Macromolecular Research, Vol. 12, No. 1, pp 78-84 (2004)

A Low-Viscousity, Highly Thermally Conductive Epoxy Molding Compound (EMC)

Jong-Woo Bae

Korea Institute of Footwear and Leather Technology, Busan 614-100, Korea

Wonho Kim*, Seungchul Hwang, and Young-Sun Choe

Department of Chemical Engineering, Pusan National University, Busan 609-735, Korea

Sang-Hyun Lee

Division of Quantum Metrology, Korea Research Institute of Standards and Science, Daejeon 305-600, Korea

Received Aug. 20, 2003; Revised Jan. 19, 2004

Abstract: Advanced epoxy molding compounds (EMCs) should be considered to alleviate the thermal stress problems caused by low thermal conductivity and high elastic modulus of an EMC and by the mismatch of the coefficient of thermal expansion (CTE) between an EMC and the Si-wafer. Though AlN has some advantages, such as high thermal conductivity and mechanical strength, an AlN-filled EMC could not be applied to commercial products because of its low fluidity and high modulus. To solve this problem, we used 2-μm fused silica, which has low porosity and spherical shape, as a small size filler in the binary mixture of fillers. When the composition of the silica in the binary filler system reached 0.3, the fluidity of EMC was improved more than twofold and the mechanical strength was improved 1.5 times, relative to the 23-μm AlN-filled EMC. In addition, the values of the elastic modulus and the dielectric constant were reduced to 90%, although the thermal conductivity of EMC was reduced from 4.3 to 2.5 W/m-K, when compared with the 23-μm AlN-filled EMC. Thus, the AlN/silica (7/3)-filled EMC effectively meets the requirements of an advanced electronic packaging material for commercial products, such as high thermal conductivity (more than 2 W/m-K), high fluidity, low elastic modulus, low dielectric constant, and low CTE.

Keywords: low viscosity, high thermal conductivity, AlN, fused silica, EMC.

Introduction

Over the last few decades, the micro-electronic industry, which needs development in computer and communication devices for fast treatment of huge amounts of data has grown rapidly in size. Thus, high performance integrated circuits are in demand and the demand has been increasing steeply.[1] The major functions of packaging are to protect devices from mechanical and chemical hazards, to distribute signals and power, and to dissipate heat. The ability of a package to adequately perform these functions depends on the properties of the device as well as the properties of package materials.

Recently, to improve performance and reduce cost, the density of package has been increased. With the increased density of package, the electrical energy consumed in a device ultimately appears as heat, elevating the temperature of the active junction and other parts of the package housing of the device. Increased temperature adversely affects the reliability of the device.[2] So, the package material should have high thermal diffusion rate, i.e., high thermal conductivity. There are several other physical properties of polymeric materials that are important to micro-electronics packaging, some of which are a low viscosity, a low dielectric constant, a low coefficient of thermal expansion (CTE), a low elastic modulus, a high water resistance, and a high flexural strength.[3,4]

However, typical epoxy molding compounds (EMC) such as fused silica filled EMC, which have low thermal conductivity, cannot effectively dissipate the heat of a silicon die. Therefore, advanced epoxy molding compounds should be considered to alleviate thermal stress problems caused by low thermal conductivity and high elastic modulus of the typical EMCs and by the mismatch of coefficient of thermal expansion (CTE) between EMC and the Si-wafer.

To more effectively solve the thermal dissipation problem, the use of high thermally conductive ceramic fillers (listed

---

*e-mail: whkim@pnu.edu

in Table I) should be considered.[5]

Though high thermally conductive fillers such as AlN (Aluminum Nitride) and alumina have higher thermal conductivity than fused silica, EMCs crack resistance often decreased because of their poor moldability and high modulus, which is caused by a high porosity and the modulus of the filler.[6,7]

To solve this problem, the filler size distribution theory[8] that provided improved fluidity for the high filler loading system, could be applied to manufacture of EMC. According to the filler size distribution theory, the maximum packing fraction is inversely proportional to the viscosity of EMC (Mooney equation; Eq. 1)

$$\ln(\eta_c/\eta_0) = (K_E\Phi_f)/(1 - (\Phi_f/\Phi_m)) \qquad (1)$$

where η_0 is the viscosity of the resin, η_c is the viscosity of the compound, Φ_m is the maximum filler fraction that can be achieved for a given filler composition, Φ_f is the packing fraction of the filler, and K_E is the Einstein constant.

The Mooney equation describes nonlinear behavior of the viscosity increase as the filler loading approaches the maximum packing fraction. It can be expected that the viscosity of the compound will be lower as the maximum packing fraction in the Mooney equation is increased. Figure 1 shows the idealized packing of a binary mixture of spheres as a function of composition using their diameter ratios as a parameter.

The thermal conductivity of EMC (65 vol% of fillers) was greatly improved up to 5.2 W/m-K by using a binary mixture of AlN (23 μm and 2 μm of Aluminum Nitrides, granule type) compared to 1.5 W/m-K of crystalline silica filled (65 vol%) EMC which is applied to packaging material as a high thermal conductive grade.[9] However, the fluidity of this mixture became worse compared to commercial products, i. e., the spiral flow length was only 33 inches compared to overflow (i. e, more than 60 inches) of commercial products.[9]

To improve fluidity and minimize the reduction of thermal conductivity of the EMC, fused silica can be considered as a small size filler because it has low porosity and spherical shape. In this study, to improve the fluidity of an AlN filled EMC, 2 μm fused silica was used as a small size filler in a binary mixture of fillers. Properties such as the spiral flow, thermal conductivity, CTE, flexural strength, elastic modulus, and dielectric properties of EMC were evaluated as a function of the volume fraction of small size silica in the binary mixture of fillers.

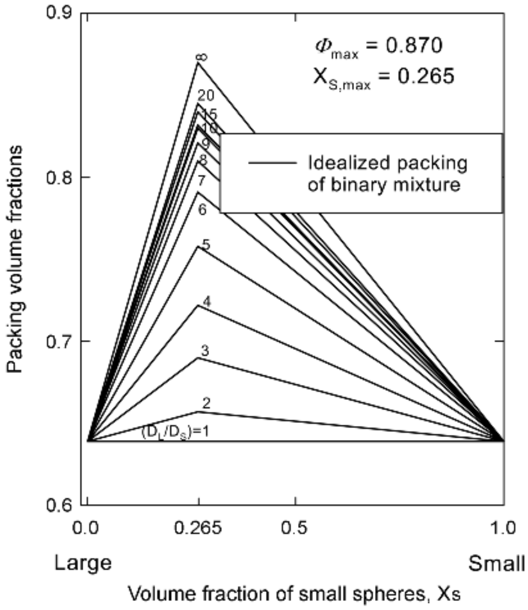


Figure 1. Idealized maximum packing of binary mixtures of spheres as a function of composition with diameter ratios as a parameter.[10]

Experimental

Raw Materials.

Matrix and Additives: Epoxy resin can be divided into two types: bisphenol-A and novolac. Solid novolac epoxy is most widely used as a package material for semi-conductors because of its excellent thermal stability. For this reason, in this study, novolac epoxy was selected as a base resin, and phenol-novolac as a hardener. Although phenol-cured epoxy is not widely used commercially, it provides excellent moldability, electrical properties, and heat and humidity resistance when compared with other cured epoxies used in microelectronic packaging.[11] The average epoxy equivalent weight of novolac epoxy is 200 g-mol^{-1}eq, and the average hydroxyl equivalent weight of phenol novolac is 106 g-mol^{-1}eq. A coupling agent, an accelerator, and natural wax were used as

Table I. Physical Properties of Ceramic Fillers and Epoxy Resin

Materials	Dielectric Constant	Coefficient of Thermal Expansion (10^{-6}/K)	Thermal Conductivity (W/m-K)	Young's Modulus (GPa)
Fused Silica	3.8	0.5	2	74
AlN	7.0	4.5	150(Practical)	330
Alumina	8.8	5.5	36	385
Epoxy	6~7	50~90	0.2~0.4	2

Table II. Basic Formulation of Epoxy Molding Compounds

Materials	wt%	Remark
Novolac Epoxy Resin	7~30	Equiv. wt of epoxy; 200
Phenol Novolac Hardner	3.5~15	Equiv. wt of phenol; 106
Catalyst (TPP)	0.75 phr	Equiv. wt ratio of
Inorganic Filler (AlN, silica)	50~85	epoxy/phenol = 1.0
Mold Release Agent (wax)	0.5	Density of AlN; 3.26
Coupling Agent	0.5	Density of fused sillica; 2.2
Stress-relief Agent	0.4	

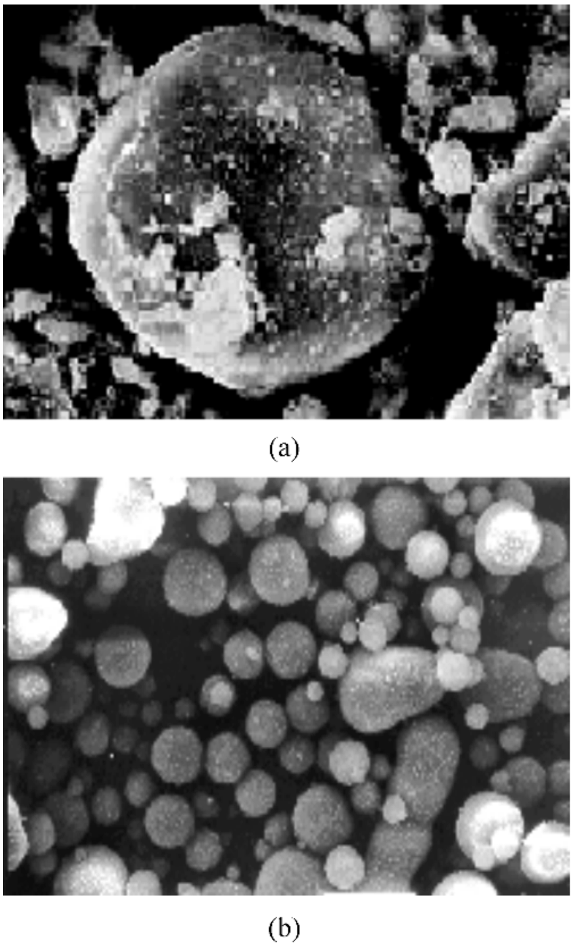
additives. The basic formulation is shown in Table II.[7,9]

Filler: Even though AlN filler leads to improvements in terms of thermal conductivity, thermal expansion and mechanical properties of EMC, it can also cause reduction of the moldability and the flowability. This study tried to suggest the proper combination of fillers considering fluidity, thermal conductivity, thermal expansion, and dielectric characteristics of the resulting EMCs. The mean particle size of an AlN (granule type) filler was 23 μm (ART Co., USA) and that of the fused silica was 2 μm (Tokuyama Co., Japan). The morphology of AlN and fused silica are shown in Figure 2.

Manufacturing Process for EMC. A two-roll mill, which can support high torque, was used as a mixer due to the high viscosity of a filled EMC (with over 50 vol% of filler). According to the formulation shown in Table II, an AlN-filled master batch was mixed at the roll surface temperature of about 90 °C for 10 min which minimize the degree of curing. After that, melt mixed EMC was cooled and crashed into granules. A disc-shaped EMC, which was made of preheated granules, was forced to flow into a heated cavity by a transfer molding machine with a pressure of 75 kg$_f$/cm^2 followed by being molded at 175 °C for 2.5 min. Molded EMC was post-cured at 175 °C for 4 hrs. Flow chart of the manufacturing process of EMC is shown in Figure 3.

Test Methods.

Spiral Flow: A spiral flow test is extensively used by IC manufacturers to evaluate the fluidity of thermosetting molding compounds, and all material vendors report this data.[3] This test consists of loading the disc of molding compound into the heated transfer pot of the press and transferring it through a spiral coil with a semi-circular cross section until the flow ceases. According to the test procedure (ASTM D3123), the recommended pressure and temperature are 110 kg$_f$/cm^2 and 150 °C, rspectively. However, in this study, the test was carried out at the pressure of 75 kg$_f$/cm^2 for 150 seconds and the temperature of 175 °C.


(a)

(b)

Figure 2. The SEM photographs of AlN and fused silica; (a) AlN and (b) fused silica.

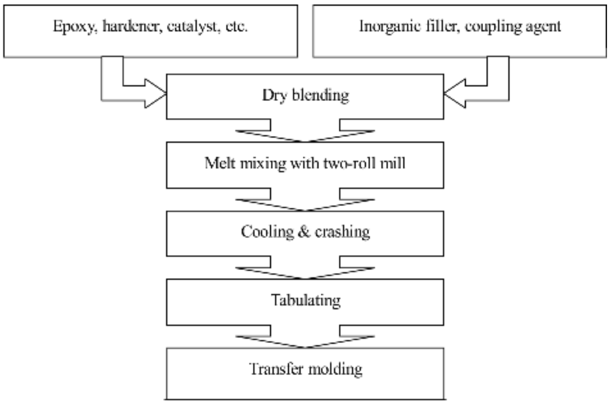


Figure 3. Flow chart of manufacturing process for epoxy molding compounds.

Thermal Conductivity: There are various equations to estimate the thermal conductivity of composite materials. However, a few equation can actually predict the thermal conductivity of composite materials because predicting thermal conductivity of composite materials depends on many factors. These equations are only useful in roughly estimating the thermal conductivity of composite materials. That implies thermal conductivity of the composite materials need to be experimentally determined.

The measurement of thermal diffusivity (δ) was carried out by a laser flash method (Sinku-Riko Co. Model TC-7000) at room temperature. The specific heat (C) was measured by a DSC (Perkin-Elmer Co, Pyris I). Also, the density of the specimen(ρ) was measured by water displacement. After that, the thermal conductivity (k) was calculated by Eq. 2[12]:

$$k = C \times \rho \times \delta \tag{2}$$

Coefficient of Thermal Expansion: The coefficient of thermal expansion (CTE) of the composite was measured by using a bar-shaped specimen in a thermo-mechanical analyzer (Perkin-Elmer, TMA 7e) with a static force (50 mN) and a heating rate of 5 °C/min from 40 to 250 °C. Eq. 3 was used to calculate CTE:

$$\alpha = \frac{\Delta L}{L_0 \Delta T} \tag{3}$$

where α is the coefficient of thermal expansion, $\Delta L/L_0$ is the thermal expansion ratio of a sample, and ΔT is the temperature difference.

Dielectric Properties: The dielectric property of the composite material was measured by using a disk shaped specimen (ϕ10 × 2 mm) with a Dielectric Analyzer (DEA, TA Instrument Co, USA) at room temperature.

Flexural Strength and Modulus: Dynamic mechanical analyzer (Perkin-Elmer Co., DMA 7e) was used to measure the flexural strength and elastic modulus of cured EMC. The sample size was 20 × 5 × 3 mm. Stress-strain property was measured using a 3-point bending apparatus with a static force scan mode (0.1 to 8 N) at room temperature. Also, elastic modulus of cured EMC was measured using a parallel plate apparatus (dynamic force: 100 mN, static force: 110 mN) with a heating rate of 5 °C/min (80 to 250 °C).

Results and Discussion

Spiral Flow. To evaluate the fluidity of EMC, the spiral flow test was carried out by a transfer molding machine with a pressure of 75 $\mathrm{kg_f/cm^2}$. The spiral flow length of EMC (65 vol% of fillers) as a function of the composition of the small size filler in the binary filler system is shown in Figure 4. In Figure 4, the best fluidity was obtained at Xs = 0.3-0.4; Xs is the composition of a 2 μm filler in the binary filler system. Those values are near the volume fraction of small sphere at which packing volume fraction becomes the maximum for a given size ratio, i. e., Xs = 0.265. The difference in the values of Xs may be due to the deviation from both monodispersity and spherical shape of the fillers. It can be concluded that fluidity is improved by the filler size distribution.

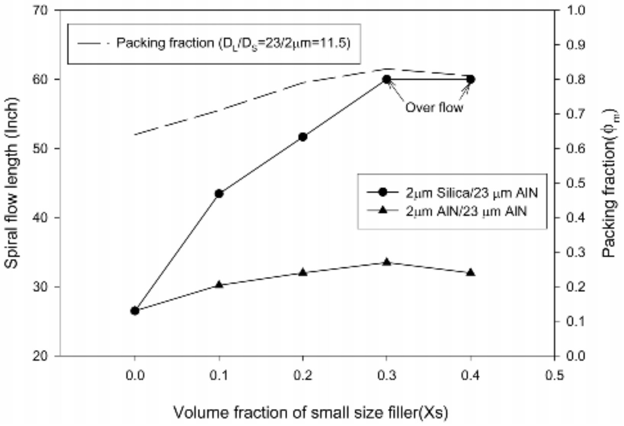


Figure 4. Spiral flow length of EMC (65 vol% of fillers) as a function of the composition of the small size filler in the binary filler systems.

Improvement of fluidity by using binary filler size distribution was pronounced for the case of a 2 μm silica/23 μm AlN filler system compared to that of a 2 μm AlN/23 μm AlN filler system. Spiral flow length of EMC (65 vol% of fillers) increased from 26.5 inches to more than 60 inches by increasing the composition of 2 μm fused silica from 0.0 to 0.3-0.4. This is mainly due to the low porosity and spherical shape of 2 μm fused silica.

Thermal Conductivity. In a multiphase system, thermal conductivity is usually estimated according to the series, parallel, or logarithmic mixing models.[5]

$$\text{Parallel model}: k_T = \Sigma\, \Phi_i k_i \tag{4}$$

$$\text{Series model}: 1/k_T = \Sigma \Phi_i / k_i \tag{5}$$

$$\text{Logarithmic model}: \ln k_T = \Sigma \Phi_i \ln k_i \tag{6}$$

where k_T is the thermal conductivity of a multiphase system, Φ_i is the volume fraction of the i component, and k_i is the thermal conductivity of the i component.

Thermal conductivity of EMC (65 vol% of fillers) as a function of the composition of silica is shown in Figure 5. Estimated values from Eqs. (4)~(6) are shown with three different lines which are based on the experimental thermal conductivity data of a 23 μm AlN filled EMC (k = 4.3W/m-K) and a 2 μm silica filled EMC (k = 0.65 W/m-K). Figure 5 shows that the thermal conductivity of EMC decreased as the composition of the silica increased, and the experimental data follows a logarithmic model quite well. To acquire more than 2.0 W/m-K of thermal conductivity of EMC, the composition of the silica in the binary filler system should be less than 0.4.

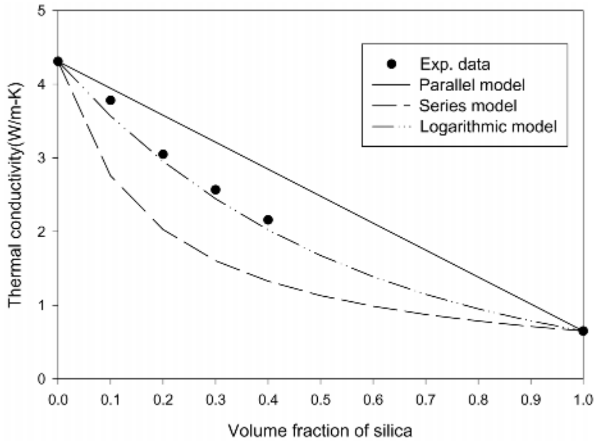


Figure 5. Thermal conductivity of EMC (65 vol% of fillers) as a function of the composition of the silica in the binary filler system.

Dielectric Constant and Dissipation Factor. High signal accumulation of device itself and higher clock rate can be obtained by using materials that has low values of the dielectric constant(see Eq. 7). Also, dissipation factor means stability of signal transfer.[13]

$$Td = (\varepsilon)^{1/2}\,(l/c) \tag{7}$$

where ε is the dielectric constant of media, c is the velocity of light, l is the distance, and Td is the delay of signal transfer.

Dielectric constant and dissipation factor of EMC(65 vol% of filler) as a function of frequency with varying the composition of the silica at room temperature are shown in Figures 6 and 7, respectively. Figures 6 and 7 show that dielectric constant and dissipation factor of EMC decreased by increasing the composition of the silica in the binary filler system because silica has lower dielectric constant than AlN as shown in Table I.

Flexural Strength and Elastic Modulus. A molded encapsulant should have sufficient mechanical strength to protect a chip from physical and thermal damage. Also, the crack resistance of an encapsulant against thermal stresses in an IC operation was increased by decreasing the elastic modulus of the materials.

The flexural strength and elastic modulus of EMC as a function of the composition of the silica in the binary filler system are shown in Figures 8 and 9, respectively. Figure 8 shows that the flexural strength of EMC was increased when the composition of the silica was increased because the wettability between the fillers and matrix, which was influenced by the fluidity of EMC, was increased. In Figure 9, the elastic modulus of EMC was decreased by increasing the volume fraction of silica because silica has lower modulus than AlN. The increased flexural strength and the decreased elastic modulus of EMC, by increasing the composition of the silica, may improve the performance of EMC as a pack-

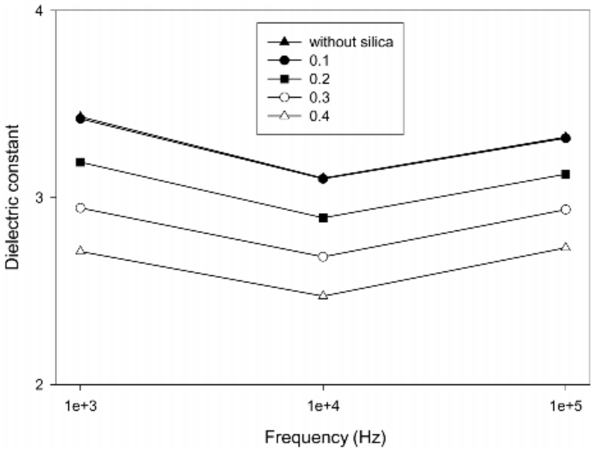


Figure 6. Dielectric constant of EMC (65 vol% of fillers) as a function of frequency with varying the composition of the silica in the binary filler system.

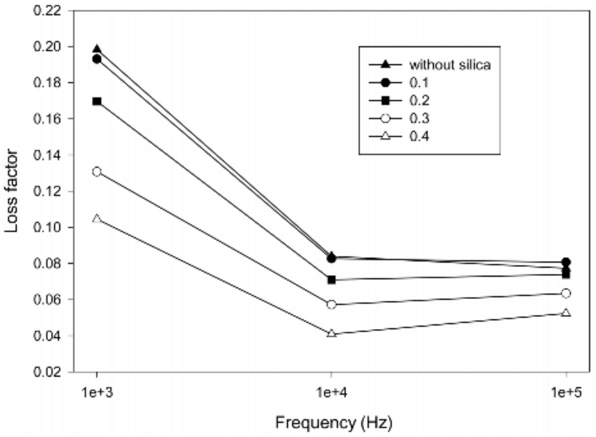


Figure 7. Dissipation factor of EMC (65 vol% of fillers) as a function of frequency with varying the composition of the silica in the binary filler system.

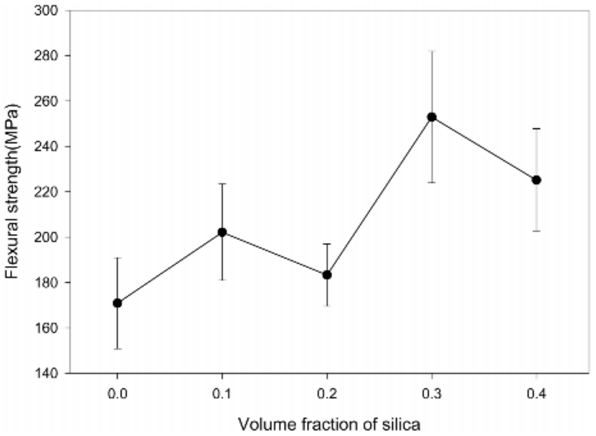


Figure 8. Flexural strength of EMC (65 vol% of fillers) as a function of the composition of the silica in the binary filler system.

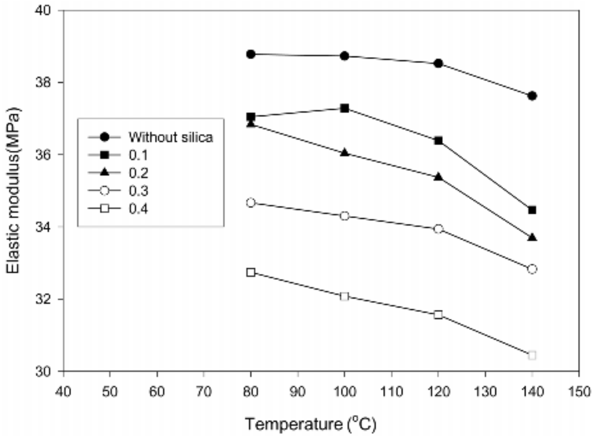


Figure 9. Elastic modulus of EMC (65 vol% of fillers) as a function of temperature with varying the composition of the silica in the binary filler system.

Table III. CTE of EMC as a Function of the Composition of the Silica in the Binary Filler System

Silica Volume Fraction	α_1 (PPM)	α_{230} (PPM)
0.0	18.4	33.7
0.1	18.5	33.8
0.2	18.9	34.0
0.3	18.8	33.7
0.4	18.3	33.4

α_1: CTE measured below glass transition temperature (T_g).
α_{230}: CTE measured from T_g to 230°C.

aging material.

CTE. If the value of CTE of EMC is similar to that of a chip or a leadframe, the packaging can withstand thermal stresses in the manufacturing processes and the thermal fatigue stress in the IC operating processes.[14] CTE of EMC as a function of the composition of the silica was measured and the values are shown in Table III. Though the value of CTE of silica(0.5 ppm) is lower than that of AlN(4.5 ppm), the value of CTE of EMC was not decreased when the composition of the silica was increased, i. e., approximately 18.5 ppm for α_1 in the range of $0.0 < Xs < 0.4$. This is due to the decreased elastic modulus of EMC by increasing the composition of the silica in the binary filler system. CTE of EMC is inversely proportional to the elastic modulus of EMC. The value of CTE of 2 μm fused silica/ 23 μm AlN filled EMC is similar to that of a copper leadframe ($\alpha_1 =$ 17 ppm). From this, matching the value of α_1 would reduce the thermal stress.

Conclusions

Though AlN shows high thermal conductivity, an AlN filled EMC could not be used in commercial products because of its low fluidity and high modulus. In this study, to solve this problem, a 2 μm silica was used as a small sized filler in the binary filler system. When the composition of the silica in the binary filler system reached 0.3, the fluidity of EMC was improved more than 2 times, and the mechanical strength was improved by 1.5 times compared with a 23 μm AlN filled EMC. Also, the elastic modulus and the dielectric constant was reduced to 90%, though thermal conductivity of EMC was reduced from 4.3 to 2.5 W/m-K compared with a 23 μm AlN filled EMC. These were due to the increase of maximum packing fraction in the binary filler system and the nature of a silica filler. So, the 2 μm silica/ 23 μm AlN filled EMC (the composition of the silica in the binary filler system is 0.3) effectively meets the requirement for commercial products as an advanced electronic packaging material, requiring a high thermal conductivity (more than 2 W/m-K), high fluidity (more than 60 inches in spiral flow length), low elastic modulus, low dielectric constant, and low CTE.

Acknowledgements. This work was supported by the Brain Korea 21 Project in 2003.

References

(1) W. J. McClean *Status 1986, A Report of the Integrated Circuit Industry*, Integrated Circuit Engineering Corporation, 1986.
(2) E. J. Rymaszewski, R. R. Tummala, and T. Watari, in *Microelectronics Packaging Handbook*, R. R. Tummala, E. J. Rymaszewski, and A. G. Klopfenstein, Eds., Chapman & Hall, New York, 1997, Part I, pp 1-128.
(3) L. T. Manzione, *Plastic Packaging of Microelectronic Devices*, VAN NOSTRAND REINHOLD, New York, 1990, pp 99-147.
(4) J. H. Lupinski and R. S. Moore, in *Polymer Materials for Electronics Packaging and Interconnection*, J. H. Lupinski and R. S. Moore, Eds., American Chemical Society, Washington DC, 1989, pp 1-24.
(5) L. T. Manzione, *Plastic Packaging of Microelectronic Devices*, VAN NOSTRAND REINHOLD, New York, 1990, pp 60-64.
(6) C. P. Wong and R. S. Bollampally, *IEEE Trans. Adv. Pack.*, **22**, 54 (1999).
(7) W. Kim, J. W. Bae, I. D. Choi, and Y. S. Kim, *Polym. Eng. Sci.*, **39**, 756 (1999).
(8) M. Mooney, *J. Colloid Sci.*, **6**, 162 (1951).
(9) J. W. Bae, W. Kim, S. H. Cho, and S. H. Lee, *J. Mater. Sci.*, **35**, 5907 (2000) .
(10) D. J. Cumberland and R. J. Crawford, *Handbook of Powder Technology*, Elsevier Science Publishers, Amsterdam, 1987, Vol. VI, Chap. 4.
(11) N. Kinjo, M. Ogata, K. Nishi, and A. Kaneda, “Epoxy Molding Compounds as Encapsulation Materials for Microelectronic Devices”, in *Advances in Polymer Science 88,*

Springer-Verlag, Berlin, Heidelberg, 1989.

(12) F. P. Incropera and D. P. Dewitt, *Introduction to Heat Transfer*, 3rd ed., John Wiley and Sons, New York, 1996, pp 50.

(13) W. O. Kingery, H. K. Bower, and D. R. Uhlmann, *Introduction to Ceramics,* Comprise, Massachusetts, 1975, Chap. 18.

(14) S. S. Manson, *Thermal Stress and Low Cycle Fatigue, Bobrowsky Thermal Shock Parameter*, Robert E. Krieger Publishing Company, Florida, 1981, Section 7. 1., pp 383.