Characterization of Pressure-Sensitive Adhesives. II. Water Vapor Permeation Studies

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Synopsis

The permeability of water vapor in a composite film [a Mylar (trademark of DuPont, Inc.) film coated with a pressure sensitive adhesive on both sides] and a Mylar film (type D) have been determined at 23°C. The water vapor permeability in the pressure sensitive adhesive, Flexbond 150 (a trademark of Air Products and Chemicals), and the Mylar film have been found to be 3.23×10^{-7} and 2.30×10^{-8} cm^3 (STP) cm · cm^{-2} · s^{-1} · (cm Hg)$^{-1}$, respectively, at 23°C.

INTRODUCTION

Flexbond 150 is a pressure-sensitive adhesive which is soft and tacky at room temperature. We have used Mylar films bonded by the adhesive in several of our products. The performance of the adhesive under high temperature and humidity has been of great concern. In a previous paper,[1] we reported the effect of temperature and humidity on the bonding strength of the adhesive. The water vapor sorption by the adhesive was also determined. It has been found that the adhesive bonding strength generally decreases with an increase in temperature, but the effect of humidity on the adhesive bonding strength is more complex. To understand the effect of humidity on the bonding strength of the adhesive, it is desirable to know the water vapor permeation rate in the adhesive. Since the glass transition temperatures (T_g) of the adhesives are generally lower than -20°C,[2] it is often desirable to use composite films (with the adhesive coated on a polymer substrate) to determine water permeability in the adhesive at room temperature.

In this article we report the results of experiments in which the water permeability in a composite film, a Mylar film double-coated with the pressure sensitive adhesive, Flexbond 150, and a single layer of Mylar film were determined. The adhesive Flexbond 150 is a vinyl acetate copolymer adhesive.[3] The results obtained indicate that the water vapor permeability in the adhesive at room temperature can be conveniently determined by use of a composite film. It has been found that water permeability in the adhesive is significantly (a factor of 14) higher than that in the Mylar film.

EXPERIMENTAL

Mylar films (type D, 3 mil thick) were obtained from DuPont, Inc. Mylar films coated with Flexbond 150 pressure sensitive adhesive on both sides

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were also obtained externally. The thickness of the adhesive is 1.5 mil. To determine the water vapor permeability, the modified weighed cell method described by Newns[4] was used. In this method, the adhesive-coated Mylar film (or the single-layer Mylar film) is sealed to the edge of a circular cylinder which contains a saturated salt solution. The cylinder is placed inside a desiccator which is filled with desiccants. The desiccator is then placed inside an oven with the temperature controlled at 23 $\pm$ 1°C. The weight loss of the water through the films was determined as a function of time. The data was taken after steady state condition was established. Five different permeation cells were used for each kind of film. The permeability data reported here represent the average of five runs.

RESULTS AND DISCUSSIONS

The gas permeability P is usually defined[5] as

$$P = (m)(l)/(\Delta P)(A)(t) = (F)(l)/\Delta P \tag{1}$$

where m is the amount of permeant which permeates through a polymer film of thickness l and area A in a period of time t, ΔP is the pressure gradient across the polymer film, and F is rate of transmission or flux.

The flux F, according to eq. (1) is equal to

$$F = (P)(\Delta P)/l \tag{2}$$

For a single layer of Mylar film, the water vapor permeability can be calculated from eq. (1) if A, ΔP, l, and m vs. t are known. To determine water permeability in the adhesive from the composite film, eq. (7) derived in the following was used.

At steady state, the rate of transmission (gas flux) across film 1 (the adhesive), film 2 (Mylar film), and film 3 (the adhesive) are the same. The flux across various films according to eq. (2) are

$$F_1 = (P_A)(\Delta P_1)/l_1 \tag{3}$$

$$F_2 = (P_M)(\Delta P_2)/l_2 \tag{4}$$

$$F_3 = (\mathrm{P_A})(\Delta P_3)/l_3 \tag{5}$$

$$F = (P)(\Delta P)/l \tag{6}$$

where F_1, F_2, F_3, and F are water vapor flux across the films 1, 2, 3, and the composite film, respectively; ΔP_1, ΔP_2, ΔP_3, and ΔP are pressure differences across films 1, 2, 3, and the composite film respectively; P_A, P_M, and P are water permeability in the adhesive, Mylar film, and composite film, respectively; l_1, l_2, l_3, and l are film thickness of films 1, 2, 3, and the composite film and are equal to 1.5, 3.0, 1.5, and 6.0 mil, respectively. Since $F_1 = F_2 = F_3 = F$, and $\Delta P_1 + \Delta P_2 + \Delta P_3 = \Delta P$, from eqs. (3)–(6), the following relationship is obtained:

$$l_1/P_A + l_2/P_M + l_3/P_A = l/P \tag{7}$$

Equation (7) was used to calculate P_A by employing experimentally determined values of P_M and P.

The water permeability of the composite film, Mylar film (type D), and the adhesive film Flexbond 150 determined at 23°C are shown in Table 1. The water permeability in Mylar film (type D) determined from the present experiment is 230×10^{-10} [cm^3 (STP) · cm · cm^{-2} · s^{-1} · $(cm\ Hg)^{-1}$] at 23°C. The permeability of water vapor in crystalline poly(ethylene terephthalate) (PETE), the main constituent of Mylar film, at 25°C is reported to be[6] 130 $\times$ 10^{-10} [cm^3(STP) · cm · cm^{-2} · s^{-1} · $(cm\ Hg)^{-1}$], which is somewhat lower than our value.

It is generally known that the degree of crystallinity has a significant effect on the permeability. It is generally recognized that transport of permeant virtually does not occur in the crystalline region. The higher the degree of crystallinity, the lower the permeability.[6] The degree of crystallinity of PETE is closely related to the thermal history of the polymers, and to a certain degree the molecular weight and molecular weight distribution. It is likely that different PETE films have different degrees of crystallinity due to different thermal history, different molecular weight, and molecular weight distribution. The water permeability in different PETE films, therefore, could be significantly different due to different degrees of crystallinity. Further, the plasticizer in the Mylar film could lower the glass transition temperature which increases the mobility of polymer chains and, therefore, resulted in an increase in permeability. Thus, the difference in permeability between our value and the value cited in *Polymer Handbook*[6] is most likely due to the difference in the degree of crystallinity and/or the plasticizing action of the additives in the Mylar films.

The water permeability in the pressure sensitive adhesive Flexbond 150 is 3230×10^{-10} cm^3 (STP) · cm · cm^{-2} · s^{-1} · $(cm\ Hg)^{-1}$, which is a factor of 14 higher than that in the Mylar film. The high water permeability in the adhesive as compared to that of Mylar film can be explained in terms of the amorphous nature of the adhesive and its low glass transition temperature (T_g). As mentioned above, Flexbond 150 is a vinyl acetate copolymer. The T_g of the Flexbond 150 is -28°C and it is amorphous at room temperature. The T_g of PETE is 67°C,[7] and it is semicrystalline at room temperature. Permeability P is usually defined as $P = DS$, where D is the diffusion coefficient and S the solubility coefficient. The solubility coefficient S in a semicrystalline polymer is approximately proportional to the amorphous volume fraction.[8] Water solubility in PETE, therefore, is expected to be smaller than in the adhesive. The diffusion coefficient of water in the glassy, semicrystalline PETE is also expected to be much smaller than in the rubbery, amorphous adhesive. According to the Bueche–Cashin–Debye equation,[9,10] the molecular diffusion coefficient D is related to the bulk viscosity η by the equation

$$D\eta = (A\rho KT/36)(R^2/M)$$

where A is Avogadro's number, ρ is the density, K is Boltzmann's constant, T is the absolute temperature, M is molecular weight, and R^2 is the mean-square end-to-end distance of a single polymer chain. While the bulk vis-

TABLE I
Water Permeability in Pressure-Sensitive Adhesive, Mylar Films, and the Composite Film at 23°C

Film	Film thickness (mil)	P [g · cm · cm^{-2} · s^{-1} · (cm Hg)$^{-1}$] × 10^{10}	P [cm^3 (STP) · cm · cm^{-2} · s^{-1} · (cm Hg)$^{-1}$] × 10^{10}
Composite	6	0.31	420
Mylar	3	0.16	230
Adhesive (Flexbond 150)	1.5	2.37	3230

cosity of most glassy polymers is 10^{13} P,[10] the viscosity of most pressure-sensitive adhesives is from 10^7 to 10^9 P.[11] Thus, it is clear from the above equation, the diffusion coefficient of water in the glassy (and semicrystalline) PETE is expected to be significantly lower than that in the soft and tacky adhesive. The large permeability difference between the adhesive and the Mylar film, therefore, can be attributed to lower water diffusivity and solubility in the semicrystalline, glassy PETE at room temperature.

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References

1. J. H. Lai, C. Knudson, L. Hallgren, R. Douglas, and P. Griffin, *Polym. Eng. Sci.*, to appear.
2. D. Satas, Ed., *Handbook of Pressure-Sensitive Adhesive Technology*, van Nostrand, New York, 1982.
3. Flexbond 150, General Purpose Pressure-Sensitive Emulsion, Air Products and Chemical, Inc., 1983.
4. A. C. Newns, *J. Text. Inst.*, **41**, 1269 (1950).
5. J. Crank, Ed., *Diffusion in Polymers*, Academic, New York, 1968.
6. J. Brandrup and E. H. Immergut, Eds., *Polymer Handbook*, Wiley, New York, 1975, p. III-238.
7. Ref. 6, p. III-160.
8. H. B. Hopfenberg and V. Stannett, in *The Physics of Glassy Polymers*, R. N. Haward, Ed., Wiley, New York, 1973, pp. 504–547.
9. F. Bueche, W. M. Cashin, and P. Debye, *J. Chem. Phys.*, **20**, 1956 (1952).
10. L. H. Lee, *J. Polym. Sci., Part A-2*, **5**, 751 (1967).
11. C. A. Dahlquist, in *Handbook of Pressure-Sensitive Adhesive Technology*, D. Satas, Ed., Van Nostrand, Princeton, N.J., 1982, pp. 78–98.

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