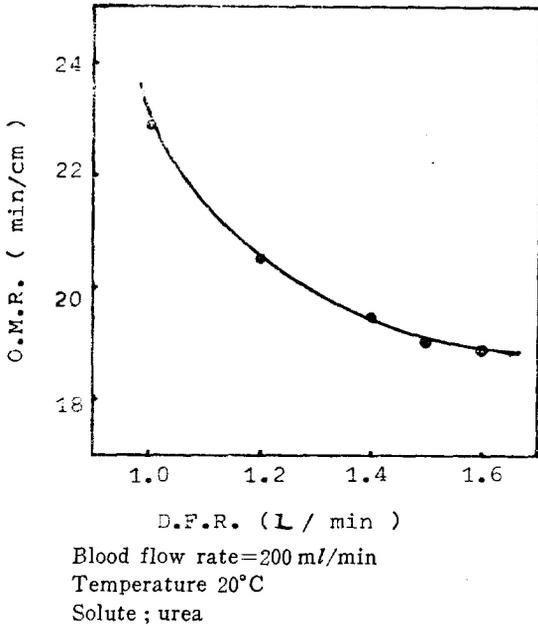


Figure 3. Overall membrane resistance (O.M.R.) against dialyzate flow rate (D.F.R.)

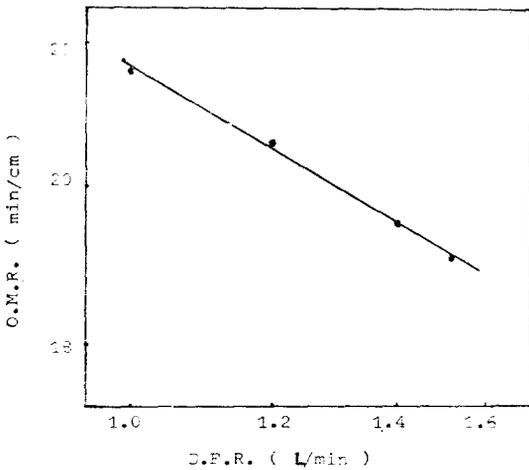


Figure 4. Overall membrane resistance (O.M.R.) vs. dialyzate flow rate on log-log coordinates

B. Effect of Blood Flow Rate(B.F.R.) on O.M.R.

The effect of B.F.R. on O.M.R. is given in Fig. 5. The empirical correlation is found to be

$$O.M.R. \propto (B.F.R.)^{-0.21}$$

In real artificial kidney system high shear rate causes "hemolysis"(rupture of red blood cell membrane). So B.F.R. is maintained at 200 to 300 ml/min.

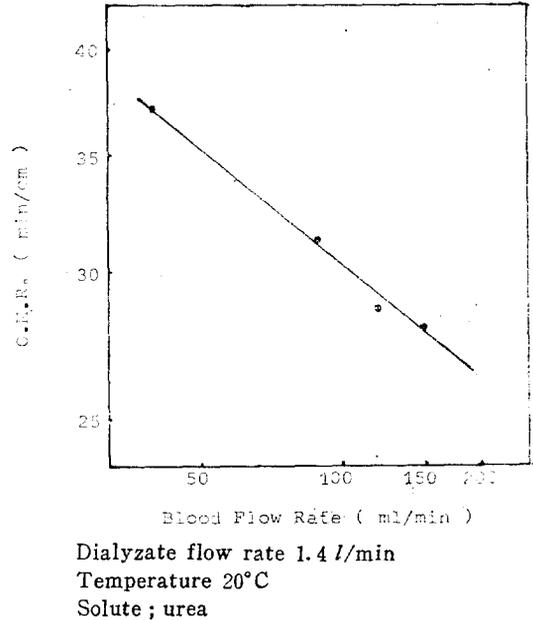


Figure 5. Overall membrane resistance (O.M.R.) against blood flow rate(B.F.R.)

C. Effect of Molecular Weight(M.W.) on O.M.R.

As solute M.W. increases, membrane permeability decreases faster than liquid diffusivity does. The O.M.R. of various solutes are listed in Table 3. Membrane 1 has smaller pore size than membrane 2. If one defines "actual mass transfer area" as number of pores multiplied by single pore area, membrane 2 has larger actual mass transfer area than membrane 1. Membrane 2 is found to be good for middle molecule removal. What albumin does not pass through the membranes is an encouraging result.

O.M.R. in Table 3 are plotted in Fig. 6 on log-log coordinates against M.W.. There is a significant trend toward increased resistance

Table 3. Diffusive Permeabilities of solutes at 20°C

Compound	M.W.	O.M.R. (min/cm)	
		Membrane 1	Membrane 2
Urea	60	21.5	19.5
Creatinine	113	44.0	31.2
Glucose	180	92.6	46.1
Naringine	580	—	122.0
Albumin	44,000	N.T.*)	N.T.

*) not detectable

as M.W. increases. This correlation provides a rough estimate of membrane resistance. Taking solute diffusivity and solvent viscosity into consideration solute radius is more reasonable parameter for estimation of membrane resistance than M.W. The Stokes-Einstein radius listed in Table 1 gives satisfactory correlation on log-log coordinates when plotted against O.M.R. in Fig. 7 The slopes and intercepts of these lines by the least square method, shown in Fig. 6 and 7 are listed in Table 4.

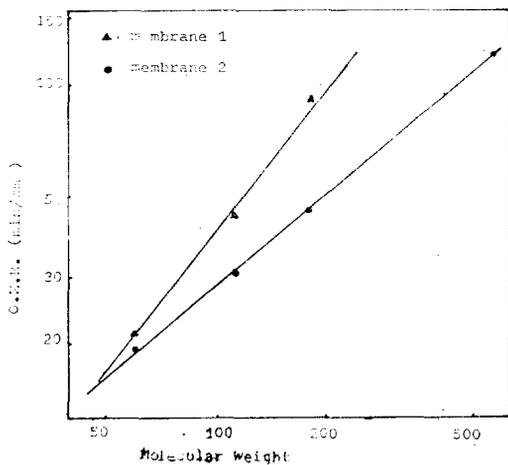


Figure 6. Overall membrane resistance (O.M.R.) against molecular weight (M.W.) on log-log coordinates
Temperature 20°C

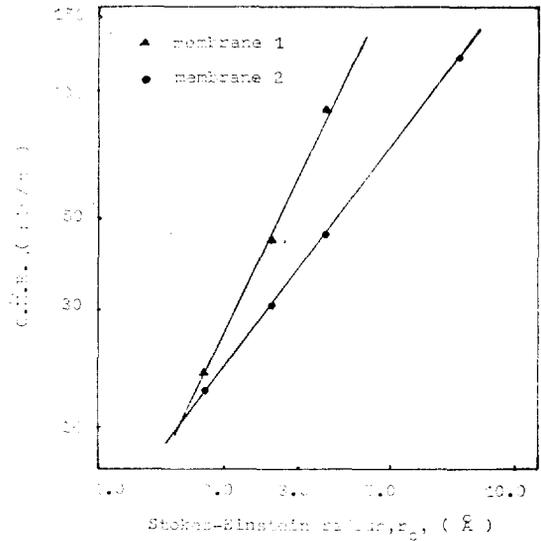


Figure 7. Overall membrane resistance (O.M.R.) vs. Stokes-Einstein radius (r_0) on log-log coordinates

Table 4. Slopes and Intercepts obtained from Figs. 6, 7

	Dependent Variable(y)	Independent Variable(x)	Slope (a)	Intercept (b)
Membrane 1	R_0^{**}	M.W.	1.321	-1.033
	R_0	r_0^{***}	2.224	0.745
Membrane 2	R_0	M.W.	0.814	-0.167
	R_0	r_0	1.326	0.945

*) Correlations are of the form; $\log y = a \log x + b$

***) The unit of R_0 is min/cm.

***) The unit of r_0 is Å.

D. The Hydraulic Permeability

The plot for volumetric flux J_v against transmembrane pressure difference Δp is shown in Fig. 8. The results are given in Table 5. The hydraulic permeability of membrane 2 is higher than that of membrane 1 because of larger actual mass transfer area. This result coincides with the previous result.

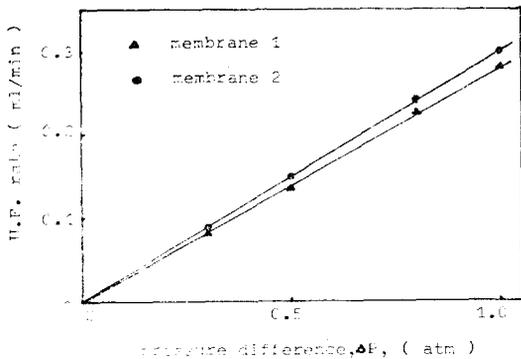


Figure 8. Ultrafiltration rate against transmembrane pressure difference

Table 5. The hydraulic permeability

membrane 1	0.26 U.F. Coeff.
membrane 2	0.29 U.F. Coeff.

1 U.F. Coefficient = 1 ml/(min)(cm²)(cmHg) × 10⁻⁴

E. Solute Rejection

Solute rejection data are plotted in Fig. 9 against M.W.. As M.W. increases, solute rejection takes a sigmoid form. For the solute of M.W. less than 180, there is no solute rejection. Albumin does not penetrate the

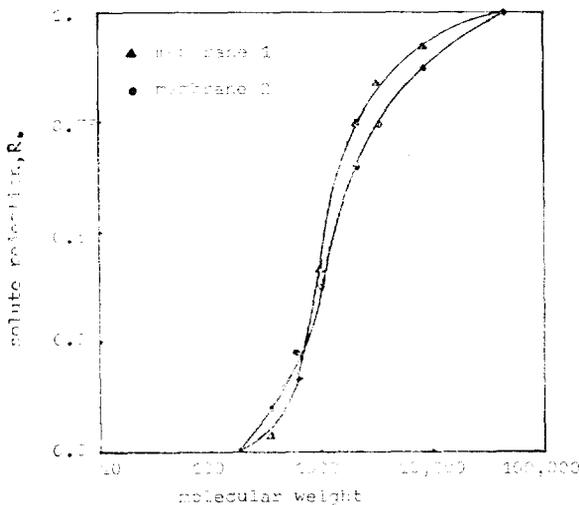


Figure 9. Solute rejection profiles against molecular weight on semi-log coordinates

membrane by ultrafiltration. In conclusion, albumin(or protein) cannot get through the membrane by diffusion(dialysis) and/or by convection(ultrafiltration). Fig. 10 shows the correlation of solute rejection with solute radius.

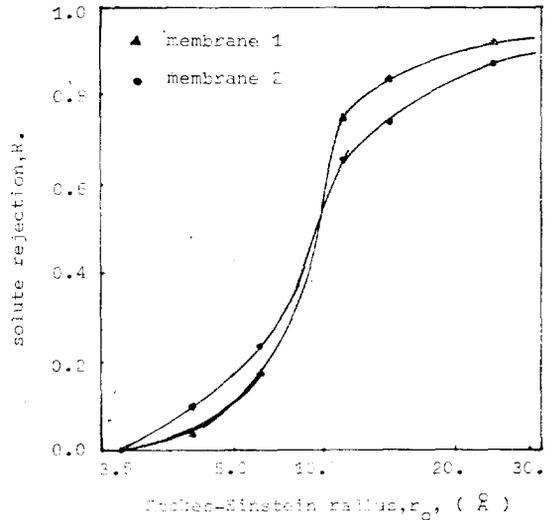


Figure 10. Solute rejection profiles against Stokes-Einstein radius (r₀) on semi-log coordinates

F. Discussion

Table 6 shows the comparison of membranes used for this study with other membranes¹¹⁾. Membrane 1 is inferior to Cuprophane used clinically in terms of mass transfer characteristics. So membrane 1 is not suitable to hemodialysis membrane because of its high solute resistance. The permeability of Membrane 2 is the same as Cuprophane(urea, 0.83 × Cuprophane; creatinine, 0.92 × Cuprophane; sucrose, 1.05 × Cuprophane; raffinose, 1.01 × Cuprophane). But the hydraulic permeability is a half of that of Cuprophane. This fact may be due to membrane thickness. The wet thickness of membrane 2 is 89 μ, and that of Cuprophane is 23 μ.

Table 6. Comparison of Membrane Properties (Solute Permeability*)

	Membrane 1	Membrane 2	Polycarb.	Cuproph.
Urea	465** (493) +	512** (543) +	665	654
Creatinine	227 (241)	321 (340)	389	370
Sucrose ^{a)}	48.5 (51.4)	127 (135)	201	129
Raffinose ^{a)}	29.0 (30.7)	92.7 (98.3)	156	97
U.F. Coeff. #	0.26	0.29	0.72	0.58

*) unit of cm/min. $\times 10^{-4}$
 **) data at 20°C
 +) data converted to 37°C
 &) Polycarbonate, Cuprophane: data at 37°C from Ref(11).
 @) estimated from the results in Table 4
 #) unit of ml/(min.) (cm²) (cmHg.) $\times 10^{-4}$

5. Kinetics of Hemodialysis; A computer simulation of the removal of metabolites(urea, creatinine)

Physiological transport kinetics during hemodialysis has been investigated for selected low M.W. metabolites. Urea transfer has been studied because of its importance to the disequilibrium syndrome.

A simple intracellular-extracellular (2 compartment) model shown in Fig. 11 is adequate to simulate urea and creatinine transport for the patient-artificial kidney system¹²⁾. The more complicated system may be required by higher M.W. metabolites¹³⁾.

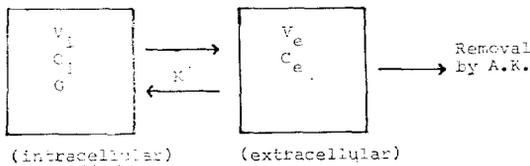


Figure 11. The 2 compartment model for the removal of urea and creatinine

The equations describing the two compartment model are easily written by considering a material balance around each compa-

rtment.

(Intracellular)

$$V_i \frac{dC_i}{dt} = G - K(C_i - C_e) \tag{21}$$

(Extracellular)

$$V_e \frac{dC_e}{dt} = K(C_i - C_e) - D(C_e - C_D) - UC_e \tag{22}$$

(Dialyrate part)

$$V_D \frac{dC_D}{dt} = D(C_e - C_D) + UC_e \tag{23}$$

(a) $t=0, C_{i0}=C_{e0}$ and $C_{D0}=0$

V_i and V_e are intracellular and extracellular fluid volume, respectively. C_i and C_e are intracellular and extracellular fluid concentration, respectively. G is metabolite production rate and K is mass transfer coefficient between intracellular and extracellular fluid. D and U are diffusive and convective clearances by the artificial kidney, respectively.

Emphasis should be placed on the realization of biologically acceptable model in a practical system. Blood is a complex heterogeneous suspension and the diffusivity for most solutes is lower than it is in water¹⁴⁾. There is also binding of some solutes to plasma proteins and

Table 7. Physiological transport parameters and membrane parameters for Urea removal kinetics

Volume of Intracellular fluid	23.24 L
Volume of Extracellular fluid	9.96 L
Volume of dialyzing fluid	100.0 L
Production Rate(G)	3.0 mg/min.
Mass Transfer Coeff(K)	0.552 L/min.
Mass Transfer Area	0.6 M ²
Diffusive Clearance(D)*)	0.44 L/min.
Convective Clearance(U)*)	0.013 L/min.
Solute rejection*)	0
Initial Concentration	1000. mg/L
Post-dialysis conc'n of extracellular fluid	300. mg/L

*) experimental values: converted to 37°C
 The rest are taken from Ref. (12).

resistance to diffusion across the red blood cell membrane. On the other hand, high M. W. substance may have a greater diffusivity in whole blood than in water, largely due to rotational activities by red cells. Physiological transport parameters are given in *Table 7* for urea and *Table 8* for creatinine.

Table 8. Physiological transport parameters and membrane parameters for creatinine removal kinetics

Production rate(G)	0.517 mg/min.
Mass Transfer coeff.(K)	0.303 L/min.
Mass Transfer Area	0.6 M ²
Diffusive Clearance(D)	0.285 L/min.
Convective Clearance	0.013 L/min.
Solute rejection	0
Initial Concentration	120 mg/L
Post-dialysis concentration of extracellular fluid	40 mg/L.

The results from computer simulation are plotted in *Fig. 12* and *13* against the treatment time. The advantage of this type of model is that it provides an estimate of the intracellular concentration which is difficult to mea-

sure experimentally, and as a result one can predict the treatment time when the concentration of extracellular fluid reaches post-dialysis concentration. The treatment time needed is 173 min. and 200 min. for urea and creatinine, respectively.

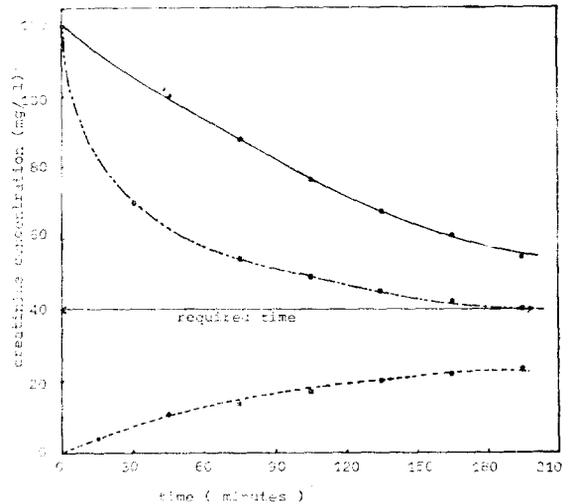


Figure 13. Creatinine concentration profiles against time ; (—●—), intracellular fluid ; (---●---), extracellular fluid ; (.....●.....), dialyzate bath

6. Conclusion

(a) For laminar range, the following correlations are obtained:

$$\text{O.M.R.} \propto (\text{D.F.R.})^{-0.31}$$

$$\text{O.M.R.} \propto (\text{B.F.R.})^{-0.21}$$

(b) Solute radius(Stokes-Einstein radius) is rather better parameter for estimating the membrane resistance than M.W..

For membrane 1(thickness 22 μ)

$$\log(R_0) = 2.224 \log(r_0) + 0.745$$

For membrane 2(thickness 38 μ)

$$\log(R_0) = 1.326 \log(r_0) + 0.945$$

(c) The hydraulic permeabilities of membrane 1 and membrane 2 are 0.26 U.F. coeff. and 0.29 U.F. coeff., respectively.

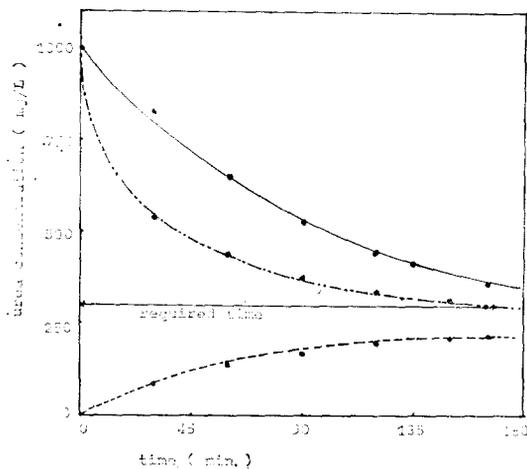


Figure 12. Urea concentration profiles against time ; (—●—), intracellular fluid ; (---●---), extracellular fluid ; (.....●.....), dialyzate bath

Nomenclature

A : mass transfer area, cm^2
 C : concentration, mg/l
 C : diffusive clearance, l/min
 D_s : diffusivity of solute, cm^2/sec
 G : production rate of metabolic wastes, mg/hr
 J : flux, $\text{mg}/\text{cm}^2/\text{min}$ or $\text{ml}/\text{cm}^2/\text{min}$
 K : mass transfer coeff., l/min
 K_D : distribution coeff., dimensionless
 k : hydraulic permeability, $\text{ml}/\text{cm}^2/\text{min}/\text{cmHg}$
 k : Boltzmann's constant, $\text{gcm}^2/\text{sec}^2/^\circ\text{K}$
 M_v : molecular weight
 P : diffusive permeability, cm/min
 Δp : transmembrane pressure difference, atm
 R : membrane resistance or gas constant, min/cm
 R_s : solute rejection, dimensionless or $\text{atm. l}/\text{gmole}/^\circ\text{K}$
 r_0 : Stokes-Einstein radius, \AA
 T : temperature, $^\circ\text{K}$
 t : time, min
 U : convective clearance, l/min
 V : volume, l
 V_s : molal volume of the solute at its normal boiling point, ml/gmole

Greek letters

μ : viscosity of solvent, $\text{g}/\text{cm}/\text{sec}$
 $\Delta\pi$: osmotic pressure difference, cmHg
 σ : reflection coeff.
 ϕ : association parameter of solvent
 ω : solute permeability parameter

Subscripts

s : solute

v : solvent
 o : overall or initial
 m : membrane
 D : dialyzate
 B : blood
 u : ultrafiltrate

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