

Effects of Storage Aging on the Properties of Epoxy Prepregs

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Changes in the properties of epoxy prepregs during storage are of both industrial and theoretical interest. It would be very helpful to have a test, suitable for production use, to determine when a prepreg has aged beyond acceptable limits. In this article, different epoxy prepregs aged under various conditions were studied by near-infrared (NIR) and mid-infrared (MIR) Fourier transform spectroscopies, dynamic mechanical analysis (DMA), rheological analysis, time-resolved light scattering (TRLS), and optical microscopy (OM). A very good relationship between storage aging time and curing conversion was obtained by NIR measurements. Furthermore, the rheological study showed that both the viscosity and the gelation time changed with storage aging. Changes in the glass transition temperatures of epoxy prepregs can result from either curing or phase separation during storage aging. The TRLS and OM results showed that structural evolution might occur during storage for some prepregs.

1. Introduction

With the rapid development of the electronics industry, printed circuit boards (PCBs) and B-stage prepregs are urgently required to have superior properties. Prepregs for PCBs are normally kept below 5 °C prior to use, but they begin to deteriorate once they are warmed to ambient or high temperature. As B-stage prepregs are commercially transported and used, storage aging during this period would have profound effects on the electrical and mechanical properties of cured PCBs.

For many years, the effects of storage^{1–4} and curing^{5,6} on the properties of epoxy prepregs have been widely studied for electronics and high-performance engineering materials. Rheological characterization⁷ and differential scanning calorimetric determination⁸ have shown that the viscosity, glass transition temperature, and curing behavior change with the aging of prepregs.^{9,10}

Mid-infrared (MIR) spectroscopy¹¹ and high-performance liquid chromatography¹² have also been used to study the concentrations of epoxy groups and curing agents with thawed age. Experimental results indicate that precuring in the epoxy resin system consumes a considerable number of epoxide groups and hardeners. However, the tedious sample preparation processes are not practical for industrial-scale monitoring.

Mechanical testing combined with microscopic investigation has shown that the prepreg surface becomes rougher and more irregular with age. These changes in composition and surface characteristics also affect the functionality of the prepreg during layout and curing.¹³

Dielectric analyses alone^{14,15} and/or combined with other methods^{16,17} have been successfully applied to monitor the curing state of epoxy prepregs. However, the high sensitivity of dielectric properties to moisture restricts the application of this method for storage study because of the complicated conditions encountered during transporting and storage. With luminescence spectroscopy under steady-state conditions, Sales

et al.¹⁸ analyzed the hygrothermal effects on uncured glass-epoxy fiber prepreg. Still, the requirement for a fluorescence probe in the resin matrix restricts the application of this method.

At the present time, industry urgently demands some convenient and precise methods, suitable for production use, to determine the validity of stored prepregs, as well as fundamental understanding of chemical and physical changes occurring during storage. In this work, we demonstrate that near-infrared (NIR) spectroscopy is a powerful tool for PCB prepreg storage judgment. Moreover, changes in physical properties such as rheological or thermal properties can come from either chemical changes during aging or physical structure changes due to phase separation induced by aging.

2. Experimental Section

2.1. Materials and Specimen Preparation. Fresh rolls of randomly selected epoxy prepregs from Panasonic, Hitachi Co., and other companies were used for this study. The prepregs for PCBs are generally composed of epoxy/phenolic resin or epoxy/dicyandiamide as specified by the manufacturer; we selected two samples as examples from each of these two kinds and herein denote them as sample a and sample b, respectively. Samples cut from the rolls were aged at 35, 50, and 70 °C for various times. For the moisture study, samples were aged at 25 and 35 °C under three selected conditions of relative humidity (RH): in a desiccator (0% RH), in a chamber having an RH of 65%, and in a chamber having an RH of 97%.

2.2. Experimental Techniques. Infrared Spectroscopy Monitoring. Mid-infrared (MIR) spectroscopy was performed with a Thermo Nicolet Nexus 440 spectrometer, and NIR spectroscopy was performed with a Nicolet NEXUS 470 FTIR spectrometer. Aged samples were tested by MIR and NIR spectroscopies at room temperature. For the in situ NIR study, fresh samples were placed in the compartment and monitored in situ in the near-infrared region (11000–4000 cm⁻¹) at 100 °C. For transmitting spectroscopy, resins from the prepregs were ground and blended with KBr powder, whereas for attenuated total reflectance (ATR), the prepregs were used directly.

Rheological Tests. The melt viscosity variations of the resin components without glass fibers were recorded on an ARES-

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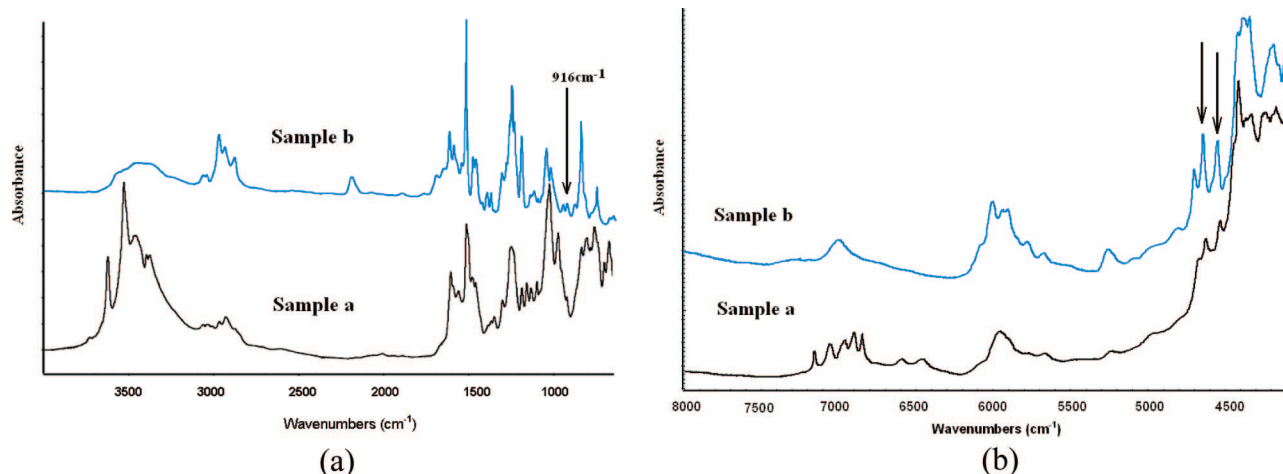


Figure 1. (a) Mid-IR and (b) near-IR spectra of fresh epoxy prepregs.

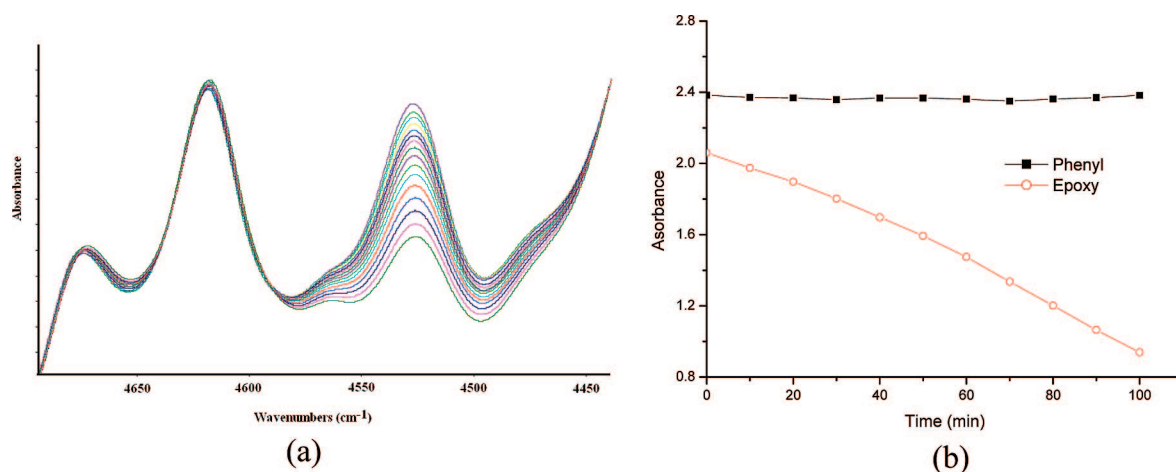


Figure 2. In situ isothermal NIR study of sample b cured at 100 °C: (a) NIR spectra, (b) evolution of the areas of the phenyl and epoxy peaks with curing.

9A rheometry instrument. All samples were tested in parallel-plate mode with a controlled strain of 1% and a test frequency of 1 Hz. The resin components, which were obtained from the prepregs by removing the glass fibers, were sandwiched between two round fixtures (diameter = 25 mm); then, the plate distance was adjusted to 1.0 mm quickly at the test temperature. As the standard industry processing temperature of PCB prepregs is 171 °C, this test temperature was chosen so as to provide the most useful information for researchers in this area.

Dynamic Mechanical Analyses (DMA). Dynamic mechanical properties were measured with a Netzsch DMA 242 instrument operating in the three-point-bending mode at an oscillation frequency of 1.0 Hz. Specimens cut from the epoxy prepregs for DMA were prepared in the form of cuboid bars with dimensions of 15 × 50 mm²; the thickness of the prepregs was measured each time. The data were collected from 0 to 150 °C at a scanning rate of 3 °C/min.

Light Scattering. The phase structures of samples were observed on a self-made time-resolved light scattering (TRLS) instrument with a controllable hot chamber at 35 °C for 7 days. The light scattering profiles were recorded at appropriate time intervals.

Optical Microscopy. The samples were observed using an optical microscope with a hot chamber (XDS-1B, Nanjing Kaier Instrument Co.).

3. Results and Discussion

3.1. Infrared Spectroscopy Study. In previous studies, mid-infrared (MIR) spectroscopy provided a reliable measure of epoxy resin age.^{1,11} The analysis, however, invariably involved dissolving the resin and was done by transmission spectroscopy with the solution dried on a salt plate.^{1,11} In fact, Cole et al.¹ also successfully used the attenuated total reflectance (ATR) technique, and they tried diffuse reflectance but found that it lacked reproducibility.

MIR Study. In this work, we also studied aged samples by MIR spectroscopy initially and found that both transmitting spectroscopy and ATR methods give quite similar spectra for the same sample. In Figure 1a, sample a shows a clear band at 916 cm⁻¹, which is assigned to the epoxy group, but another strong peak visibly overlaps the epoxy peak. For sample b, the epoxy peak is also quite clear, but another peak at long wavenumbers is located close to it. Therefore, to calculate the curing conversion with aging, one needs to split the epoxy signal from the other peaks and correct the baseline. One must also ensure that no other peaks are near the phenyl ring vibrations at either 1610 or 1500 cm⁻¹, which are always defined as the internal standard for curing conversion in ordinary FTIR studies. From the industrial point of view, MIR spectroscopy might be unfeasible for studying the aging of some prepregs for these reasons.

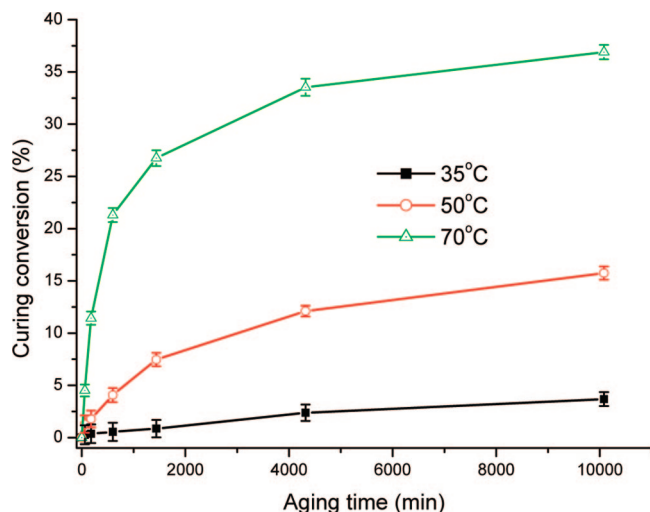


Figure 3. Evolution of the curing conversion of sample a aged at various temperatures as determined by NIR spectroscopy.

Internal Standard Selection of NIR Spectroscopy. Recently, Wang et al.¹⁹ studied the isothermal curing process of epoxy prepregs with NIR spectroscopy, and they found that this method gives reliable results related to those obtained by differential scanning calorimetry. Jiang et al.²⁰ also successfully applied the NIR method to resin and volatile-content quality control. Mijovic and Andjelic²¹ and Kortaberria et al.²² established that optical-fiber-connected NIR spectroscopy can be used for the in situ study of epoxy reaction processes. These curing studies by NIR spectroscopy would certainly be very helpful for extending the applications of NIR spectroscopy in our aging study.

In the NIR spectra of samples a and b (Figure 1b), the band at 7000 cm^{-1} can be assigned to the overtones of OH-related groups. The overlapping bands from 6100 to 5500 cm^{-1} are assigned to the CH/CH₂/CH₃ overtones of epoxy groups, benzene groups, and the backbone.^{23–25} The band at 4530 cm^{-1} is assigned to the combination vibrations of epoxy groups and is the band that is always used to analyze curing conversion.

For studying storage by NIR spectroscopy, the primary work is to select suitable bands as internal standards for the calculation of epoxy variations with aging. An in situ isothermal NIR study could be the most feasible way to achieve this goal because of the easy comparison of different bands.

In Figure 2a, the NIR spectra of sample b as an example represent the typical evolution of the relevant functional groups

with isothermal curing. It is quite clear that the areas of all of the phenyl peaks near 4620 cm^{-1} remain almost unchanged, whereas the consumption of the epoxy groups is evidently demonstrated by the decrease of the characteristic epoxy peak at 4530 cm^{-1} . The complete disappearance of the epoxy peak at 4530 cm^{-1} is a common phenomenon for every fully cured prepreg. Although the other peaks such as that at 6073 cm^{-1} are also assigned to epoxy, they are not as obvious as the one at 4530 cm^{-1} and thus are not shown here. Because the integral intensity of the selected reference peak will be used to demonstrate epoxy conversion with aging, this peak must have a universal applicability for a broad range of commercial epoxy prepregs. To choose the right reference peak, we studied different types of epoxy prepregs from several companies and carefully analyzed the variations of different bands by in situ isothermal NIR spectroscopy.

As shown in Figure 2b, the integral intensity of the epoxy peak and the band at 4623 cm^{-1} , which is assigned to the CH stretching of the benzene ring, were plotted as a function of isothermal curing time at $100\text{ }^{\circ}\text{C}$. Clearly, the integral intensity of the phenyl peak remains almost constant; the very slight fluctuations might come from either instrument deviation or sample flow, but these fluctuations have quite minimal effects on the calculation of the epoxy conversion. Therefore, we chose the phenyl peak at 4623 cm^{-1} as the reference for further studying the storage aging of different prepregs.

Aging Study by NIR Spectroscopy. For the storage aging study, the samples were either kept at ambient temperature or heated to accelerate the aging process. With ATR-NIR spectroscopy, the curing conversions of aged samples were calculated according to the equation

$$\text{conversion} = \frac{\text{area}(e, t)/\text{area}(s, t)}{\text{area}(e, 0)/\text{area}(s, 0)} \quad (1)$$

where $\text{area}(e, t)$ is the peak area of epoxy at a given time t , $\text{area}(s, t)$ is the peak area of the internal standard (phenyl peak) at the same given time, $\text{area}(e, 0)$ is the initial peak area of epoxy without any conversion, and $\text{area}(s, 0)$ is the initial peak area of the internal standard. However, for commercial reasons, it is almost impossible to obtain the IR spectra of initial uncured resin mixtures (in stage A). Therefore, the fresh epoxy prepregs from the manufacturers were used instead. As the ultimate goal of a storage study for industry is to make sure that the conversion of aged samples remains below a critical value, a relative conversion has the same importance as the absolute value in this case.

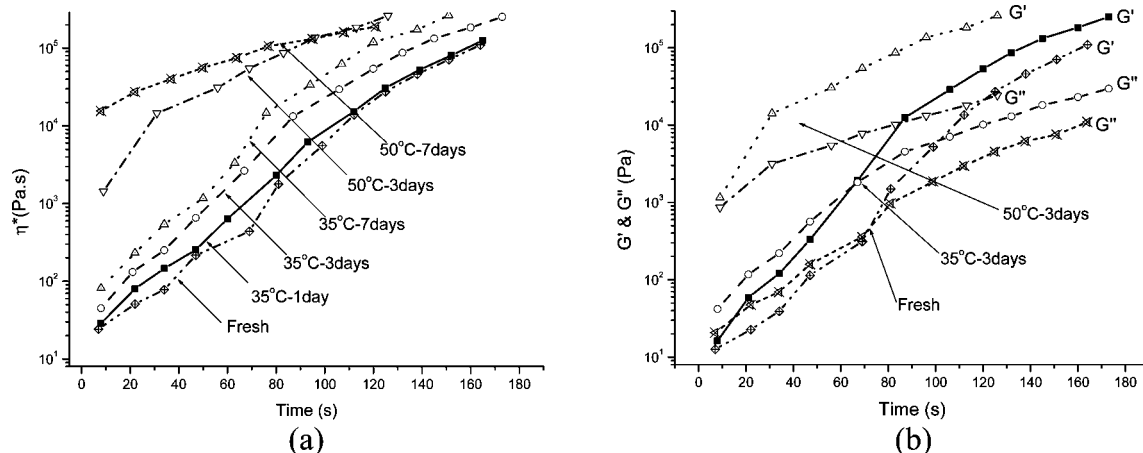


Figure 4. Rheological behaviors of aged sample a at $171\text{ }^{\circ}\text{C}$: (a) viscosity versus time, (b) storage and loss moduli versus time.

Table 1. Rheological Properties of Sample a Aged under Different Conditions

sample	viscosity (Pa s) ^a	gel time (s) ^b	α (%) ^c
fresh	24 ± 2	73 ± 4	0
35 °C, 1 day	29 ± 3	72 ± 4	1.12 ± 0.06
35 °C, 3 days	45 ± 5	66 ± 3	2.54 ± 0.08
35 °C, 7 days	81 ± 6	59 ± 3	3.70 ± 0.04
50 °C, 3 days	1440 ± 50	—	11.8 ± 0.12
50 °C, 7 days	15590 ± 400	—	15.6 ± 0.17

^a Initial viscosity at 171 °C. ^b Gel time of samples cured at 171 °C.^c Average relative curing conversion of aged samples.

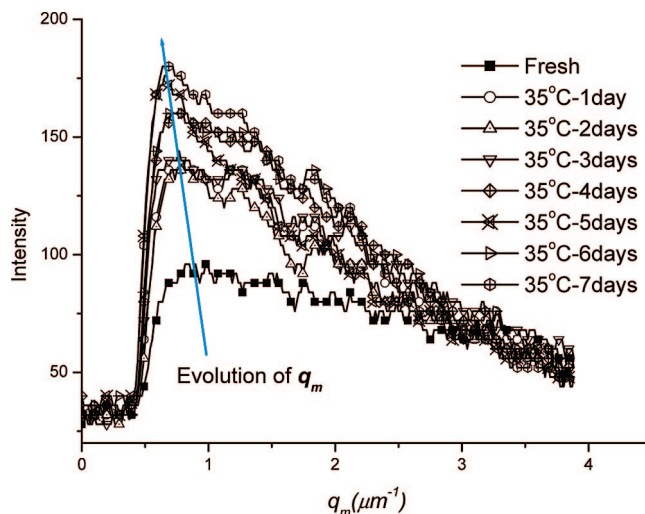
However, one of the most important difficulties for IR studies comes from the unevenness of preregs at different sites either in quality or in conversion. As a result, the curing conversion at different test sites varies, even though sites on the same sample were aged for the same period and at the same temperature. By selecting at least 20 sites for the same sample, we obtained an average value of the conversion. As shown in Figure 3 for sample a as an example, the conversion of the sample obtained by this statistical method exhibits a quite smooth curve with aging time at various temperatures.

During prepreg storage, moisture is another factor in need of investigation. Samples aged at 25 and 35 °C under three humidity conditions were studied. As there are quite a few differences between the spectra of these samples, the NIR results are not shown here. The moisture was found to have no effect on the IR band positions of either phenyl or epoxy groups, although humid conditions increased the curing conversion slightly as determined by calculation of the peak areas. Because the preregs used for PCBs are highly reactive with strong accelerators and have to be preserved below 5 °C, the accelerating effects of moisture are not as apparent as for ordinary slow-curing preregs.³

In conclusion, NIR spectroscopy can be a reliable and convenient method for the storage characterization of epoxy preregs because it is a relatively quick test and provides accurate results under different conditions for a broad range of commercial products.

3.2. Physical Properties. For commercial preregs, storage aging will always bring complicated changes in both physical properties and structures. To identify the connections between properties and aging, the rheological properties, thermal transitions, and phase structure changes were investigated.

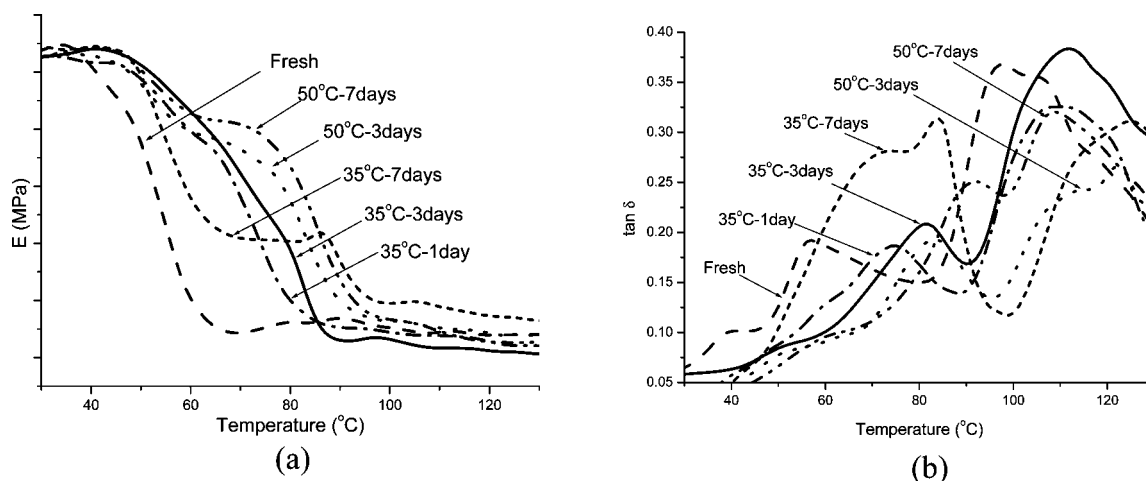
Rheological Behavior. Figure 4 shows the evolution of the viscosity and moduli of the aged samples, and Table 1 lists the

**Figure 6.** In situ TRLS study of sample a aged at 35 °C for 7 days.

initial viscosities and gelation times of these samples. The test temperature of 171 °C was chosen here because of the standard processing conditions of the PCB industry. As one can see, with increased aging time or higher temperature, the aged samples have higher viscosities in the initial stage and throughout the curing process, whereas the gel time was shortened at the same time. Here, the gelation point was determined by the crosspoint of the storage modulus (G') and the loss modulus (G''). This type of increase in viscosity with aging has been reported earlier and is believed to result from an increase of curing conversion.⁷

Glass Transition Temperatures. Another important parameter for the physical properties is the glass transition temperature (T_g), which is a combined result of different physical and chemical factors and would certainly influence the subsequent processing and PCB properties. Frigione et al.¹⁰ previously studied the relationship between the aging time and the glass transition temperature, reactivity, and processability by calorimetry. Their results showed an increase in T_g and a decrease in processability with aging time.

In Figure 5 are shown the results of DMA performed to study the change in modulus and $\tan \delta$ with aging temperature and time. For samples aged at low temperatures and for short periods, such as 35 °C for 1 day and 3 days, T_g increased slowly with aging time, as one can see from both the modulus and $\tan \delta$. This increase in T_g agrees with previous studies¹⁰ for one-phase systems. However, when the aging time was prolonged,

**Figure 5.** Test results of dynamic mechanical analyses of aged sample a: (a) modulus versus temperature, (b) $\tan \delta$ versus temperature.

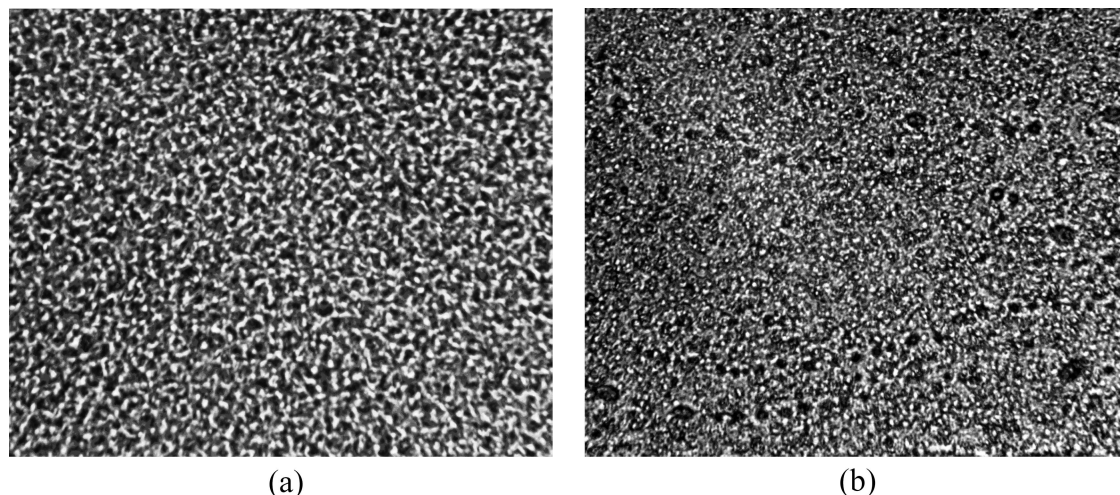


Figure 7. Optical microscopy image of sample aged at 35 °C for 7 days ($\times 400$): (a) sample without glass fiber, (b) solvent-extracted sample.

for example, to 35 °C for 7 days, another glass transition at higher temperature appeared, whereas the first T_g remained almost unchanged. Increasing the aging temperature, for example, to 50 °C for 3 and 7 days, increased the modulus plateau before the second glass transition, and the aging time also increased both the modulus and temperature of the second T_g . However, fully cured PCB prepregs in industry have only one T_g , which is located in the range of 170–178 °C by DMA for the studied samples.

Given that multiple glass transition temperatures always mean heterogeneous structures of polymer systems, the DMA curves with two T_g values might indicate a kind of phase separation in the prepregs during the aging process for some prepregs. As sample a is representative of epoxy/phenolic resin prepregs, this type of thermal behavior would be of great interest for an industry study.

Phase Separation. As time-resolved light scattering (TRLS) has already been used for studying structure in phase separation process,²⁶ we employed this in situ method to observe the structure change of prepregs during aging. For TRLS, the scattered light intensity is shown as a function of the magnitude of the scattering vector q_m , which is defined by

$$q_m = (4\pi/\lambda) \sin(\theta/2) \quad (2)$$

where λ is the wavelength of light in the blend (dimension of the phase structure) and θ is the scattering angle between the incident and scattered light. As q_m is inversely proportional to the interdomain distance and the scattering light intensity is closely related to the concentration difference between the phases, TRLS provides information on both the phase dimension and concentration change.

In Figure 6, the light intensity grows with aging time, and the peaks shift to low q_m value. These types of plots definitely indicate a phase separation process of the prepreg. During the aging process, sample a shows bright scattering rings in TRLS, which indicates phase separation following the spinodal decomposition (SD) mechanism, in which the regular morphology by phase separation can give a clear profile in TRLS. The shift of q_m toward a smaller angle indicates that the interdomain distance becomes longer during phase separation, i.e., the dimension of the phase structure grows with aging. At the same time, the increase in scattering light intensity, I_m , evidently indicates an increase in concentration difference due to phase separation.

Optical microscopy (OM) was used to observe the morphology of the aged samples. Figure 7a shows aged samples without glass fibers, where the phase separation was not the fillers because the fresh samples show a kind of cloudy opaqueness under OM. Then, we dissolved the samples in tetrahydrofuran and filtered the inorganic filler content. The resulting transparent solutions were poured onto glass slides and aged. Although the solvent extraction process would change some properties of the epoxy resins in prepregs, Figure 7b still shows the heterogeneous phase structure in the resulting mixture.

Combining the TRLS results and DMA observations, we can provide the following explanations for the storage aging: As some commercial prepregs are always composed of a large variety of epoxy, phenolic resin, and high-molecular-weight additive polymers, multiphase structures are possible during aging for many prepregs. Before aging, the epoxy resins in the fresh prepregs had an almost homogeneous structure; the TRLS profiles and DMA results of only one T_g are evidence for this structure. With the aging (curing) process, a reaction-induced phase separation takes place as a result of an increase in entropy with the molecular weight of the epoxy resins.²⁷

It is well-known that, for thermosetting blend systems, phase separation is controlled by the curing reaction and the diffusion of epoxy molecules.^{26,27} Phase separation might take place above a certain curing conversion at suitable conditions. At low curing rate, the diffusion of epoxy molecules is the controlling factor (as the T_g of the epoxy-rich material is quite low). Prolonged aging time at low temperature such as 35 °C would facilitate phase separation because of the low-rate conversion during this period. This would result in a higher- T_g epoxy-rich phase because of relatively complete phase separation. In contrast, at a higher curing rate, the curing reaction is the controlling factor, and quick gelation would freeze the phase structure in the initial stage; therefore, only one T_g was found for prepregs cured at 171 °C. Under intermediate conditions, aging (curing) and phase separation might take place at the same time. Aging (curing) would increase the T_g of the epoxy-rich phase, and phase separation could enhance the T_g of the other phase. For a higher aging temperature such as 50 °C, a higher curing rate would compete with phase separation to block some low-molecular-weight epoxy in the higher- T_g phase; as a result, the modulus of the higher- T_g phase increased quickly, whereas the T_g values changed little at the same time.

In general, storage aging would cause both chemical and physical changes. Chemical changes affect the subsequent

process and curing conditions, and physical changes such as morphology evolution can have a profound influence on the resulting materials. However, below certain chemical changes (curing conversion), there would be no phase separation during storage. Therefore, curing conversion monitoring is of essential importance for property control, and near-infrared spectroscopy is a powerful tool in prepreg tests for PCBs and other applications.

4. Conclusions

Near-infrared spectroscopy can be applied in investigations of storage aging of prepreps in industrial applications, whereas mid-infrared spectroscopy is restrained in some systems because of overlapping bands. Both the physical and chemical properties of epoxy prepreps are obviously influenced by storage aging. Rheological tests show clearly an increase in viscosity and a decrease in gel time with aging time and temperature. The results from dynamic mechanical analyses (DMA), time-resolved light scattering (TRLS), and optical microscopy (OM) show morphology variations with both aging temperature and period for some prepreps, which would have profound effects on the processing of prepreps and the properties of PCB materials.

Acknowledgment

This research work was supported by the National Natural Science Foundation of China (NNSFC 20704008), the Specialized Research Fund for the Doctoral Program of Higher Education of China (SRFDP 20070246001), and the Key Project Science Foundation of Shanghai Municipal Commission of Education (06zz78).

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Received for review November 24, 2008

Revised manuscript received January 12, 2009

Accepted March 9, 2009

IE8018005