



CHAPTER III EXPERIMENTAL

3.1 Materials

- Derakane®470-300 (Epoxy vinyl ester based resin) from Dow Chemical Company.
- Derakane®8084 (Epoxy vinyl ester based resin) from Dow Chemical Company.
- Teflon® sheet (Engineering grade) from EnFlo Inc.

3.2 Experiments

3.2.1 Membrane Preparation

A sandwich membrane was prepared by firstly mixing Derakane® resin with methyl-ethyl-ketone peroxide (MEKP) catalyst and allowing the mixture to polymerize for 10-20 minutes*. Then, a small amount of the mixture was poured on a Teflon® sheet. Another Teflon® sheet was placed on top of the mixture to make a sandwich membrane with Derakane® resin in between. After that, the Derakane® mixture was spread all over the two Teflon® sheets. A load of approximately 10 kg was put on the sandwich membrane and the sandwich membrane was allowed to dry for at least one day. A digital micrometer (Digical III from Sylvac) was used to measure the thickness of the sandwich membrane.

*The period of time required for Derakane® to form polymer chains is called the gel time. It depends on the type of Derakane®, for Derakane® 470-300 the gel time is 10 minutes, but it is 20 minutes for Derakane® 8084.

3.2.2. The Determination of Hydrogen and Chlorine Permeability Coefficients

The hydrogen and chlorine permeability coefficients of sandwich membranes in H₂-Ar atmosphere and pure chlorine atmosphere at 25, 50 and 80°C were determined following standard method ASTM F739-91. The set up of the apparatus to determine the H₂ permeability coefficient of the sandwich membrane is shown in Figure 3.1. It was composed of a test cell (ASTM Test cell from A.A. Pesce Glass Company) consisting of two chambers; the challenge side and the collection side, separated by the sandwich membrane. The test cell was placed in an oven held at constant temperature (at 25, 50 or 80°C). The challenge gas (H₂-Ar) was flowed to the inlet of the challenge side of the test cell and argon, which was used as the collection gas (or sweep gas), was flowed to the inlet of the collection side. The gas from the outlet of the collection side of the test cell was analyzed by a gas chromatograph (GC Varian 3600).

In the case of the determination of the chlorine permeability coefficient, a method using detector tubes in conjunction with ASTM F739-91 described by Sarner and Henry (1989) was applied. Figure 3.2 illustrates the set up of the apparatus for this experiment. It was quite similar to the set up of the determination of the hydrogen permeability coefficient but, instead of using a gas chromatograph, Cl₂ detector tubes (No.109SB from Kitagawa) were used. Cl₂ detector tubes contain a chemical which changes color when reacting with chlorine. The stain length indicates the cumulative amount of chlorine permeating through the membrane. The chlorine permeation was recorded with time and then the permeation flux of chlorine was determined by the slope of the graph of the amount of Cl₂ permeation (mg/m²) versus time (s).

For the mixed gas system, the determination of both hydrogen and chlorine permeability coefficients was made by setting up the apparatus as illustrated in Fig.3.3. Both analytical devices; the gas chromatograph and chlorine detector tubes were used in series. The gas from the collection side of the test cell was connected with the chlorine detector tubes first so that the chemical in the tube could absorb the chlorine before it entered the gas chromatograph. The operating temperature for the mixed gas experiments was at 80°C.

3.3 Procedure

3.3.1 The Determination of Hydrogen Permeability Coefficient in H₂ – Ar Atmosphere

1.1 The test cell was set up with H₂/Ar mixture as the challenge gas.

Challenge gas: Gas mixture of 95% Ar and 5% H₂

Collection gas: Ar

1.2 The tested material was mounted in the test cell and assembled as shown in Figure 3.1

1.3 The test cell was installed in an oven, held at the desired temperature.

1.4 The outlet of collection side was connected to the gas chromatograph.

1.5 The collection gas was passed into the collection chamber continuously at a flow rate of 10 cm³/min.

1.6 Sampling of the collection side gas was initiated and analyzed by a gas chromatograph. Samples were analyzed every 10 minutes.

1.7 The challenge gas was allowed to flow into the challenge chamber at the rate of five times the chamber volume (volume of the chamber = 100 cm³) for 1 minute. Then, the flow rate was reduced to 10 cm³/min.

1.8 The timing of the test was started when the challenge gas flow rate was reduced to 10 cm³ / min. The concentration of hydrogen in the challenge gas and the time of the analysis were recorded.

1.9 The test was continued until a constant concentration of H₂ in the collection stream was achieved.

3.3.2 The Determination of Chlorine Permeability Coefficient in Pure Chlorine Gas

The test apparatus was set up as shown in Figure 3.2.

Challenge gas : Pure chlorine

Collection gas: Ar

The same procedure as the previous section was followed but instead of using a gas chromatograph, chlorine detector tubes were used. The cumulative amount of chlorine permeating through the membrane was recorded with time.

3.3.3 Test of Materials with Mixed Hydrogen and Chlorine Gas

The same procedure described in section 3.3.1 and 3.3.2 was used for hydrogen and chlorine permeability coefficient determination. The set up of the apparatus was shown in Figure 3.3. The challenge gas mixtures used were 1%, and 5% hydrogen in chlorine.

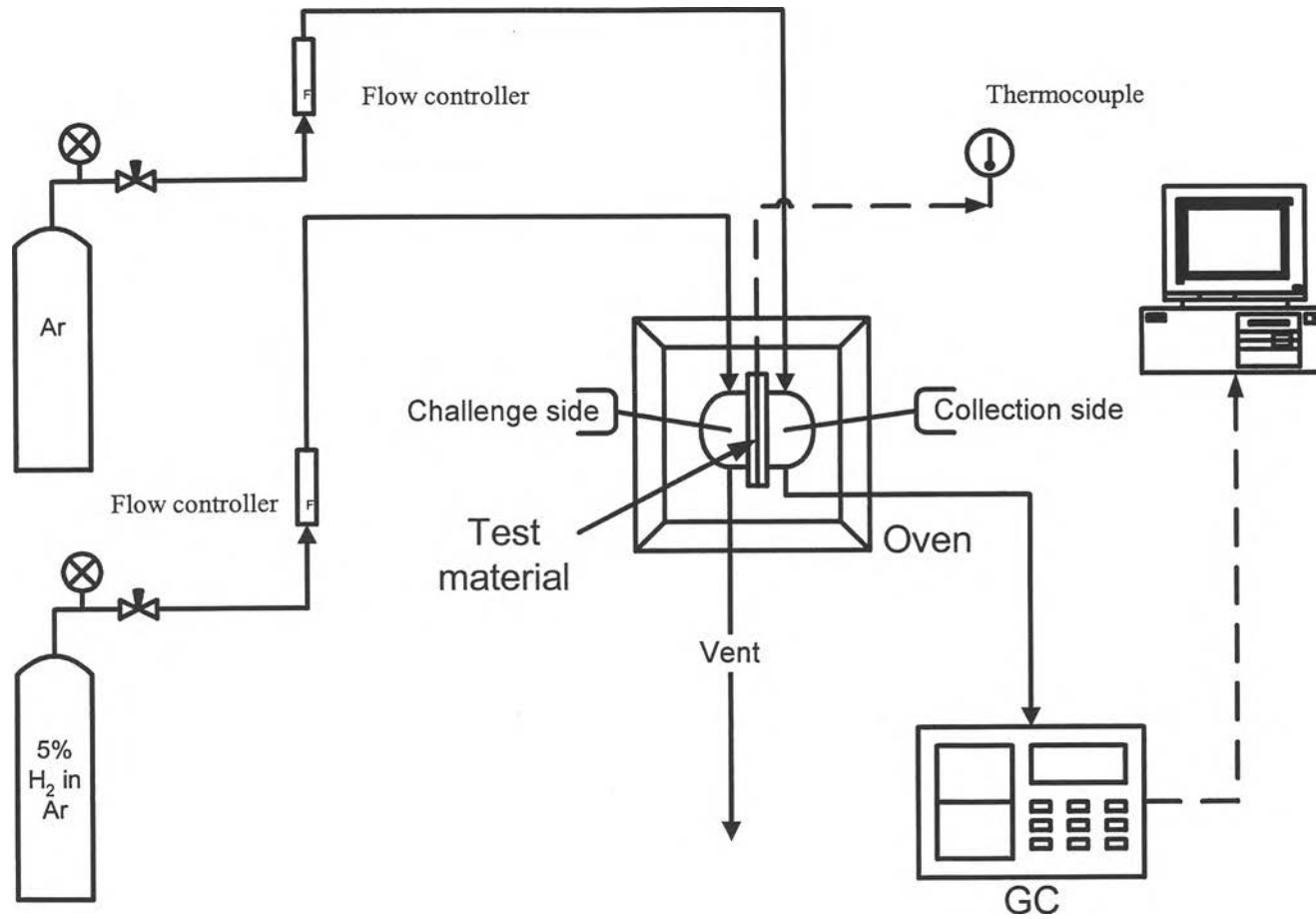


Figure 3.1 Schematic of the apparatus for the determination of H₂ permeability coefficients in H₂-Ar gas system.

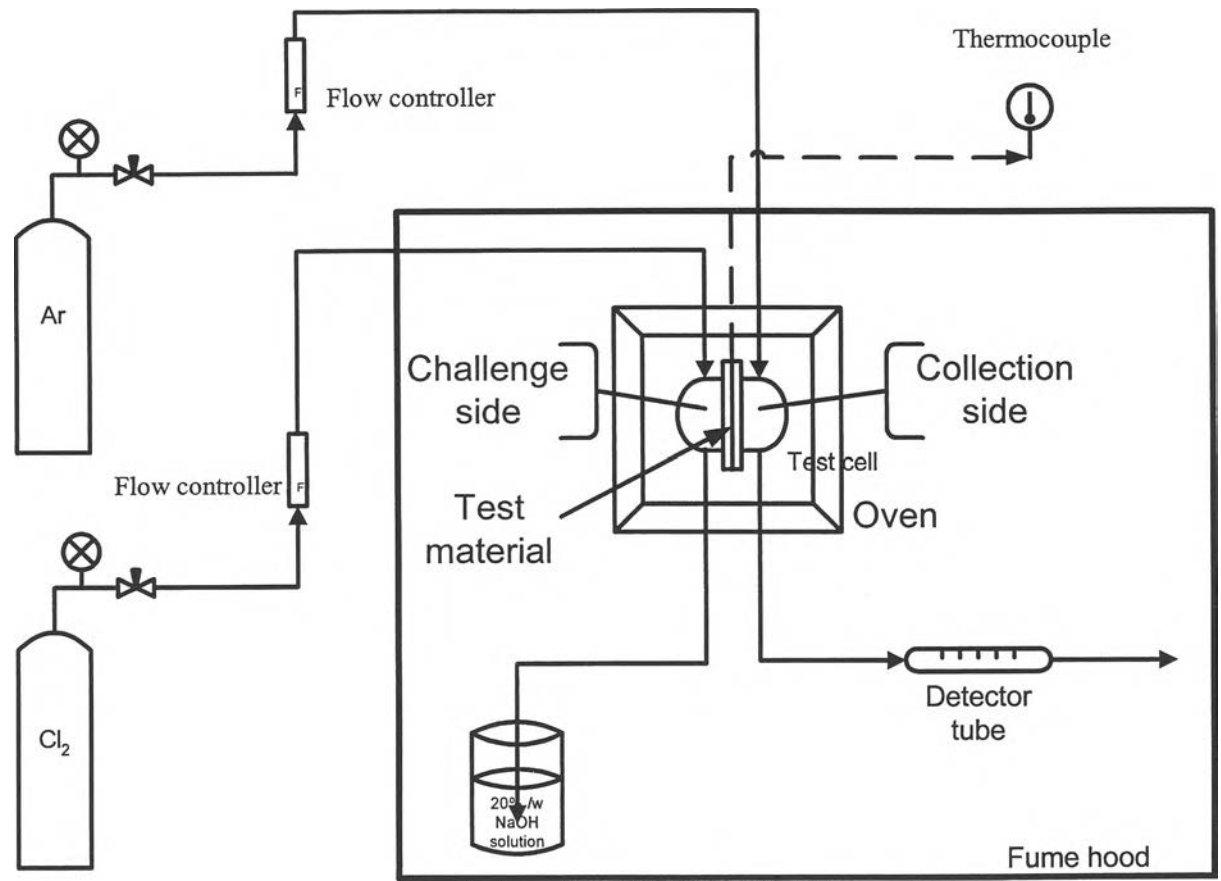


Figure 3.2 Schematic of the apparatus for the determination of Cl₂ permeability coefficients in pure chlorine gas system.

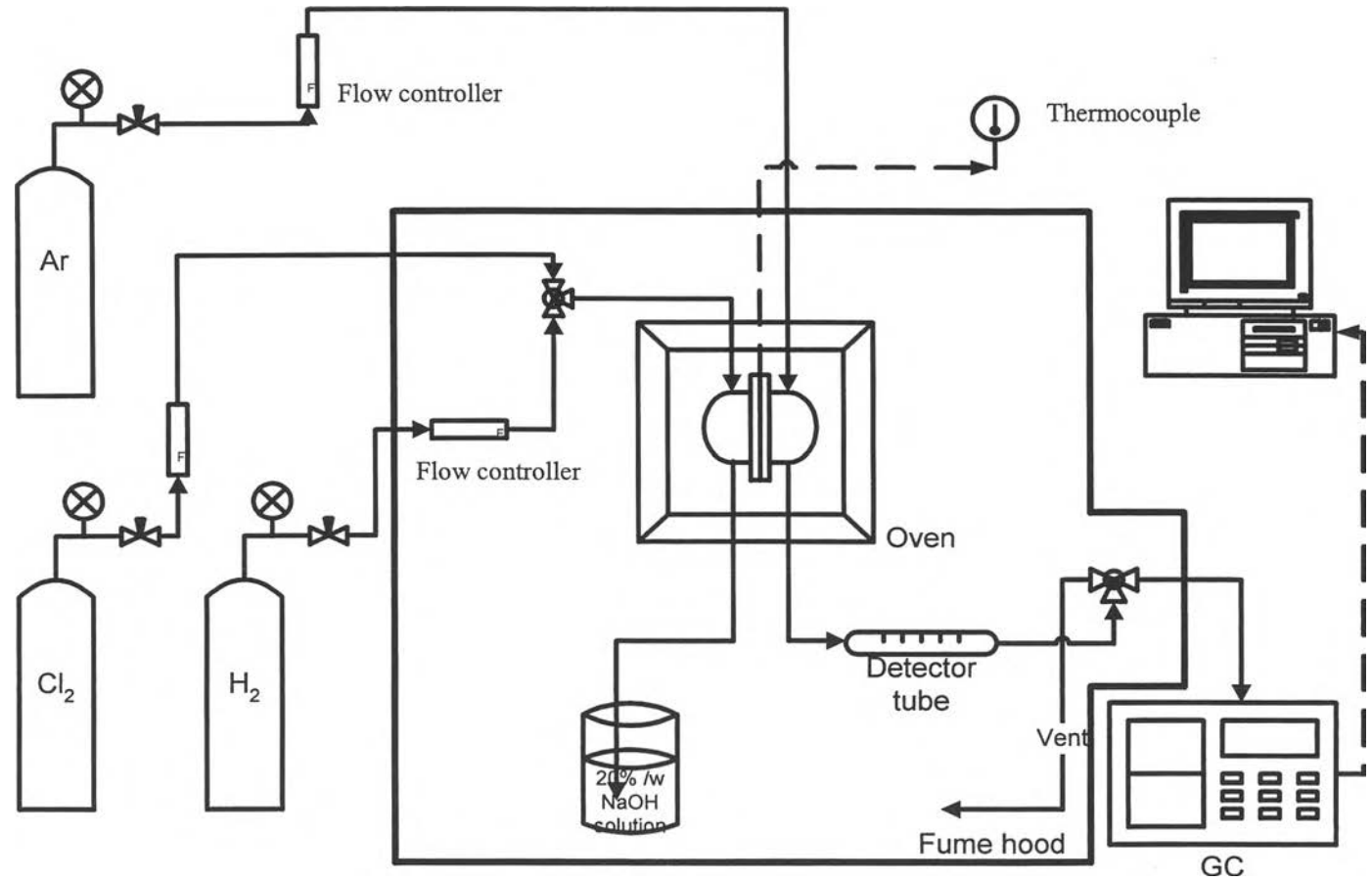


Figure 3.3 Schematic of the apparatus for the determination of H₂ and Cl₂ permeability coefficients in mixed gas.

3.4 Calculation of Permeability Coefficient

Permeability coefficient describes the amount of gas permeating through a membrane at constant temperature per unit time, per unit area, per unit pressure difference. At steady state, the permeability coefficient of a gas through a membrane can be calculated by the following equation:

$$P_i = \frac{J_i}{(p_1 - p_2)/l} \quad (28)$$

where J_i is the flux of component i at steady state, p_1 and p_2 are the feed and permeate side pressure and l is the thickness of the membrane.

In this study, the permeate side was swept continuously with fresh Ar so p_2 can be eliminated and Eq.(28) can be written as follow:

$$P_i = \frac{J_i l}{p_1} \quad (29)$$

The permeation flux, also called the permeation rate, J_i , was calculated from the concentration data obtained from the analytical devices. Eq.(30) describes the permeation rate of hydrogen in the collection gas at any time.

$$J_i(t_i) = \frac{C_{H_i} F_{Ar}}{A} \quad (30)$$

where $J_i(t_i)$ is permeation flux, ($\text{mole} \cdot \text{m}^{-2} \cdot \text{min}^{-1}$);

t_i is time at which concentration of hydrogen in collection gas was C_{H_i} , min;

C_{H_i} is concentration of hydrogen at time t_i , $\text{mole} \cdot \text{L}^{-1}$;

F_{Ar} is flow rate of fresh argon through the collection side of the test cell, $\text{L} \cdot \text{min}^{-1}$;

A is Cross sectional area of flow, m^2 .

For chlorine, the detector tube indicates the cumulative amount of chlorine permeating through the membrane by a calibrated scale marked on the tube. The absolute amount of permeation at one time, P_M , can be known by;

$$P_M = C_b (\text{conversion factor}) V_L \quad (31)$$

where C_b = tube calibration concentration as indicated on the scale, ppm

$$\text{conversion factor} = \frac{\text{molecular weight of challenge gas}}{\text{gas molecular volume}}, g.L^{-1};$$

The molecular volume of a gas using an ideal gas law is 24.46 L at 25°C.

V_L = volume of gas that is used to generate the calibration, (100 mL for Kitagawa detector tube).

Then, the chlorine permeation in mg/m^2 unit is determined using Eq.(32);

$$\text{permeation} = \frac{P_M}{A} \quad (32)$$

The slope of the graph of permeation versus time represents the permeation flux of chlorine through the membrane (shown in Figure 3.4). Then, the Cl_2 permeability coefficient can be determined using Eq.(29).

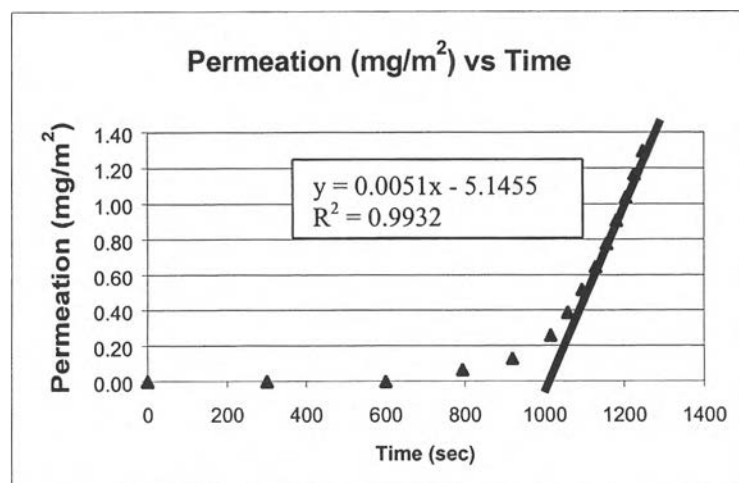


Figure 3.4 An example of the plot of the permeation of chlorine versus time.