Low-Stress Encapsulants by Vinylsiloxane Modification

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SYNOPSIS

Dispersed silicone rubbers were used to reduce the stress of cresol–formaldehyde novolac epoxy resin cured with phenolic novolac resin for electronic encapsulation application. The effects of structure, molecular weight, and contents of the vinylsiloxane oligomer on reducing the stress of the encapsulant were investigated. Morphology and dynamic mechanical behavior of rubber-modified epoxy resins were also studied. The dispersed silicone rubbers effectively reduce the stress of cured epoxy resins by reducing flexural modulus and the coefficient of thermal expansion (CTE), whereas 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 device use life. © 1994 John Wiley & Sons, Inc.

INTRODUCTION

Epoxy molding compounds (EMCs) have been widely used as an encapsulation material for semiconductor devices. *o*-Cresol–formaldehyde novolac epoxy (CNE) is the resin typically employed to encapsulate microelectronic devices, owing to its excellent heat, moisture, solvent, and chemical resistance, superior electrical and mechanical properties, and good adhesion to many substrates. Upon cure, this multifunctional epoxy resin provides a densely cross-linked protective layer; however, it is relatively brittle.

The scale of integration of LSIs 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 (SMT) also generates heat stress to devices.[1,2] Internal stress causes package cracking, passivation layer cracking, aluminum pattern deformation, etc.[3–5] Therefore, the development of a low-stress EMC is required for high-reliability semiconductor devices. The sources of internal stress with plastic encapsulants are considered to be (1) shrinkage due to the curing process and (2) differential shrinkages between the device and the resin. In the case of epoxy resin encapsulation, the first source (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 of thermal expansion coefficient is expressed in the following equation:

$$S = K \int (\alpha_r - \alpha_s)\, E_r \, \mathrm{dT}$$

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, 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.[6–26] Increasing the amount of silica filler used in an encapsulant effectively lowers the thermal expansion coefficient; however, this approach not only increased the elastic modulus, but also increased 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 rubber particles and an epoxy matrix, is considered to be more desirable than a one-phase structure in view of the thermal property needs.[3,27] Traditional modifiers include functionalized rubber such as car-

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Journal of Applied Polymer Science, Vol. 51, 2047–2055 (1994)
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boxyl-terminated butadiene–acrylonitrile copolymers (CTBN). When CTBN modifiers are incorporated into resin compositions, a two-phase morphology consisting of relatively small (−0.1–5 μm) rubber particles dispersed in a resin matrix is generated that toughens epoxy resins.[28–31] However, phase separation depends upon the formulation, processing, and curing conditions.[32–36] 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.

Organosiloxane polymers are known for their excellent thermal and thermooxidative stabilities, moisture resistance, good electric properties, and low stress. In this article, the preformation of a stable dispersion of rubbery particles in the uncured epoxy resin via *in situ* polymerization of vinylsiloxane oligomers in the epoxy resin was investigated. The devices encapsulated by the vinylsiloxane-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 *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 curing agent with an average hydroxyl functionality of 6 and a hydroxyl equivalent weight of about 104 (Schnectady Chemical, HRJ-2210). The vinylsiloxanes were purchased from HULS America Inc. or Shin-Etsu Chemical Co.; their structure and molecular weight (MW) are shown in Table I. The polymer chain was predominantly dimethylsiloxy units with partial side-chain vinyl or terminal vinyl groups. Glycidyl methacrylate (GMA) was used as a compatibilizer and cross-linker for the elastomer. The A-1 catalyst was the (ethyl)triphenylphosphonium acetate acetic acid complex, Ph_3P was the triphenylphosphine that was used as curing accelerator, and *t*-butyl peroxy-2-ethyl hexanoate and 1,1-bis-(*t*-butyl peroxy)-3,3,5-trimethylcyclohexane were used as free-radical initiators.

General Procedure for the Preparation of Silicone Rubber-modified Epoxy Resin

To a four-neck round-bottom flask, equipped with a heating mantle, stirrer, reflux condenser, thermocouple, and temperature controller were added 384 g of *o*-cresol–formaldehyde novolac epoxy resin (CNE). 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 mix-

Table I Vinylsiloxane Employed for the Modification of CNE

Abbreviation	Structural Formula	MW	Supplier
VS1		5,970	HULS
VS2	$H_2C{=}CH{-}Si(Me)_2{-}O{-}[{-}Si(Me)_2{-}O{-}]_n{-}Si(Me)_2{-}CH{=}CH_2$	10,810	Shin-Etsu
VS3		28,000	HULS
VS4		62,700	HULS
VS5		15,286	Shin-Etsu
VS6	$Me_3Si{-}O{-}[{-}Si(Me)_2{-}O{-}]_n{-}[{-}Si(Me)(CH{=}CH_2){-}O{-}]_m{-}SiMe_3$	28,000	HULS

ture of methacrylic acid (MAA, 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 MAA 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, MAA, in the reaction mixture. The resultant epoxy resin thus had a radically polymerizable double bond in the molecule. Under a nitrogen atmosphere, a mixture consisting of vinylsiloxanes (45 g), GMA (5 g), and free-radical initiator (1.2 g) was then added dropwise over a 4 h period via a metering pump while maintaining the reaction temperature at 120°C. Upon completion of the vinylsiloxane addition, the reaction temperature was held at 120°C for an additional 3 h. The reaction mixture was then heated at 150°C under a full vacuum for 30 min to remove unreacted monomers and to decompose the radical initiator. When it was cooled to room temperature, a creamy silicone rubber-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_3P in a mill at moderate temperature to give a thermosettable epoxy resin powder. The resin powder was cured in a mold at temperature of 150°C and pressure of 50 kg/cm^2 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

Epoxy equivalent weight (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) mm^3. The storage modulus G' and tan δ were determined. The peak of tan δ was identified as the glass transition temperature (T_g) because a large decrease in G' occurred at this point. The JEOL JSM-6400 scanning electron microscope (SEM) was employed to examine the morphology of rubber-modified cured samples fractured cryogenically in liquid nitrogen. The fracture surfaces were vacuum-coated with gold. Mechanical 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 E790-84a. The coefficient of thermal expansion (CTE) was measured with a DuPont 943 thermal mechanical analyzer in accordance with ASTM E831-86. The specimens were heated at a rate of 5°C/min. The thermal expansion was proportional to the temperature and the CTEs were calculated from the slope. The thermal shock cycling test was carried out by the following procedure.[37,38] 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 is counted as the failure of that device. The percentage of devices that failed (cracked), as a function of cycles, is plotted.

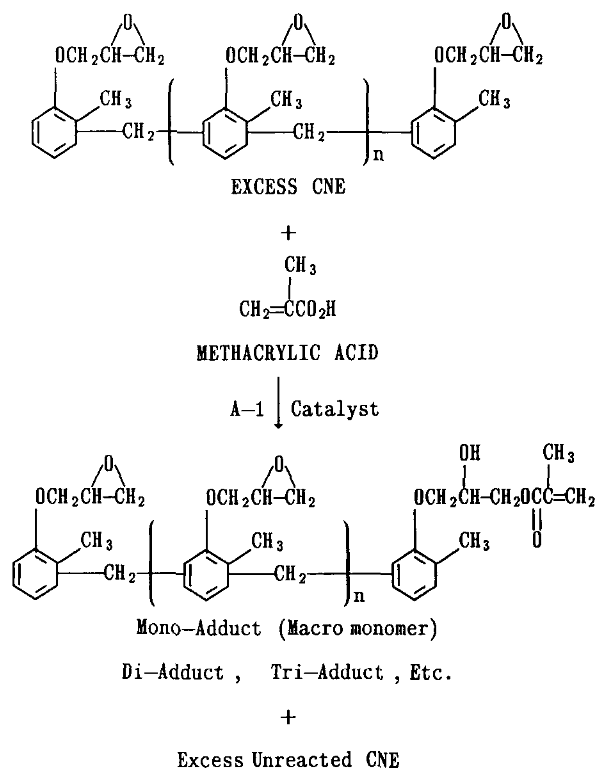

Figure 1 Vinylization of an epoxy resin to form a macromonomer.

RESULTS AND DISCUSSION

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 cresol–formaldehyde novolac epoxy resin (CNE) with methacrylic acid to form a reactive macromonomer as shown in Figure 1. The second step is the polymerization of the resultant macromonomer with various vinylsiloxane oligomers in the presence of a radical polymerization initiator to form stable silicone rubber particles dispersed in the matrix resin in a "sea-island" structure ("islands" of silicone rubber dispersed in the "sea" of an epoxy resin as shown in Fig. 2).

Dynamic Viscoelastic Analysis of the Modified Resins

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

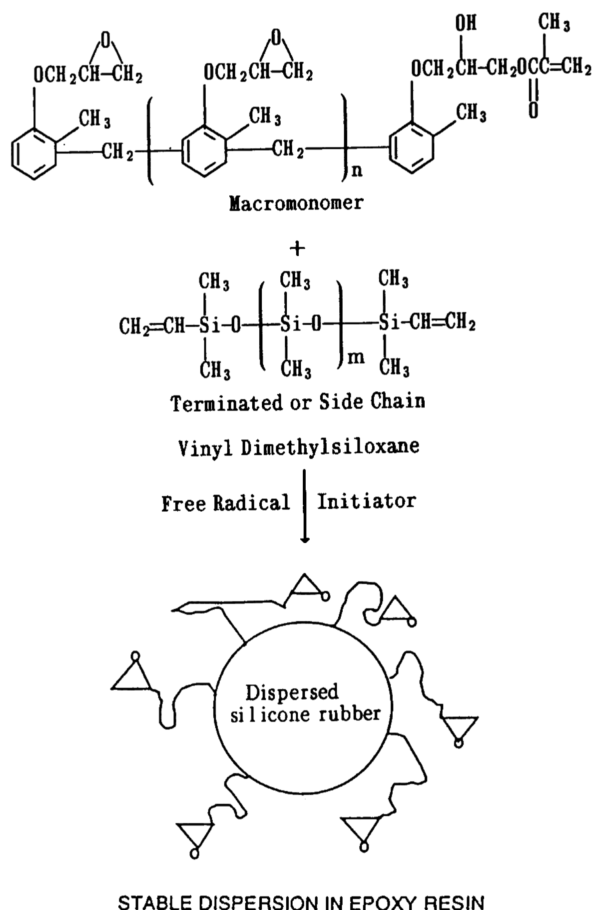

Figure 2 Vinyl *in situ* polymerization to form a dispersion.

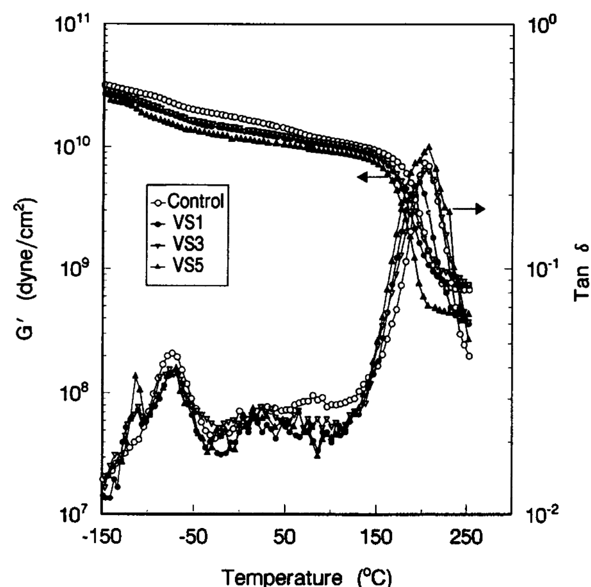

Figure 3 Dynamic viscoelastic analysis for various modified epoxy resins (no filler). The curing agent is phenol–formaldehyde novolac (HRJ-2210).

epoxy resins. The tan δ curves for the control epoxy network exhibit the two major relaxations observed in most epoxy polymers[39]: 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 3 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 small peak

Table II T_g's of Control and Various Silicone Rubber-Modified Epoxy Resin Cured with Phenolic Novolac

Modifier	T_g[a] (°C)	T_g[b] (°C)
Control[c]	—	202
VS1	−110	197
VS2	−111	200
VS3	−112	202
VS4	−110	200
VS5	−114	204
VS6	−113	205

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

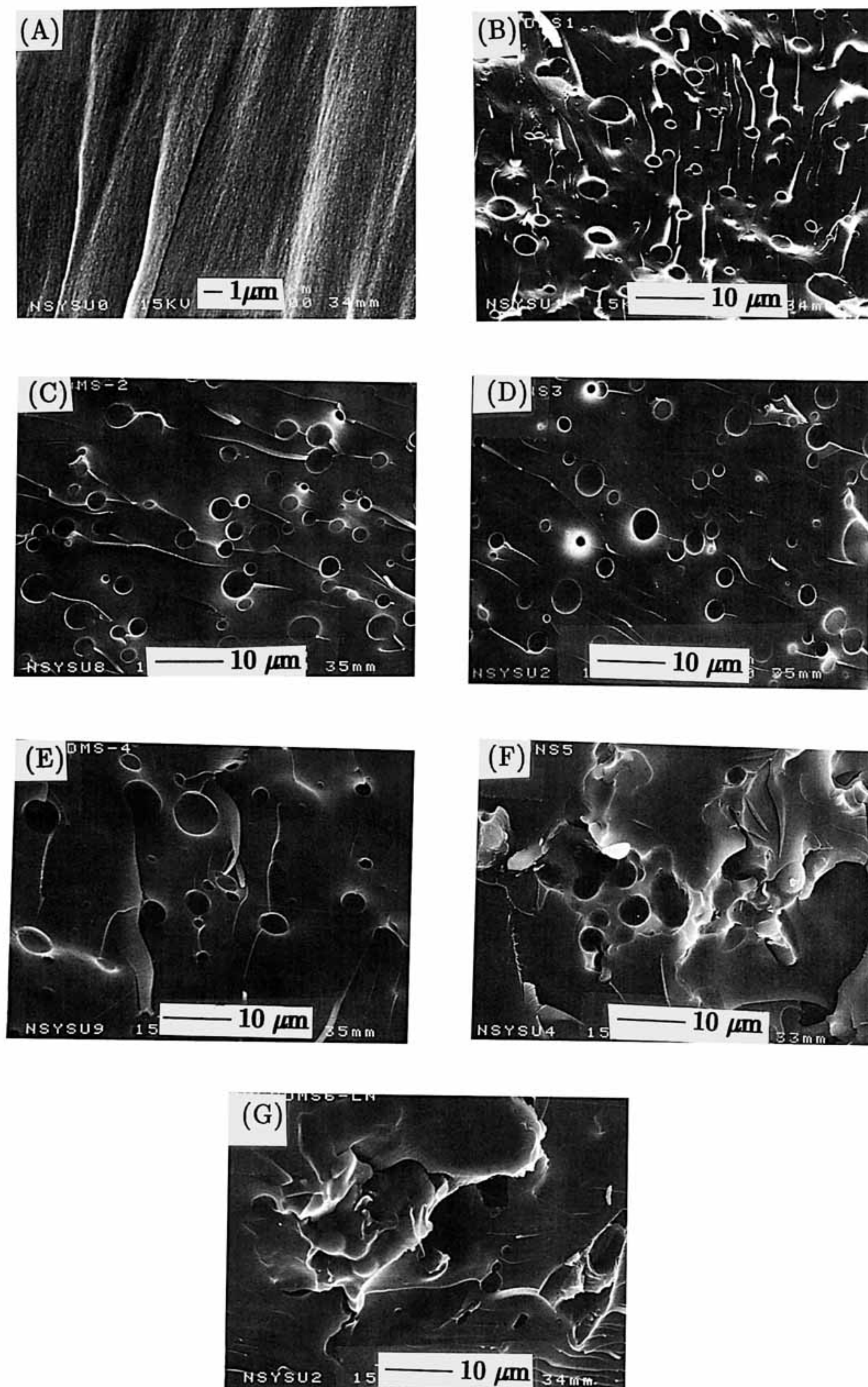

Figure 4 Morphology of cold-snap fracture surfaces of cured epoxy resins modified with various vinylsiloxanes (10 wt %): (A) unmodified; (B) VS1; (C) VS2; (D) VS3; (E) VS4; (F) VS5; (G) VS6.

from −130 to −100°C with a center near −110°C corresponding to the glass transition temperature of the siloxane phase. This small peak further supports the multiphase separation. This is in agreement with SEM observations. It is interesting to note that the side-chain vinylsiloxane (VS5) modification has higher siloxane peak than that of the terminal vinylsiloxane modifications. This could be attributed to a more complete phase separation of side-chain vinylsiloxane rubber particles from the matrix resin than that of the terminal vinylsiloxane rubber-modified systems. In addition, the α-relaxation peak in the tan δ curve became broader but the peak position has hardly changed except for VS1 modification with respect to the control resin. The major T_g and rubber T_g of the cured modified epoxy resins are shown in Table II. Whereas the side-chain vinylsiloxane-modified CNEs (VS5, VS6) have slightly higher T_g's than the unmodified resin, however, terminal vinylsiloxane-modified CNEs have approximately equal to or slightly lower T_g's than that of the unmodified resin depending on the MW of the siloxane modifiers. The solubility of siloxane rubbers in epoxy resin increases when the MW of the siloxane modifier becomes smaller and this greater solubility has resulted in a depression of the glass transition temperature of the modified epoxy resin.

Morphology

SEM photomicrographs of cold-snap surfaces for the control and six vinylsiloxane-modified resins are given in Figure 4. The two-phase structure is observed in all rubber-modified resins, but the shapes of rubber particles are quite different between side-chain and terminal vinylsiloxane modification. The sizes of the rubber particle for the terminal vinylsiloxanes are 1–5 μm and slightly proportional to the MW of the siloxane modifiers. These can be attributed to the compatibility of siloxane and CNE.

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 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

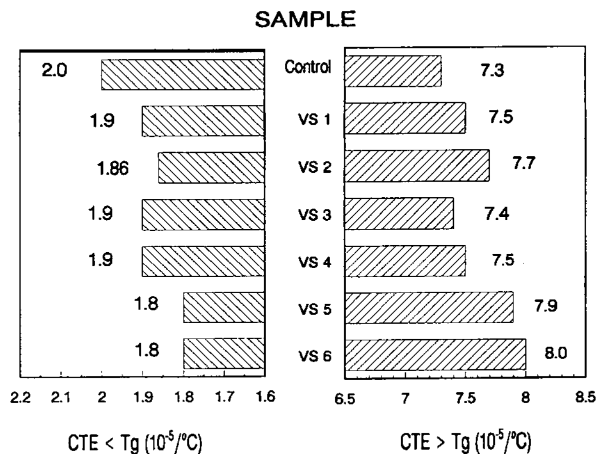

Figure 5 Coefficient of thermal expansion (CTE) for various rubber-modified epoxy resins.

Encapsulation Formulation

A control resin and CNE modified with various silicone rubbers or silicone powder were formulated into eight 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:

Coefficient of Thermal Expansion (CTE)

Figure 5 shows the thermal mechanical properties. The CTE in the glassy state below the glass transition temperature (T_g) was taken from 60 to 100°C and the CTE above the T_g was taken from 200 to 240°C. Below the T_g, the rubber-modified encapsulants all have slightly lower CTE than that of the unmodified resin and this will result in a small difference in CTEs between encapsulant and silicon chip. However, above the T_g, the addition of silicone rubber will increase the CTE.

Glass Transition Temperature (T_g)

Glass transition temperatures (T_g) were determined from tangents of the CTE as a function of temper-

ature at 100 and 200°C. The results are shown in Figure 6. The T_g of cured CNE is relatively unaffected by the modification with side-chain vinylsiloxanes (VS5, VS6), but slightly depressed when modified with terminal vinylsiloxanes, especially for the one with low MW (VS1). This result suggests that the side-chain vinylsiloxane rubber is not soluble in the epoxy matrix and remains phase-separated throughout the cure process. This observation is consistent with the viscoelastic investigation. This result is quite different from silicone powder modification wherein an increase in toughness is accompanied by a decrease in the T_g, indicating that chemical toughening is better than physical toughening.

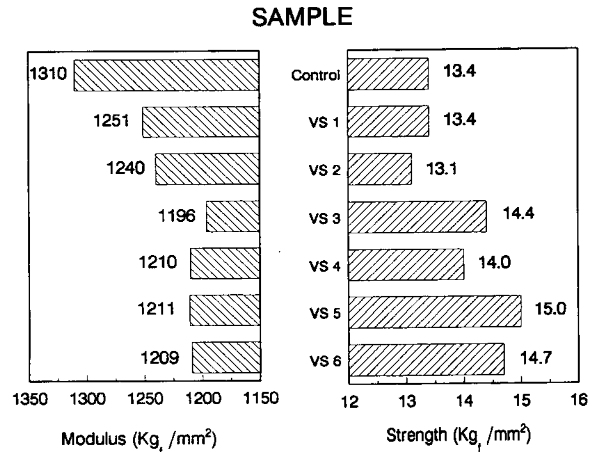

Figure 7 Flexural test properties for various rubber-modified epoxy resins.

Flexural Test Properties

Figure 7 shows the result of the flexural test. The flexural moduli of the cured resins were reduced markedly with various silicone rubber modifications; however, the flexural strength remained equal to or larger than that of the control resin. The amount of reduction in flexural modulus by silicone rubbers is dependent on the structure and MW of vinylsiloxanes and is greater with side-chain vinylsiloxane modification.

Thermal Stress

Thermal stress of cured encapsulants can be approximated by the product of flexural modulus and CTE ($E \cdot \alpha_{r1}$). Figure 8 shows the dependence of stress on structure and MW of vinylsiloxane. The results clearly indicate that the side-chain vinylsiloxanes are better modifiers than are the main-chain terminal vinylsiloxanes in reducing stress and the optimal MW of modifier is around 20,000–30,000.

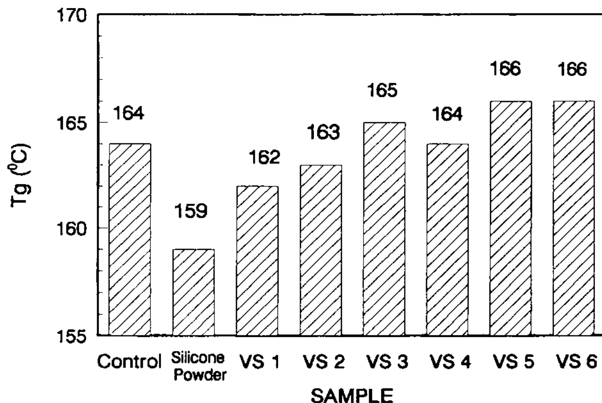

Figure 6 Glass transition temperature for various rubber-modified epoxy resins.

The Effect of Rubber Content on Stress

Since VS6 modification has the lowest stress value, the effect of rubber content on stress was evaluated

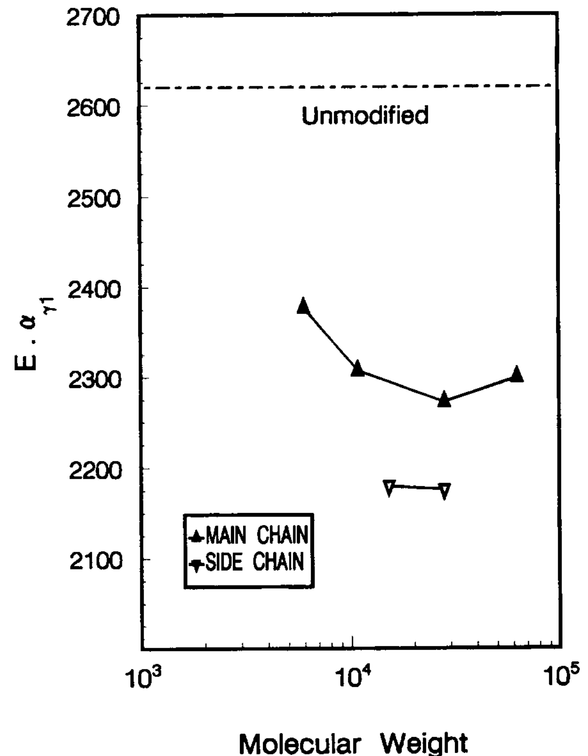

Figure 8 Dependence of stress on vinylsiloxane molecular weight and structure.

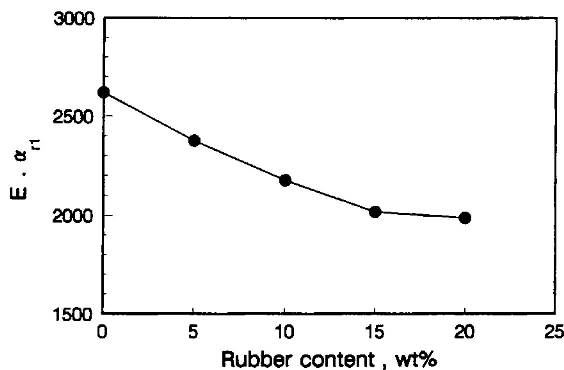

Figure 9 Stress of the cured encapsulant as function of silicone rubber (VS6) content.

with that system. The stress of cured encapsulants as a function of silicone rubber (VS6) contents in CNE is shown in Figure 9. The stress reduced gradually with the increase in rubber content up to 15 wt % and then leveled off. Figure 10 shows that the flexural modulus of the cured encapsulant decreased with the increase in the rubber content of CNE, while T_g's were hardly depressed. These can be attributed to the complete separation of rubber phase from CNE.

From the test results of thermal mechanical properties, it is concluded that the structure and MW of vinylsiloxane oligomer greatly affect the T_g, flexural modulus, and CTE of the cured resins. VS6-modified CNE, which has a lower stress than that of CNE and has the same glass transition temperature as that of CNE, was chosen to encapsulate the

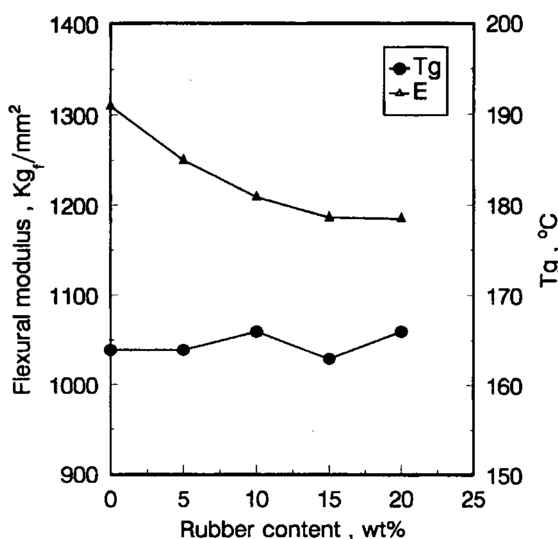

Figure 10 Flexural modulus and T_g of the cured encapsulant as function of silicone rubber (VS6) content.

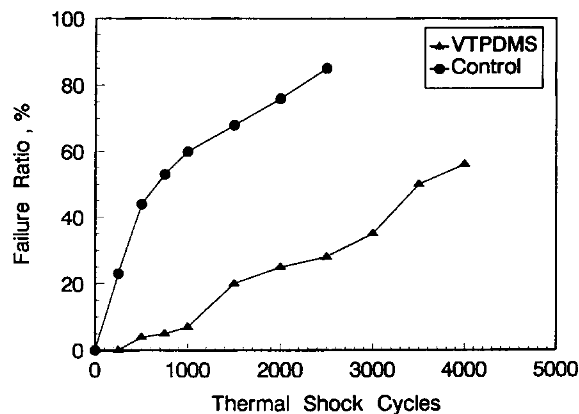

Figure 11 Thermal shock cycling test results. Temperature cycle: −65°C × 15 min and 150°C × 15 min.

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 of 50% of the device failures happened after 3600 cycles for the VS6-modified CNE compared to after 750 cycles for the control resin. VS6-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 reduces the stress of cured epoxy resins. Reduction in stress was greatly affected by the structure and MW of the vinylsiloxane modifier and the rubber content. The most suitable concentration of rubber in the epoxy resin was around 10–15 wt %. The side-chain vinylsiloxane is the best modifier for epoxy resins, which significantly reduces the flexural modulus while maintaining the T_g of cured epoxy resins. In electronic encapsulation applications, the encapsulant from VS6-modified CNE has exhibited excellent thermal shock resistance and resulted in an extended device use life.

Financial support of this work by the National Science Council of the Republic of China is gratefully appreciated (NSC 82-0405-E006-77).

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Received May 25, 1993
Accepted August 2, 1993