

Modification of epoxy resins with polysiloxane thermoplastic polyurethane for electronic encapsulation: 1

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A stable dispersion of polysiloxane thermoplastic polyurethane (TPU) particles in an epoxy resin matrix was achieved via epoxy ring opening with the isocyanate groups of urethane prepolymer to form an oxazolidone. The effects of the structure and molecular weight of the polysiloxane TPU in reducing the stress of electronic encapsulants were investigated. The mechanical and dynamic viscoelastic properties of polysiloxane TPUmodified epoxy networks were also studied. A phase-separated structure was observed via scanning electron microscopy. The dispersed polysiloxane TPU rubbers effectively reduce the stress of cured epoxy resins by reducing the flexural modulus and the coefficient of thermal expansion, while the glass transition temperature is increased because of the formation of the rigid oxazolidone structure. Electronic devices encapsulated with the polysiloxane TPU-modified epoxy moulding compounds have exhibited an excellent resistance in the thermal shock cycling test and have resulted in an extended device used life. Copyright © 1996 Elsevier Science Ltd.

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INTRODUCTION

Epoxy moulding compounds (EMCs) have been widely used as encapsulation materials for semiconductor devices in order to protect them from their surroundings and connect them to printed circuit boards¹. Because of its excellent heat, moisture, solvent and chemical resistance, superior electrical and mechanical properties, and good adhesion to many substrates, o-cresolformaldehyde novolac epoxy (CNE) is the resin typically employed to encapsulate microelectronic devices. Upon cure, this multifunctional epoxy resin provides a densely crosslinked protective layer; however, it is relatively brittle.

The trend of electronics equipment is towards being miniaturized and becoming thinner; at the same time, the scale of integration of large-scale integrated circuits (LSICs) is continuing upward, forcing the design towards larger chips, finer patterns and higher pin counts that are more susceptible to internal stress failure. The prevailing surface mount technology (SMT) also causes thermal stress to devices. Internal stress may cause package cracking, passivation layer cracking and aluminium pattern deformation, etc. 2^{-4} . Therefore, the development of a low-stress EMC is required for highreliability semiconductor devices. The sources of internal stress that result from the use of plastic encapsulants are

considered to be shrinkage of the plastic upon curing and thermal mismatch between the resin and the device. In the case of EMC encapsulation, the first source of shrinkage has been relatively minor, whereas the second one, which is caused by the difference in thermal expansion coefficients between resin and silicon chip, is the dominant factor. The internal stress on IC devices caused by the difference in thermal expansion coefficients is expressed by the following equation³:

$$
S = K \int (\alpha_{\rm r} - \alpha_{\rm s}) E_{\rm r} dT
$$

where S is the internal stress in the encapsulant; K is a constant; α_r is the thermal expansion coefficient of the encapsulant; α_s is the thermal expansion coefficient of the silicon chip; and E_r is the flexural modulus of the encapsulant. Therefore, it is necessary to reduce the thermal expansion coefficient and the flexural modulus of EMCs to reduce internal stress.

Reductions of internal stress by lowering of either the thermal expansion coefficient or the flexural modulus of the encapsulant have been reported $6-14$. Increasing the amount of silica filler used in an encapsulant effectively lowers the thermal expansion coefficient; however, this approach not only increases the elastic modulus, but also increases the viscosity of the resin composition, resulting in poor mouldability. Lowering the flexural modulus by modification with a rubber in a phase-separated two-

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phase structure is considered to be more desirable than a one-phase structure in view of the thermal property needs^{15} . Traditional modifiers that can reduce the elastic modulus of the cured epoxy resins, including reactive liquid rubber such as carboxy-terminated butadieneacrylonitrile copolymer (CTBN), have been reported^{10,17}. When CTBN modifiers are incorporated into resin compositions, a two-phase morphology consisting of relatively small rubber particles dispersed in a resin matrix is generated that toughens epoxy resins. However, phase separation depends upon the formulation, processing and curing conditions. Incomplete phase separation can result in a significant lowering of the glass transition temperature $(T_g)^{18}$. Moreover, the presence of CTBN modifiers in EMCs raises the thermal expansion coefficient of the resultant EMC. The relatively high T_{g} of butadiene-acrylonitrile copolymer also limits their lowtemperature applications. Polysiloxanes are known for their excellent thermal and thermo-oxidative stabilities, moisture resistance, good electrical properties, low stress and lower $T_{\rm g}$ (-123°C) values than conventional elastomers. Low-stress EMCs modified by vinyl- or hydride-terminated polydimethylsiloxanes reported by the authors^{19,20} have effectively reduced the stress of cured epoxy resins by reducing the flexural modulus and the coefficient of thermal expansion; however, the T_{g} values were slightly sacrificed. Therefore, developing a material that not only reduces the stress of cured EMC but also increases the T_g of the resultant EMC is a major task for the plastic packaging industry.

In this work, a stable dispersion of thermoplastic polyurethane (TPU) or polysiloxane TPU particles in an epoxy resin matrix was achieved via epoxy ring opening with the isocyanate groups of urethane prepolymer to form an oxazolidone. The effects of the structure and molecular weight of TPU in reducing the stress of electronic encapsulants were investigated. The mechanical and dynamic viscoelastic properties of TPU-modified epoxy networks were also studied. Devices encapsulated by the TPU-modified EMC were also evaluated by the thermal shock cycling test.

Table 1 Polyols used in the manufacture of polyurethanes

EXPERIMENTAL

Materials

The control epoxy resin was o-cresol-formaldehyde novolac epoxy resin (CNE) (Quatrex 3330, Dow Chemical Co.; epoxy equivalent weight $EEW = 192$). A phenol-formaldehyde novolac resin was used as a curing agent with an average hydroxyl functionality of 6 and a hydroxyl equivalent weight of about 104 (Schenectady Chemical, HRJ-2210). Methylene bis(4 phenylisocyanate) (MDI) (Multrathane, Mobay Chemical) was distilled under reduced pressure (170°C at 0.05 mmHg). Poly(tetramethylene ether) glycol (PTMG) (Polymer, Quaker Oats) was degassed under vacuum at 65°C and 2 mmHg for 3 h to remove any absorbed water, then stored over type 4 A molecular sieves. Poly(propylene ether) glycol (PPG) and poly(ethylene ether) glycol (PEG) (Naclai Tesque Inc., Kyoto, Japan) were degassed in the same manner as the PTMG. Polydimethylsiloxane- α , ω -diols (PDMS-diols) with hydroxyl numbers 62 (XF-6001) and 112 (X-22-160AS), giving a calculated *MW* of 1810 and 1002 respectively, were purchased from Shin-Etsu Chemical Co. Ltd, Japan. The structure, *MW* and designations of the polydiols used in the manufacture of the polyurethanes are listed in *Table 1.* The structural formulae of CNE and MDI are also shown later. Stannous octoate was used as a catalyst in the syntheses of isocyanate-terminated PDMS-diols. 2-Phenylimidazole was used as a catalyst in the epoxideisocyanate reaction to form oxazolidone. Triphenylphosphine (Ph_3P) was used as a curing accelerator.

General procedure for the synthesis of urethane prepolymer

Urethane prepolymer. To a flame-dried 500 ml fournecked round-bottomed flask, equipped with a watercooled condenser with a capped $CaCl₂$ drying tube, a thermometer, N_2 inlet, a 150ml addition funnel and mechanical stirrer, was charged 1.0mol of MDI and heated to 85° C. To the MDI was added 0.5 mol of polyol

(PEG, PPG or PTMG) dropwise (i.e. $NCO:OH = 2:1$) under a nitrogen atmosphere. The mixture was stirred and maintained at 85°C until the absorption peak of the OH group in infra-red (i.r.) spectra had disappeared.

PDMS-based urethane prepolymer. The reaction flask was equipped in the same manner as mentioned above. The PDMS-based urethane prepolymer was synthesized by adding 0.5mol of PDMS with 0.15wt% stannous octoate dropwise to 1.0 mol of MDI over a period of 1 h while maintaining the reaction temperature at 65°C under a nitrogen atmosphere. Completion of the reaction was confirmed by i.r. for the disappearance of the absorption peak of the OH group.

General procedure for the preparation of TPU-modified CNE

To a 1 litre four-necked round-bottomed flask, equipped with a CaCl₂ drying tube, heating mantle, N_2 inlet, stirrer, thermocouple and temperature controller, was added 400 g of o-cresol-formaldehyde novolac epoxy resin (CNE). The epoxy resin was heated to 150°C and then vigorously stirred and dehydrated under vacuum $\left($ <10 mmHg) until the water content was less than 0.01% (measured by Karl Fischer). The reaction temperature was then raised to 160° C, and 350 ppm (based on CNE) of 2-phenylimidazole was added. To the stirring CNE was added 57.2 g of urethane or PDMSbased urethane prepolymer. The reaction temperature was held at 160°C for 2h. Completion of the epoxideisocyanate reaction was confirmed by infra-red spectroscopy for the disappearance of the NCO group absorption. The resulting TPU- or polysiloxane TPU-modified epoxy resin contained ca. 12.5 wt% dispersed rubber and had an *EEW* of ca. 230.

Curing procedure of epoxy resins

Various rubber-modified epoxy resins were mixed with a stoichiometric amount of curing agent and Ph_3P in a mill at moderate temperature to give a thermosettable epoxy resin powder. The resin powder was cured in a mould at a temperature of 150° C and 50 kg cm^{-2} for a period of 1 h and then post-cured at 180°C for 2h and 210° C for 3 h to obtain a cured specimen.

Measurement and testing

Infra-red spectra were recorded with a Perkin-Elmer 16PC *FTi.r.* spectrophotometer operated with a dry air purge. Signals of four scans at a resolution of 4 cm^{-1} were averaged before Fourier transformation. The *EEW* of epoxy resins were determined by the $HClO₄/potentio$ metric titration method. Dynamic viscoelastic properties were studied on a Rheometrics RDA-II rheometer between -150 and 250° C, with a heating rate of 5 or 2° C/step at a frequency of 1 Hz. The rectangular torsion mode was chosen and the dimensions of the specimen were 51 mm \times 12.7 mm \times 0.76 mm. The storage modulus G' and tan δ were determined. 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. The flexural properties of cured resins were obtained 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 three-point loading system was chosen and rectangular bar specimens, $80 \text{ mm} \times 10 \text{ mm} \times 4 \text{ mm}$, were moulded directly by a transfer moulding process. The coefficient of thermal expansion *(CTE)* was measured with a DuPont 943 thermal mechanical analyser in accordance with ASTM E831-86. A specimen 4 mm in length was used at a heating rate of 5° Cmin⁻¹. Normally, the thermal expansion increases with increase in temperature and the *CTE* values 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 pre-weighed 3 mm thick \times 50 mm diameter cured discs in boiling water for 100 h. The discs were removed and were cooled to ambient temperature and then wiped dry and weighed to determine any weight gain. A thermal shock cycling test was carried out by the following procedure²¹: 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 moulding process and subjected to a thermal cycling test. A cycle consisted of -65° C \times 15 min and 150° C \times 15 min. The devices were inspected by an optical microscope for cracks after 250, 500, 750, 1000, 1500, 2000, 2500, 3000, 3500, 4000 and 4500 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, is plotted.

RESULTS AND DISCUSSION

The incorporation of TPU or polysiloxane TPU as a soft segment into an epoxy resin matrix can be achieved in a two-step reaction. The first step involves the synthesis of urethane prepolymer or PDMS-based urethane prepolymer are shown in *Schemes 1* and 3. The equation for the first step indicates the formation of an isocyanateterminated soft segment, which is then grafted onto epoxy resin in the second step via epoxy ring opening in the presence of a catalyst (2-phenylimidazole) to form an oxazolidone to give a stable dispersion of TPU particles in an epoxy resin matrix as shown in *Schemes 2* and 4. The resulting TPU-modified CNEs are multifunctional epoxy resins, which show similar reactivity to other epoxy resins and can be cured with various curing agents.

Infra-red spectroscopy

Typical *FTi.r.* survey spectra of PDMS-diol (X22- 160AS), PDMS-based urethane prepolymer and polysiloxane TPU-modified CNE are shown in *Figure 1.* The completion of isocyanate-terminated prepolymer formation was confirmed by the disappearance of the hydroxyl group absorption peak at 3500 cm^{-1} and the appearance of the absorption peak of urethane at 3316 cm^{-1} (N-H) and 1736 cm^{-1} (C=O). The resulting isocyanateterminated urethane prepolymer was then grafted onto epoxy resin via epoxy ring opening in the presence of a catalyst (2-phenylimidazole) at 160°C to form an oxazolidone. The completion of this reaction was confirmed by the disappearance of the -NCO group absorption peak at $2250-2270 \text{ cm}^{-1}$. The trimerization of-NCO to form an isocyanurate, a side reaction that can take place simultaneously with oxazolidone formation, is known to be favoured at a low reaction

Scheme 1 The synthesis of urethane prepolymer

temperature. However, at a reaction temperature of 160°C and with 2-phenylimidazole catalyst, the oxazolidone formation is predominant. A typical i.r. spectrum of polysiloxane TPU-modified CNE is given in *Figure lc,* which shows the i.r. characteristic absorption peak of oxazolidone at 1756 cm^{-1} and without any absorption peak of isocyanurate at $1700-1710 \text{ cm}^{-1}$.

Dynamic viscoelastic properties

The dynamic viscoelastic spectra are shown in *Figures*

2, 3 and 4. The sample designations correspond to those in *Table 1.* Dynamic viscoelastic analysis can give information on the microstructure of cured rubbermodified epoxy resins. *Figure 2* shows the storage modulus G' and tan δ curves for the resins modified with various TPU particles, and *Figure 4* shows those for cured polysiloxane TPU-modified CNE. Storage moduli decreased with modification for all samples. The tan δ curves for the control epoxy network exhibit two major relaxations observed in most epoxy polymers²²: the high-

Scheme 2 The preparation of TPU-modified CNE via epoxide–isocyanate reaction

MDI-PDMS urethane prepolymer

Where

Scheme 4 The preparation of polysiloxane TPU-modified CNE via epoxide-isocyanate reaction

Figure 1 *FTi.r.* spectra of (A) polydimethylsiloxane- α , ω -diol (X-22-160AS), (B) PDMS-based urethane prepolymer and (C) polysiloxane TPU-modified CNE

Figure 2 Dynamic viscoelastic analysis for the control and various polyol TPU-modified epoxy resins (no filler). The curing agent is phenol-formaldehyde novolac (HRJ-2210)

temperature or α transition corresponds to the major $T_{\rm g}$ of the cured epoxy resins, above which significant chain motion takes place; the low-temperature or β transition is attributed predominantly to the motion of the $CH₂$ - $CH(OH) - CH₂-O$ (hydroxy ether) group of the epoxy. *Figure 2* shows that the α -relaxation peak in the tan

Figure 3 Low-temperature transitions for the control and various polyol TPU-modified epoxy resins (no filler). The curing agent is phenol-formaldehyde novolac (HRJ-2210)

Figure 4 Dynamic viscoelastic analysis for the control and various polysiloxane TPU-modified epoxy resins (no filler). The curing agent is phenol-formaldehyde novolac (HRJ-2210)

 δ curve became broader with PEG or PPG modification, but the peak position and the magnitude of the α -relaxation peak hardly changed with respect to the control resin. However, the peak position of the α relaxation peak in the tan δ curve for the resin modified with PTMG shifted markedly towards higher temperature. This result can be attributed to the solubility between the epoxy resin and the TPU particles. The solubility of TPU particles in the epoxy resin decreases with increasing carbon number in the polyol, and this lowering in solubility has resulted in complete phase separation. The β -relaxation peak in the tan δ curves at ca. -100 to -50° C was observed without an additional peak for TPU. These results can be explained in that the T_g values of the pure TPUs are around the β relaxation peak of the cured epoxy resin. For the sake of separating the TPU's T_g from the β -relaxation peak of the cured epoxy resin, the heating rate of the rheometer was decreased from 5°C to 2°C/step. The

Table 2 Dynamic viscoelastic properties of cured neat rubbermodified epoxy resin system

Sample designation	T_g^a (°C) matrix	Shear modulus ^b at 20° C (\times 10 ⁹ dyn cm ⁻²)	T_g^c (°C) rubber
Control ^{d}	204	16.0	
PEG	204	9.1	-67
PPG	206	9.1	-68
PTMG	217	9.0	-69
S1810	220	9.0	-119
S ₁₀₀₂	218	13.5	-113

^a Peak of tan δ at higher temperature

^b Units: 10^9 dyncm⁻² $\equiv 0.1$ GPa

Peak of tan δ at lower temperature

 d Control: unmodified CNE

results are shown in *Figure 3* in which a double peak was observed markedly for the resins modified with TPU. *Figure 4* shows that the peak position of the α relaxation peak in the tan δ curves for the resins modified with polysiloxane TPU shifted markedly towards higher temperature. This result can be attributed not only to complete phase separation of polysiloxane TPU particles but also to the formation of oxazolidone structure via epoxy ring opening with the isocyanate groups of urethane. In addition to α and β peaks, these curves all show an additional markedly small peak from -130 to -105 °C with a centre near -120° C corresponding to the T_g of the polysiloxane phase. This small peak further supports the multiphase separation. The dynamic viscoelastic properties of the cured rubber-modified epoxy resin systems including the major T_g and the rubber's T_g are shown in *Table 2.*

Morphology

SEM photomicrographs of freeze fractured surfaces for the control and five TPU-modified resins are given in *Figure* 5. A phase-separated structure is observed in all

Figure 5 Morphology of cold-snap fracture surfaces of cured epoxy resins modified with various TPU particles (12.5 wt%): (A) unmodified; (B) PEG; (C) PPG; (D) PTMG; (E) S1810; (F) S1002

Table 3 Typical encapsulating formulation

Ingredients	Amount $(wt \ \%)$
Cresol epoxy novolac	17.5
Phenolic hardener (HRJ-2210)	9.1
Brominated epoxy resin	2.5
Fused silica (GP-71 Harbison-Walker)	68.5
Mould release (Hoechst Wax OP&E)	04
Carbon black	0.4
Silane coupling agent (DC Z-6040)	04
Antimony trioxide	1 ₀
Triphenylphosphine accelerator	0.2

Procedure

1. B-Stage melt mix at 95'C for approx. 7 min

2. Cool and grind to a uniform powder
3. Transfer moulding of nowder at 175°

Transfer moulding of powder at 175° C for 90 s

4. Post-cure at 175°C for 4h

Figure 6 Coefficient of thermal expansion *(CTE)* for various TPUmodified epoxy resin encapsulants

rubber-modified resins. No matter whether it is TPU or polysiloxane TPU modification, the spherical shape of the rubber particles is quite the same. The sizes of rubber particle are $0.2-1 \mu m$ and slightly proportional to the *MW* and the structure of the TPU modifiers. The polysiloxane TPU particles have slightly larger size than the polyol TPU particles. These can be attributed to the compatibility of TPU and CNE.

Encapsulation Jormulation

A control resin and CNE modified with various TPU rubbers or silicone powder were formulated into seven electronic encapsulating formulations. The formulations were each cured at 175°C for 4h. The details of the encapsulating formulations are given in *Table 3.* The thermal mechanical properties of the cured encapsulating formulations were determined by the following tests.

Thermal mechanical properties

Figure 6 shows the thermal mechanical properties. The *CTE* in the glassy state below the $T_{\rm g}$ was taken from 60 to 100 $^{\circ}$ C and the *CTE* above the T_g was taken from 200 to 240 $^{\circ}$ C. For the *CTE* below T_{g} , all rubber-modified

encapsulants have *CTE* approximately equal to or slightly lower than that of the unmodified resin and this will result in a small difference in *CTEs* between encapsulant and silicon chip.

 T_{g} values were determined from the tangents of the *CTE* as a function of temperature at 100 and 200°C. The results are shown in *Figure 7*. The T_g are slightly lower for polyol TPU modification, while T_g are slightly higher for polysiloxane TPU modification compared to the unmodified encapsulant. This observation is consistent with the viscoelastic investigation.

Flexural test properties

Figure 8 shows the result of the flexural test. The flexural moduli of the cured encapsulants were reduced markedly no matter whether it is modified with polyol TPU or polysiloxane TPU. The flexural strengths of polysiloxane TPU-modified CNE were approximately equal to or slightly larger than that of the control resin. However, the flexural strength was significantly reduced with silicone powder modification. This result

Figure 7 Glass transition temperature for various TPU-modified epoxy resin encapsulants

Figure 8 Flexural test properties for various TPU-modified epoxy resin encapsulants

Figure 9 Comparison of stress for various TPU-modified epoxy resin encapsulants

Figure 10 Moisture absorption for various TPU-modified epoxy resin encapsulants

indicates that physical blending, although it improves the toughness of cured epoxy resin, lowers its strength.

Internal stress

The internal stress of IC devices encapsulated by EMCs is closely related to the product of flexural modulus (E_{r1}) and thermal expansion coefficient (α_{r1}) below the T_g of the cured EMCs²⁰. *Figure 9* shows the internal stress of the control and various rubbermodified encapsulants. The polysiloxane TPU-modified EMC (S1810) and TPU-modified EMC (PTMG) have a lower stress than that of the control.

Moisture absorption

Absorbed moisture in the package was found not only to plasticize the epoxy resin, causing a lowering of the $T_{\rm g}$ and in turn affecting mechanical response, but also to cause package cracking²³. This crack (so-called popcorn phenomenon) is caused by evaporation and expansion of absorbed moisture in the package at the temperature of reflow soldering (215 to 260°C) for mounting on printed

Figure 11 Thermal shock cycling test results. Temperature cycle: -165° C × 15 min and 150°C × 15 min

circuits²⁴. The moisture absorption results are given in *Figure 10.* The S1810 encapsulant consisting of polysiloxane moiety absorbed the least moisture.

The polysiloxane TPU-modified EMC (S1810) and TPU-modified EMC (PTMG) have not only a lower stress but also a higher T_g than that of the control. Moreover, S1810 absorbs the least moisture. Thus they were chosen to encapsulate the semiconductor devices. The encapsulated devices were subjected to a thermal shock cycling test in comparison with the control resin.

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, 4000 and 4500 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 *Figure 11.* The result indicates that, for 50% of the devices, failure happened after 3500 cycles for the S1810 modified EMC and after 2500 cycles for the PTMGmodified CNE compared to after 750 cycles for the control resin. S1810-modified EMC has much better thermal shock resistance resulting from its lower stress, higher T_g , and lower moisture absorption than the others.

CONCLUSION

A process was developed to incorporate stable dispersed TPU or polysiloxane TPU particles in an epoxy resin matrix that not only greatly reduces the stress of cured EMCs but also increases the T_g of the resultant EMC. Reduction in internal stress of the encapsulant was greatly affected by the structure and molecular weight of the TPU. Electronic devices encapsulated with the polysiloxane TPU-modified EMCs have exhibited excellent resistance to the thermal shock cycling test and have resulted in an extended device used life.

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