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Recycling of epoxy resin compounds for moulding electronic components

MASATOSHI IJI
Ecology-Based Systems Research Laboratory, Resources and Environment Protection Research Laboratories, NEC Corporation, 1-1 Miyazaki, 4-Chome, Miyamae-ku, Kawasaki, Kanagawa 216, Japan

This study reports the recycling of the cured epoxy resin compounds containing silica filler and additives for moulding electronic components, which is generated as a mould residue in moulding process. The pulverized residue (moulding resin powder) showed good surface reactivity due to the functional groups contained (silanol, hydroxy and epoxy) and reacted with polar resins such as epoxy resin and phenol resin in a similar manner to silica powder. Recycling a low-stress-type moulding resin powder containing silicone elastomer into a standard moulding resin yielded a new moulding resin that has far better thermal impact resistance than that made with the original standard moulding resin. Moreover, the moulding resin powder was found to be suitable as a filler for epoxy resin products such as insulating materials, paints and adhesives to supply them with sufficient insulating, strength and adhesive properties. Use of the powder as a decorating agent for an acrylic-resin-type construction material also produced a marble-like appearance and improved the surface hardness of the material.

1. Introduction

Electronic components such as integrated circuits (ICs) are mainly moulded with epoxy resin compounds consisting of epoxy resin, silica particle filler and additives (moulding resins). These are used because of their high thermal resistance and good insulating properties. Increased production of electronic components in recent years has greatly increased the amount of mould residue of moulding resins, which is generated in the moulding process of electronic components. However, recycling this residue has been difficult because the epoxy resin in the residue cannot be remelted for curing in the moulding process. Most of the residue is therefore disposed of at landfill sites.

Material recycling that re-uses pulverized waste materials by blending with the original materials or other materials has been studied for thermosetting resins as a practical method. Studies for general thermosetting resins, such as polyester, polyurethane and phenol, have indicated that recycled resins composed of these cured resin powders and the original resins have sufficient strength at a lower recycling ratio [1–5], but few studies have been made on recycling the epoxy resin. In one study, a cured epoxy resin compound powder consisting of epoxy resin and amine hardener was recycled into the original compound. The strength of the resulting material was found to be sustainable at a low recycling ratio [6]. In another study, a powder of cured epoxy resin board reinforced with glassfibre that is used as a printed-circuit board for electronic products was tested as a filler for plastics products. The strength of the resulting material was comparable with that of materials with conventional fillers [7]. The present author and a co-worker [8–10] found that using this resin board powder as a filler for epoxy resin products employed as paints and adhesives can yield products that have better strength and thermal expansion property than those made with conventional fillers. However, no study has been reported on the recycling of cured epoxy resin compounds containing silica particle filler and various additives, such as the mould residue of moulding resins for electronic components.

This paper reports on a newly developed technology for recycling the mould residue of epoxy resin compound used as moulding resin for electronic components. First, to determine the basic properties of the residue powder (moulding resin powder) for recycling, the powder's chemical structure and reactivity towards polar resins were studied. Then the recycling of the moulding resin powder into the original moulding resin was tested. Moreover, the possibility was examined of employing it as a filler for epoxy resin products used for insulating materials, paints and adhesives, and also as a decorating agent for an acrylic-resin-type construction material.

2. Experiment

2.1. Materials and sample preparation

2.1.1. Moulding resin sample

This study used two model-moulding resins for standard resin and low-stress-type resin. The model standard resin was composed of *o*-cresol Novolak-

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TABLE I Compositions of tested epoxy resin compound for moulding resin

Component	Composition (wt%)	
	Standard resin	Low-stress-type resin
Epoxy resin (*o*-cresol Novolak)	15.3	13.3
Phenol resin hardener	9.0	7.9
Silica powder	70.0[a]	70.0[b]
Curing catalyst	0.2	0.2
Releasing agent (wax)	0.7	0.7
Carbon black powder	0.3	0.3
Silane coupling agent	0.5	0.6
Fire retardants		
Brominated epoxy resin	2.0	2.0
Diantimony trioxide	2.0	3.0
Silicone elastomer	—	2.0

[a] Pulverized silica (average, 15 μm).
[b] Pulverized silica (average, 15 μm) + spherical silica (average, 22 μm): 50 + 50 (wt%).

type epoxy resin (melting point, 70 °C), phenol Novolak resin hardener (melting point, 80 °C), fused silica particle filler, curing catalyst (triphenilphosphine), a releasing agent (Carnauba wax), carbon black powder, a silane coupling agent (γ-glycidoxypropyltrimethoxysilane) and fire retardants (brominated bisphenol A type epoxy resin and diantimonytrioxide). The model low-stress-type resin additionally contained silicone elastomer as a low-stress agent. The blending ratios of these components in the resins are listed in Table I, and these components were supplied by the Sumitomo Bakelite Co. The moulding resins were produced by pre-mixing the components in a mixer, mixing the resulting pre-mixture by using a roll blender heated to 100–105 °C and then cooling, crushing and tabulating. The moulding resins were moulded into an IC package (16-pin dual-in-line-type package (DIP)) by using a conventional transfer moulding machine under the curing condition of 5 min at 170 °C. The mould residues (Fig. 1) of the moulding resins were then recovered and pulverized into powders (average particle size; 16 μm) with a roll mill and a ball mill.

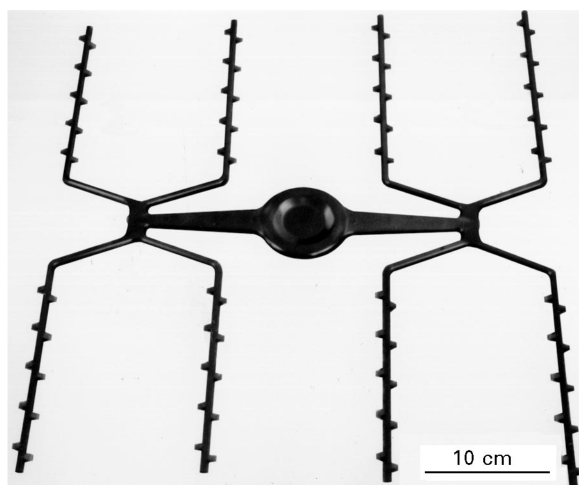

Figure 1 Cured epoxy resin compound generated as mould residue in the moulding process of electronic components.

2.1.2. Recycled moulding resin
The standard moulding resin powder and the low-stress-type moulding resin powder were mixed with the components of the standard moulding resin and moulded in the same way mentioned above and then after-cured under the condition of 6 h at 170 °C.

2.1.3. Epoxy resin products with moulding resin powder
Epoxy resin compounds for insulating materials, paints and adhesives were prepared as epoxy resin product models. The insulating material consisted of the standard moulding resin powder as a filler (60 wt%), *o*-cresol Novolak epoxy resin (20.8 wt%), phenol Novolak resin (12.2 wt%), a releasing agent (0.9 wt%), carbon black powder (0.4 wt%), a curing catalyst (0.3 wt%), brominated epoxy resin (2.7 wt%) and diantimony trioxide (2.7 wt%). Fused silica powder (average particle size, 20 μm: Denki Kagaku Co.) was used as a conventional filler. These components were the same materials as listed in Table I. These were mixed, moulded and after-cured in the same way as mentioned above. For surface treatment of the moulding resin powder, a silane coupling agent such as γ-glycidoxypropyltrimethoxysilane and aminopropyltriethoxysilane (Shinetsu Kagaku Co.) was pre-mixed with the moulding resin powder in a ball mill at weight ratios of 1.0 : 100 and 0.5 : 100 and were heated to 100 °C for 1 h.

The paint consisted of the standard moulding resin powder as a filler, bisphenol A-type epoxy resin (Epicoat 828, Yuka Shell Epoxy Co.), a diluting agent (propylene glycol ether; Eponit 028, Asahidenka Co.), a levelling agent (acrylic copolymer; Polyflow S, Kyoeisha Kagaku Co.), fine silica powder (Aerogiru 200, Nippon Aerogiru Co.), a silane coupling agent (γ-glycidoxypropyltrimethoxysilane) and an amine-type hardener (X-4199, ACR Co.). The blending ratio is listed in Table II. As conventional fillers, talc powder (average particle size, 11 μm; Takehara Kagaku Co.), calcium carbonate powder (average particle size, 9.2 μm; Shiraishi Calcium Co.) and fused

TABLE II Compositions of tested epoxy resin compounds for paints and adhesives

Component	Composition (wt%)	
	Paint	Adhesive
Epoxy resin	39.7	38.8
Diluting agent	7.9	7.8
Levelling agent	0.2	0.2
Fine silica powder	0.2	0.2
Silane coupling agent	0.2	0.2
Filler	31.8	31.0
Amine hardener	20.0	21.9

silica powder (average particle size, 10 μm; Denki Kagaku Co.) were used. These components were mixed in a mixer and then degassed in a vacuum at room temperature. The compounds were moulded in a mould and cured at 23 °C for 7 days.

The adhesive consisted of the above-mentioned components for the paint but another amine-type hardener (Tormide 245, Fujikasei Co.). The blending ratio is also listed in Table II. These components were mixed and degassed by the same method mentioned above. The resulting compounds were bonded between two ferric boards (SPCC-SB) and cured at 23 °C for 7 days.

2.1.4. *Construction material with moulding resin powder*

An acrylic resin compound was prepared as a model construction material. It consisted of acrylic resin and aluminium hydroxide powder (maximum powder size, 150 μm) at a weight ratio of 35 : 65. These components were supplied by Tsutsunaka Plastics Co. The standard moulding resin powders with maximum particle sizes of 1 and 5 mm were added to the components in the weight ratios listed later in Table VI and then mixed in a mixer and compression moulded in a mould under a curing condition of 190 °C for 35 min.

2.2. Measurement methods of properties

2.2.1. *Basic properties of moulding resin powder*

The average particle size and size distribution of powder samples such as moulding resin powder and other fillers were measured on a laser-diffraction-type particle-size-distribution measurement apparatus (SALD-2000, Shimazu Seisakusho Co.). The surface area of the powders was determined on a surface area measurement apparatus (Flow-sorb ‖ 2300, Shimaze Seisakusho Co.).

The chemical surface structure of the moulding resin powder was determined by means of scanning electron microscopy (SEM) and electron probe X -ray microanalysis (EPMA) which were obtained with a Shimazu EPMA-8705 analyser. Fourier-transform infrared (FT–IR) spectra recorded on a FTS-15E/D spectrophotometer (Bio Rad Digilab Co.), and ^{13}C nuclear magnetic resonance (NMR) spectra obtained with a JNM-EX270W spectrometer (Japan Electron Co.) were also used for the chemical analysis of the powder.

The standard moulding resin powder's reactivity towards polar resins such as *o*-cresol Novolak epoxy resin (ESCN 195 XF, Sumitomo Chemical Co.) and phenol resin (BRG 556, Showa Kobunshi Co.) was measured by a gravimetric method. The moulding resin powder (average particle size, 16 μm; surface area, 1.9 $m^2 g^{-1}$) and the epoxy resin were mixed in the weight ratio of 20 : 1 in a solvent (acetone), dried at room temperature for 1 day and then heated at 170 °C for two time periods (2.5 and 5.0 h). The resulting samples were washed twice with a solvent (mixing a 5 g sample with 50 ml of acetone, stirring and centrifuging) to remove the unreacted eopxy resin, dried at 120 °C for 2 h and then measured for the resulting weight loss, which is the unreacted epoxy resin's weight. Finally, the reaction ratio of the moulding resin powder with the epoxy resin was obtained by evaluating the ratio of the reacted epoxy resin's weight to the originally added epoxy resin's weight and was then compared with that of the fused silica powder (average particle size, 16 μm; surface area, 2.0 $m^2 g^{-1}$; Denki Kagaku Co.) with the epoxy resin treated in the same way. The same experiment was also performed using the phenol resin in place of the epoxy resin.

2.2.2. *Mouldability of moulding resin in the moulding process*

To determine the mouldability of recycled moulding resin in the moulding process of electronic components, the resin's flow property in the metal mould (spiral flow) and Barcol hardness of the moulded resins were measured under a curing condition of 170 °C for 2 min according to EMMI normalization and JIS K 6911 method, respectively. Also, burr, which is penetrating resin in the air vent (depth, about 100 μm) in the metal mould used for a 16-pin DIP-type IC package, was measured for its maximum length. The gelation time for the resins was determined at 170 °C.

2.2.3. *Reliability of moulding resin*

The reliability of the recycled moulding resins was measured in terms of moisture resistance and thermal impact resistance using a 16-pin DIP-type IC package. A silicon test chip (4.5 mm × 9.0 mm) with an aluminium test circuit (width, 10 μm; interval, 10 μm) was mounted on a lead frame made of Alloy 42 and moulded into the package with the moulding resins under a curing 170 °C for 2 min. The package was after-cured at 170 °C for 6 h. The moisture resistance was determined using a pressure-cooker test, which measures the ratio of defective packages with opened circuits after boiling at 125 °C under 2.3 atm. The thermal impact resistance was determined using a temperature-cycling test, which measures the ratio of defective packages with outside cracks after temperature cycles over a range from − 60 to + 150 °C at a frequency of 3 cycles h^{-1}.

2.2.4. General properties of moulded resin sample

The strength properties of the moulded resin samples, such as the tensile strength, flexural strength and Charpy impact strength, were measured as specified in JIS K 7111 and JIS K 7203. Their thermal expansion ratio was tested at a heating rate of 5 °C min^{-1} by the method specified in JIS K 7197, the volume resistance was measured according to JIS K 6911, the surface hardness was measured by comparison with pencil hardness, and the thermal deformation temperature was determined as specified in JIS K 7207. The viscosity of the resin samples before adding the hardener was measured, at a rotation rate of 2 rev min^{-1}, on a type BH-7 viscometer (Tokyo Keiki Co.). The adhesive strength of the resin samples for the ferric board (SPCC-SB) was measured by tearing the specimens according to JISK 6850 method.

3. Results and discussion

3.1. Basic properties of cured epoxy resin compound (moulding resin) powder

The chemical structure of the pulverized cured epoxy resin compound for standard moulding resin (moulding resin powder) was studied. As shown by the SEM photograph in Fig. 2 and the carbon distribution in the EPMA photograph in Fig. 3, the moulding resin powder consisted mainly of silica powder and cured epoxy resin, and the silica surface was partly covered with the resin. FT–IR spectrum analysis indicated that the moulding resin powder has silanol groups from the silica in the resin. However, the amount of silanol groups was smaller than that in the original silica powder as shown by the difference spectrum between the powder and the original silica powder (Fig. 4). Therefore, some of the silica powder in the moulding resin powder was found to react with the epoxy resin. The ^{13}C NMR spectrum of the moulding resin powder, which was almost the same as that of the low-stress-type moulding resin powder shown in later Fig. 8a, indicated that the powder has epoxy groups from the epoxy resin unreacted with the phenol resin hardener and also hydroxyl groups resulting from the reaction between the epoxy resin and the hardener.

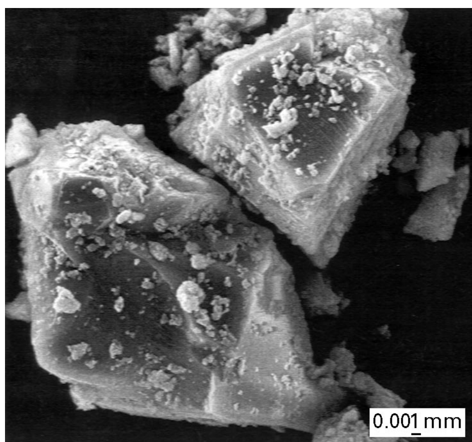

Figure 2 SEM photograph of cured epoxy resin powder (moulding resin powder).

From these results, the chemical structure of the moulding resin powder is proposed to be as shown in Fig. 5. The powder can react with polar resins because of these reactive functional groups (epoxy, hydroxyl and silanol).

The reactivity of the moulding resin powder towards polar resins such as epoxy resin and phenol resin was measured by the gravimetric method and also compared with that of the fused silica powder toward these polar resins. Fig. 6 shows the reaction ratios of the moulding resin powder and the fused silica powder with the epoxy resin, and Fig. 7 shows the same with the phenol resin. The moulding resin powder maintained a good level of reactivity towards the epoxy resin but was a little less reactive than the

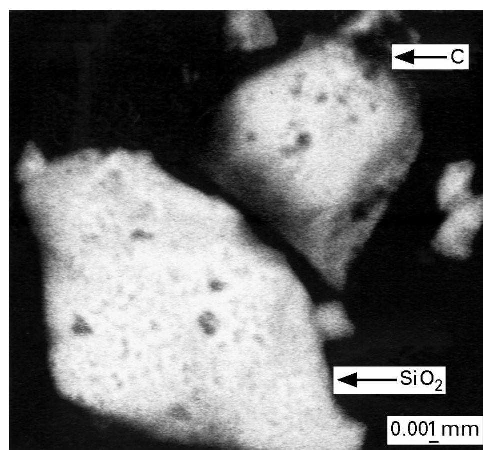

Figure 3 EPMA photograph of moulding resin powder. The white part is the Si in silica particles, and the black part on the silica particle is C in the resin.

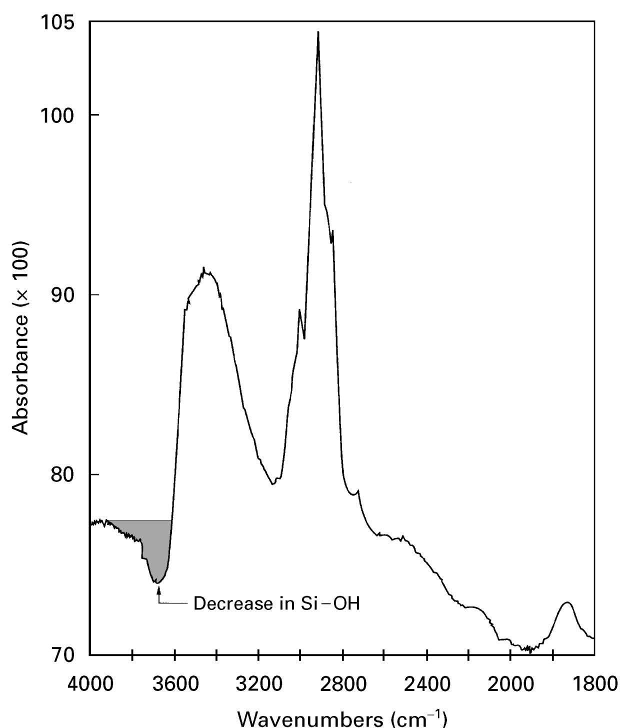

Figure 4 Difference FT–IR spectrum between the moulding resin powder and silica powder (silica powder spectrum – moulding resin powder spectrum).

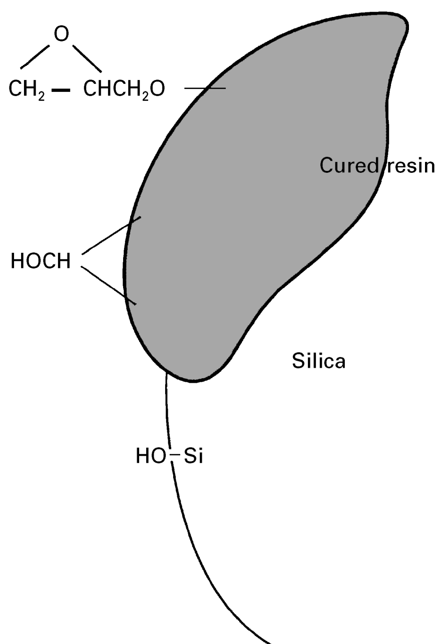

Figure 5 Chemical structure of the moulding resin powder. The powder consisted of silica particles partly covered with cured resin and has functional groups (epoxy, silanol and hydroxy).

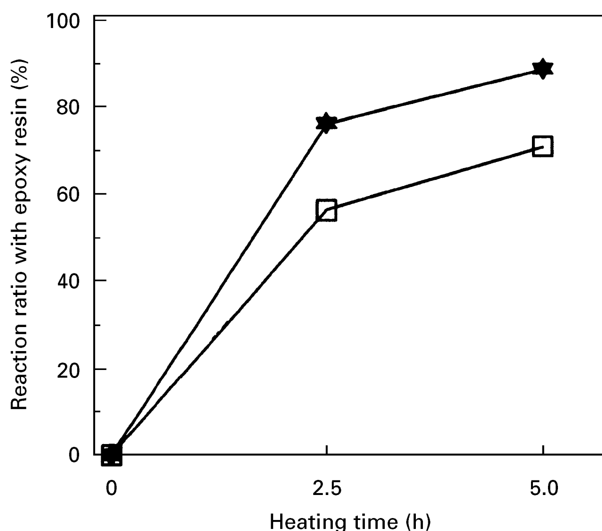

Figure 6 Reactivity of moulding resin powder (□) and silica powder (★) with epoxy resin.

original silica powder. This was probably because the moulding resin powder has fewer silanol groups than the original silica powder has; these groups effectively reacts with the epoxy groups in epoxy resin. On the other hand, the moulding resin powder was a little more reactive with the phenol resin than the original silica powder was. This was mainly due to the remaining epoxy groups in the moulding resin powder; these groups show good reactivity towards hydroxy groups in the phenol resin.

These results show that moulding resin powders can sufficiently bind to polar resins such as epoxy resin and phenol resin by reacting with them in a similar manner to that in which silica powder reacts with them.

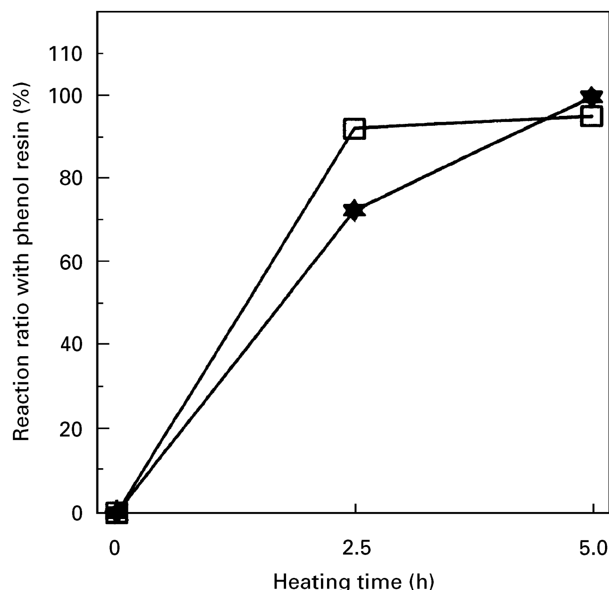

Figure 7 Reactivity of moulding resin powder (□) and silica powder (★) with phenol resin.

3.2. Recycling into original moulding resin

Moulding resins are broadly classified into two types: standard resins used for general IC packages and low-stress-type resins used for large scale integrated-circuit packages. The latter resins contain elastomers represented by silicone compounds in addition to the components of the standard resins in order to reduce the stresses which are generated in the interface between the moulding resin and the IC chip owing to differences between their elastic modulus and thermal expansions. Table III lists the properties of these model moulding resins and indicates that the thermal impact resistance of low-stress-type resin was better than that of the standard resin. This property is mainly affected with the interface stress.

Table III also shows the properties of the recycled resins obtained by recycling these two moulding resin powders into the original standard moulding resin. When the standard resin powder was recycled into the original standard resin at a recycling ratio of 10 wt%, the flow property of the recycled resin in the moulding process was considerably reduced (68 → 22 cm). Using an epoxy resin with a melting temperature lower (55 °C) than that of the original epoxy resin (70 °C) for the recycled resin improved the flow property, but the moisture resistance and thermal impact resistance of the recycled resin were inferior to those of the original standard resin. This is probably due to insufficient solubility between the resin components and the solid components in the recycled resin by adding the cured moulding resin powder.

When the low-stress-type moulding resin powder was recycled into the original standard resin, the flow property was also insufficient. By using a spherical silica powder (average powder size, 20 μm) which was mixed into the original pulverized silica at 50 wt%, in addition to using the above-mentioned epoxy resin with a low melting temperature, the flow property of the resulting product was improved to a sufficient

TABLE III Properties of original moulding resin and recycled moulding resin

Property	Original moulding resin		Recycled moulding resin		
	Standard resin	Low-stress-type resin	Standard resin[a] with standard resin powder (10 wt%)	Standard resin[b] with low-stress-type resin powder (25 wt%)	Standard resin with low-stress type resin powder heated at 170 °C for 8 h
Mouldability					
Flow (cm)	69	73	68	47	61
Barcol hardness	74	58	58	55	62
Burr (mm)	0.6	1.0	1.3	1.3	2.3
Strength properties					
Flexural strength (kgf mm^{-2})	14.7	14.2	17.3	15.8	14.2
Flexural modulus (kgf mm^{-2})	1418	1240	1505	1391	1353
Charpy impact strength (kgf mm^{-1})	2.3	2.3	2.8	3.6	2.7
Thermal mechanical properties					
Glass transition temperature (°C)	155	171	139	154	154
Thermal expansion coefficient α_1/α_2 ($10^{-5} \times$ °C^{-1})	2.0/6.5	1.8/7.4	2.2/6.3	2.0/6.6	2.1/6.3
Moisture resistance reliability					
(Defected package ratio), 120 °C, 2.3 atm, 200 h	0/10	0/10	0/10	0/10	0/10
520 h	1/10	0/10	4/10	0/10	0/10
Thermal impact resistance reliability					
(Defected package ratio), 150 °C–60 °C, 100 cycles	3/10	0/10	7/10	0/10	0/10
150 cycles	5/10	0/10	9/10	0/10	0/10
200 cycles	8/10	0/10		1/10	0/10

[a] An epoxy resin with a low melting temperature was used.
[b] An epoxy resin with low melting temperature and a spherical silica (50 wt% of total silica) were used.

level up to the recycling ratio of 25 wt%. Furthermore, heating the moulding resin powder (at 170 °C for 8 h) improved the flow property. The ^{13}C NMR spectra of the low-stress-type moulding resin powder before and after heating show that the amount of the epoxy groups in the powder decreased by heating (Fig. 8). This heating effect is therefore due to the lowering of the moulding resin powder's surface reactivity, which influences the main factors (viscosity and thixotropy) for the flow property of the resulting products. Moreover, the thermal impact resistance of the resulting product was found to be far better than that of the original standard resin. This improvement was mainly caused with the silicone elastomer in the low-stress-type resin powder.

From these results, it was concluded that there is a distinct possibility of using the residue of low-stress-type moulding resins to make a new moulding resin that has thermal impact resistance higher than original standard moulding resins and better cost performance.

3.3. Use as a filler for epoxy resin products

The present study examined using the standard moulding resin powder as a filler for epoxy resin products, such as insulating materials, paints and adhesives. Since these products use epoxy resins similar to that in the moulding resin powder, strong affinity between the matrix epoxy resin and the moulding resin powder is expected.

3.3.1. Filler for insulating materials

The mouldability in the moulding process, strength and insulating property of epoxy resin-type insulating material with the moulding resin powder were compared with those of the reference material with a fused silica filler. Table IV lists the properties of the resulting material. The flexural strength and insulating property of the moulded material with the moulding resin powder were almost the same as those of the material with the silica powder. The flow property of the material with the powder in the moulding process was not as good as that of the material with the silica powder but was sufficient for moulding. As will be discussed later, this difference is because the volume of moulding resin powder is higher than that of silica powder.

The effects of treating the moulding resin powder's surface was also examined by using silane coupling agents that are usually utilized for improving and adjusting the surface reactivity of inorganic fillers. The surface treatment of moulding resin powder with the coupling agents, especially aminopropyltriethoxysilane (amino silane) improved the strength and insulating property of the moulded material with the powder. This is caused by the improvement in the interface adhesion between the matrix epoxy resin and the moulding resin powder by treating with the silane. Treating the moulding resin powder with γ-glycidoxypropyltrimethoxysilane (epoxy silane) had little effect on the strength and insulating property but improved the flow property in the moulding process of the resulting material. These different effects of these

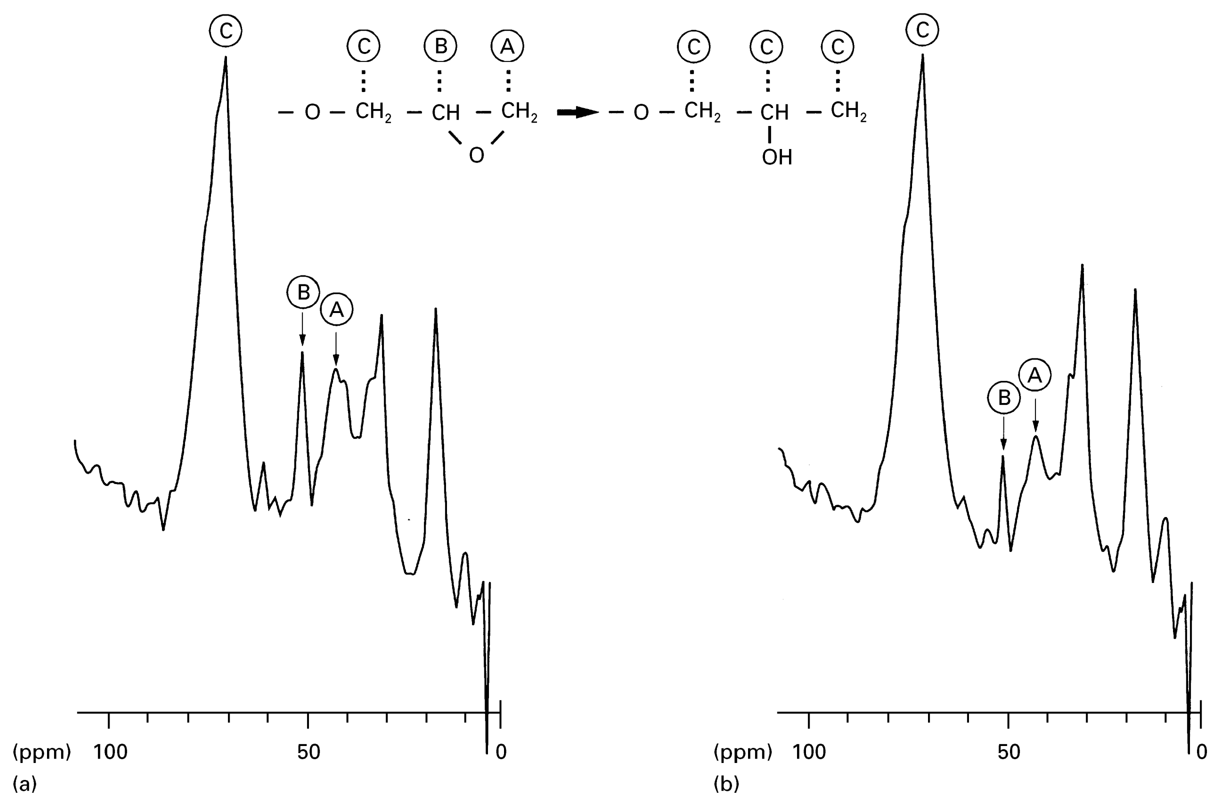

Figure 8 ^{13}C NMR spectrum of moulding resin powder: (a) before heating; (b) after heating.

TABLE IV Properties of epoxy resin compounds with moulding resin powders for insulating materials

Property	Silica powder 60 wt%	Moulding resin powder 60 wt%	Moulding resin treated with amino silane 0.5 wt%	Moulding resin treated with amino silane 1.0 wt%	Moulding resin treated with epoxy silane 1.0 wt%
Mouldability					
Flow (cm)	199	76	78	75	89
Gell time (s)	105	72	67	70	68
Burr (mm)	7.2	1.0	0.8	0.5	0.3
Strength property					
Flexural strength (kgf mm^{-2})	14.6	14.8	16.1	16.1	14.0
Flexural modulus (kgf mm^{-2})	1057	696	722	742	678
Insulation resistance					
at 150 °C (10^{13} Ω cm)	4.0	2.0	24	22	4.0
After boiling at 100 °C for 24 h (10^{14} Ω cm)	14	15	32	57	16
Glass transition temperature (°C)	141	142	138	143	141
Thermal expansion coefficient α_1/α_2 (10^{-5} °C^{-1})	2.8/8.6	3.9/11	4.1/11	4.1/11	4.1/11

silanes were mainly attributed to the lower reactivity of the epoxy silane towards the moulding resin powder and the matrix resin, and the higher plasticity of the silane.

3.3.2. Filler for paints and adhesives

The strength and thermal expansion properties of the epoxy resin compound with the moulding resin powder were compared with those of compounds with conventional fillers such as calcium carbonate powder, talc powder and silica powder. Table V shows the results. The strength of moulded compound with the moulding resin powder was comparable with that of compounds with silica powder and was superior to those of compounds with calcium carbonate powder and talc powder. The thermal expansion of moulded compound with the moulding resin powder was comparable with that of compounds with these conventional fillers. These results were mainly caused by ensuring that the moulding resin powder contains silica filler and by good binding between the moulding resin powder and the matrix epoxy resin.

In addition, the viscosity of the compounds with the moulding resin powder and the conventional fillers was measured before adding the hardener. As also

TABLE V Properties of epoxy resin compounds with moulding resin powder for paints and adhesives

Property	None	Moulding resin powder	Silica powder	Calcium carbonate powder	Talc powder
Average particle size of filler (μm)	—	16	10	9.2	11
Viscosity (Pa s)	2.5	48	14	12	206
Flexural strength (kgf cm^{-2})	817	800	787	610	577
Charpy impact strength (kgf cm^{-1})	2.3	2.4	1.6	2.5	2.5
Tensile strength (kgf cm^{-2})	440	474	428	336	280
Thermal expansion (10^{-5} C^{-1})	13	11	11	14	9.5
Adhesive strength (kgf cm^{-2})	183	161	151	172	138

TABLE VI Properties of acrylic-resin-type construction materials with moulding resin powder

Property	Acrylic resin + aluminium hydroxide (65 wt%)	Acrylic resin + moulding resin powder (less than 1 mm; 10 wt%)	Acrylic resin + moulding resin powder (less than 1 mm; 20 wt%)	Acrylic resin + moulding resin powder (less than 5 mm; 10 wt%)
Flexural strength (kgf cm^{-2})	550	450	430	360
Charpy impact strength (kgf cm cm^{-2})	1.5	1.6	1.7	1.6
Tensile strength (kgf cm^{-2})	270	290	290	230
Thermal deformation temperature (°C)	94	93	93	93
Surface hardness	6	7	8	7

listed in Table V, the viscosity of compound with the moulding resin powder was higher than those of compounds with calcium carbonate powder and silica powder and was lower than that of compound with talc powder. This was mainly due to the difference in filler volumes resulting from the difference in the specific gravities (talc, 1.8; moulding resin, 1.9; silica, 2.2; calcium carbonate, 2.9) and also the structures of these fillers. This effect on the viscosity indicates that the moulding resin powder is a suitable filler for improving paintability by preventing dripping.

Table V also shows the adhesive strength of the epoxy resin compounds with the moulding resin powder and the above-mentioned conventional fillers for the ferric board. The adhesive strength of the cured compound with the moulding resin powder was almost equal to that of the cured compounds with these conventional fillers.

These results show that the moulding resin powder is a usable filler for epoxy resin products such as insulating materials, paints and adhesives.

3.4. Use as a decorating agent for an acrylic-resin-type construction material

Since the moulding resin is carbon blackened, the standard moulding resin powder was tested as a decorating agent for an acrylic-resin-type construction material.

As shown in Fig. 9, the moulded construction material including 10 wt% of the moulding resin powder (maximum particle size, 1 mm) has a good appearance, looking rather like a certain type of marble. The moulding resin powder in the moulded material also had high stability during cutting the material because of the strong binding between the powder and the matrix acrylic resin.

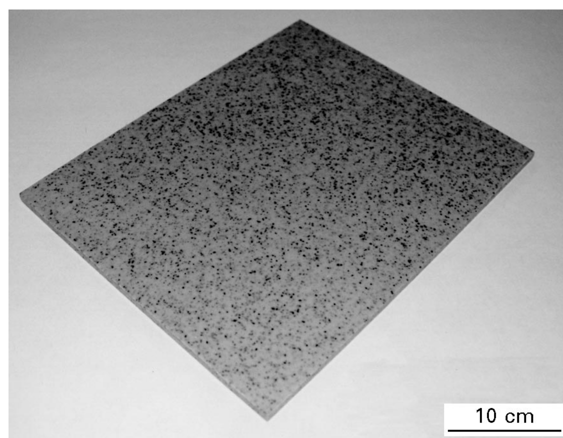

Figure 9 Acrylic-resin-type construction material with moulding resin powder. The black parts are moulding resin powders.

Table VI lists the properties of moulded materials with the powders (maximum particle sizes, 1 and 5 mm). The strength of materials with the powders were not as good as that of the original material, particularly when large-sized powder was used, but they are sufficient for the construction materials to be used according to JIS regulations. The thermal deformation temperature of materials with the powders was comparable with or superior to that of the original material. The surface hardness of materials with the powders was higher than that of the original material. This improvement is due to the hard silica filler in the moulding resin.

This indicates that the moulding resin powder can be a useful decorating agent for resin-type construction materials.

4. Conclusion

Recycling cured epoxy resin compound containing silica filler and additives used as moulding resin for electronic components was studied. The following results were obtained.

1. The moulding resin powder (cured epoxy resin powder) was reactive with functional groups (epoxy, hydroxyl and silanol). The powder reacted with polar resins such as epoxy resin and phenol resin in a similar way to the original silica powder.

2. Recycling a standard moulding resin powder into the original standard resin did not provide a product with sufficient resistance to moisture and thermal impact. On the other hand, when a powder of low-stress-type moulding resin containing a silicone elastomer was recycled into the standard moulding resin, the resulting product had far better thermal impact resistance than that made from the original standard resin.

3. Using the moulding resin powder as a filler for epoxy resin products such as insulating materials, paint and adhesives was successful. The strength, adhesive and insulating properties of the resulting products were similar to those of products with the conventional fillers.

4. The cured moulding resin powder, which is carbon blackened, was also shown to be a useful decorating agent for an acrylic-resin-type construction material. The powder produced an appearance much like that of a certain type of marble, sufficiently maintained the strength and thermal stability of the material and also improved its surface hardness.

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