

## REFERENCES

- ASTM F739-91. (1991). Standard Test Method for Resistance of Protective Clothing Materials to Permeation by Liquids or Gases Under Conditions of Continuous Contact. Annual Book of ASTM Standards. Philadelphia: The American Society for Testing and Materials.
- Austin, G.T. (1984). Shreve's Chemical Process Industries. McGraw-Hill, Inc.
- Barrer, R.M. (1951). Diffusion in/and through Solids / by Richard M. Barrer. Cambridge Eng.: The University Press.
- Barrer, R.M. (1968). Simple gases. In Crank, J. Diffusion in Polymers, edited by J. Crank and G.S. Park. New York: Academic Press.
- Barrett, C.R., Nix, W.D., and Tetelman, A.S. (1973). Chapter 12. In The Principles of Engineering Materials. New Jersey: Prentice Hall.
- Carslaw, H.S., and Jaeger, J.C. (1959). Linear flow of heat in the solid bounded by two parallel planes. In Conduction of Heat in Solids. Oxford: The Clarendon Press.
- Center for Nuclear Energy Research (2000). Status Report for Development of a Solid State Sensor for Monitoring Hydrogen in Humid Chlorine Gas, Phase II: Testing of Sensors.
- Clark, L.C. (1956). Trans. Am. Soc. Artif. Internal. Organs, 2, 41-46.
- Henglein, F.A. (1969). Chemical Technology / F.A. Henglein ; translated by R.F. Lang. New York: Pergamon Press.
- Holleck B., and Wicke D. (1968). Diffusion data. Diffusion in Palladium and its Alloys, 2, 28-33.
- Kalvoda, R. (1987). Electroanalytical Methods in Chemical and Environmental analysis. New York: Plenum Press.
- Kay, B.D., Peden, C.H.F., and Goodman, D.W. (1986). Kinetics of hydrogen absorption by Pd (110). Physical Review B, 34(2), 817-822.

- Lasser, R., and Powell, G.L. (1986). Solubility of H, D and T in Pd at low concentration. Physical Review B, 34(2), 578-586.
- Lewis, F.A. (1967). Palladium and Hydrogen System by F.A. Lewis. New York: Academic Press.
- Matsuyama, M., Miyake, H., Ashida, K., and Watanabe, K. (1982). Permeation, diffusion and dissolution of hydrogen isotopes, methane and inert gases through/in a tetrafluoroethylene film. Journal of Nuclear Materials, 110, 296-300.
- Meares, P. (1976). Membrane Separation Processes / edited by Patrick Meares. New York: Elsevier Scientific Publishing Company.
- Miura, N., Yamazoe, N., and Seiyama, T. (Eds.). (1988). Amperometric sensor: Development of a solid-state gas sensor using proton conductor operative at room temperature. Chemical Sensor Technology, 1, 123-139.
- Morris, D.R., and Sun, X. (1993). Water sorption and transport properties of Nafion 117H. Journal of Applied Polymer Science, 50, 1445-1452.
- Morris, D.R., Yang, L., Sun, D., and Steward F.R. (1995). Measurement of Henry's law constant for hydrogen in water at high temperature with an in-situ palladium resistivity probe. Paper presented at Canadian Meeting, Center of Nuclear Energy Research, Canada.
- Nakatsuji, H., and Hada, M. (1985). Interaction of a hydrogen Molecule with Palladium. Journal of American Chemical Society, 107, 8264-8266
- Perry, R.H. (1984). Perry's Chemical Engineers's Handbook. 6<sup>th</sup> ed, McGraw Hill.
- Rakotoarevelo, J. (July 1995). Literature Search for Diffusion Coefficient or Permeability of Hydrogen through Teflon and Palladium Membranes at Elevated Temperatures. Paper presented at Center of Nuclear Energy Research, Canada.

- Raymond, K.E. (1985). Alkali and chlorine products. Kirk-Othmer Concise Encyclopedia of Chemical Technology, 59-61.
- Raymond, K.E. (1998). Alkali and chlorine products. Encyclopedia of Chemical Technology, 1, 984-989.
- Sarner, S.F., and Henry III, N.W. (1989). The use of detector tubes following ASTM method F-739-85 for measuring permeation resistance of clothing. American Industrial Hygiene Association Journal, 50(6), 298-302.
- Tufts, L.E. (1992). Reaction Characteristics of hydrogen admixed with chlorine, oxygen and inert gases. In Explosive Properties of Gaseous Mixtures containing Hydrogen and Chlorine. Washington, DC: The Chlorine Institute, Inc.
- Zolandz, R.R., and Fleming, G.K. (1992). Definitions. In Winston Ho, W.S., and Sirkar, K.K. Membrane Handbook. New York: Van Nostrand Reinhold.

## APPENDICES

### Appendix A Properties of polymers

The following polymers were tested to determine the permeability coefficients of hydrogen and chlorine.

#### A.1 Teflon

Common Names: Polytetrafluoroethylene, PTFE  
Density = 2.280-2.290 g/cm<sup>3</sup>  
Viscosity (at 25°C) = 3000 centipoises  
Upper Working Temperature = 260°C  
Limiting Oxygen Index = 95%  
Molecular Weight = 99.96

#### A.2 Viton

Common Name:  
Hexafluoropropylenevinylidene fluoride copolymer  
Density = 2 g/cm<sup>3</sup>  
Flammability = Self Extinguishing  
Resistance to Ultra-violet = Excellent  
Upper Working Temperature = 220 to 300°C

#### A.3 Derakane Resin

Common Name: Epoxy vinyl ester resins  
Viscosity, cps at 25°C = 350  
Specific gravity = 1.045  
Tensile strength, psi = 12,100  
Heat Distortion Temperature = 99-104°C

### Appendix B Properties of normal-hydrogen gas

Hydrogen gas was tested to determine the permeation of hydrogen to the test materials.

#### Normal-Hydrogen gas

Physical properties  
Formula: H<sub>2</sub>  
Atomic number: Z=1  
Molecular weight: 2.016 g/mole  
Ionization potential: 15.427 eV  
Density S.T.P: 0.08989 kg/m<sup>3</sup>

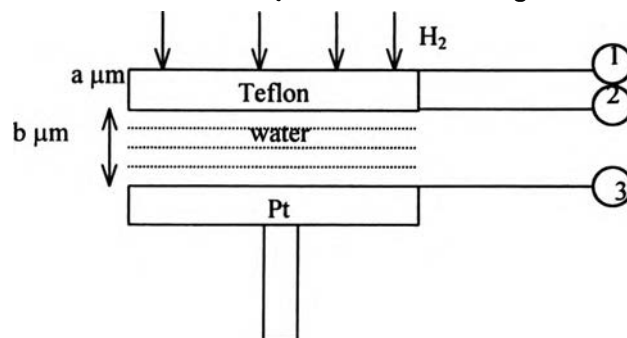
@ Operational condition: 80°C and 1atm  
Density: 0.06956 kg/m<sup>3</sup>  
Heat capacity at constant pressure (Cp): 3.452 kcal/kg K  
Heat capacity at constant volume (Cv): 2.463 kcal/kg K  
Viscosity: 9.92×10<sup>-5</sup> poises  
Thermal conductivity (k): 49.1276×10<sup>-5</sup> cal/cm sec K  
Melting point: -259.1°C  
Boiling point: -252.7°C

**Appendix C An example of current determination from electrochemical amperometric sensor using heat conduction in solids method**

Using 0.15 mm thick Teflon as a membrane in the amperometric sensor in 5% $H_2$  gas mixture at 25°C. Consider two situations which produce different amount of the current.

**C.1 Under steady state condition: the current is independent of time**

Two dimensional system as showed in Figure C.1 is considered.



**Figure C.1** Two dimensional Teflon-water system in an amperometric sensor. The mass flux of  $H_2$  can be determined from Fick's law.

$$J_1 = D_1 \frac{C_{H_2,1} - C_{H_2,2 \text{ in Teflon}}}{a} \quad (C.1)$$

and

$$J_2 = D_2 \frac{C_{H_2,2 \text{ in water}}}{b} \quad (C.2)$$

where:

$D_1$	=	$D_{H_2\text{-Teflon}}$
$D_2$	=	$D_{H_2\text{-water}}$
$a$	=	thickness of Teflon [ $\mu\text{m}$ ]
$b$	=	thickness of water layer [ $\mu\text{m}$ ].

Using the relationship of solute in gas and liquid phase at interface 2,

when 
$$C_{H_2,2 \text{ in Teflon}} = Sp_{H_2,2} \quad (C.3)$$

then

$$p_{H_2,2} = \frac{C_{H_2,2 \text{ in Teflon}}}{S} \quad (C.4)$$

Using Henry's law, when: 
$$p_{H_2,2} = H C_{H_2,2 \text{ in water}}$$

where:

$S$	=	Solubility of $H_2$ in Teflon.
$H$	=	Henry's Constant.

Which is, 
$$\frac{C_{H_2,2 \text{ in Teflon}}}{S} = H C_{H_2,2 \text{ in water}}$$

Thus, 
$$C_{H_2,2 \text{ in Teflon}} = SH C_{H_2,2 \text{ in water}} \quad (C.5)$$

At Steady State:  $J_{H_2} = J_1 = J_2$

Therefore, Eq(C.1) = Eq(C.2) and becomes,

$$D_1 \frac{C_{H_2,1} - C_{H_2,2 \text{ in Teflon}}}{a} = D_2 \frac{C_{H_2,2 \text{ in water}}}{b} \quad (C.6)$$

Substitute Eq(C.5) into Eq(C.6), Eq(C.6) becomes,

$$D_1 \frac{C_{H_2,1}}{a} = D_2 \frac{C_{H_2,2 \text{ in water}}}{b}$$

$$D_1 \frac{C_{H_2,1}}{a} = \frac{D_2 C_{H_2,2 \text{ in water}}}{b} + \frac{D_1 S H C_{H_2,2 \text{ in water}}}{a}$$

$$D_1 \frac{C_{H_2,1}}{a} = C_{H_2,2 \text{ in water}} \left[ \frac{D_2}{b} + \frac{D_1 S H}{a} \right]$$

$$D_1 \frac{C_{H_2,1}}{a} = C_{H_2,2 \text{ in water}} \left[ \frac{a D_2 + b D_1 S H}{a b} \right]$$

Then,

$$C_{H_2,2 \text{ in water}} = D_1 \frac{C_{H_2,1}}{a} \left[ \frac{a b}{a D_2 + b D_1 S H} \right]$$

$$C_{H_2,2 \text{ in water}} = \frac{b D_1 C_{H_2,1}}{a D_2 + b D_1 S H} \quad (C.7)$$

Substitute Eq(C.7) into Eq(C.2) and Eq(C.2) then becomes,

$$J_2 = \frac{D_2}{b} \frac{b D_1 C_{H_2,1}}{a D_2 + b D_1 S H}$$

Or

$$J_2 = \frac{D_1 D_2 C_{H_2,1}}{a D_2 + b D_1 S H} \quad (C.8)$$

At steady state, the response current,  $I_s$  is given as,

$$i_s = 2 F J_{H_2} \quad (C.9)$$

Substitute  $J_{H_2}$  in Eq(C.9) by  $J_2$  in Eq(C.8) to get

$$I_s = 2 F \frac{D_1 D_2 C_{H_2,1}}{a D_2 + b D_1 S H} \quad (C.10)$$

Evaluation of constants in Eq(C.10)

1)  $F$  = Faraday's constant =  $96500 \text{ A sec mol}^{-1}$

2)  $a$  = thickness of Teflon = [m]

3)  $b$  = thickness of water layer = [m]

4)  $D_2$  = Diffusivity of  $H_2$  in water at  $25^\circ\text{C}$  (Perry's Handbook) =  $58.5 \text{ m}^2 \text{ sec}^{-1}$

5)  $D_1$  = Diffusivity of  $H_2$  in Teflon at  $25^\circ\text{C}$  (Matsuyama *et al.*, 1982) =  $14.7 \pm 1.54 \times 10^{-8} \text{ cm}^2 \text{ sec}^{-1}$   
 $D_1 = 14.7 \pm 1.54 \times 10^{-4} \text{ m}^2 \text{ sec}^{-1}$

6)  $C_{H_2,1}$  = concentration of  $H_2$  in gas feed

$$p_{H_2,1} = 0.05 \text{ atm}$$

Using Ideal Gas Law: 
$$C_{H_2,1} = \frac{p_{H_2,1}}{RT}$$

Therefore, 
$$C_{H_2,1} = \frac{0.05 \text{ atm}}{0.08206 \times 10^{-3} \text{ m}^3 \text{ atm}} \left| \frac{\text{mol K}}{298.15 \text{ K}} \right|$$
  

$$= 2.044 \text{ mol m}^{-3}$$

7)  $S$  = Solubility of  $H_2$  in Teflon at  $25^\circ\text{C}$

From  $S = P/D$

Where  $P$  = Permeability Coefficient =  $3.347 \times 10^{-10} \text{ mol m}^{-1} \text{ atm}^{-1} \text{ sec}^{-1}$

$D$  = Diffusivity Coefficient =  $14.7 \times 10^{-4} \text{ m}^2 \text{ sec}^{-1}$

Thus, 
$$S = \frac{3.347 \times 10^{-10} \text{ mol}}{\text{m atm sec}} \left| \frac{\text{sec}}{14.7 \times 10^{-4} \text{ m}^2} \right|$$

$$S = 2.277 \times 10^{-7} \text{ mol m}^{-3} \text{ atm}^{-1}$$

8)  $H$  = Henry's constant of  $\text{H}_2$  in water at  $25^\circ\text{C}$  (Perry's Handbook)  
 $= 7.07 \times 10^{-4} \text{ atm (mol solution) (mol solute)}^{-1}$

Assumption: For dilute solution when mole fraction of solute,  $x \ll 1$  then mol solution = mol solvent.

Where: mol solute = mol  $\text{H}_2$  and mol solution = mol  $\text{H}_2\text{O}$

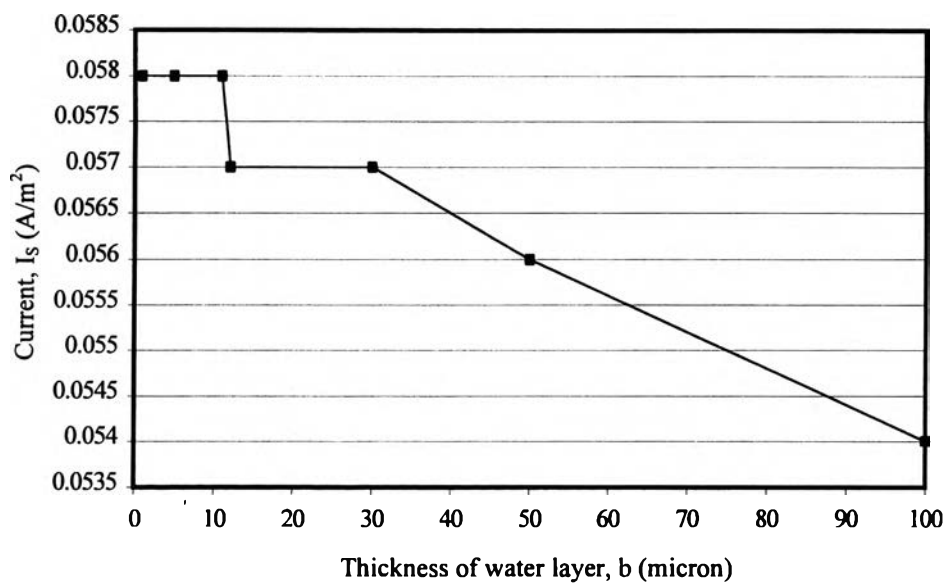
Then  $H = 7.07 \times 10^{-4} \text{ atm (mol H}_2\text{O) (mol H}_2\text{)}^{-1}$

$$= 7.07 \times 10^{-4} \frac{\text{atm mol H}_2\text{O}}{\text{mol H}_2} \left| \frac{1.8 \times 10^{-2} \text{ kg}}{\text{mol H}_2\text{O}} \right| \frac{\text{m}^3}{1000 \text{ kg}}$$

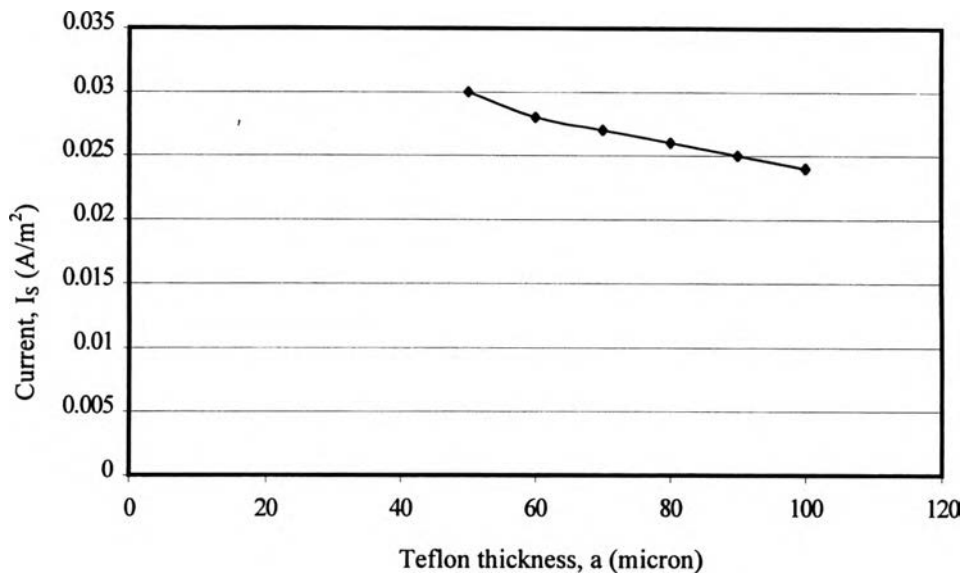
$$= 1.2726 \times 10^{-8} \text{ atm m}^3 \text{ mol}^{-1}$$

Substitute each constant into Eq.(C.10) and let  $n = m = 1 \mu\text{m} = 10^{-6} \text{ m}$

Therefore,  $I_s = 5.8 \times 10^{-8} \text{ A m}^{-2}$



**Figure C.2** The relationship between current and thickness of water layer under steady state condition when Teflon thickness is  $100 \mu\text{m}$ .



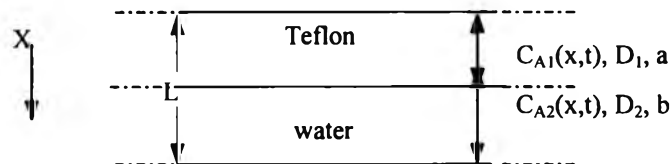
**Figure C.3** The relationship between current and Teflon thickness under steady state condition when thickness of water layer is 2 mm.

From the calculation of Teflon-water system in an amperometric sensor, Figure C.2 and C.3 show the current at the steady state as a function of thickness of water layer when Teflon thickness is constant at 100  $\mu\text{m}$  and a function of Teflon thickness when thickness of water layer is constant at 2 mm, respectively.

It is cleared that the response current is decreasing with an increasing in thickness of the water layer and Teflon.

#### C.2 Under unsteady state condition: the current is a function of time

Consider the diffusion of hydrogen into two-layer slab. Initially the entire region is at zero. For  $t > 0$ , the outer boundary surfaces at  $x = 0$  and  $x = L$  are kept at constant concentration  $C_{A0}$  and zero respectively.



**Figure C.4** Two-region of Teflon-water slab.

- Let
- $C_{A1}$  represents hydrogen concentration in Teflon
  - $C_{A2}$  represents hydrogen concentration in water
  - $D_1$  represents diffusivity coefficient of hydrogen in Teflon
  - $D_2$  represents diffusivity coefficient of hydrogen in water.
  - $a$  is thickness of Teflon
  - $b$  is thickness of water layer
  - $L$  is total thickness as shown in Figure C.4.

When there is no reaction within the system, the mass transfer differential equations are

$$\frac{\partial^2 C_{A1}}{\partial x^2} = D_1 \frac{\partial^2 C_{A1}}{\partial t} \quad (\text{C.11})$$

$$\frac{\partial^2 C_{A2}}{\partial x^2} = D_2 \frac{\partial^2 C_{A2}}{\partial t} \quad (\text{C.12})$$



The initial boundary conditions are

$$C_{A1} = 0 \quad \text{in region } 0 \leq x \leq a, t = 0 \quad (\text{C.13})$$

$$C_{A2} = 0 \quad \text{in region } a \leq x \leq L, t = 0 \quad (\text{C.14})$$

Since there is no contact resistance at the interface the boundary conditions are

$$D_1 \frac{\partial C_{A1}}{\partial x} = D_2 \frac{\partial C_{A2}}{\partial x} \quad \text{at } x = a, t > 0 \quad (\text{C.15})$$

$$C_{A1} = C_{A0} \quad \text{at } x = a, t > 0 \quad (\text{C.16})$$

$$C_{A2} = 0 \quad \text{at } x = L, t > 0 \quad (\text{C.17})$$

Using Henry's law to determine the relationship between  $p_{A1}$  and  $C_{A2}$  at the interface to obtain

$$p_{A1} = H C_{A2} \quad (\text{C.18})$$

where  $H$  is Henry's constant of hydrogen dissolved in water at 25°C, equals to 1.2726 atm<sup>3</sup> mol<sup>-1</sup> and  $p_{Ai}$  is partial pressure of hydrogen in layer  $i$ , which the value can be evaluated from the relationship of hydrogen in gas and liquid phase at interface

$$p_{A1} = \frac{C_{A1}}{S} \quad (\text{C.19})$$

and  $S$  is the solubility of hydrogen in Teflon at 25°C equals to 22.767 mol atm<sup>-1</sup> m<sup>-3</sup>.

Thus the relationship of  $C_{A1}$  and  $C_{A2}$  at  $x = a$  can be determined as

$$C_{A1} = k C_{A2} \quad (\text{C.20})$$

where  $k$  is constant equals to  $S$  multiplied by  $H$ .

The general solution of the system is in the form of

$$C_{Aj}(x, t) = C_{Aj}(x, t) + \sum_{n=1}^{\infty} A_n X_{jn}(x) e^{-D_j \beta_{jn}^2 t} \quad (\text{C.21})$$

where  $j = 1$  for region  $0 \leq x \leq a$

$j = 2$  for region  $a \leq x \leq L$

The eigenfunction  $X_{jn}(x)$  in cartesian co-ordinate system are

$$X_{1n}(x) = \sin(\beta_{1n} x) \quad (\text{C.22})$$

$$X_{2n}(x) = B_n [\sin(\beta_{2n} x) + C_n \cos(\beta_{2n} x)] \quad (\text{C.23})$$

Using the boundary condition at  $x = L$ , i.e., Eq. C.17 to evaluate the unknown coefficient  $C_n$ ,

$$\begin{aligned} X_{2n}(x) &= 0 \quad \text{at } x = L \\ B_n [\sin(\beta_{2n} L) + C_n \cos(\beta_{2n} L)] &= 0 \\ C_n &= -\tan(\beta_{2n} L) \end{aligned} \quad (\text{C.24})$$

Since there is no mass storage at the interface, the time behavior of concentration should be the same on either side of the interface. This condition is satisfied when

$$D_1 \beta_{1n}^2 = D_2 \beta_{2n}^2$$

Then

$$\beta_{1n} = \sqrt{\frac{D_2}{D_1}} \beta_{2n} \quad (\text{C.25})$$

The unknown coefficient  $B_n$  is determined using the boundary condition at the interface, i.e., Eq. C.15,

$$\begin{aligned} D_1 \frac{\partial X_{1n}(x)}{\partial x} &= D_2 \frac{\partial X_{2n}(x)}{\partial x} \quad \text{at } x = a \\ D_1 \frac{\sin(\beta_{1n} x)}{x} &= D_2 \frac{B_n [\sin(\beta_{2n} x) + C_n \cos(\beta_{2n} x)]}{x} \quad \text{at } x = a \end{aligned}$$

$$D_1 \beta_{1n} \cos(\beta_{1n} x) = D_2 B_n \beta_{2n} [\cos(\beta_{2n} x) - C_n \sin(\beta_{2n} x)] \quad \text{at } x = a$$

$$B_n = \left( \frac{D_1 \beta_{1n} \cos(\beta_{1n} x)}{D_2 \beta_{2n} \cos(\beta_{2n} x) - C_n \sin(\beta_{2n} x)} \right) \quad \text{at } x = a$$

So,

$$B_n = \left( \frac{D_1 \beta_{1n} \cos(\beta_{1n} a)}{D_2 \beta_{2n} \cos(\beta_{2n} a) - C_n \sin(\beta_{2n} a)} \right) \quad (\text{C.26})$$

Using the boundary condition at the interface, i.e., Eq. C.20, to determine the eigenvalues  $\beta_{1n}$  and  $\beta_{2n}$ , that is

$$\begin{aligned} X_{1n}(x) &= k X_{2n}(x) \quad \text{at } x = a \\ \sin(\beta_{1n} x) &= k B_n [\sin(\beta_{2n} x) + C_n \cos(\beta_{2n} x)] \quad \text{at } x = a \end{aligned}$$

$$\sin(\beta_{1n} a) = k B_n [\sin(\beta_{2n} a) + C_n \cos(\beta_{2n} a)] \quad (C.27)$$

Substitute value of  $B_n$  and  $C_n$  into Eq. C.27, then Eq. C.27, becomes,

$$\sin(\beta_{1n} a) = k \frac{D_1 \beta_{1n}}{D_2 \beta_{2n}} \frac{\cos(\beta_{1n} a)}{\cos(\beta_{2n} a) + \tan(\beta_{2n} L) \sin(\beta_{2n} a)} [\sin(\beta_{2n} a) - \tan(\beta_{2n} L) \cos(\beta_{2n} a)] \quad (C.28)$$

After that substitute  $\beta_{1n}$  from Eq. C.25 into Eq. C.28, Eq. C.28 becomes,

$$\sin\left(\sqrt{\frac{D_2}{D_1}} \beta_{2n} a\right) = k \sqrt{\frac{D_1}{D_2}} \frac{\cos\left(\sqrt{\frac{D_2}{D_1}} \beta_{2n} a\right)}{\cos(\beta_{2n} a) + \tan(\beta_{2n} L) \sin(\beta_{2n} a)} \times [\sin(\beta_{2n} a) - \tan(\beta_{2n} L) \cos(\beta_{2n} a)] \quad (C.29)$$

Evaluation of  $\beta_{2n}$  can be made from Eq. C.29, by trial and error method.  $A_n$  is an arbitrary constant determined by

$$A_n = \frac{W_1^2 \int_0^a F_1(x) X_{1n}(x) w(x) dx + W_2^2 \int_a^L F_2(x) X_{2n}(x) w(x) dx}{W_1^2 \int_0^a X_{1n}^2(x) w(x) dx + W_2^2 \int_a^L X_{2n}^2(x) w(x) dx} \quad (C.30)$$

where, for two-layer slab problem:

$W_1, W_2$  are the discontinuous-weighting function, equals to 1

$w(x)$  is the Sturm-Liouville weighting function for the cartesian co-ordinate system, equals 1

$F_1(x)$  and  $F_2(x)$  are the expanded function in an infinite series of eigenfunctions at  $t = 0$  and are determined by substituting  $C_{Aj}(x, t)|_{t=0} = 0$  for  $j = 1, 2$  in the general solution, i.e., Eq. C.21.

$$\text{For } j = 1, \quad C_{A1}(x, t)|_{t=0} - C_{A1}(x, t) = A_n X_{1n}(x) e^{-D_1 \beta_{1n}^2 t} \\ -C_{A1}(x, t) = A_n X_{1n}(x) e^{-D_1 \beta_{1n}^2 t} \quad \text{in } 0 \leq x \leq a \quad (C.31)$$

$$\text{For } j = 2, \quad C_{A2}(x, t)|_{t=0} - C_{A2}(x, t) = A_n X_{2n}(x) e^{-D_2 \beta_{2n}^2 t} \\ -C_{A2}(x, t) = A_n X_{2n}(x) e^{-D_2 \beta_{2n}^2 t} \quad \text{in } a \leq x \leq L \quad (C.32)$$

$$\text{Then} \quad F_1(x) = -C_{A1}(x, \infty) \quad (C.33)$$

$$F_2(x) = -C_{A2}(x, \infty) \quad (C.34)$$

where  $C_{A1}(x, \infty)$  and  $C_{A2}(x, \infty)$  are the steady-state solutions defined from boundary conditions Eq.

C.16 and C.17, to get  $C_{A1}(x, \infty)$  and  $C_{A2}(x, \infty)$  are  $C_{A0}$  and zero respectively.

Eq. C.33 and C.34, then, have a form of,

$$F_1(x) = -C_{A0} \quad (C.35)$$

$$F_2(x) = 0 \quad (C.36)$$

Substitute  $W_1(x), W_2(x), w(x), F_1(x), F_2(x), X_{1n}(x)$  and  $X_{2n}(x)$  into Eq. C.30,

$$A_n = \frac{\int_0^a -C_{A0} \sin(\beta_{1n} x) dx}{\int_0^a \sin^2(\beta_{1n} x) dx + \int_a^L \{B_n [\sin(\beta_{2n} x) + C_n \cos(\beta_{2n} x)]\}^2 dx}$$

$$A_n = \frac{-C_{A0} \int_0^a \sin(\beta_{1n} x) dx}{\int_0^a \sin^2(\beta_{1n} x) dx + B_n^2 \int_a^L [\sin(\beta_{2n} x) + C_n \cos(\beta_{2n} x)]^2 dx}$$

$$\frac{A_n}{C_{A0}} = \frac{\int_a^0 \sin(\beta_{1n} x) dx}{\int_0^a \sin^2(\beta_{1n} x) dx + B_n^2 \int_a^L [\sin(\beta_{2n} x) + C_n \cos(\beta_{2n} x)]^2 dx} \quad (C.37)$$

Let  $Q = \int_a^0 \sin(\beta_{1n} x) dx = \frac{\cos(\beta_{1n} a) - 1}{\beta_{1n}}$

$$R = \int_0^a \sin^2(\beta_{1n} x) dx = \frac{1}{2} \left[ a - \frac{\sin(2\beta_{1n} a)}{2\beta_{1n}} \right]$$

$$\begin{aligned} S &= B_n^2 \int_a^L [\sin(\beta_{2n} x) + C_n \cos(\beta_{2n} x)]^2 dx \\ &= B_n^2 \left\{ \frac{1}{2} [(L-a) + \frac{\sin(2\beta_{2n} a) - \sin(2\beta_{2n} L)}{2\beta_{2n}}] + \frac{C_n}{\beta_{2n}} [\sin^2(\beta_{2n} L) - \sin^2(\beta_{2n} a)] \right. \\ &\quad \left. + \frac{C_n^2}{2} [(L-a) + \frac{\sin(2\beta_{2n} L) - \sin(2\beta_{2n} a)}{2\beta_{2n}}] \right\} \end{aligned}$$

Summarizing, the concentration distribution in the two-layer Teflon-water slab from Eq. C.21, is shown as,

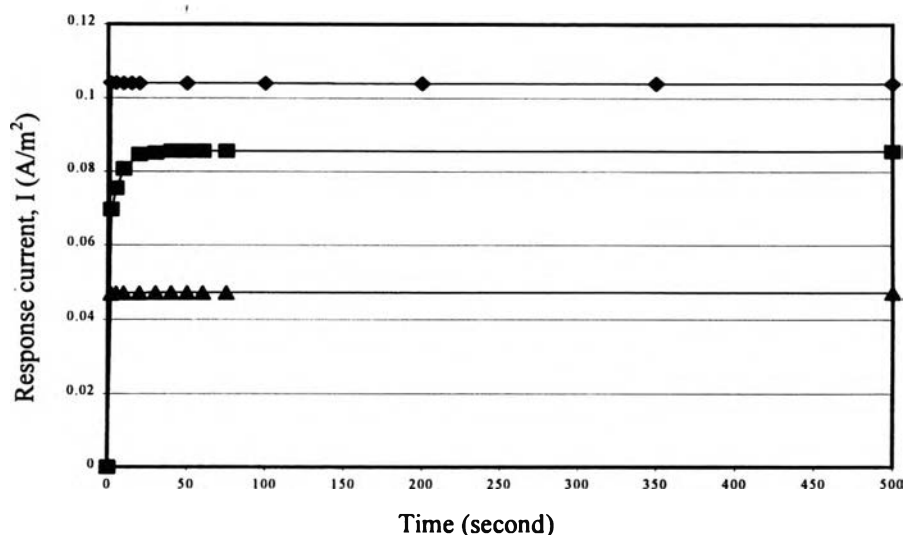
$$\begin{aligned} C_{A1}(x, t) &= C_{A0} + A_n X_{1n}(x) e^{-D_1 \beta_{1n}^2 t} \\ C_{A2}(x, t) &= A_n X_{2n}(x) e^{-D_2 \beta_{2n}^2 t} \end{aligned}$$

or,

$$\begin{aligned} \frac{C_{A1}(x, t)}{C_{A0}} &= 1 + \frac{A_n}{C_{A0}} X_{1n}(x) e^{-D_1 \beta_{1n}^2 t} \\ \frac{C_{A2}(x, t)}{C_{A0}} &= \frac{A_n}{C_{A0}} X_{2n}(x) e^{-D_2 \beta_{2n}^2 t} \end{aligned} \quad (C.38)$$

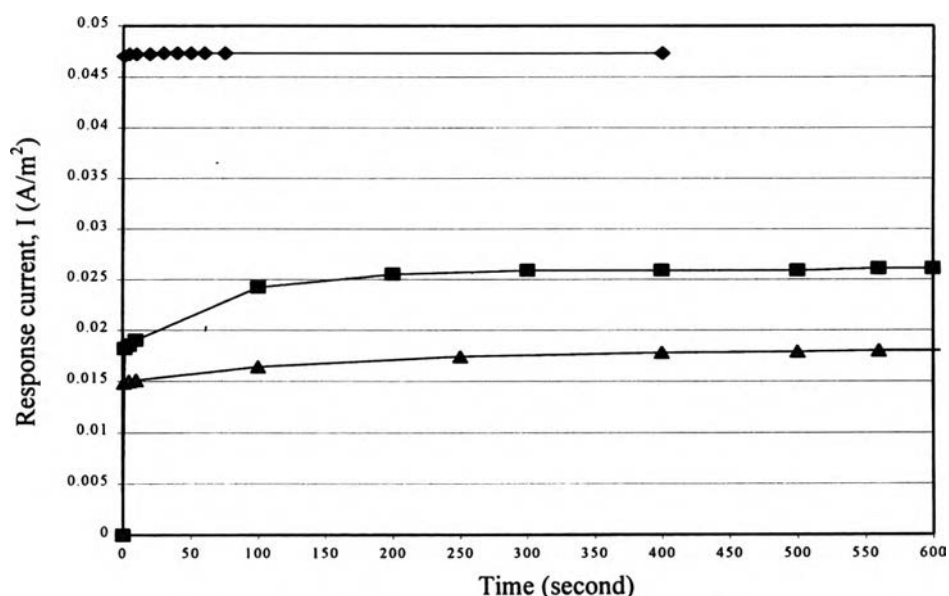
Using  $C_{A1}(x, t)$  and  $C_{A2}(x, t)$  obtained from Eq. C.38 to determine the mass flux of hydrogen from Fick's law, i.e., Eq. C.1 and C.2. Under the unsteady state condition,

$$J_{\text{total}} = J_1 + J_2 \quad (C.39)$$



**Figure C.5** The relationship between the response current of Teflon-water system of an amperometric sensor under unsteady state as a function of time when the Teflon thickness is constant at 100  $\mu\text{m}$  where  $\blacklozenge$ ,  $\blacksquare$ ,  $\blacktriangle$  represent 50  $\mu\text{m}$ , 0.5 mm and 2 mm thick of water layer, respectively.

Then the response currents under unsteady state condition at various thickness of water layer and Teflon were plotted as a function of time to study the effect of the thickness of polymer coating on an electrical amperometric sensor on the Breakthrough Time (B/T) as shown in Figure C.4 and C.5.



**Figure C.6** The relationship between the response current of Teflon-water system of an amperometric sensor under unsteady state as a function of time when the thickness of water layer is constant at 2 mm where  $\blacktriangle$ ,  $\blacksquare$ ,  $\blacklozenge$  represent 0.1 mm 0.3 mm and 0.5 mm, thick of water layer, respectively.

From Figure C.4 and C.5, the response currents increase with an increasing distance from the Teflon surface to the platinum surface that detected the hydrogen concentration in a form of an electron. In addition, the Breakthrough Time (B/T) of each thickness is concluded in Table C.1.

**Table C.1** The Breakthrough Time (B/T) of hydrogen concentration of Teflon-water system in an amperometric sensor under unsteady state condition.

Teflon thickness (mm)	Thickness of water layer (mm)	Hydrogen Breakthrough Time (B/T), (second)
0.1	0.05	2
0.1	0.50	40
0.1	2.00	75
0.3	2.00	560
0.5	2.00	600

The advantage of an amperometric sensor relative to the Pd/H electrical resistance is the relative rapidly of response to a change of hydrogen pressure. It follows from the fact that the detection principle is not dependent upon the absorption of hydrogen. However, it is believed that the amperometric sensor may require periodic recalibration or regeneration of the platinum anode due to the progressive loss of the electrochemical sensor.

#### Appendix D Theoretical Analysis for gas diffusion through solids

In the case where

1. The slab region is  $-L < x < L$
2. Initial concentration  $c = f(x) = 0$
3. The ends are kept at constant concentration  $C$

In the region  $-L < x < L$

The diffusion equation is

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} \quad (D.1)$$

$$c = C \text{ when } x = -L \text{ and } x = L$$

$$\text{and } c = f(x) \text{ at } t = 0$$

The solution of this differential equation has the following form (Carslaw, and Jaeger, 1959, ch.3);

$$c = C - \frac{4C}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^n}{2n+1} e^{-D(2n+1)^2 \pi^2 t / 4L^2} \cos \frac{(2n+1)\pi x}{2L} \quad (D.2)$$

Introducing the dimensionless parameters

$$T = \frac{Dt}{L^2} \quad \text{and} \quad E = \frac{x}{L} \quad (D.3)$$

$$c = C - \frac{4C}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^n}{2n+1} e^{-T(2n+1)^2 \pi^2 / 4} \cos \frac{(2n+1)\pi E}{2} \quad (D.4)$$

The solution becomes,

$$\frac{c}{C} = 1 - \frac{4}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^n}{2n+1} e^{-T(2n+1)^2 \pi^2 / 4} \cos \frac{(2n+1)\pi E}{2} \quad (D.5)$$

Or  
When  $n = 1$ ;

$$\frac{c}{C} = 1 - \frac{4}{\pi} \left[ e^{-0.25 \pi^2 \tau} \cos \frac{\pi E}{2} - \frac{1}{3} e^{-2.25 \pi^2 \tau} \cos \frac{3 \pi E}{2} \right] \quad (D.6)$$

**Nomenclature**

$c$  = concentration of permeant  $H_2$  through the membrane at any time,  $t$ .

$n$  = number of the slabs.

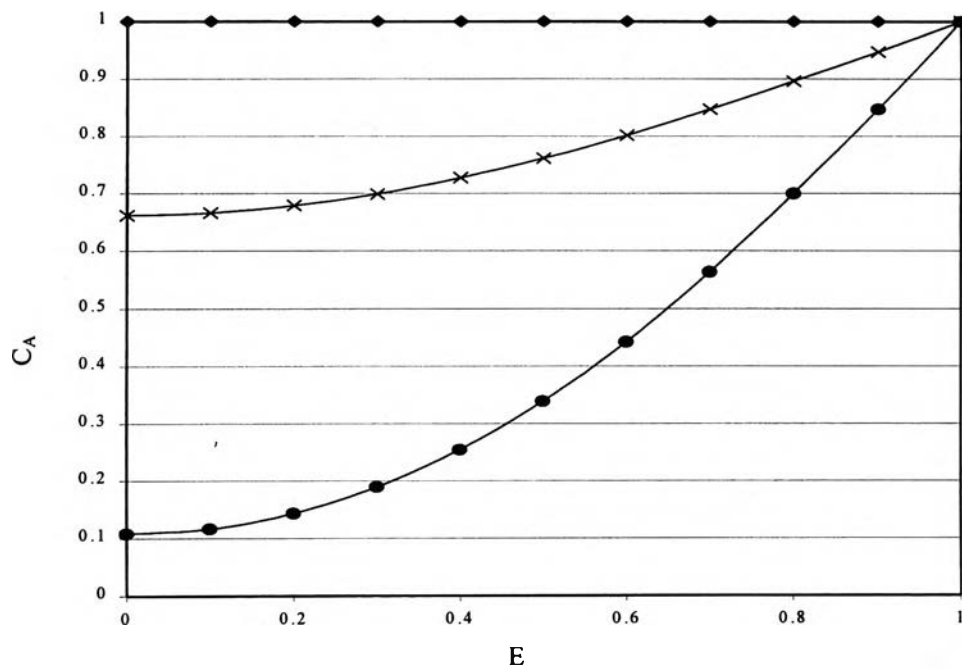
$L$  = thickness of the slab.

$D$  = Diffusivity coefficient of hydrogen to the membrane.

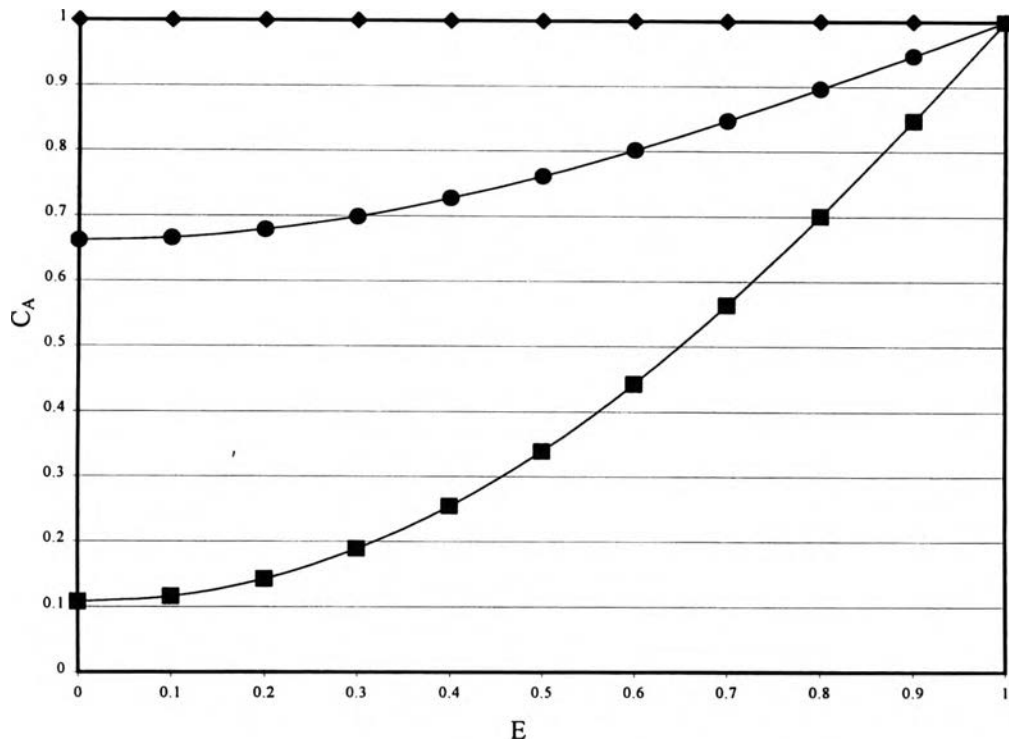
**Assumption:**

$$D_{H_2\text{-Derakane resin}} = 4.31 \times 10^{-12} \text{ m}^2/\text{sec}$$

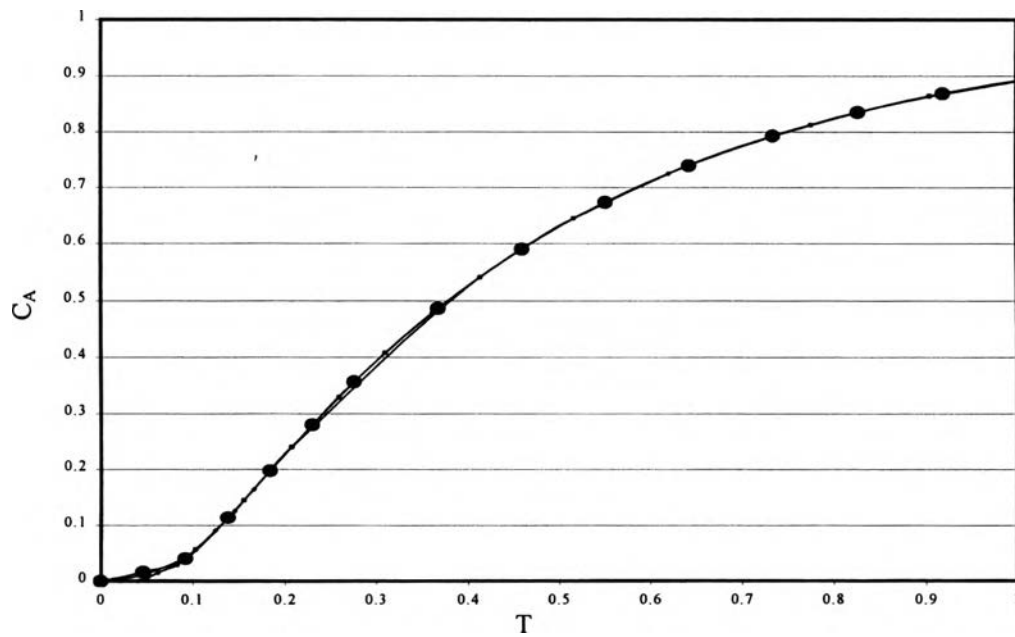
$$D_{H_2\text{-Viton}} = 5.60 \times 10^{-11} \text{ m}^2/\text{sec}$$



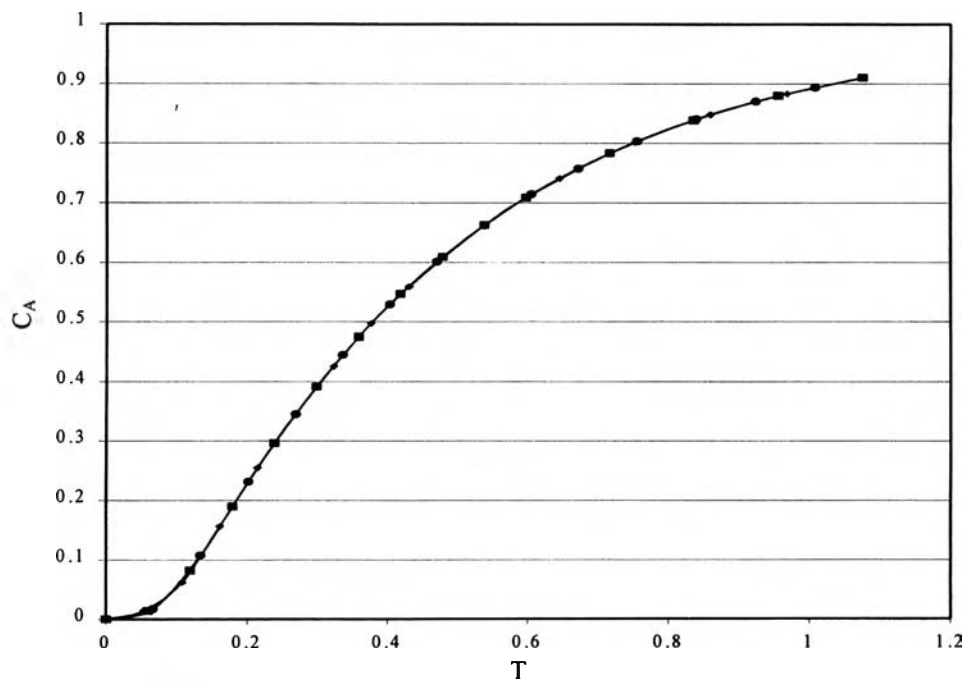
**Figure D.1** The concentration distribution at various thickness of epoxy vinyl ester resin at  $t = 10$  minute, \* = 0.05mm, ■ = L is 0.1 mm and ▲ = L is 0.25 mm.



**Figure D.2** The concentration distribution at various thickness of Fluorodyn solution at  $t = 10$  minute,  $\blacklozenge = L$  is 0.1 mm,  $\bullet = L$  is 0.25 mm,  $\blacksquare = L$  is 0.5 mm.



**Figure D.3** The center concentration at various thickness of epoxy vinyl ester resin slab as a function of time,  $\bullet = L$  is 0.075,  $\blacklozenge = L$  is 0.1 mm,  $\blacksquare = L$  is 0.25 mm.



**Figure D.4** The center concentration at various thickness of Fluorodyn solution slab as a function of time, ■ = 0.075 mm, ● = 0.1 mm, ◆ = L is 0.25 mm.



