Modification of Epoxy Resins by Hydrosilation for Electronic Encapsulation Application

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SYNOPSIS

Hydride terminated polydimethyl siloxanes were used to reduce the stress of cresol–formaldehyde novolac epoxy resin cured with phenolic novolac resin for electronic encapsulation application. The effects of the structure and molecular weight of the hydride terminated polydimethyl siloxane in reducing the stress of encapsulant were investigated. The mechanical and dynamic viscoelastic properties and morphologies of rubber modified epoxy networks were also studied. A "sea-island" structure ("islands" of silicone rubber dispersed in the "sea" of an epoxy resin) was observed via SEM. The dispersed silicone rubbers effectively reduce the stress of cured epoxy resins 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 use life for the devices. © 1994 John Wiley & Sons, Inc.

INTRODUCTION

Organosiloxane polymers are known for their excellent thermal and thermooxidative stabilities, very low glass-transition temperature (T_g, $-123°C$), moisture resistance, good electric properties, and low stress.[1] 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 initiated.

Because of its excellent heat, moisture, solvent, and chemical resistance, superior electrical and mechanical properties, and good adhesion to many substrates, o-cresol-formaldehyde novolac epoxy (CNE) is the resin typically employed in the encapsulation of microelectronic devices. Upon cure, this multifunctional epoxy resin provides a densely cross-linked protective layer; however, it is relatively brittle.

The scale of large scale integrated circuits (LSICs) is continuing upward, forcing the design of large chips and finer patterns that are more susceptible to internal stress failure. The prevailing surface mount technology also generates heat stress to devices.[2,3] Internal stress causes package cracking, passivation layer cracking, aluminum pattern deformation, etc.[4,5] Therefore, the development of low-stress epoxy molding compounds (EMCs) is required for high-reliability semiconductor devices. The sources of internal stress with plastic encapsulants are considered to be shrinkage due to the curing process and differential shrinkages between the device and the resin. In the case of epoxy resin encapsulation, the first source of shrinkage is relatively minor, whereas the second one, which is the difference of thermal expansion coefficients, is the dominant effect. The thermal stress as produced by the differences between thermal expansion coefficients is expressed in the following equation[6]:

$$S = K \int (\alpha_r - \alpha_s) E_r dT$$

where S is thermal stress; K, a constant; α_r, the thermal expansion coefficient of the resin; α_s, the thermal expansion coefficient of the silicon chip, and E_r, the flexural modulus of the resin. Reductions of thermal stress by lowering of either the thermal expansion coefficient or the flexural modulus of the encapsulant have been reported.[7–29] Increasing the amount of silica filler used in an encapsulant effec-

tively 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 moldability. The lowering of the elastic modulus by modification with a rubber in a "sea-island" structure (consisting of "islands" of rubber particles dispersed in the "sea" of an epoxy matrix) is considered to be more desirable than a one-phase structure in view of the thermal property needs.[6,30–32] Traditional modifiers include functionalized rubber such as carboxyl terminated butadiene–acrylonitrile copolymers (CTBN). When CTBN modifiers are incorporated into resin compositions, a two-phase morphology consisting of relatively small ($\sim$ 0.1–5 μm) rubber particles dispersed in a resin matrix is generated that toughens epoxy resins.[33–36] However, phase separation depends upon the formulation, processing, and curing conditions.[37–41] Incomplete phase separation can result in a significant lowering of the glass-transition temperature (T_g). Moreover, the rubber phase that separates during cure is difficult to control and may result in uneven particle size. The differences in the morphology and volume of the separated phase affect the mechanical performance of the product. To minimize these problems, the preformation of a stable dispersion of rubbery particles in the uncured epoxy resin via hydro-silation of epoxy resin using hydride terminated siloxane oligomers was investigated. The devices encapsulated by the hydride siloxane modified EMC were also evaluated by the thermal shock cycling test.

EXPERIMENTAL

Materials

All reagents and solvents were reagent grade or were purified by standard methods before use. The control epoxy resin was CNE (Quatrex 3330, Dow Chemical Co., epoxy equivalent weight, EEW, 192). A phenol–formaldehyde novolac resin was used as curing agent with an average hydroxyl functionality of 6 and a hydroxyl equivalent weight of about 104 (Schenectady Chemical, HRJ-2210). The hydride terminated siloxanes were purchased from HULS America Inc. or Shin–Etsu Chemical Co.; their structure and molecular weight (M_w) are shown in Table I. 2-Allyl-phenyl (2-AP) was purchased from Aldrich Chemical Co. The A-1 catalyst was (ethyl)triphenyl phosphonium acetate acetic acid complex. Ph$_3$P was triphenylphosphine that was used as curing accelerator. H$_2$PtCl$_6$ (5 wt % in isopropanol) was used as a catalyst in hydrosilation.

Table I   Hydride Terminated Siloxanes Employed for Modification of CNE

Abbrev.	Structural Formula	M_w	Supplier
HS1		400	HULS
HS2		17500	HULS
HS3		28000	HULS
HS4		62000	HULS

$$H-\underset{\underset{Me}{|}}{\overset{\overset{Me}{|}}{Si}}-O-(-\underset{\underset{Me}{|}}{\overset{\overset{Me}{|}}{Si}}-O-)_n-\underset{\underset{Me}{|}}{\overset{\overset{Me}{|}}{Si}}-H$$

| HS5 | | 15286 | Shin–Etsu |

$$H-\underset{\underset{Me}{|}}{\overset{\overset{Me}{|}}{Si}}-O-(-\underset{\underset{Me}{|}}{\overset{\overset{H}{|}}{Si}}-O-)_m-(-\underset{\underset{Me}{|}}{\overset{\overset{Me}{|}}{Si}}-O-)_n-\underset{\underset{Me}{|}}{\overset{\overset{Me}{|}}{Si}}-H$$

$m = 5.7, n = 12.3$

$$Me-\underset{\underset{Me}{|}}{\overset{\overset{Me}{|}}{Si}}-O-(-\underset{\underset{Me}{|}}{\overset{\overset{H}{|}}{Si}}-O-)_m-(-\underset{\underset{Me}{|}}{\overset{\overset{Me}{|}}{Si}}-O-)_n-\underset{\underset{Me}{|}}{\overset{\overset{Me}{|}}{Si}}-Me$$

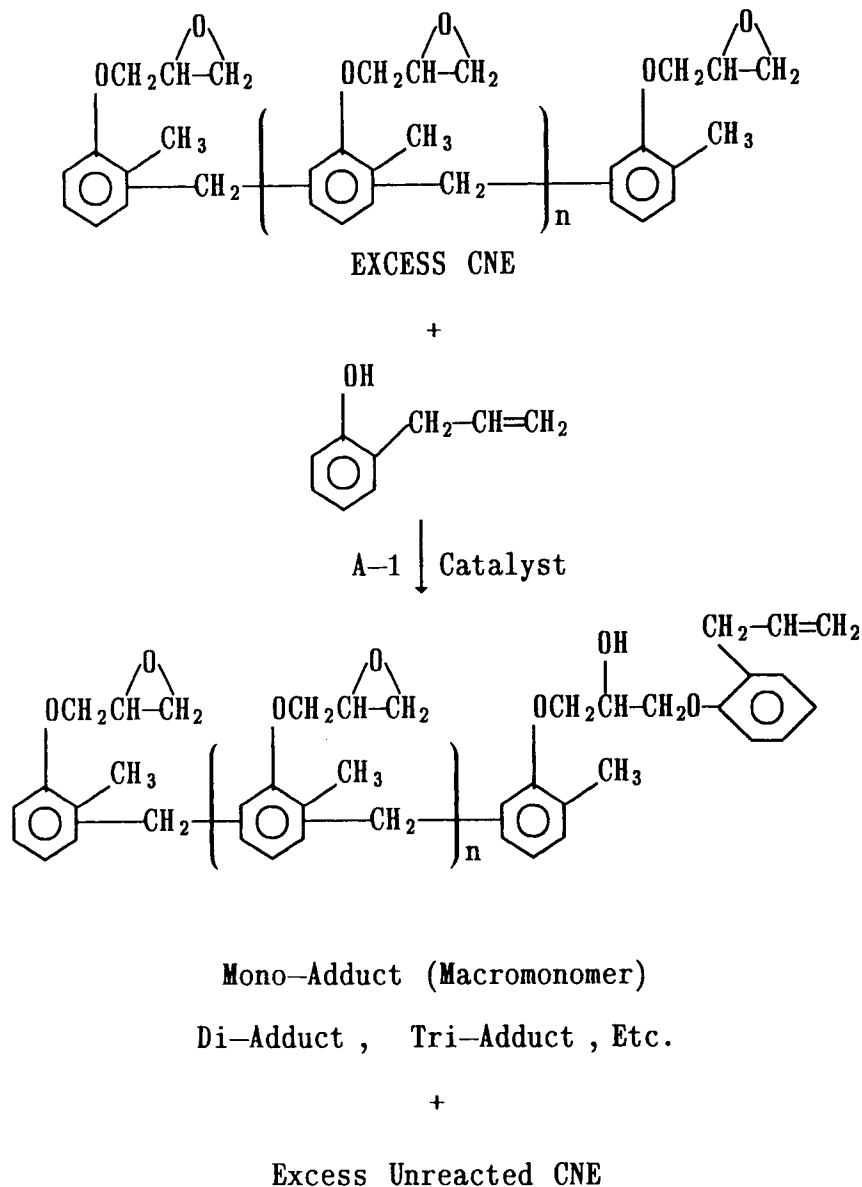| HS6 | $m = 38, n = 10$ | 3182 | Shin–Etsu |
| HS7 | $m = 18, n = 6.7$ | 2722 | Shin–Etsu |
| HS8 | $m = 30, n = 54.7$ | 6000 | Shin–Etsu |

Figure 1 Vinylization of an epoxy resin to form a macromonomer.

General Procedure for Preparation of Silicone Rubber Modified Epoxy Resin

CNE, 384 g, was added to a four-neck round-bottom flask, equipped with a heating mantle, stirrer, reflux condenser, thermocouple, and temperature controller. The epoxy resin was heated to 120°C, and then vigorously stirred and dehydrated under vacuum (< 10 mmHg) until the water content was less than 0.01% (measured by Karl Fischer). The mixture of 2-AP (5.5 g) and A-1 catalyst (0.8 g) was added dropwise via a metering pump over a period of 30 min while maintaining the reaction temperature at 120°C. After the completion of 2-AP addition, the reaction temperature was maintained at 120°C for an additional 1.5 h. Completion of the reaction was confirmed by GC for the disappearance of the reactant, 2-AP, in the reaction mixture. The resultant epoxy resin thus had a radically polymerizable double bond in the molecule. The above resultant epoxy resins, 400 mL of toluene, and 40 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 was 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
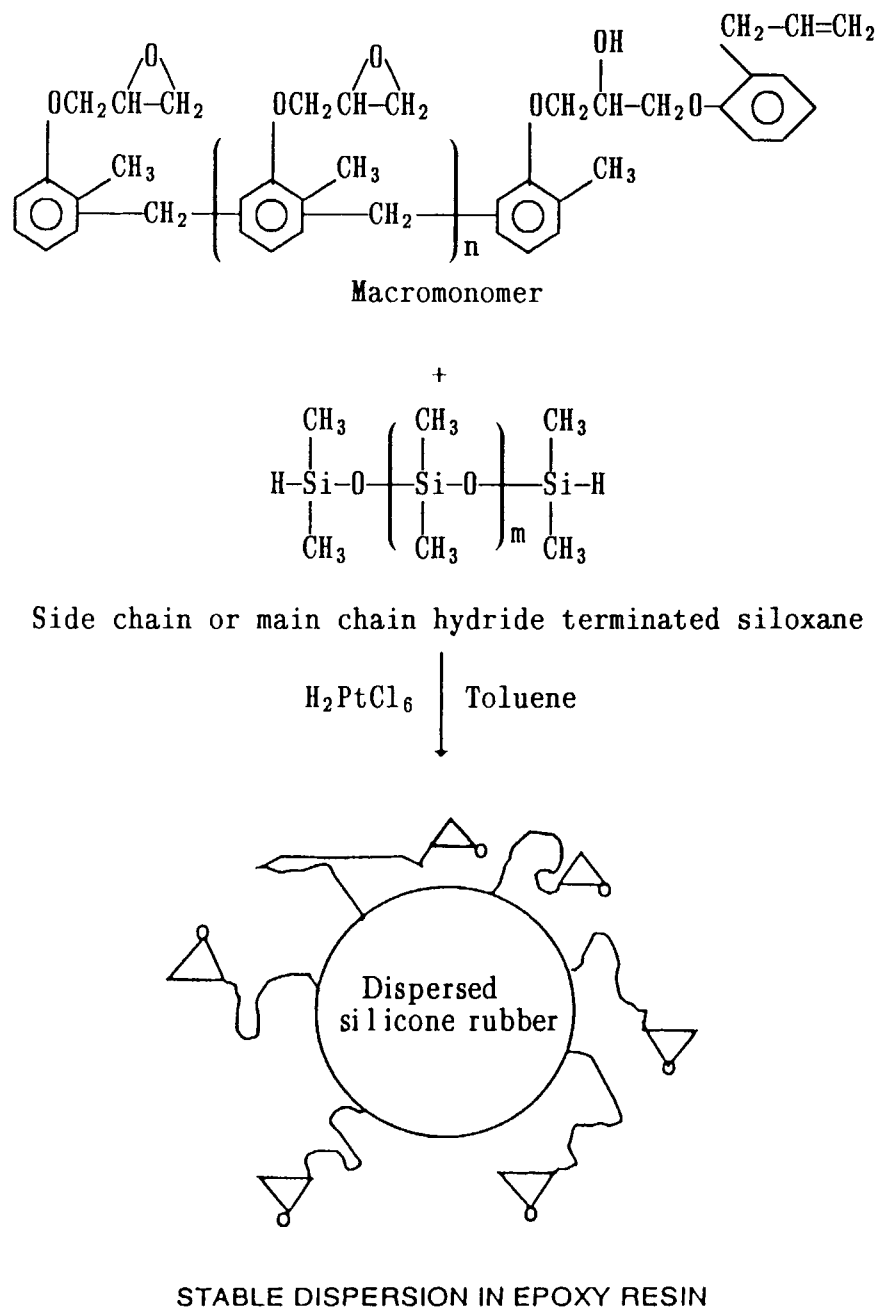
CH_2-CH=CH_2

OH

O

O

OCH_2CH-CH_2 OCH_2CH-CH_2 OCH_2CH-CH_2O

CH_3 CH_3 CH_3

-CH_2- [-CH_2-]$_n$

Macromonomer

+

CH_3 CH_3 CH_3

H-Si-O- [Si-O-]$_m$ Si-H

CH_3 CH_3 CH_3

Side chain or main chain hydride terminated siloxane

H_2PtCl_6 | Toluene

Dispersed silicone rubber

STABLE DISPERSION IN EPOXY RESIN

Figure 2 Macromonomer and hydride siloxane via "hydrosilation" to form a dispersion.

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 150°C under a full vacuum for 2 h to remove solvents. Upon cooling to room temperature, a creamy silicone modified epoxy resin was obtained. The resulting epoxy novolac contained ca. 10 wt % dispersed silicone rubber and had an EEW of ca. 230.

Curing Procedure of Epoxy Resins

Various silicone rubber modified epoxy resins were mixed with a stoichiometric amount of curing agent and Ph$_3$P in a mill at moderate temperature to give a thermosettable epoxy resin powder. The resin
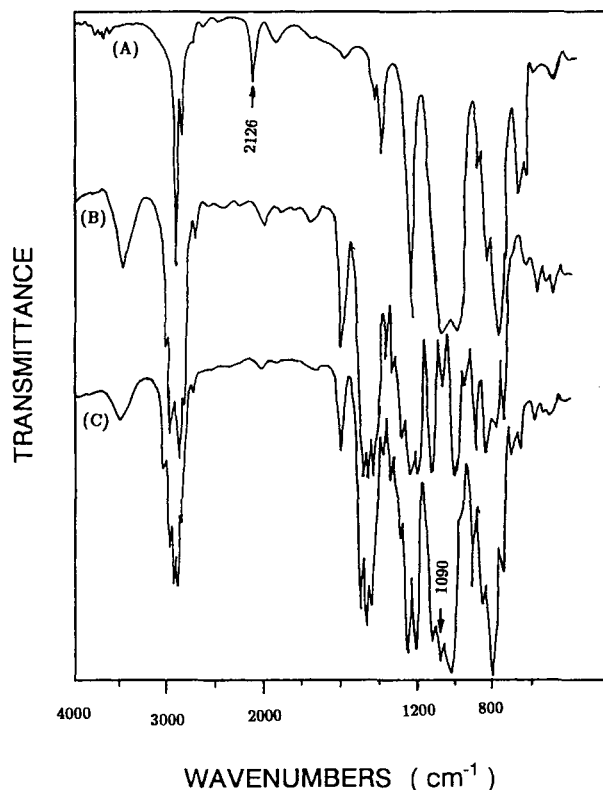
Figure 3 FTIR spectra of (A) hydride siloxane (HS2); (B) unmodified CNE; (C) HS2 modified CNE.

powder was cured in a mold at 150°C and pressure of 50 kg/cm² for a period of 1 h, and then postcured at 180°C for 2 h and 210°C for 3 h to obtain a cured specimen.

Measurement and Testing

Infrared spectra were recorded with a Perkin–Elmer 16PC FTIR spectrophotometer operated with a dry air purge. Signals of four scans at a resolution of 4 cm⁻¹ were averaged before Fourier transformation. All IR spectra covered the range 400–4000 cm⁻¹. EEW of rubber modified epoxy resin was determined by the $HClO_4$/potentiometric titration method. Dynamic viscoelastic properties were performed on a Rheometrics RDS-2 rheometer between −150 and 250°C, with a ramp rate of 5°C/step at a frequency of 1 Hz. The rectangular torsion mode was chosen and the dimensions of the specimen were 51 (L) × 12.7 (W) × 0.76 (T) mm³. The storage modulus G' and tan δ were determined. The peak of tan δ was identified as the T_g because a large decrease in G' occurred at this point. The JEOL JSM-6400 scan-

ning 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 with a Shimadzu AGS-500 universal testing machine. Flexural strength and modulus were obtained at a crosshead speed of 2 mm/min according to ASTM D790-86. A three-point loading system was chosen and the rectangular bar specimens, 80 (L) × 10 (W) × 4 (T) mm³, were molded directly by a transfer molding process. The coefficient of thermal expansion CTE was measured with a DuPont 943 thermal mechanical analyzer (TMA) in accordance with ASTM E831-86. A specimen 4-mm in length was used at a heating rate of 5°C/min. 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. A thermal shock cycling test was carried out by the following procedure[42,43]: 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 then 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, 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.

Table II T_gs of Control and Various Hydride Siloxane Modified Epoxy Resins Cured with Phenolic Novolac

Modifier	T_g^a (°C)	T_g^b (°C)
Control	—	202
HS1	−108	192
HS2	−111	201
HS3	−109	199
HS4	−112	201
HS6	−114	203
HS7	−112	201
HS8	−112	202

Control: unmodified CNE.
ª Peak of tan δ at lower temperature.
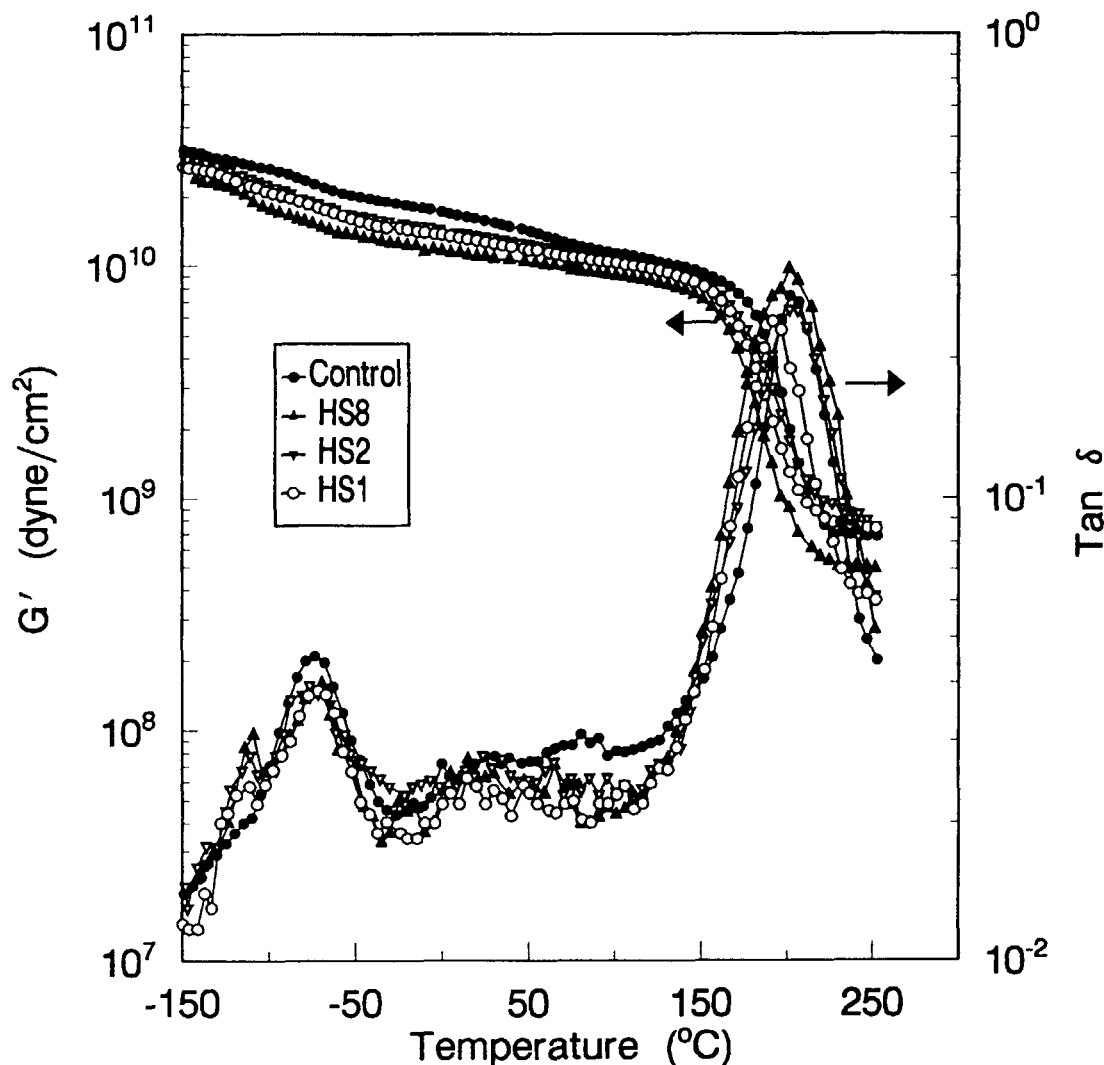ᵇ Peak of tan δ at higher temperature.

Figure 4   Dynamic viscoelastic analysis for various hydride siloxane modified epoxy resins (no filler). The curing agent is phenol–formaldehyde novolac (HRJ-2210).

RESULTS AND DISCUSSION

Characteristic of Siloxane Modified Epoxy Resins

Dispersed silicone rubbers in epoxy resins can be prepared in two steps. The first step involves the vinylization of part of the epoxy groups in the CNE with 2-allylphenol to form a reactive macromonomer as shown in Figure 1. The second step is the hydrosilation of the resultant macromonomer with various hydride terminated siloxanes 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 shown in Figure 2. The completion of hydrosilation is observed by the

FTIR spectrum shown in Figure 3. 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 broad Si—O—Si peak at about 1090 cm^{-1}. It is worth noting that gellation takes place in the case of the HS5 modifier. Gellation results from too many Si—H functional groups in HS5 causing the formation of a cross-linked macromolecule.

Dynamic Viscoelastic Analysis of Rubber Modified Resins

Dynamic viscoelastic analysis can give information on the microstructure of cured rubber modified

epoxy resins. The tan δ curves for the control epoxy network exhibit the two major relaxations observed in most epoxy polymers[44]: a high-temperature or α transition corresponds to the major T_g of the cured epoxy resin above which significant chain motion takes place; the low temperature or β transition is attributed predominantly to the motion of the $CH_2 — CH(OH) — CH_2 — O$ (hydroxyether) group of the epoxy. Figure 4 shows the storage modulus G' and tan δ curves for the resins modified with various silicone rubbers. The storage modulus decreased with modification. In addition to the epoxy α and β peaks, these curves show an additional markedly small peak from -130 to $-100°C$ with a center near $-110°C$ corresponding to the T_g of the siloxane phase. This small peak further supports the multiphase separation. It is interesting to note that the magnitude of the siloxane peaks in the tan δ curves for the side chain hydride terminated siloxane (HS8) modified product is higher than the main chain hydride terminated siloxane modified product; however, the peak positions are unchanged. It can be attributed to a more complete phase separation of side chain hydride siloxane rubber particles from the matrix resin than for the main chain terminated hydride siloxane modified systems. In addition, the α-relaxation peak in the tan δ curve became broader but the peak position hardly changed except for HS1 modification with respect to the control resin. The major T_g and the rubber T_g of the cured epoxy resins are shown in Table II, wherein the major T_g's of the

Table III Typical Encapsulating Formulation

Ingredients	Weight (%)
Cresol epoxy novolac	17.5
Phenolic hardener (HRJ-2210)	9.1
Brominated epoxy resin	2.5
Fused silica (GP-71 Harbison–Walker)	68.5
Mold release (Hoechst Wax OP&E)	0.4
Carbon black	0.4
Silane coupling agent (DC Z-6040)	0.4
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
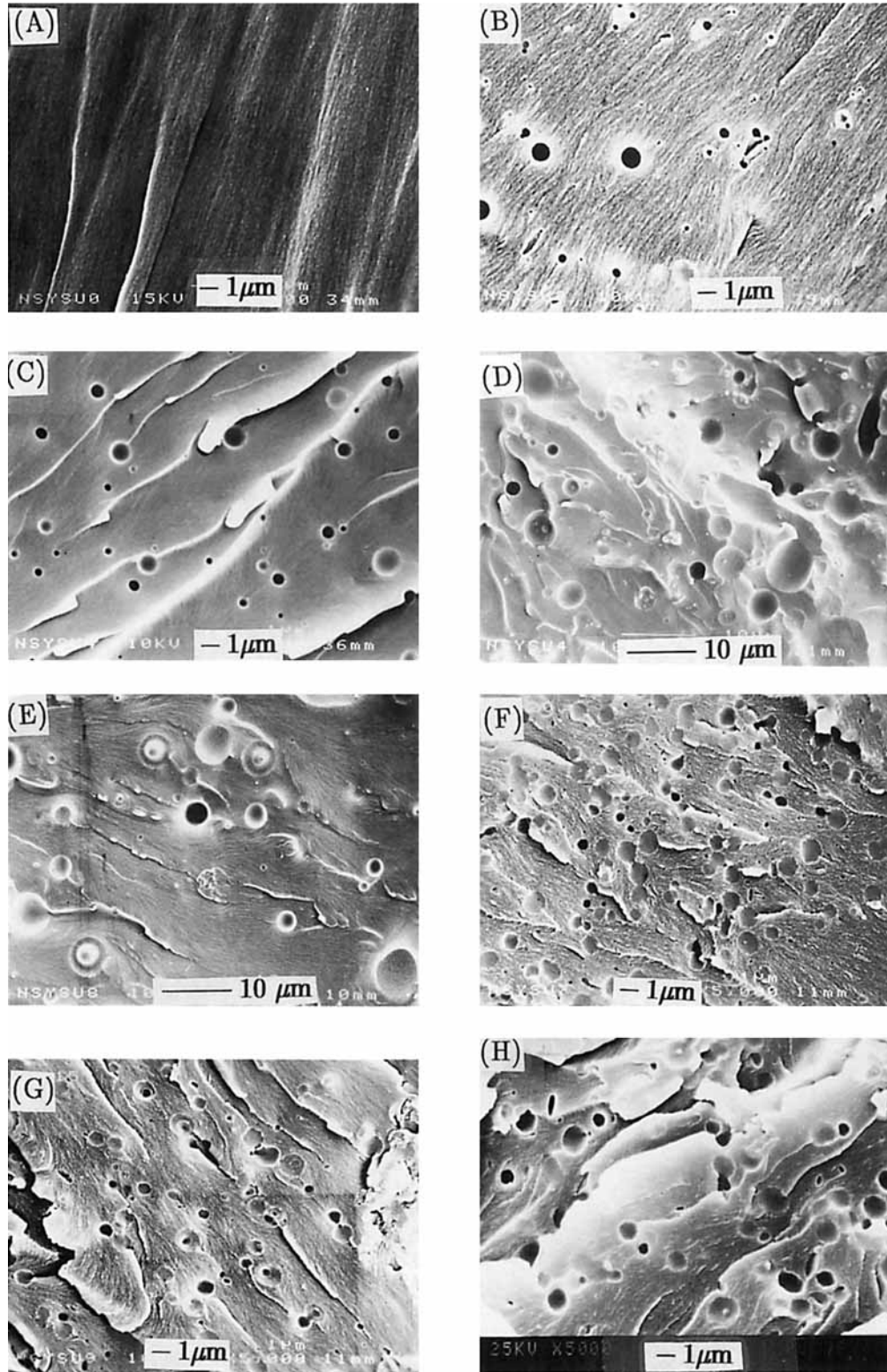3. Transfer molding of powder at 175°C for 90 s
4. Postcure at 175°C for 4 h

cured rubber modified resin are equal to or slightly lower than the unmodified resin no matter whether it was modified with side chain or main chain hydride terminated siloxanes. The HS1 modified resin especially has the lowest T_g. This result can be explained as: incomplete phase separation of rubber and/or a small amount of unreacted hydride siloxane remained in the resin and acted as a plasticizer. Incomplete phase separation could result from the higher solubility of low M_w HS1 (M_w 400) in the matrix resin.

Morphology

SEM photomicrographs of cold snap surfaces for the control and seven hydride siloxane modified resins are given in Figure 5. A sea-island structure is observed in all rubber modified resins. No matter whether it is side chain or terminal hydride siloxane modification, the spherical shape of rubber particles are quite the same. The sizes of rubber particle are 0.5–5 μm and slightly proportional to the M_w of the siloxane modifiers. These can be attributed to the compatibility of siloxane and CNE.

Encapsulation Formulation

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

CTE

Figure 6 shows the thermal mechanical properties. The CTE in the glassy state below the T_g was taken from 60 to 100°C and the CTE above the T_g was taken from 200 to 240°C. For the CTE below the T_g, all rubber modified encapsulants have slightly lower CTE than the unmodified resin and this will result in a small difference in CTEs between encapsulant and silicon chip.

T_g

T_gs were determined from tangents of the CTE as a function of temperature at 100 and 200°C. The results are shown in Figure 7. The T_g of cured CNE is relatively unaffected by modification with the side

Figure 5   Morphology of cold-snap fracture surfaces of cured epoxy resins modified with various hydride siloxanes (10 wt %). (A) unmodified; (B) HS1; (C) HS2; (D) HS3; (E) HS4; (F) HS6; (G) HS7; (H) HS8.
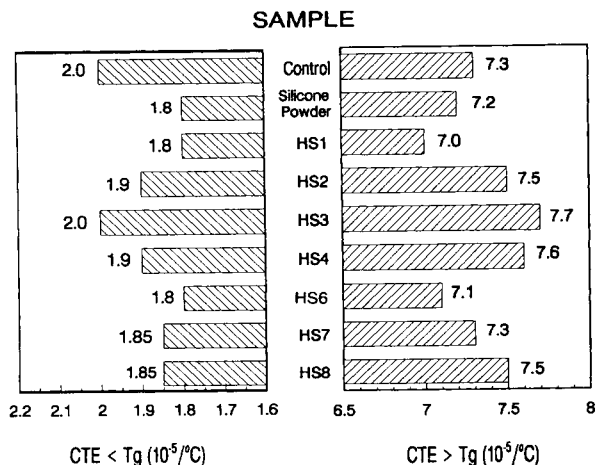
SAMPLE

Figure 6 Coefficient of thermal expansion (CTE) for various hydride siloxane modified epoxy resin encapsulants.

SAMPLE

Figure 8 Flexural test properties for various hydride siloxane modified epoxy resin encapsulants.

chain hydride terminated siloxanes (HS5, HS7, HS8). However, the HS1 modified resin has a significantly lower T_g and this may result from the greater solubility of HS1 modifier in the matrix resin than for the others. 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 resins were reduced markedly no matter whether modified with silicone powder or various hydride siloxanes. The flexural strengths of hydride siloxane-modified CNE were approximately equal to or slightly larger than that of the control resin, however, the flexural strength was significant reduced with the silicone powder
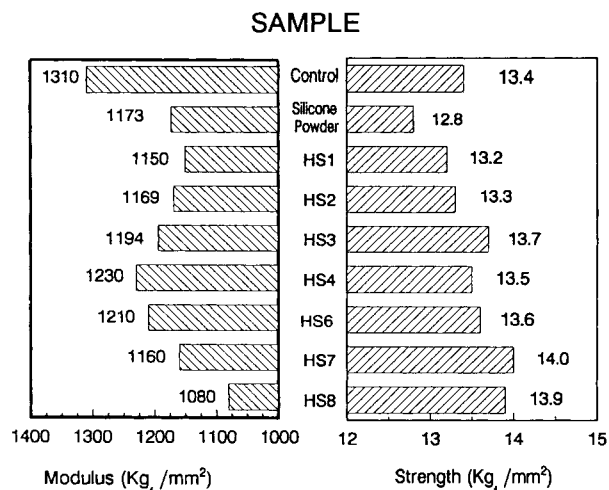
modification. This result can be explained by the fact that physical blending, although it effectively improves the toughness of cured epoxy resin, lowers it's strength.

Internal Stress

Internal stress of the cured encapsulants can be approximated by the product of flexural modulus and the CTE below the T_g ($E \cdot \alpha_{r1}$). Figure 9 shows the stress of the control and various rubber modified encapsulants. The dispersed silicone rubber particles effectively reduce the flexural modulus and the CTE but the reduction is the greatest with HS8 modification. This can be attributed to the complete separation of the rubber phase from epoxy matrix which acts as a stress reliever.
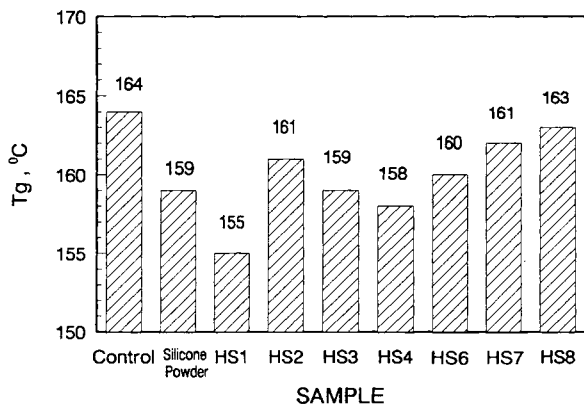

Figure 7 Glass-transition temperature for various hydride siloxane modified epoxy resin encapsulants.
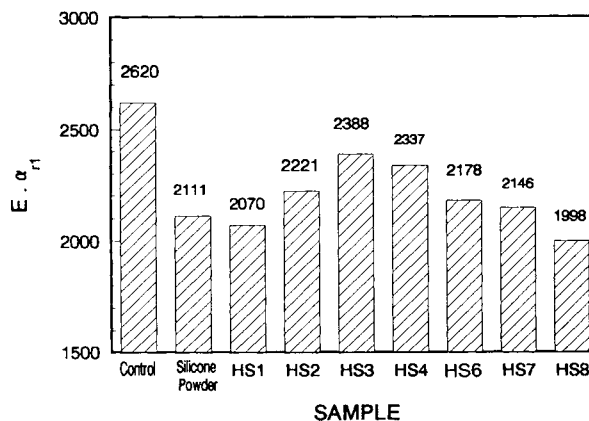

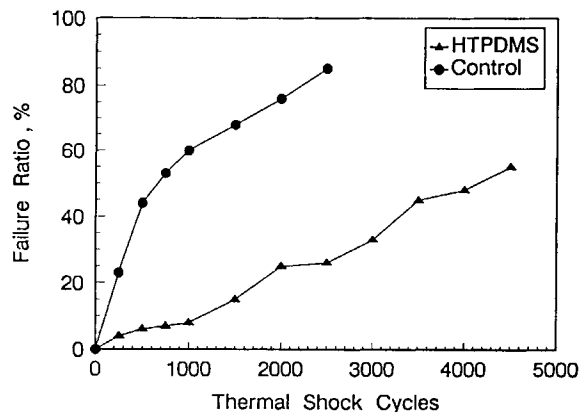**Figure 9** Comparison of stress for various hydride siloxane modified epoxy resin encapsulants.

Figure 10 Thermal shock cycling test results. Temperature cycle: $-65°C \times 15$ min and $150°C \times 15$ min.

From the test results of thermal mechanical properties, it is concluded that the structure and the molecular weight of the hydride terminated siloxanes greatly affect the T_g, flexural modulus, and CTE of the cured resins. HS8 modified CNE network, which has the lowest stress among all modifications and has the same T_g as that of the unmodified CNE network, was 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 10. The result indicates that for 50% of the devices, failure happened after 4100 cycles for the HS8 modified CNE compared to after 750 cycles for the control resin. HS8 modified CNE has much better thermal shock resistance.

CONCLUSION

A process was developed to incorporate stable dispersed silicone rubber particles in an epoxy resin matrix that greatly reduce the stress of cured epoxy resins. Reduction in internal stress of the encapsulant was greatly affected by the structure and mo-

lecular weight of the hydride siloxane. The side chain hydride terminated siloxane is slightly better than the main chain terminated hydride siloxane for the modification of CNE in electronic encapsulation in view of thermal stress. HS8 is the best modifier for epoxy resins, which significantly reduces the flexural modulus and the CTE while maintaining the T_g of the cured epoxy resin. In electronic encapsulation applications, the encapsulant from HS8 modified CNE has exhibited excellent thermal shock resistance and resulted in an extended device use life.

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