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Modification of epoxy resin with siloxane containing phenol aralkyl epoxy resin for electronic encapsulation application

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Abstract

A process was developed to incorporate stable dispersed polysiloxane particles into a phenol aralkyl novolac epoxy resin, which was used as an ingredient in the encapsulant formulation to withstand the thermal stress. The mechanical and dynamic viscoelastic properties and morphologies of rubber-modified epoxy networks were studied. A "sea-island" structure ("islands" of silicone rubber dispersed in the "sea" of an epoxy resin) was observed in cured rubber-modified epoxy networks via SEM. The dispersed silicone rubber-modified aralkyl novolac epoxy resin effectively reduces the stress of cured epoxy molding compounds by reducing flexural modulus and the coefficient of thermal expansion (CTE), while the glass-transition temperature (T_g) was hardly depressed. Electronic devices encapsulated with the dispersed silicone rubber-modified epoxy molding compounds have exhibited excellent resistance to the thermal shock cycling test and have resulted in an extended use life for the devices. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Aralkyl novolac epoxy resin; Polysiloxane; Encapsulant

1. Introduction

Epoxy molding compounds (EMCs) are most popularly used as encapsulation materials for semiconductor devices to provide protection of the integrated circuits (IC) devices from moisture, mobile-ion contaminants, and adverse environmental conditions such as temperature, radiation, humidity, and mechanical and physical damage [1]. Such EMCs are composed of many raw materials. Nowadays, *o*-cresol-formaldehyde novolac epoxy (CNE) is the resin typically employed to encapsulate microelectronic devices because of its well-balanced properties including excellent heat, solvent, moisture, and chemical resistance, superior mechanical and electrical properties, and good adherence to many substrates. Upon cure, this multifunctional epoxy resin provides a densely crosslinked protective layer; however, it is relatively brittle.

Recently, for EMC-sealed IC, the chip size has increased rapidly while the packages, on the other hand, are designed to be smaller and thinner in outer dimensions to meet the demands of compactness and light weight for electronic equipment [2]. These trends increase thermal stress in the device package system which often causes package cracking during thermal shock testing under accelerating ambient [3,4]. The prevailing surface mount technology (SMT) also causes thermal stress to devices. Therefore, it is necessary to design the device package so as to reduce the thermal stress and to increase the strength and toughness of the cured EMCs for high-reliability semiconductor devices.

Reductions of thermal stress by lowering of either the coefficient of thermal expansion (CTE) or the flexural modulus of the encapsulant have been reported [5–8]. Increasing the filler loading in an encapsulant effectively lowers the CTE [9]; however, this approach not only increases the elastic modulus, but also decreases the molding properties due to high viscosity at melting condition and increases the possibility of wire deformation.

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Two principal methods of lowering the flexural modulus of an epoxy molding compound – one is to start with materials which by themselves have a low flexural modulus and the other is to incorporate a rubber in a "sea-island" two-phase structure [4,10]. In our previous study [11], we have successfully synthesized a series of aralkyl novolac epoxy resins that have a low internal stress resulting from a low flexural modulus.

Organosiloxane polymers are known for their excellent thermal and thermooxidative stabilities, very low T_g (−123°C), moisture resistance, good electric properties, and low stress [6]. Therefore, reduction of stress in thermosetting resins, via incorporation of organosiloxane moiety into the uncured thermosetting resins, and inducing a phase separation before curing to form small discontinuous rubber particles in the resin matrix, was used most oftenly [12,13]. In this work, a new process was developed to incorporate stable dispersed polysiloxane particles into a phenol aralkyl novolac epoxy resin, which was used as an ingredient in the encapsulant to withstand the thermal stress. The mechanical and dynamic viscoelastic properties and morphologies of rubber-modified epoxy networks were studied. The devices encapsulated by the polysiloxane-modified EMC were also evaluated by the thermal shock cycling test.

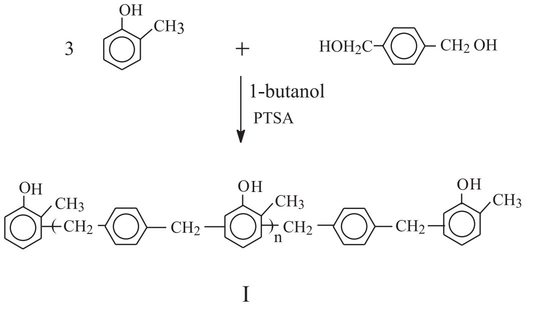
2. Experimental

2.1. Materials

o-Cresol and allyl chloride were purchased from Aldrich and were vacuum-distilled prior to use. *p*-Toluene sulfonic acid 1-hydrate (PTSA) was purchased from Ferak and vacuum-dried prior to use. *p*-Xylylene glycol obtained from Aldrich and epichlorohydrin(ECH) purchased from Ferak, were used without further purification. All solvents were purified by standard methods before use. Hydride-terminated polydimethylsiloxane ($M_w = 17500$) was purchased from HULS America Inc. The control epoxy resin was CNE (Quatrex 3330, Dow Chemical Co., epoxy equivalent weight, EEW, 192). A phenol–formaldehyde novolac resin with an average hydroxyl functionality of 6 and a hydroxyl equivalent weight of about 104 (Schenectady Chemical, HRJ-2210) was used as a hardener. Triphenyl-phosphine (Ph_3P) was used as a curing accelerator. H_2PtCl_6 (5 wt.% in isopropanol) was used as a catalyst in hydrosilation.

2.2. Synthesis of aralkyl novolac resin

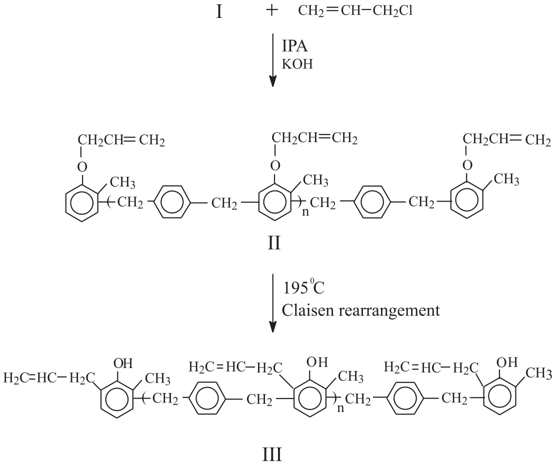
Into a 1-l three-necked flat-bottomed flask was added a solution of 69 g (0.5 mol) of *p*-xylylene glycol in 200 ml of 1-butanol and 1.5 mol of *o*-cresol (159 g). After purging with nitrogen for 10 min, PTSA, (1.0 g) was added to the above solution at room temperature. The mixture was allowed to react by stirring at 115–120°C for 10 h under a reduced pressure (ca. 200 mm Hg), in the meantime, removing water by azeotroping with 1-butanol and returning the distilled 1-butanol to the system. The resulting mixture was neutralized with a dilute aqueous potassium hydroxide solution and washed with deionized water until the wash solution was neutral. The organic phase was finally "rotavapped" at 195°C under full vacuum for 3 h to remove the excess phenol till less than 0.3% (as analyzed by LC). Measurements by gel permeation chromatography (GPC) and infrared (IR) absorption spectroscopy were made on the resin. The synthetic scheme is shown in Scheme 1.



Scheme 1. The synthesis of aralkyl novolac resin.

2.3. Preparation of the allyl aralkyl novolac resin

To a 500 ml four-neck round-bottomed flask was added a solution of 84 g of the above synthesized aralkyl novolac resin in 180 ml of isopropanol (IPA) and 25 g of potassium hydroxide. After purging with nitrogen for 10 min, allyl chloride (41 g) was continuously added to the above solution at room temperature via a metering



Scheme 2. The preparation of the allyl aralkyl novolac resin.

pump over a period of 20 min. Upon completion of the addition, the reaction temperature was raised to 40°C and maintained for 1 h, and then raised to 70°C and maintained for 6 h. The mixture was filtered to remove potassium chloride and then rotavapped at 175°C under reduced pressure to remove IPA. The residue was dissolved in ethyl acetate and washed with deionized water. The organic phase was "rotavapped" at 175°C under full vacuum over 3 h to remove the solvent to obtain the allyl ether of aralkyl novolac resin (II). The resultant product was further heated to 195°C with stirring while maintaining the reaction temperature at 195°C for 3 h to complete the Claisen rearrangement. The product, allyl aralkyl novolac resin (III), was obtained. The synthetic scheme is shown in Scheme 2.

2.4. Preparation and dehydrohalogenation of polyglycidyl ether of the aralkyl novolac resin

Preparation and dehydrohalogenation of polyglycidyl ether of the allyl aralkyl novolac resin (IV) was carried out in accordance with the reported procedures of our previous study [11]. The synthetic scheme is shown in Scheme 3.

2.5. Preparation of silicone rubber-modified aralkyl novolac epoxy resin

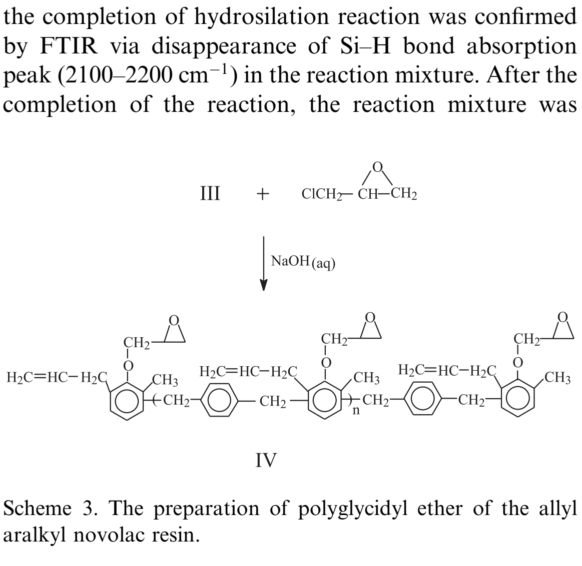
The above resultant epoxy resin (IV) 50 g, 400 ml of toluene, and 150 g of hydride-terminated siloxane were added to a 1-l flask, equipped with a hot plate, thermocouple, and temperature controller, magnetic stirrer, and a Dean–Stark trap with a reflux condenser. The contents were mixed thoroughly and the temperature was raised to remove a minor amount of water by azeotroping with toluene for 1 h. H_2PtCl_6 solution (5%, 1 g) was added and the mixture was allowed to reflux until the completion of hydrosilation reaction was confirmed by FTIR via disappearance of Si–H bond absorption peak (2100–2200 cm^{-1}) in the reaction mixture. After the completion of the reaction, the reaction mixture was then heated to 160°C under a full vacuum for 3 h to remove solvents. Upon cooling to room temperature, a creamy silicone modified epoxy resin was obtained. The synthetic scheme is shown in Scheme 4.

2.6. Preparation of cured epoxy resins

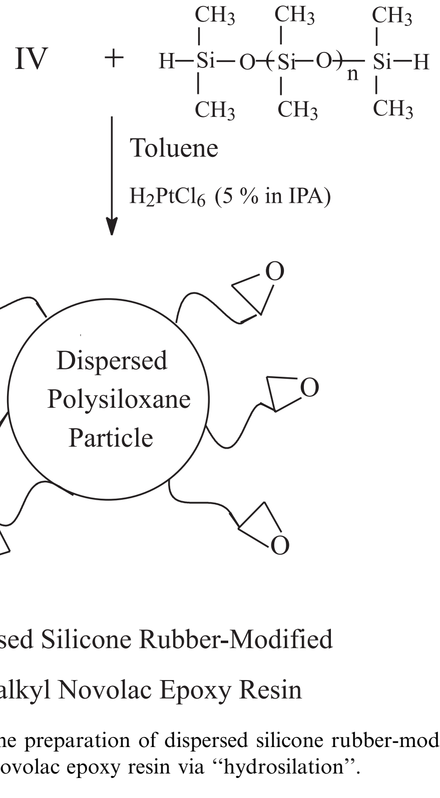
The siloxane-modified aralkyl epoxy resin was mixed with CNE, a stoichiometric amount of curing agent (HRJ-2210), and accelerator (Ph_3P) in a mill at moderate temperature to give a thermosettable epoxy resin powder. The amount of siloxane modifier was 12.5 wt.% of total resin components. The resin powder was cured in a mold at a temperature of 150°C and 50 $kg\,cm^{-2}$ for a period of 1 h and then postcured at 180°C for 2 h and 200°C for 3 h to obtain a cured specimen for dynamic viscoelastic analysis and SEM.

2.7. Preparation of epoxy molding compounds

Control resin and aralkyl novolac epoxy resin modified with silicone rubber were formulated into two electronic encapsulating formulations. The details of encapsulating formulations are given in Table 1. The formulations were each cured at 175°C for 4 h. The thermal mechanical properties of the cured encapsulating formulations were determined by the following tests.



Scheme 3. The preparation of polyglycidyl ether of the allyl aralkyl novolac resin.



Scheme 4. The preparation of dispersed silicone rubber-modified aralkyl novolac epoxy resin via "hydrosilation".

Table 1
Encapsulation formulation

Ingredients	Weight (%)	
	Control	Siloxane-modified
Cresol novolac epoxy	17.5	15.3
Siloxane-modified aralkyl epoxy	0	3.0
Phenolic hardener (HRJ-2210)	9.1	8.3
Brominated epoxy resin (diglycidyl ether of tetrabromobisphenol A)	2.5	2.5
Fused silica (GP-71 Harbison–Walker)	68.5	68.5
Mold release (Hoechst Wax OP and E)	0.4	0.4
Carbon black	0.4	0.4
Silane coupling agent (γ-aminopropyl triethoxysilane)	0.4	0.4
Antimony trioxide	1.0	1.0
Triphenylphosphine accelerator	0.2	0.2

Procedure
1. B-Stage melt mix at 95°C for approximately 7 min
2. Cool and grind to a uniform powder
3. Transfer molding of powder at 175°C for 90 s
4. Postcure at 175°C for 4 h

2.8. Measurement and testing

EEWs of epoxy resins were determined by the $HClO_4$/potentiometric titration method [14]. Infra-red spectra were recorded with a Perkin–Elmer 16PC FTIR spectrophotometer operated with a dry air purge. Signals of 32 scans at a resolution of 4 cm^{-1} were averaged before Fourier transformation. Mass spectrometric analyses were performed on a VG70-250S GC/MS spectrometer. Dynamic viscoelastic properties were studied on a Perkin–Elmer 7 series thermal analyzer with DMA mode between −150 and 300°C with a heating rate of 5°C min^{-1} at a frequency of 1 Hz. The temperature/time scan in a three-point bending mode was chosen and the dimensions of the specimen were $15 \times 5.5 \times 1.5$ mm^3 according to ASTM D790-95a, the distance of the support span was 10 mm. The elastic modulus E' and $\tan\delta$ were determined. The molecular weight of aralkyl novolac resin was obtained on a Shimadzu C-R4A gel permeation chromatograph (GPC) using a Shimadzu GPC-8025 column. Samples were analyzed by a Shimadzu RID-6A reflex indicator. A JEOL JSM-6400 scanning electron microscope was employed to examine the morphology of cured rubber-modified samples fractured cryogenically in liquid nitrogen. The fracture surfaces were vacuum-coated with gold. Flexural properties of cured resins were measured using a Shimadzu AGS-500 universal testing machine. Flexural strength and modulus were obtained at a crosshead speed of 2 mm min^{-1} according to ASTM D790-86. A 3-point loading system was chosen and rectangular bar specimens, $80 \times 10 \times 4$ mm^3, were molded directly by a transfer molding process. The CTE was measured with a DuPont 943 thermal mechanical analyzer (TMA) in accordance with ASTM E83-186. A specimen 4 mm in length was used at a heating rate of 5°C min^{-1}. Normally, the thermal expansion increases with the increase in temperature and the CTEs were calculated from the slope. An abrupt change in slope of the expansion curve indicates a transition of the material from one state to another. The moisture pick up was determined by placing preweighed 3 mm thick × 50 mm diameter cured disks in boiling water for 100 h. The disks were removed, cooled to ambient, wiped dry, and weighed to determine any weight gain.

2.9. Thermal shock cycling test

The thermal shock cycling test was carried out by the following procedure [15]. The device used was a 14-pin LM 324 quad operational amplifier with a single passivation layer. The device was encapsulated with an encapsulation formulation by a transfer molding process and subjected to a thermal cycling test. A cycle consisted of −65°C × 15 min and 150°C × 15 min. The devices were inspected by an optical microscope for cracks after 250, 500, 750, 1000, 1500, 2000, 2500, 3000, and 4000 cycles. Any crack observed in the encapsulated device was counted as the failure of that device. The percentage of devices that failed (cracked), as a function of cycles, was plotted.

3. Results and discussion

3.1. Synthesis and characterization

Phenol aralkyl novolac (I) was synthesized by condensing *p*-xylylene glycol with excess *o*-cresol using *p*-toluene sulfonic acid as a catalyst as shown in Scheme 1.

GPC measurement of the novolac was carried out and the average molecular weights ($\overline{M}_n = 753$ and $\overline{M}_w = 908$) were calculated by use of GPC calibration curve. Dispersed silicone rubber in aralkyl novolac epoxy resin was prepared through several steps. The first step involves the preparation of an allyl ether of aralkyl novolac resin (II) resulted from the aralkyl novolac resin with allyl chloride in the presence of potassium hydroxide followed by the Claisen rearrangement at 195°C to form an allyl aralkyl novolac resin (III) as shown in Scheme 2. The resultant allyl aralkyl novolac resin was further reacted with a halohydrin in the presence of an alkali metal hydroxide to obtain an allyl aralkyl epoxy resin (IV) as shown in Scheme 3. The last step is the hydrosilation of the resultant allyl aralkyl epoxy resin with hydride-terminated siloxane in the presence of a catalyst (5% H_2PtCl_6 in isopropanol) to form a stable silicone rubber particles dispersed in the matrix resin in a sea-island structure as shown in Scheme 4. The structure of the synthesized aralkyl novolac was confirmed by infrared (IR) and mass spectra (MS). From the IR spectra (Fig. 1) of I and its poly(allyl ether) (II), we can see a strong absorption peak at 3400–3600 cm^{-1} representing the –OH functional groups of the synthesized aralkyl novolac, and a peak appeared at 990 cm^{-1} representing the ether groups absorption and a peak appeared at 1640 cm^{-1} representing the allyl double bond absorption of the allyl ether of aralkyl novolac whereas the –OH functional groups absorption peak disappeared. The Claisen rearrangement was observed from the IR spectra shown in Fig. 2(A), we can see the reappearance of –OH functional groups absorption while the ether groups absorption peak disappeared. In addition, a peak at 1020 cm^{-1} shown in Fig. 2(B) represents the oxirane ring absorption of allyl aralkyl epoxy resin. Furthermore, the completion of hydrosilation was confirmed by the disappearance of the Si–H bond absorption peak at 2126 cm^{-1} and the appearance of a Si–O–Si peak at about 1088 cm^{-1} as shown in Fig. 2(C).

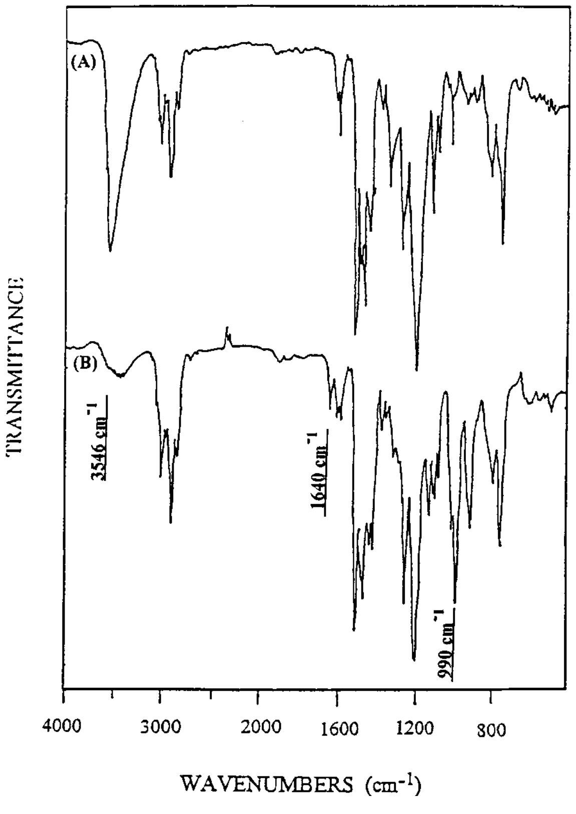


Fig. 1. FTIR spectra of (A) aralkyl novolac resin, I and (B) allyl ether of aralkyl novolac resin, II.

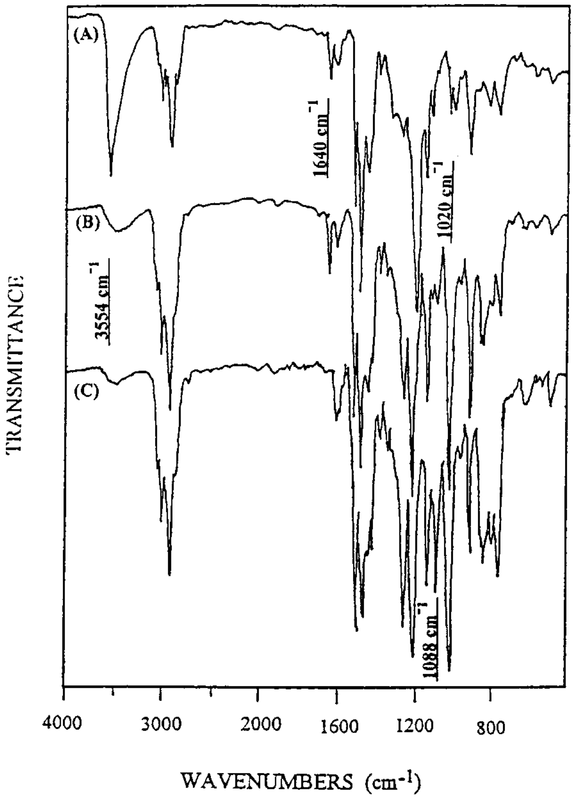


Fig. 2. FTIR spectra of (A) allyl aralkyl novolac resin, III, (B) allyl aralkyl epoxy resin, IV and (C) hydride terminated siloxane modified epoxy resin.

3.2. Dynamic viscoelastic analysis

Dynamic viscoelastic analysis can give information on the microstructure of cured epoxy resins. The dynamic viscoelastic spectra are shown in Fig. 3, which shows the storage elastic modulus E' and $\tan\delta$ curves for

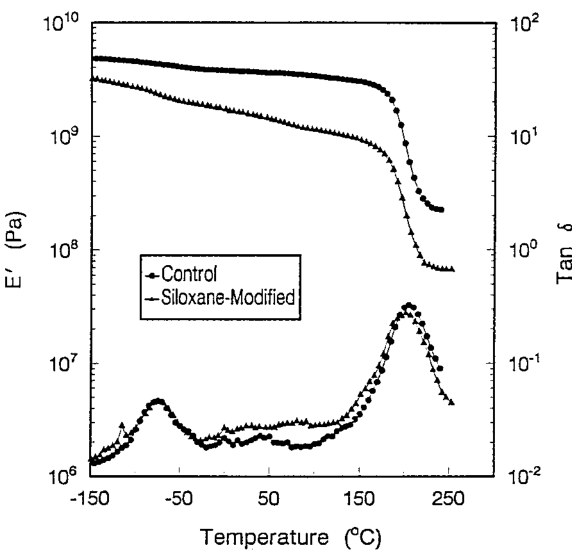


Fig. 3. Comparison of dynamic viscoelastic properties for cured siloxane-modified and unmodified epoxy resins (no filler). The curing agent is phenol–formaldehyde novolac (HRJ-2210).

the control resin and CNE mixed with siloxane-modified aralkyl epoxy resin. It can be seen that the elastic modulus decreases dramatically with rubber modification. The tan δ curves for the cured epoxy resins exhibit two major relaxations observed in most epoxy polymers [16]: a high-temperature tan δ peak corresponds approximately to the T_g of the cured epoxy resins, above which significant chain motion takes place; the low-temperature or β-transition is attributed predominately to the motion of the CH_2–CH(OH)–CH_2–O (hydroxy ether) group of the epoxy. It can be seen from Fig. 3 that the peak position of α-relaxation peak in the tan δ curve of the siloxane-modified epoxy networks is slightly lower than that of unmodified. This can be attributed to the addition of aralkyl epoxy resin whose T_g is lower than that of CNE. In addition to the epoxy α and β peaks, the tan δ curve for the siloxane-modified product shows an additional markedly small peak around −115°C corresponding to the T_g of the polysiloxane phase. The dynamic viscoelastic properties of the cured epoxy resins including the major T_g and the rubbers T_g are shown in Table 2.

Table 2
Dynamic viscoelastic properties of cured neat rubber-modified and unmodified epoxy resins

Sample	Matrix, T_g[a] (°C)	$E'_{50°C}$ (GPa)	Rubber, T_g[b] (°C)
Control[c]	204.9	3.61	–
Siloxane-modified	202.4	1.44	−115

[a] Peak of tan δ at higher temperature.
[b] Peak of tan δ at lower temperature.
[c] Control, unmodified CNE.

3.3. Morphology

SEM photomicrographs of freeze fractured surfaces for the control and the siloxane-modified epoxy networks are given in Fig. 4. Electron micrographs reveal that rubber particles with 1 μm or less in size have been dispersed in the rubber-modified resin's matrix in a "sea-island" structure ("island" of polysiloxane particles dispersed in the "sea" of an epoxy resin).

The thermal mechanical properties and moisture absorption of the cured encapsulating formulations were determined and the results are shown in Table 3.

3.4. Coefficient of thermal expansion

The CTE in the glassy state below the T_g was taken from 60°C to 100°C and the CTE above the T_g taken from 200°C to 240°C. For the CTE below the T_g, siloxane-modified encapsulant has slightly lower CTE than the unmodified resin and this will result in a small dif-

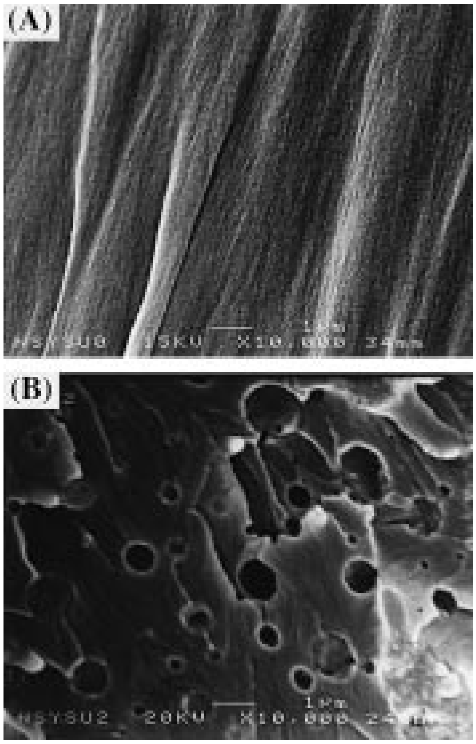


Fig. 4. Morphology of cold-snap fracture surfaces of cured epoxy resins modified with or without polysiloxane rubber particles (12.5 wt.%). (A) unmodified and (B) siloxane-modified.

Table 3
Some thermal and mechanical properties and moisture absorption of epoxy molding compounds

	Control	Siloxane-modified
Thermal mechanical analysis		
α_1 ($\times 10^{-5}$ K^{-1})[a]	2.0	1.8
α_2 ($\times 10^{-5}$ K^{-1})[b]	7.3	7.5
T_g (°C)	164	163
Flexural test[c]		
Flexural modulus (GPa)	12.8	10.7
Flexural strength (MPa)	131.3	128.1
Thermal stress parameter		
$E' \times \alpha_1$ (kPa K^{-1})	256.0	192.6
Moisture absorption	0.32	0.29

[a] Thermal expansion coefficient below T_g.
[b] Thermal expansion coefficient above T_g.
[c] Three-point bending flexural test.

ference in CTEs between encapsulant and silicon chip. This result is in agreement with that reported in the earlier works [17]. It is known that the addition of a modifier to the epoxy matrix, with a higher thermal expansion than the matrix, leads to an increase in the local free volume in the epoxy matrix and thus to increase the linear CTE [18,19]. On the contrary, the addition of silica fillers, with lower values of CTE than the resin matrix, has the opposite effect on stress compared to the case of silicon modifier in the epoxy; that is, radial compressive stresses occur at the interface of the resin and the filler, and tangential tensile stress occur in the surrounding matrix from the cure temperature to room temperature. This stress should reduce the local free volume in the resin matrix and thus decrease the value of CTE of the material [20]. Thus, the siloxane modifier is considered to cause an opposite effect on CTE in the molding compound as compared to the epoxy matrix.

3.5. Glass-transition temperature

T_gs were determined from tangents of the CTE as a function of temperature at 100 and 200°C. The results are shown in Table 3. The T_g of cured siloxane-modified epoxy molding compound is slightly lower than that of the control resin. This result is consistent with the viscoelastic investigation.

3.6. Flexural test properties

From the result of the flexural test shown in Table 3, the flexural modulus of the cured epoxy encapsulant was reduced markedly with silicone rubber modification. The flexural strength of hydride siloxane-modified epoxy encapsulant was slightly lower than that of the control resin.

3.7. Internal stress

Internal stress of the cured encapsulants can be approximated by the product of flexural modulus and the CTE below the T_g, ($E\alpha_1$) [3,13], where E and α_1 represent flexural modulus and CTE, respectively. From Table 3, we can see that the stress of rubber-modified encapsulant is markedly lower than that of the control resin. The dispersed silicone rubber particles effectively reduce the stress of cured EMC by reducing flexural modulus and the CTE.

3.8. Moisture absorption

Absorbed moisture in the package was found not only to plasticize the epoxy resin, causing a lowering of the T_g and in turn affecting mechanical response, but also to cause the package crack during the soldering process [21]. This cracking (the so-called popcorn phenomenon) is caused by evaporation and expansion of absorbed moisture in the package at the temperature of reflow soldering (215–260°C) for mounting of encapsulated IC on printed circuit board. The moisture absorption result shown in Table 3 indicates that the siloxane-modified EMC absorbed less moisture than did the unmodified EMC.

3.9. Thermal shock cycling test

The control and the siloxane-modified EMC were used to encapsulate the semiconductor devices and the encapsulated devices were subjected to a thermal shock cycling test. The thermal shock cycling test involves cycling the encapsulated devices at −65 and 150°C and observing the crack (failure) of encapsulated devices at various intervals (after 250, 500, 750, 1000, 1500, 2000, 2500, 3000, 3500, and 4000 cycles). Any crack that occurred in a device is counted as failure for that device. The percentage of devices that failed vs. test cycles is given in Fig. 5. The result indicates that for 50% of the devices, failure happened after 3000 cycles for the siloxane modified EMC compared to after 700 cycles for the control resin. Siloxane modified EMC has much better thermal shock resistance, lower stress, and lower moisture absorption than the control, that has resulted in an extended lifetime for the devices.

4. Conclusions

Phenol aralkyl novolac was synthesized by condensing p-xylylene glycol with excess o-cresol using p-toluene sulfonic acid as a catalyst. A process was developed to incorporate stable dispersed polysiloxane particles into the phenol aralkyl novolac epoxy resin, which was used as an ingredient in the encapsulant formulation to

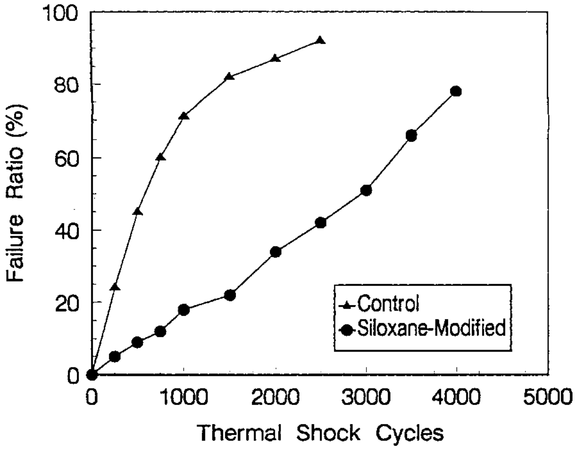


Fig. 5. Thermal shock cycling test results. Temperature cycle: −65°C × 15 min and 150°C × 15 min.

withstand the thermal stress. A "sea-island" structure was observed in cured rubber-modified epoxy networks via SEM. The dispersed silicone rubber-modified aralkyl novolac epoxy resin effectively reduced the stress of cured epoxy molding compounds by reducing flexural modulus and the coefficient of thermal expansion, while the glass-transition temperature was hardly depressed. Electronic devices encapsulated with the dispersed silicone rubber-modified epoxy molding compounds have exhibited excellent resistance to the thermal shock cycling test and have resulted in an extended lifetime for the devices.

Acknowledgements

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