

Hygrothermal effects of epoxy resin. Part I: the nature of water in epoxy

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Abstract

This study assesses the nature of sorbed water and the related hygrothermal effects in epoxy resins. This paper is the first of a two-paper series. Three epoxy systems, DGEBA + mPDA, TGDDM + DDS, and Fiberite 934, were used in the investigation. Water sorption was achieved by immersing the materials in distilled water at constant temperature of 45°C, 60°C, 75°C and 90°C for 1530 h. Water absorption and desorption profiles were analyzed to determine the diffusion parameters. Solid state nuclear magnetic resonance (NMR) was conducted to determine the mobility of water in epoxy. The study shows that water molecules bind with epoxy resins through hydrogen bonding. Two types of bound water were found in epoxy resins. The binding types are classified as Type I or Type II bonding, depending on difference in the bond complex and activation energy. The activation energy of Type I and Type II bound water is ~ 10 and ~ 15 kcal/mol, respectively. Type I bonding corresponds to a water molecule which forms a single hydrogen bond with the epoxy resin network. This water molecule possesses a lower activation energy and is easier to remove from the resin. Type II bonding is as a result of a water molecule forming multiple hydrogen bonds with the resin network. This water molecule, therefore, possesses a higher activation energy and is correspondingly harder to remove. Type I bound water is the dominant form of the total amount sorbed water. The amount of Type II bound water depends strongly on the exposure temperature and time. Higher immersion temperature and longer exposure time result in a greater amount of Type II bound water. © 1999 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Knowledge of the bonding character of water molecules in epoxy resins is the most important and fundamental to understand hygrothermal effects. Even though considerable research has focused on hygrothermal effects [1–7], water diffusion mode and related mechanisms in epoxy resin are still not fully understood. This is because in part to the reality that the hygrothermal effects in epoxy are quite complex. Two mechanistic approaches have emerged that generally characterize the nature of water in epoxy. One is the free volume approach which presumes that water diffuses into epoxy resin and resides in the free volume of the material. For this approach bonding between water molecule and epoxy resin network is deemed insignificant [8,9]. The other approach is the interaction concept that suggests water molecules couple strongly with certain hydrophilic functional groups such as hydroxyl or amine in epoxy resin [7,10–12].

Based on dielectric experiments, Woo and Piggott [8]

suggested that the water molecules were not bound to polar groups or to hydrogen bonding sites in epoxy resin. Clustering of water molecules in the polymer was reported rather than complete molecular separation. Adamson [7] investigated thermal expansion and swelling by using epoxy resin and graphite/epoxy (Gr/Ep) composites. He determined that some water molecules interacted by forming hydrogen bonds with hydrophilic groups in epoxy resin while other water molecules are retained in free volume of the material. Apicella et al. [12] studied water absorption in glassy epoxy resin and proposed that there are three absorption modes: (i) bulk dissolution of water in the polymer network, (ii) moisture absorption onto the surface of vacuoles which define the excess free volume of the glassy structure, and (iii) hydrogen bonding between polymer hydrophilic groups and water. Jelinski and collaborators [13] investigated the nature of the epoxy–water–molecule interaction using quadrupole echo deuterium nuclear magnetic resonance (NMR) spectroscopy. They concluded that: (i) water in epoxy resin was impeded in its movement, (ii) no free water existed, (iii) there was no evidence of tightly bound water, and (iv) sorbed water molecules disruption of the hydrogen-bonded network in the epoxy resin was unlikely. The water molecules migrated from site to site, but

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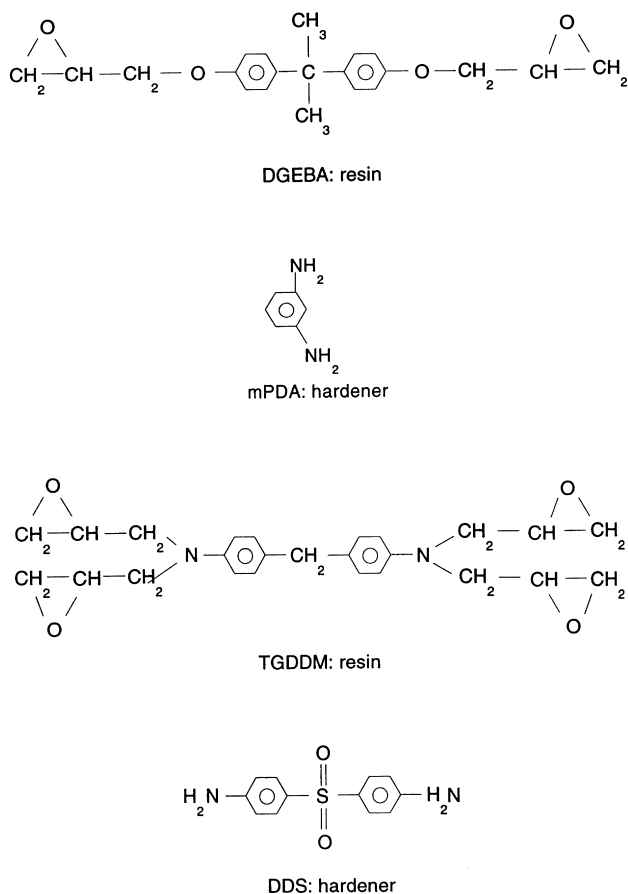


Fig. 1. Chemical structures of DGEBA, mPDA, TGDDM and DDS.

apparently such a jumping motion did not involve a particular hydrogen exchange mechanism.

Although there is considerable agreement among some models, substantial disagreement still persists. No single theory or model exists with sufficient experimental support to explain all hygrothermal phenomena. In this study, water absorption and desorption profiles were analyzed to determine pertinent diffusion parameters. Solid state nuclear magnetic resonance (NMR) was conducted to assess the bonding characteristics of water in epoxy. The aim of this study is to discern the interaction nature of sorbed water and the related hygrothermal effects in epoxy resins. This paper is the first part of a two-paper series.

2. Materials and experimental procedure

Three epoxy systems that have been used extensively for high performance polymer matrix composites (PMCs) were used in this study. One epoxy system consists of diglycidyl ether of bisphenol-A (DGEBA, Shell Epon828) and meta-phenylene diamine (mPDA) hardener. The other is tetraglycidyl-4,4'-diaminodiphenyl methane resin (TGDDM, Ciba Geigy MY720) with 4,4'-diaminodiphenyl sulfone (DDS, DuPont) hardener. The third is Fiberite 934 epoxy resin

that consists mainly of TGDDM resin, DDS hardener, with small amounts of additives of which the types and concentrations are proprietary. The chemical structures of these resin systems are shown in Fig. 1. Materials for test coupons were prepared using the following procedures.

DGEBA resin and 14.5 phr (part per hundred resin by weight) mPDA hardener were heated separately to 75°C in an oven until the mPDA was melted. Then the epoxy resin and hardener were mixed thoroughly and degassed for 10 min. The degassed mixture was poured into a mold for curing. To cure the resin the following heating sequence was used: 75°C for 2 h followed by 125°C for 2 h, postcured at 180°C for 8 h, and then cooled down to 60°C . Specimens were taken out of the oven after temperature ramped down to 60°C . The temperature rate in all ramping stage was $1.5^\circ\text{C}/\text{min}$.

TGDDM resin and 44 phr DDS hardener were heated to 135°C respectively, and then combined and stirred until DDS was melted and a clear brown liquid was obtained. The material was degassed for 15 min. The curing procedure was: 135°C (mixing and degassing) followed by 80°C for 1 h + 100°C for 2 h + 150°C for 4 h + 200°C for 7 h (postcuring) and then cooled down to 60°C .

The frozen Fiberite 934 resin was put in a mold and heated to 135°C . The material was degassed for 10 min at 135°C . The curing procedure was 135°C for 2 h followed by 177°C for 4 h, postcured at 190°C for 6 h, and then followed by a 60°C cool down. The temperature ramping rate was the same as that used for the DGEBA + mPDA systems.

After the materials were prepared, infrared (IR) spectroscopy testing was performed to determine the extent of curing. During the curing process, the intensity of the 910 cm^{-1} wavenumber peak that corresponds to the epoxide group diminishes gradually in the IR spectrum. Infrared results showed that the 910 cm^{-1} peak of all three epoxy resins vanished therefore confirming that the epoxy systems were fully cured. Incomplete curing in epoxy resin is not desirable for the present study as secondary curing during hygrothermal exposure may render experimental observations ambiguous and promote misinterpretation of results.

3. Results and discussions

3.1. Water absorption

The fully cured epoxy resin panels for use in this experiment were cut into samples with dimensions of $38 \times 25.4 \times 1.15\text{ mm}$ using a fine-grit, diamond-blade saw. The thickness of the water absorption test samples was made small, deliberately, compared to its width and length. The ratio of the edge surface area to the total surface area is only 5%, and as such, edge effects could be ignored and simple one-dimensional diffusion model analysis can be applied without incurring significant error.

After conditioning samples at 110°C for 24 h to ensure

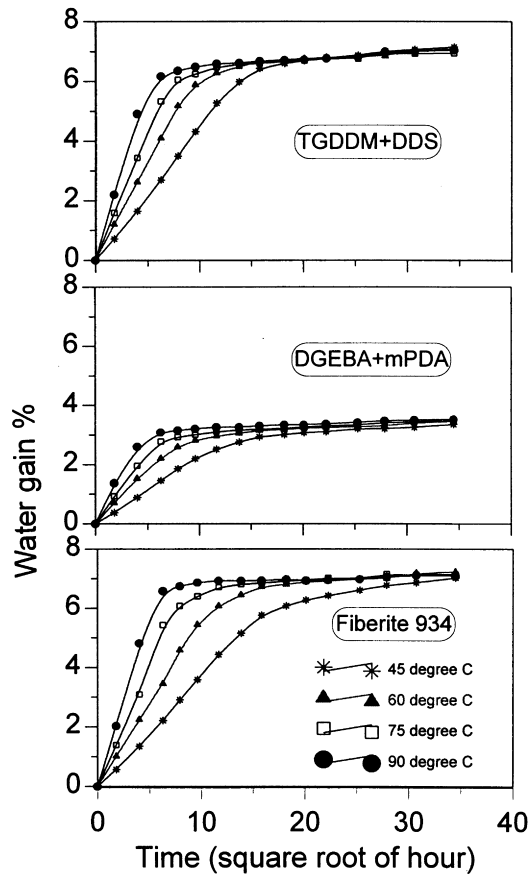


Fig. 2. Water absorption profiles of the three epoxy systems at different temperatures for 1530 h. The symbols are experimental data.

complete dryness and the amelioration of residual stresses induced during sample processing and preparation, the specimens were placed into distilled water chambers maintained at constant temperatures of 45°C, 60°C, 75°C, and 90°C. The specimens were weighed periodically using a digital balance with 0.01 mg resolution to determine the percent weight change, and, thus, water uptake. The water gain percentage, $M\%$, was determined from the equation:

$$M\% = \frac{(W - W_d)}{W_d} \times 100\% \quad (1)$$

Here, W is the weight of the water-sorbed epoxy specimen and W_d is the initial weight of the dry specimen. To ensure the removal of excessive surface (superficial) water, specimens were gently wiped dry using clean, lint-free tissue

paper and allowed to stand free at ambient lab environment for ~ 2 min.

The aforementioned absorption procedure yielded a series of water gain versus time curves. The diffusivity, D , was determined from the initial slope of the percentage water gain M_t versus $t^{1/2}$ curve [1],

$$D = \frac{\pi}{16} \left(\frac{h}{M_m} \right)^2 \left(\frac{M_{t2} - M_{t1}}{\sqrt{t_2} - \sqrt{t_1}} \right)^2 \quad (2)$$

where h is the thickness of the specimen, t is exposure time and M_m is the maximum water gain (saturation). The activation energy Q and diffusion constant D_0 were obtained by plotting $\ln D$ versus $1/T$ according to the Arrhenius rate equation given,

$$D = D_0 \exp\left(-\frac{Q}{RT}\right) \quad (3)$$

The water sorption profiles of the three hydrothermal-exposed epoxy systems are shown in Fig. 2. At the initial stage water gain is linearly proportional to $t^{1/2}$. After exposure for 1530 h, specimens reached full saturation for all hydrothermal conditions. For a given material, the saturation water content (M_m) is virtually the same irrespective of the immersion temperatures. This behavior indicates that water diffusion in the three materials is Fickian in nature. Not unexpectedly, Fiberite 934 and TGDDM + DDS systems show similar water gains of 6.95 and 6.80 wt% because of their similar resin chemistry. The DGEBA + mPDA system exhibited a much lower saturation level (3.35 wt%) compared with to Fiberite 934 and TGDDM + DDS systems. The DGEBA + mPDA system did not show two-stage water absorption behavior as reported by other researchers [9]. This difference may be owing to difference in material preparation since they did not report the degree of cure of the material.

The diffusivity D is obtained according to Eq. (2) and the activation energy Q is obtained by plotting $\ln D(T)$ versus $1/T$ following Eq. (3). All water absorption related parameters of the three epoxy systems are shown in Table 1.

3.2. Water desorption

After exposure in distilled water at temperatures, 45°C, 60°C, 75°C, and 90°C for 1530 h, all specimens reached full saturation, M_m . Subsequently, water-saturated specimens were placed into a dry chamber in order to desorb water from the specimens. During desorption testing, specimens

Table 1

The diffusion-related parameters of the three epoxy systems: diffusivity D at different exposure temperatures, activation energy Q and maximum water content M_m

	D_{45} ($\text{mm}^2 \text{s}^{-1}$)	D_{60} ($\text{mm}^2 \text{s}^{-1}$)	D_{75} ($\text{mm}^2 \text{s}^{-1}$)	D_{90} ($\text{mm}^2 \text{s}^{-1}$)	Q (kcal/mol)	M_m (%)
TGDDM + DDS	3.13×10^{-7}	6.61×10^{-7}	1.15×10^{-6}	2.35×10^{-6}	11.1	6.80
DGEBA + mPDA	3.35×10^{-7}	7.93×10^{-7}	1.35×10^{-6}	3.14×10^{-6}	12.2	3.35
Fiberite 934	2.08×10^{-7}	5.03×10^{-7}	8.96×10^{-7}	1.34×10^{-6}	10.4	6.95

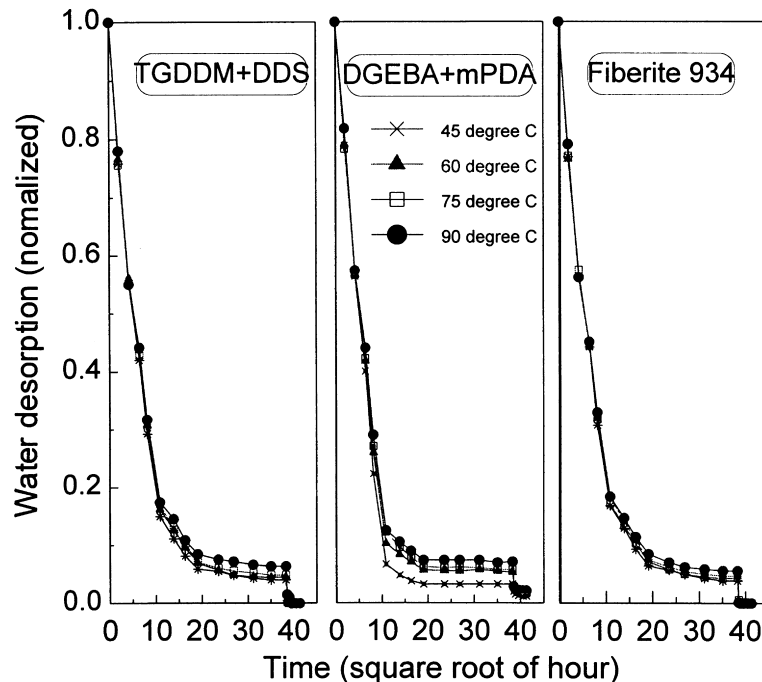


Fig. 3. Water desorption profiles of the three epoxy systems at 60°C for 1450 h and then 140°C 240 h. The symbols are experimental data and represent the samples with different bath temperatures in water absorption process.

are weighed periodically to determine the percent weight change. Each datum of the desorption profile was the average value of three samples.

Water desorption profiles of the three epoxy systems are shown in Fig. 3 resulting from thermal soaking at 60°C for 1450 h, and then at 140°C for 240 h. The initial desorption rate of absorbed water was rapid. With time, however, the desorption slowed. Even after long-time desorption at 60°C for 1450 h, it was observed that some residual water remained in the specimens. The retained water could not be expediently removed at 60°C regardless of how long the materials were thermally soaked. Much higher temperatures (i.e., 140°C) had to be used for removing this residual water from the resins.

The amounts of retained water of the three epoxy systems exposed at different temperatures are shown in Table 2. The data in Table 2 are taken directly from Fig. 3. The trend suggests that more residual water retention occurs at higher exposure temperature. To investigate the relationship between the amount of the residual water and exposure

time, Fiberite 934 resin, immersed in water at 90°C, was taken out at different exposure time and then desorbed at 60°C to determine the amount of residual water. Fig. 4 shows an increase in the amount of retained water with exposure time at the fixed absorption exposure temperature (90°C).

The existence of residual water in polymer materials is not an unfamiliar phenomenon as Moy [10], Marsh [14], and Xiang [15] have reported that some of the absorbed water could not be removed unless a higher desorption temperature was used. Moy [10] investigated epoxy–water interaction by DSC, IR, NMR spectroscopy, and water absorption/desorption experiments using TGDDM + DDS resin system. He reported a strong hysteresis related to the desorption process indicating residual amounts of water that could only be removed by heating the polymer to temperatures above 100°C. Marsh and Springer [14] reported moisture solubility and diffusion results for epoxy-glass composite. They too showed that the sorbed water could not be fully removed during desorption at temperatures

Table 2

The amount of retained water in the three water-saturated epoxies after desorption at 60°C for 1450 h. The trend is that higher immersion temperature induces greater levels of retained water

	Residual water of sample bathed at 45°C then desorp. at 60°C 1450 h (w%)	Residual water of sample bathed at 60°C then desorp. at 60°C 1450 h (w%)	Residual water of sample bathed at 75°C then desorp. at 60°C 1450 h (w%)	Residual water of sample bathed at 90°C then desorp. at 60°C 1450 h (w%)
TGDDM + DDS	0.29	0.32	0.34	0.44
DGEBA + mPDA	0.11	0.18	0.20	0.25
Fiberite934	0.27	0.30	0.33	0.39

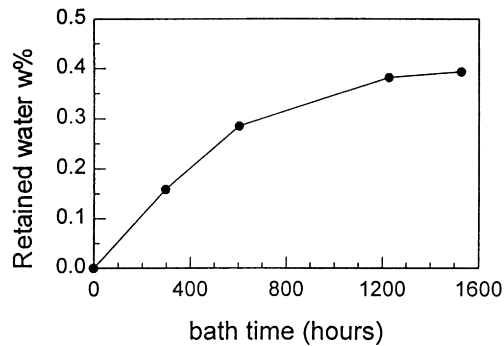


Fig. 4. The amount of residual water of the epoxy Fiberite 934 changes with exposure time at 90°C.

used typically for water absorption. Attempts to remove the residual water in the material at temperatures of 85°C, 100°C and 110°C were unsuccessful. Temperatures in excess of the glass transition temperature (i.e., 120°C) were required to remove the residual water.

Previous studies [10,14,15] have suggested the existence of the residual water, but none pursued this matter further with regard to the mechanisms of such occurrence. The present study not only reveals the existence of the residual water but also shows the amount of residual water varies with hygrothermal exposure history. The amount of residual water depends strongly on the exposure time and temperature. As shown in Table 2 and Fig. 4, both higher immersion temperatures and longer immersion times enhance the amount of residual water in the resin.

3.3. Desorption activation energy

It is evident that at least two bound water–epoxy resin complex exist in the epoxy resins as a result of long-time hygrothermal exposure. Retained water molecules which are easily removed by thermal desorption at lower temperatures are consistent with a so-called Type I bound water complex. Moreover, retained water molecules which are considerably more difficult to remove by thermal desorption are referred to as a Type II bound water complex. Higher temperatures are required to remove Type II bound

water from the epoxy resin network. To determine the activation energies of Type I and Type II sorbed water more detailed desorption experiments were conducted. Saturated TGDDM + DDS samples were thermally desorbed to determine diffusivity and activation energy of Type I and Type II bound water. Fig. 5 schematically illustrates the circumstance for the desorption of Type I and Type II bound water. Both fields indicative of the removal of Type I and Type II bound water are illustrated in Fig. 5. The activation energy, Q , for desorption Type I bound water was determined using fully-saturated, M_m , specimens as the initial condition of the desorption diffusion process. The activation energy for desorption Type II bound water was determined using specimens that still contained residual water after low-temperature (75°C) bake out in dry air. The activation energies for both Type I and Type II bound water are shown in Fig. 6. Also, the activation energy for absorption of water in the resin is shown. The results in Fig. 6 show that activation energy necessary to desorb Type II bound water is significantly higher (~ 15 kcal/mol). In contrast, the activation energy necessary for desorption of Type I bound water is less (~ 10 kcal/mol). It is of interest to note, and perhaps fortuitous, that the difference between Q for Type II and Q for Type I water is approximately 5 kcal/mol which is on order of the energy for hydrogen bonding. Such differences in the activation energies for desorption of Type I and Type II bound water suggest that the binding strengths bound complexes between water molecules and the resin vary.

3.4. NMR test

NMR testing provides a sensitive probe to assess the interaction between sorbed water and epoxy resin since the NMR peak width directly reflects the mobility of molecular species. Broad spectral NMR peaks are indicative of tight bonding between interacting species. Although many NMR reports document various aspects of sorbed water in epoxy resins, scant detailed NMR reports were found by the authors on water in epoxy at different hygrothermal stages. In this study NMR was carried out to determine water–resin bonding characteristics. A Varian VXR 400 solid state NMR spectroscopy was used for ^1H NMR testing. The

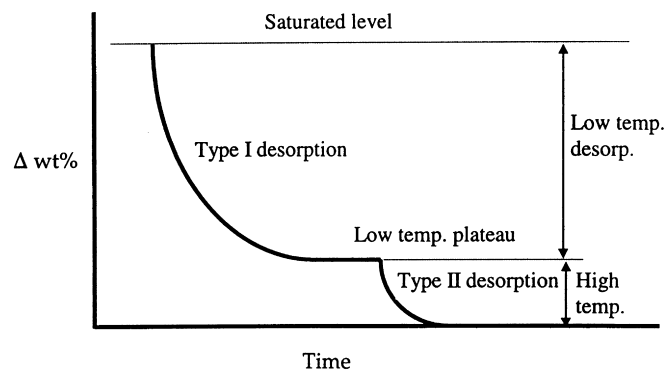


Fig. 5. Schematic of water desorption process.

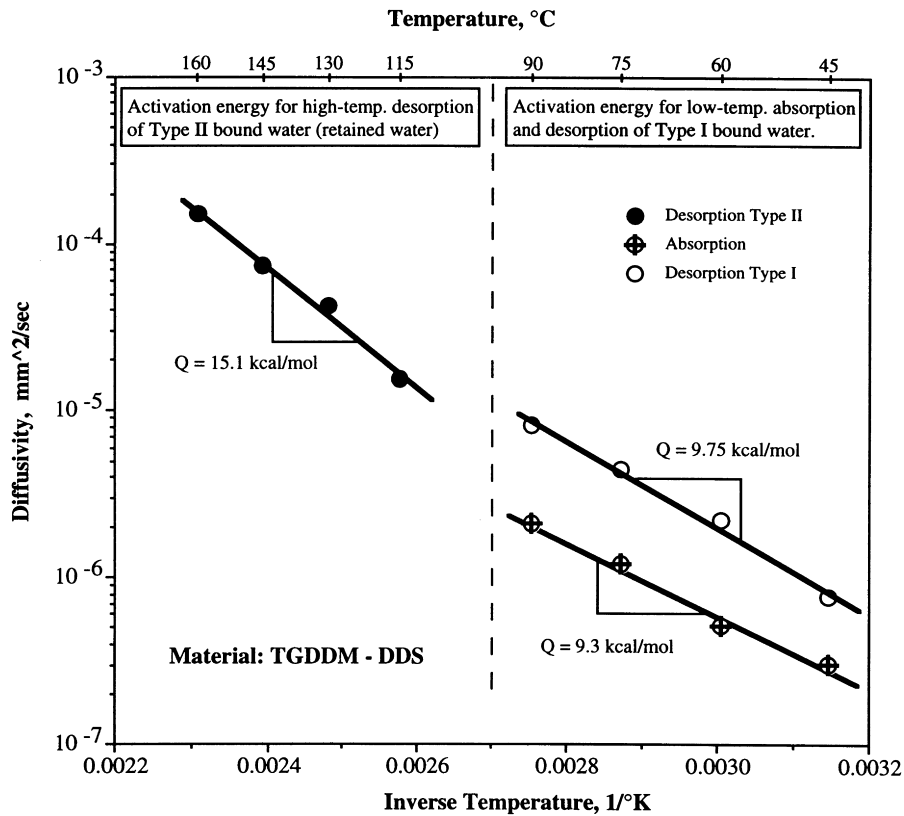


Fig. 6. Activation energy of TGDDM + DDS system. $Q_{\text{type I}}$ was obtained by desorption of saturated specimens at 45°C, 60°C, 75°C and 90°C. $Q_{\text{type II}}$ was obtained by desorption of semi-dried specimens at 115°C, 130°C, 145°C and 160°C. For comparison absorption activation energy is also shown.

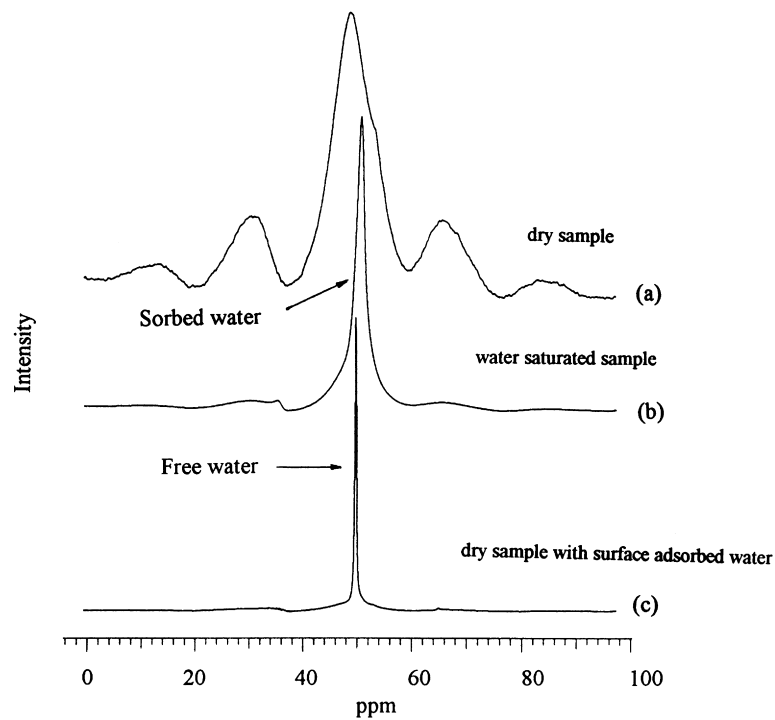


Fig. 7. ¹H NMR spectra for TGDDM + DDS system. (a) dry epoxy, (b) water-saturated epoxy and (c) free water mixed with dry epoxy. The results indicate that the mobility of sorbed water in epoxy is between free-water and solid states.

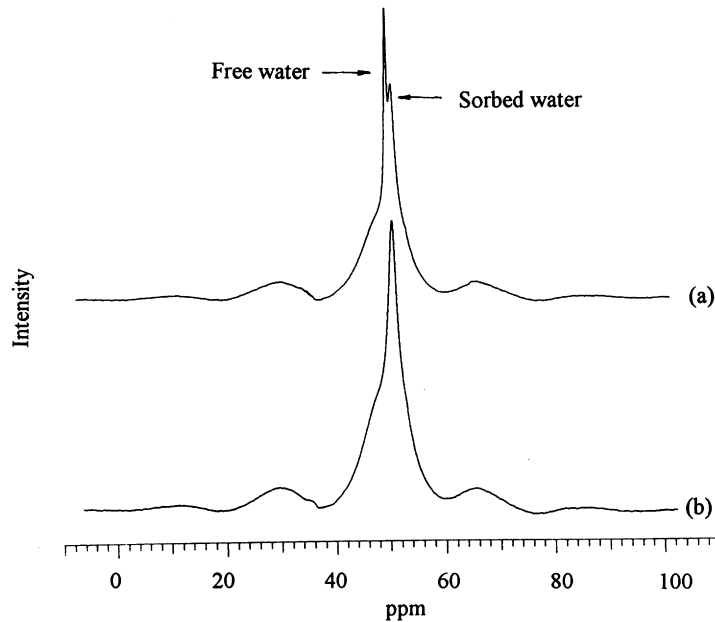


Fig. 8. ^1H NMR spectra of Fiberite 934 show free water changes into impeded sorbed water with time. (a) dry resin mixed with 1% liquid water. Liquid water and sorbed water can be seen clearly. (b) 10 min later, liquid water disappeared and all sorbed water molecules were impeded.

resin examined in the NMR tests was ground into small particles where the particle diameter on average was (1 mm. The spin frequency used was 4000 and 6000 Hz.

Three ^1H NMR spectra of the TGDDM + DDS epoxy system are shown in Fig. 7: (a) dry epoxy sample, (b) water-saturated epoxy sample, and (c) dry epoxy sample with adsorbed water on its surface. The peak width of sorbed water (7b) is broader than that of liquid water (7c) and much narrower than that of tightly bonded hydrogen and hydroxyl group in epoxy resin (7a). The mobility of bound water in epoxy is between free-water (liquid) and solid states. Fig. 8 represents the ^1H NMR spectra of Fiberite 934 (TGDDM + DDS) and it indicates how quickly the free water is absorbed and changed into impeded water. Fig. 8a is a spectrum of dry resin which was acquired right after mixing with 1 wt% water. Free water and a small amount of bound water are exhibited. However, after just 10 min, free water molecules abates revealing that sorbed water molecules have become impeded (8b). This result also reveals the rate in which bound water complexes form in epoxy resins.

3.5. Postulation of the two types of bound water

The results from this study indicate that water in epoxy resin can be classified into two types of bound water according to their activation energies for desorption and NMR spectral evidence. Bonding for these bound water molecules is not nearly as strong as main-chain bonding (60–100 kcal/mol) of the polymer network structure [16], but for Type II bound water, it is significantly higher than physical bonding (0.5–2 kcal/mol) denoted by Van der Waals and dipole–dipole bonding. Antoon et. al. proposed that the sorbed water was held by hydrogen bonding [17]. Atkins reported

that breaking hydrogen bonding of water requires energy between 5 and 20 kcal/mol [18]. The mobility of sorbed water ranges between that of nearly-free water and the tightly-bonded hydrogen molecules on the backbone. Moreover, the activation energy of the two types of bound water is in the energy range of hydrogen bonding. Therefore, water molecules bond with epoxy resin network through hydrogen bonding.

A model of bonding between water and epoxy network is

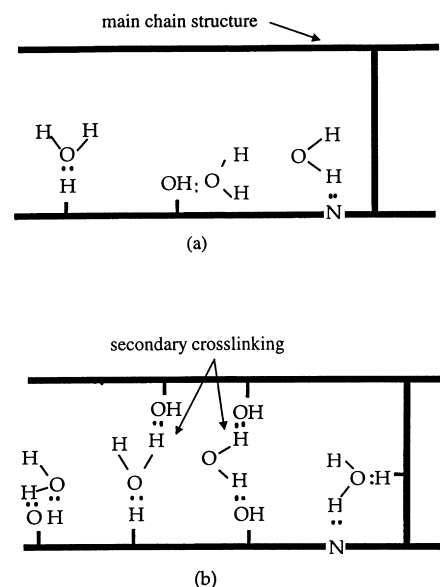


Fig. 9. Possible bound water complexes in epoxy network: (a) water molecules form one hydrogen bond with resin network and have lower activation energy, (b) water molecules form more than one hydrogen bonds with resin network and have higher activation energy.

proposed in Fig. 9. Fig. 9a presents single hydrogen bonding between water and epoxy network. This corresponds to the Type I bound water which has lower activation energy and is more easily to remove. Fig. 9b shows multi-site interconnective hydrogen bonding model. The bonding complexes of the multi-site/interconnective model corresponds to the Type II bound water which has a higher activation energy and is harder to remove by desorption. Type I bound water diffuses into the epoxy network and breaks the initial interchain Van der Waals force resulting in the increase of chain segment mobility and swelling. As the amount of the Type I bound water is dominant in the water sorption process, it readily plasticizes the epoxy resins. Water molecules which form multi-site interconnective bond complexes (Type II) do not contribute significantly to plasticization of the resin. Moreover, these bond complexes create bridging between chain segments resulting in (pseudo crosslinking) secondary crosslinking. The formation of the Type I bonded water occurs readily in the moisture sorption process, particular at low temperature hygrothermal exposure, however, the impetus to form Type II bonding is substantially less. Longer time and higher temperatures provide the stimulus for the formation of multi-site interconnective bonding. Consequently, the amount of Type II bound water increases with exposure time and temperature. The phenomenon of water retention is not limited to epoxy resins. Residual water or so-called locked-in water has been observed in other polymers [19].

4. Conclusion

The degree of sorbed water interaction in epoxy resins was assessed from the results obtained by water desorption measurement, residual water contents, desorption activation energy analysis, and ^1H NMR studies on the epoxy resins at different hygrothermal stages. Depending on the difference in bond structures and activation energy, the bond states have been classified as Type I or Type II bonding. Type I bonding involves water molecules forming a single hydrogen bond and or dispersion bonding with the epoxy resin network. Type I bound water possesses a thermal desorption activation energy of ~ 9.5 kcal/mol and, thus, is more easily desorbed from the resin. Type I bound water manifested by disrupting the interchain Van der Waals force resulting in the increase of segment mobility. Type I

bound water acts as a plasticizer. Type II bonding results from water molecules forming multiple hydrogen bonds with the resin network. The activation energy in Type II bonding is 15.1 kcal/mol. These water molecules possess a higher activation energy and are correspondingly harder to remove from the resin by desorption. Type II bound water molecules do not act as a plasticizer but rather form bridges between structural segments resulting in secondary crosslinking. The amount of Type II bound water strongly depends on the hygrothermal temperature and time history. Higher exposure temperature and longer exposure time result in the formation of a greater amount of Type II bound water. Hygrothermal effects on physical properties such as the T_g in epoxy resins will be discussed in Part II of this series.

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