Electrically conducting powder filled polyimidesiloxane

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Electrically conducting powders (nickel, silver and graphite) were used as fillers in polyimidesiloxane to form polymer-matrix composites with low electrical resistivity and low thermal expansion. Nickel particles of size 1–5 μm provided composites of electrical resistivity almost as low as that provided by silver particles. Furthermore, nickel-filled composites had superior electromagnetic interference shielding effectiveness, lower thermal expansion and superior mechanical properties, which were attained at 40 weight % Ni. Nickel particles of size 78–127 μm were far less effective than those of size 1–5 μm. These composites may be used in electronic packaging.

Key words: *composite materials; particulate fillers; electrical resistivity; EMI shielding effectiveness; thermal expansion coefficient; mechanical properties; polyimidesiloxane*

Traditionally, polymer-matrix composite materials have been thought of primarily as insulating materials and, as such, they have been used in applications like power tool handles, cable jackets, capacitor films and electronic packaging materials. More recently, however, there is a growing market need for polymer-matrix composite materials with increased electrical conductivity. Electrically conductive composites can be used as a replacement for metals in applications such as business machine housings, medical electronics, die attach adhesives, solders and automotive parts. The flexible characteristic and easy manufacturing processes of polymer-matrix composites make these materials attractive.

Electrically conductive polymer-matrix composites typically comprise electrically insulating polymer matrices and electrically conductive fillers. The conductive fillers may be particulate or fibrous. The electrical conductivity of a polymer-matrix composite material is related to the filler volume fraction, size, shape and structure. A critical filler volume fraction is necessary to achieve low electrical conductivity, which is a consequence of the formation of a continuous electrical path through the composite material. Electrically conductive polymer-matrix composite materials not only can be used as electrically conducting materials, but they can also be used as shielding materials to attenuate electromagnetic interference (EMI) arising from natural and man-made sources. With the rapid growth of the electronics industry, man-made electromagnetic 'noise' has already become a dominant pollution source. Therefore, there is a big market in shielding materials.

Polyimidesiloxane (Occidental Chemical Corporation, SIM–2030M) was chosen as the polymer matrix in this investigation. It is a soluble, fully imidized polyimidesiloxane, i.e., a silicone–imide copolymer (SIM). This new kind of thermoplastic polymer exhibits much higher glass transition temperature (T_g), thermal stability and chemical and flame resistance[1,2]. It also has attractive dielectric properties and processing conditions. In this work, several kinds of electrically conducting powder, including silver, nickel and graphite flakes, were used as the conducting fillers. This paper presents the mechanical properties, electrical conductivities, EMI shielding effectiveness and thermal expansion properties of these three electrically conducting, powder-filled, polyimidesiloxane composite materials.

EXPERIMENTAL METHODS

Materials and sample preparation

Three types of conducting powder were used in this investigation: silver powder from Metz Refining Co in South Plainfield, NJ, USA; nickel powder (two particle sizes) from Atlantic Equipment Engineers in Bergenfield, NJ, USA; and graphite flakes from Nanshu Graphite Mine in Shantong, People's Republic of China. The basic properties of these three powders are shown in Table 1. The SIM–2030M polymer used was a dry fine powder. The glass transition temperature (T_g) is about 220°C. Table 2 lists the basic properties of this polymer.

The completely mixed polymer and filler powders were placed into a matched metal die and then compressed

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Table 1. Properties of conducting powders

Material	Particle size (μm)	Density (g cm^{-3})	Electrical resistivity (Ω cm)	CTE* (10^{-6}K^{-1})
Silver	0.8–1.35	10.49	1.49×10^{-6}	19.2
Nickel	1–5	8.80	6.14×10^{-6}	12.7
	78–127			
Graphite flakes	25.4	2.25	5.0×10^{-3}	27.3 (∥c) −0.5 (⊥c)

* 30–100°C

Table 2. Properties of SIM-2030M polymer

Density	1.25 g cm^{-3}
Particle size	50–100 μm
Tensile strength	39.5±2.0 MPa
Tensile modulus	2.1±0.11 GPa
Elongation at break	(2.1±0.13) %
Electrical resistivity	$> 10^{10}$ Ω cm
Coefficient of thermal expansion	74.4×10^{-6}K^{-1} (30–100°C) 86.4×10^{-6}K^{-1} (30–150°C)

at 5.90 MPa and 300°C for 30 min. For different tests, moulded samples of various shapes were made.

Electrical testing

Electrical resistivity measurements were made on composite materials which had been moulded into bars. The four-probe method was used. Four specimens of each composition were tested. Five data points were obtained for each specimen.

The EMI shielding effectiveness was measured by using the coaxial cable method. The sample was in the form of an annular disc. The outside and inside diameters were 97.4 and 28.8 mm, respectively. The thickness ranged from 2.82 to 2.95 mm. In order to obtain a continuous metallic contact between the sample and the steel shielding tester chamber, conductive silver paint was applied to the inner surface of the centre hole of the sample and the outer rim of the annular disc.

Thermal expansion measurements

A Mettler TMA40 thermal mechanical analyser was used for measuring the coefficient of thermal expansion (CTE). The temperature was scanned from 30 to 170°C and the heating rate was 10°C min^{-1}. The specimens were annealed in a furnace at 200°C for 2 h and then furnace cooled before the measurement. Two specimens of each composition were tested. The CTE values were averaged over two temperature ranges, namely 30–100°C and 30–150°C. The error in the average CTE was ± 5% for 30–100°C and ± 8% for 30–150°C.

Mechanical testing

The specimens were tested mechanically using standard methods and a Materials Testing System (MTS). An IBM-XT computer with OPUS-200 software and a data acquisition board were used for the data collection. For tensile tests, EA–13–120LZ–120 strain gauges (from Measurements Group, Inc) were applied on the specimens to measure the strain. Four specimens of each composition were tested.

RESULTS AND DISCUSSION

Electrical properties

The electrical resistivities of the various composite materials are listed in Table 3. It can be seen that the resistivities are similar for equal loading (in volume %) when the particle size is close. For example, 40 weight % (wt %) silver-filled SIM–2030M has the same level of resistivity as 40 wt % nickel-filled polymer when the filler particle sizes are less than 5 μm. However, for the same kind and content of conductive powder but different filler particle sizes, such as 40 wt % nickel powder of size 1–5 μm and 78–127 μm, the composite with a smaller filler particle size gave a lower resistivity. The critical volume loading required for conductivity is greatly dependent on the particle size ratio, that is, the ratio of the radius of the polymer particle to that of the metal particle. The critical volume loading for a small particle size powder, like nickel or silver, is seen from Fig. 1 to be 6 vol %. This loading is very close to the results of Kusy and Turner[3] when the particle size ratio is equal to or larger than 30. Indeed, the average particle size ratio is 30 for the nickel filler of particle size 1–5 μm. This ratio is even larger for the silver filler. For 78–127 μm nickel-powder-filled polymer, even when a high loading was used, the resistivity was still very high ($3.72 \times 10^{6}\Omega$ cm at 17.6 vol %). This is because a high critical volume loading is required for continuity in the electrical path when the particle size ratio is equal to or less than two[3]. The average particle size ratio is only 0.7 for the nickel filler of particle size 78–127 μm. On the other hand, the graphite-flake-filled polymer attained a relatively low resistivity at a low filler volume content, even though the particle size ratio is approximately three. This is because the particle shape has an important effect on the resistivity. Flakes can form a continuous path more easily than round particles.

The microstructure of silver-, nickel- and graphite-flake-filled polymer materials is shown by micrographs of polished (not etched) surfaces (Fig. 2). It can be seen that the smaller the size of the particles, the easier it is to form a continuous electrically conductive path. The continuity was present in Figs 2(a), 2(b) and 2(d) for silver, small nickel powder and graphite flakes as

Table 3. Resistivity of conducting powder-filled SIM-2030M composite materials

Filler	Particle size (μm)	Filler content Wt %	Filler content Vol %	Resistivity (Ω cm)	
Silver	0.8–1.35	10	1.3	2.42×10^{8}	($\pm 0.28 \times 10^{8}$)
		20	2.9	2.12×10^{8}	($\pm 0.21 \times 10^{8}$)
		30	4.9	0.131	(±0.021)
		40	7.4	2.26×10^{-3}	($\pm 0.17 \times 10^{-3}$)
		50	10.7	9.76×10^{-4}	($\pm 0.75 \times 10^{-4}$)
Nickel	1–5	10	1.6	7.53×10^{8}	($\pm 0.30 \times 10^{8}$)
		20	3.4	5.34×10^{8}	($\pm 0.04 \times 10^{8}$)
		30	5.7	0.155	(±0.005)
		40	8.7	6.75×10^{-3}	($\pm 0.74 \times 10^{-3}$)
		50	12.4	2.31×10^{-3}	($\pm 0.14 \times 10^{-3}$)
Nickel	78–127	10	1.6	2.11×10^{8}	($\pm 0.16 \times 10^{8}$)
		20	3.4	1.78×10^{8}	($\pm 0.18 \times 10^{8}$)
		30	5.7	1.72×10^{8}	($\pm 0.17 \times 10^{8}$)
		40	8.7	1.07×10^{8}	($\pm 0.05 \times 10^{8}$)
		60	17.6	3.72×10^{6}	($\pm 1.45 \times 10^{6}$)
Graphite flakes	25.4	2	1.1	1.19×10^{8}	($\pm 0.16 \times 10^{8}$)
		5	2.8	1.57×10^{6}	($\pm 0.46 \times 10^{6}$)
		10	5.8	1.62×10	($\pm 0.45 \times 10$)
		20	12.2	5.57×10^{-1}	($\pm 0.15 \times 10^{-1}$)

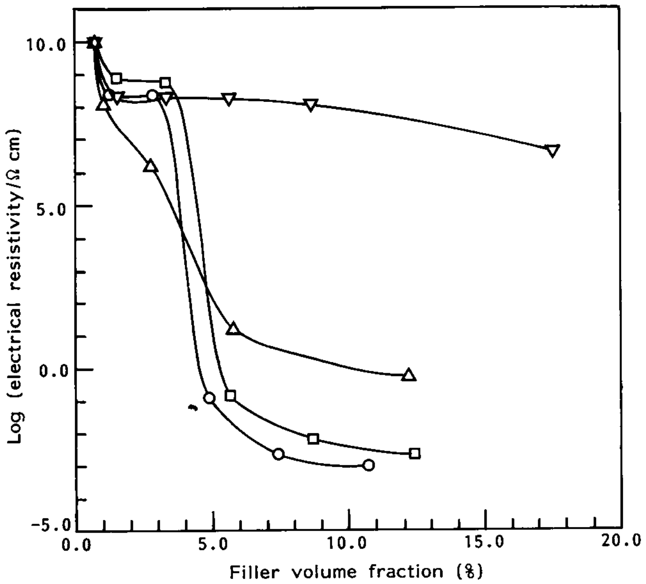

Fig. 1 Variation of the logarithm of the electrical resistivity with filler volume fraction: ○, silver; □, nickel (1–5 μm); ▽, nickel (78–127 μm); △, graphite flakes

fillers. It was absent in Fig. 2(c) for large nickel powder as the filler.

The EMI shielding effectiveness for the silver- and nickel-powder-filled polymer is presented in Table 4 for filler loadings of 30 and 40 wt %. A shielding effectiveness of 20 to 30 dB (99 to 99.9 percent attenuation) would meet 50 percent of the need of the electronics industry, while one of 30 to 40 dB (99.9 to 99.99 percent attenuation) would meet 95 percent of the requirements[4,5]. From Table 4, it can be seen that the 40 wt % nickel-filled polymer can meet almost all requirements of the electronics industry because its EMI shielding effectiveness is about 50 dB in the frequency range of 1.0 to 2.0 GHz.

For reducing electromagnetic waves with a metallic shield, reflection and absorption are the main mechanisms. The absorption part of the attenuation of high frequency electromagnetic fields is proportional to the thickness of the sample. It also increases with increasing frequency and increasing conductivity of the shielding material. The reflection part decreases with increasing frequency and increases with increasing

Table 4. EMI shielding effectiveness (dB) of nickel- and silver-powder-filled SIM-2030M composite materials

Filler	Filler content Wt %	Filler content Vol %	Thickness (mm)	Frequency (GHz) 1.0	1.1	1.2	1.3	1.4	1.5	1.6	1.7	1.8	1.9	2.0
Nickel	30	5.7	2.85	42.5	40.5	39.0	37.5	38.5	37.5	39.0	41.5	43.2	45.0	44.0
1–5 (μm)	40	8.7	2.95	50.0	>50	>50	>50	>50	>50	>50	>50	>50	>50	>50
Silver	30	4.9	2.82	2.6	3.2	2.7	1.9	0.9	1.4	0.7	0.5	0.5	0.4	1.2
0.8–1.35 μm	40	7.4	2.85	8.7	8.2	7.4	6.2	5.8	6.2	7.3	9.2	10.5	12.1	12.5

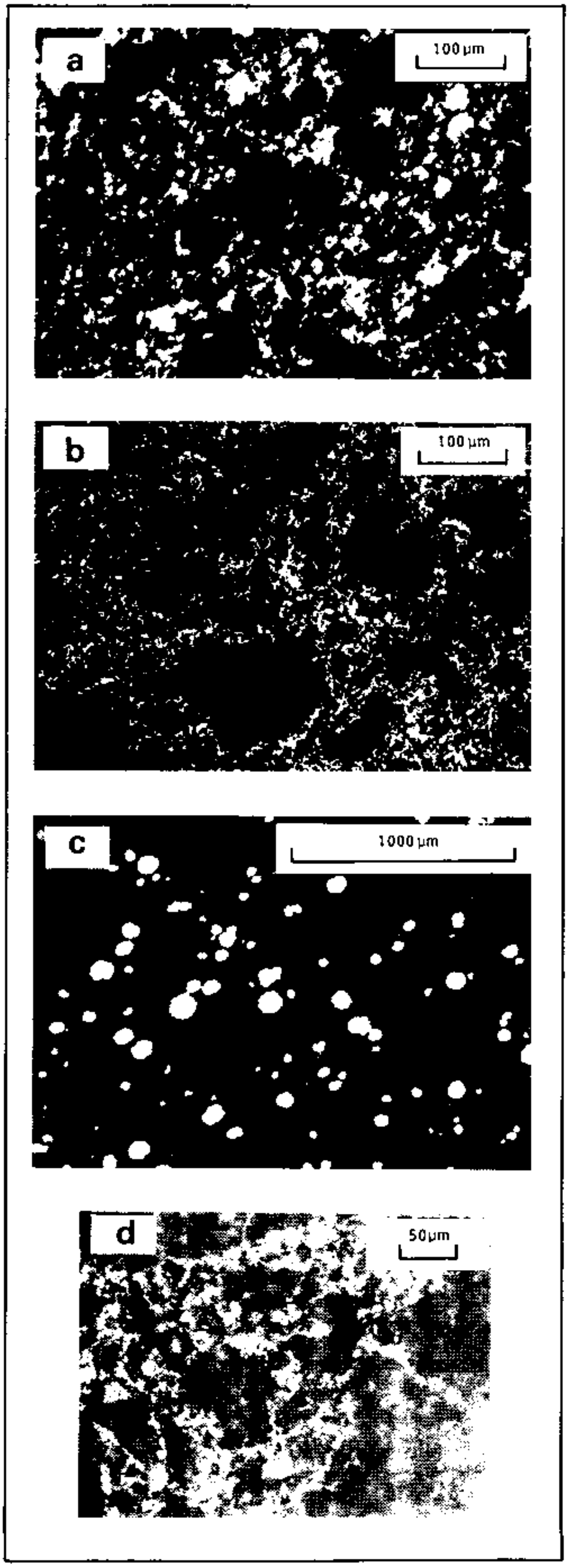

Fig. 2 Microscope photographs of polished sections of composites containing: (a) 40 wt % silver powder; (b) 40 wt % nickel powder (1–5 µm); (c) 40 wt % nickel powder (78–127 µm); and (d) 20 wt % graphite flakes

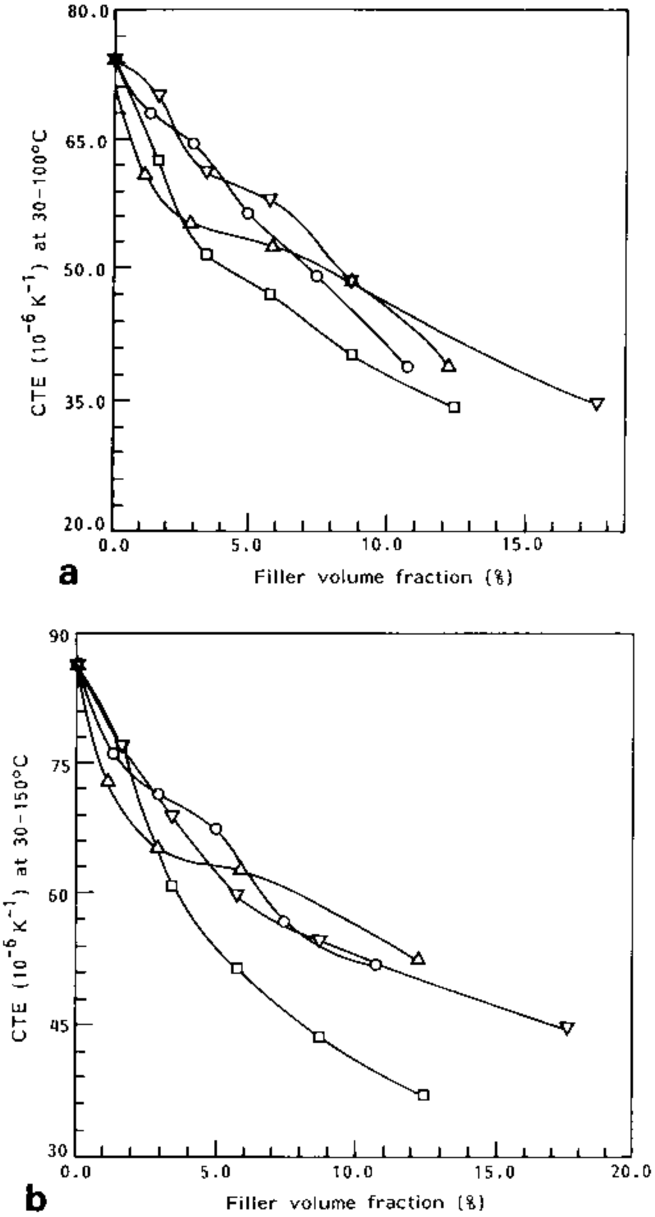

Fig. 3 Variation of the coefficient of thermal expansion (CTE) with filler volume fraction: ○, silver; □, nickel (1–5 µm); ▽, nickel (78–127 µm); △, graphite flakes. (a) CTE averaged at 30–100°C, (b) CTE averaged at 30–150°C

conductivity of the material. For the polymer filled with 40 wt % nickel powder (1–5 µm), both absorption and reflection act as important mechanisms for the EMI shielding because the composite has a low electrical resistivity (6.75×10^{-3} Ω cm). For the silver-powder-filled polymer, it is unclear why the EMI shielding effectiveness is lower than that of the nickel-powder-filled polymer even though its electrical resistivity is lower than that of the nickel-filled polymer.

Thermal expansion properties

The coefficients of thermal expansion (CTE) of silver-, nickel- and graphite-flake-filled polymer materials are shown in Fig. 3. The CTE was dramatically decreased with increasing filler volume loading. For nickel particles of the two sizes (1–5 µm and 78–127 µm), the

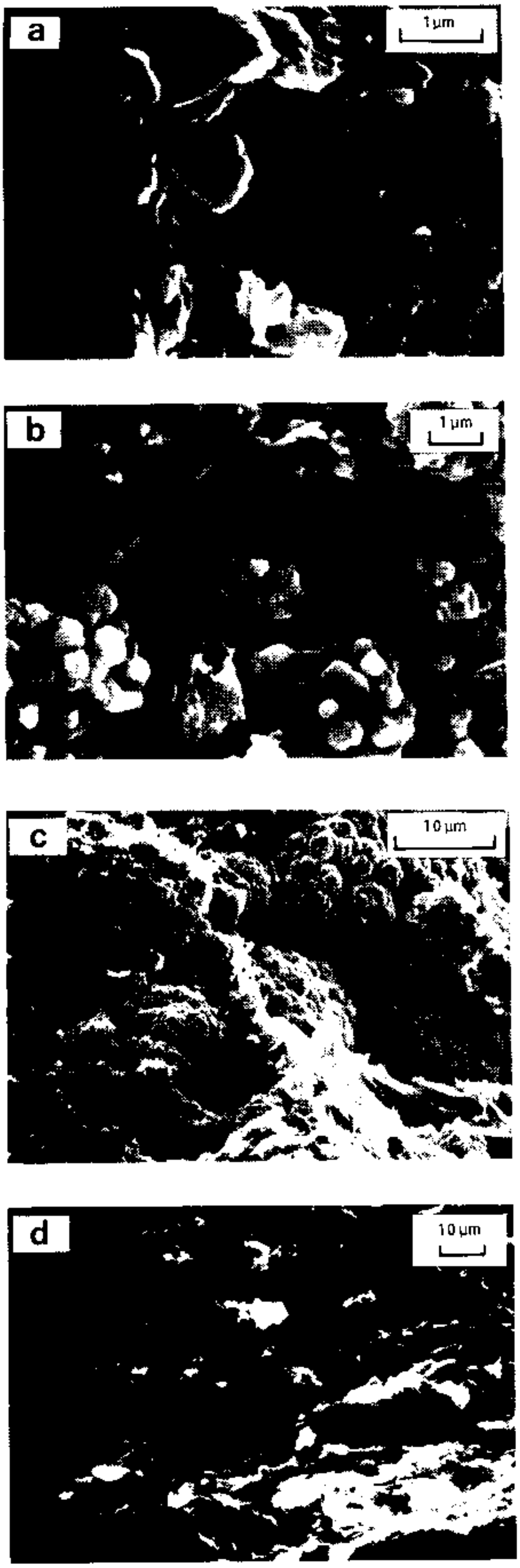

Fig. 4 SEM photographs of tensile fracture surfaces of composites containing: (a) 40 wt % silver powder; (b) 40 wt % nickel powder (1–5 µm); (c) 40 wt % nickel powder (78–127 µm); and (d) 20 wt % graphite flakes

smaller particles gave a lower CTE than the bigger particles at the same volume loading. The particle size had been reported to affect indirectly the thermal expansion through adhesion quality[6]. The particle properties, such as the particle shape, surface energy, etc., also play a role. It is well known that low thermal expansion, conducting polymer composite materials can be technologically used as die attach adhesives and electronic packaging materials. From Fig. 3, it can be seen that nickel (1–5 µm) filled polymer exhibits the lowest CTE.

Mechanical properties

The mechanical properties, including tensile strength, modulus and elongation at break, of conducting powder-filled polymer composite materials are given in Table 5. For every filler, with the increase of the filler content, the modulus increased and the elongation at break decreased. For the silver-powder-filled polymer, the tensile strength increased as the silver content was increased from 0 to 1.3 volume % (vol %) and then decreased with further increase in the silver content. The tensile fracture surface was observed using scanning electron microscopy (SEM), as shown in Fig. 4(a). Near the centre of this photograph (in the upper left quadrant) is a silver particle. The surface of the silver particle is quite smooth and the contact between the particle and the polymer matrix is not tight. The poor interfacial bonding is the origin for the degradation of the tensile strength for silver contents beyond 1.3 vol %.

For the polymer filled with nickel powder (either particle size), the tensile strength increased with increasing filler volume fraction from 0 to 8.7 vol %, though it decreased when the volume fraction was further increased. This behaviour is more attractive than that of the silver-powder-filled polymer and arises because of the rough surface of the nickel powder. Figs 4(b) and 4(c) show the tensile fracture surfaces of 40 wt % nickel-filled polymer composites of two different particle size ranges. These photographs show that the surfaces of Ni particles of both particle size ranges are rough.

The graphite-flake-filled polymer was found to be the most brittle of the three conducting, powder-filled polymer composite materials, even at a low filler volume fraction. The tensile strength decreased monotonically as the filler volume fraction was increased from 0 to 12.2 vol %. This is due to the poor wetting of the graphite flakes by the polymer. Fig. 4(d) shows the tensile fracture surface, which reveals a flaky microstructure.

Comparison between nickel powder and nickel fibres

The small nickel powder (1–5 µm) is the most attractive particulate filler among all those investigated here for obtaining SIM composites of low electrical resistivity, high EMI shielding effectiveness, low thermal expansion and good mechanical properties. The attraction of

Table 5. Mechanical properties of conducting-powder-filled SIM-2030M composite materials

Filler	Size (μm)	Filler content		Density (g cm^{-3})	Tensile strength (MPa)	Modulus (GPa)	Elongation (%)
		Wt %	Vol %				
Silver	0.8–1.35	0	0	1.25	39.5 (±2.0)	2.11 (±0.11)	2.10 (±0.13)
		10	1.3	1.35	49.6 (±1.7)	2.31 (±0.10)	2.55 (±0.12)
		20	2.9	1.48	47.5 (±0.7)	2.39 (±0.03)	2.28 (±0.11)
		30	4.9	1.64	46.7 (±1.4)	2.42 (±0.05)	2.10 (±0.06)
		40	7.4	1.91	42.5 (±0.8)	3.14 (±0.11)	2.01 (±0.07)
		50	10.7	2.19	39.5 (±1.9)	3.16 (±0.09)	1.91 (±0.07)
Nickel	1–5	0	0	1.25	39.5 (±2.0)	2.11 (±0.11)	2.10 (±0.13)
		10	1.6	1.39	40.2 (±0.8)	2.27 (±0.10)	1.94 (±0.11)
		20	3.4	1.52	45.4 (±1.9)	2.46 (±0.06)	2.05 (±0.19)
		30	5.7	1.65	49.9 (±1.5)	2.67 (±0.04)	2.28 (±0.06)
		40	8.7	1.88	51.2 (±1.5)	2.98 (±0.03)	2.08 (±0.09)
		50	12.4	2.19	45.4 (±3.0)	3.54 (±0.15)	1.60 (±0.19)
	78–127	0	0	1.25	39.5 (±2.0)	2.11 (±0.11)	2.10 (±0.13)
		10	1.6	1.40	41.0 (±1.6)	2.23 (±0.11)	2.20 (±0.15)
		20	3.4	1.59	44.7 (±1.4)	2.30 (±0.08)	2.52 (±0.10)
		30	5.7	1.61	49.3 (±1.4)	2.39 (±0.07)	2.89 (±0.16)
		40	8.7	1.77	50.9 (±1.3)	2.48 (±0.07)	3.05 (±0.13)
		60	17.6	2.56	36.5 (±2.1)	2.68 (±0.14)	1.79 (±0.20)
Graphite flakes	25.4	0	0	1.25	39.5 (±2.0)	2.11 (±0.11)	2.10 (±0.13)
		2	1.1	1.26	39.1 (±2.6)	2.41 (±0.14)	1.56 (±0.12)
		5	2.8	1.27	30.5 (±2.4)	2.85 (±0.17)	1.38 (±0.15)
		10	5.8	1.30	27.3 (±2.3)	3.25 (±0.17)	1.35 (±0.10)
		20	12.2	1.32	13.2 (±1.3)	5.10 (±0.51)	0.71 (±0.16)

nickel powder led us to the investigation of nickel fibres.

The nickel fibres were of diameter 20 μm and of lengths 260, 700 and 1000 μm, as kindly provided by National-Standard.

Table 6 shows the electrical resistivity of the nickel-fibre-filled SIM–2030M composite materials compared with those of the small nickel powder (1–5 μm) filled counterpart. The resistivity generally decreased with increasing fibre length for the same fibre content, but, even for the composite with the longest fibres (1000 μm), the resistivity was higher than that with the nickel powder of the same filler content.

Table 6. Electrical resistivity of nickel-fibre-filled SIM-2030M composite materials

Fibre length (μm)	Filler content		Resistivity (Ω cm)
	Wt %	Vol %	
260	30	5.7	3.08×10^4 ($\pm 0.50 \times 10^4$)
	40	8.7	1.15×10^3 ($\pm 0.20 \times 10^3$)
700	30	5.7	3.34×10^3 ($\pm 1.08 \times 10^3$)
	40	8.7	1.32 (±0.02)
1000	30	5.7	3.45×10^2 ($\pm 0.42 \times 10^2$)
	40	8.7	7.09×10^{-2} ($\pm 0.22 \times 10^{-2}$)
1.5 μm powder	30	5.7	0.155 (±0.005)
	40	8.7	6.75×10^{-3} ($\pm 0.74 \times 10^{-3}$)

Table 7 compares the EMI shielding effectiveness of the nickel fibre composites and the nickel powder (1–5 μm) counterpart. The shielding effectiveness generally increased with increasing fibre length for the same fibre content, but, even for the composite with the longest fibres (1000 μm), the shielding effectiveness was lower than that with the nickel powder of the same filler content. This is consistent with the resistivity results.

Table 8 shows the tensile properties of the nickel fibre composites and the nickel powder (1–5 μm) counterpart. The positive effects of fibres (700 and 1000 μm long) and powder on the strength and modulus are comparable and are larger than those of fibres of length 260 μm. On the other hand, fibres of all three lengths have the advantage over the powder in increasing the elongation.

Fig. 5 shows the tensile fracture surface of the composite containing 1000 μm nickel fibres in the amount of 40 wt %. Much fibre pull-out was observed, as shown by the holes and tracks in the matrix left behind by the pulled-out fibres, in addition to the protrusion of fibres from the fracture surface. The fracture surface also shows that the fibres were uniformly distributed in the composite. The fibre pull-out indicates poor bonding between the fibres and the matrix. This explains the fact that the fibres are comparable to the powder in their effectiveness in strengthening the polymer.

Overall, the nickel fibres are not as attractive as the nickel powder in providing composites with low electri-

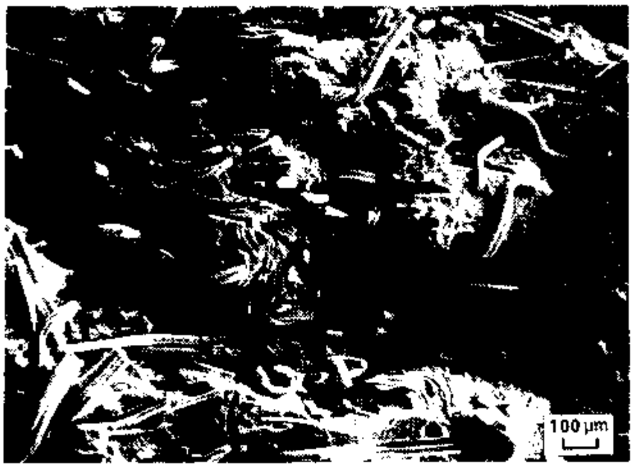

Fig. 5 SEM photograph of tensile fracture surface of composite containing 40 wt % nickel fibres of length 1000 μm

cal resistivity, high EMI shielding effectiveness and good mechanical properties. The poor mechanical effect of the fibres is attributed to the poor bonding between the fibres and the matrix; this poor bonding is probably due to the smooth surface of the fibres compared with the rough surface of the powder particles. The poor electrical effects (both resistivity and EMI shielding) of the fibres are probably due to the large size of the fibres (20 μm diameter) compared with the small size of the powder particles (1–5 μm).

CONCLUSIONS

The electrical, mechanical and thermal expansion properties of the various types of filled SIM polymer show that the small nickel powder (1–5 μm) is the most attractive filler for obtaining SIM composites of low electrical resistivity, high EMI shielding effectiveness, low thermal expansion and good mechanical properties. This combination of properties is desirable for applications such as a die attach adhesive in electronics and for EMI shielding.

In comparison with the nickel fibres, the small nickel powder (1–5 μm) as filler gave SIM composites of lower resistivity and higher EMI shielding effectiveness.

Silver powder as filler gave SIM composites of slightly lower resistivity, much lower EMI shielding effectiveness and higher CTE than composites containing small nickel powder. In addition, the high cost of silver makes nickel even more attractive.

Table 7. EMI shielding effectiveness (dB) of nickel-fibre-filled SIM-2030M composite materials

Fibre length (μm)	Filler content		Thickness (mm)	Frequency (GHz)										
	Wt %	Vol %		1.0	1.1	1.2	1.3	1.4	1.5	1.6	1.7	1.8	1.9	2.0
260	30	5.7	2.92	4.2	7.2	6.0	10.5	3.9	1.5	1.8	0.2	0.1	6.4	0
	40	8.7	2.81	20.3	21.3	19.5	21.0	15.3	12.5	19.6	21.2	26.2	19.8	23.6
700	30	5.7	2.98	15.0	17.5	17.3	22.4	12.5	10.5	15.5	15.5	19.4	14.6	18.7
	40	8.7	2.90	42.5	40.0	37.1	40.5	29.0	26.8	33.5	33.5	42.0	26.6	33.5
1000	30	5.7	2.98	22.0	23.0	20.9	21.8	15.3	12.6	17.6	17.8	21.7	16.4	21.0
	40	8.7	3.00	42.5	43.4	>50	>50	>50	>50	>50	>50	>50	34.0	43.0
1–5 μm powder	30	5.7	2.85	42.5	40.5	39.0	37.5	38.5	37.5	39.0	41.5	43.2	45.0	44.0
	40	8.7	2.95	50.0	>50	>50	>50	>50	>50	>50	>50	>50	>50	>50

Table 8. Tensile properties of nickel-fibre-filled SIM-2030M composite materials

Fibre length (μm)	Filler content		Strength (MPa)	Modulus (GPa)	Elongation (%)
	Wt %	Vol %			
260	0	0	39.5 (±2.0)	2.11 (±0.11)	2.10 (±0.13)
	30	5.7	44.34 (±0.59)	2.57 (±0.09)	2.57 (±0.05)
	40	8.7	46.49 (±0.41)	2.63 (±0.10)	2.69 (±0.05)
700	0	0	39.5 (±2.0)	2.11 (±0.11)	2.10 (±0.13)
	30	5.7	51.00 (±0.60)	2.72 (±0.05)	3.06 (±0.03)
	40	8.7	52.23 (±1.33)	2.79 (±0.01)	2.96 (±0.13)
1000	0	0	39.5 (±2.0)	2.11 (±0.11)	2.10 (±0.13)
	30	5.7	50.06 (±1.11)	2.71 (±0.07)	2.92 (±0.09)
	40	8.7	48.45 (±0.36)	2.48 (±0.02)	3.00 (±0.06)
1–5 μm powder	0	0	39.5 (±2.0)	2.11 (±0.11)	2.10 (±0.13)
	30	5.7	49.9 (±1.5)	2.67 (±0.04)	2.28 (±0.06)
	40	8.7	51.2 (±1.5)	2.98 (±0.03)	2.08 (±0.09)

Compared with the small nickel particles (1–5 μm), the large nickel particles (78–127 μm) are poor as a filler; composites with the large nickel particles have a high electrical resistivity, a large CTE and poorer mechanical properties. This is due to the higher critical volume fraction required for electrical conduction for larger particles as a filler, and the more effective reinforcement provided by the smaller particles.

The graphite flakes are the worst among all the fillers investigated here. Composites containing the graphite flakes have a high electrical resistivity (relative to those containing small nickel powder or silver powder) and the poorest mechanical properties, although the CTE is low (not the lowest). This is related to the large size of the graphite flakes, the poor bonding between graphite and the polymer, and the nearly zero CTE of graphite itself perpendicular to the *c*-axis.

The superior mechanical properties and low CTE of the composites containing the small nickel powder are partly due to the good bonding between the rough surface of the nickel particles and the polymer matrix.

Maiti and Mahapatro[7] found that the tensile strength and modulus of nickel-powder-filled polypropylene (PP) composites were decreased by the presence of the filler. This is due to the nickel powder affecting the crystallinity and crystal size of the polypropylene[7]. However, Kusy and Turner[8] reported that 8 vol % nickel particles of size around 30 μm increased the tensile strength of polyvinyl chloride (PVC) by about 150% and that the nickel particles reduced porosity and crack formation. For nickel particles of size 1–5 μm in SIM, we observed a tensile strength increase of 30% at 8.7 vol % Ni.

El-Tawansi *et al.*[9] and Sandrolini *et al.*[10] discussed the effect of nickel powder content on the electrical resistivity of filled polymers. The particle sizes they used were 44 μm[10] and 50 μm[9]. For the 44 μm nickel-particle-filled polyethylene adipate (PEA), even at 60 wt % loading, the electrical conductivity of the composites was still 10^{12} Ω cm[10]. For 50 μm nickel-particle-filled polystyrene, the electrical resistivity was 1.07×10^{13} Ω cm at 37.5 wt %[9]. These high resistivity values are because large particles require a high critical volume fraction in order to form a continuous electrical path.

For the small nickel particle (1–5 μm) filled SIM, the optimum nickel content is 40 wt % (8.7 vol %). As shown in Table 5, the tensile strength and elongation are decreased when the nickel content is increased from 40 to 50 wt % and the highest strength is achieved at 40 wt %. The degradation of the mechanical properties overshadows the desirable decrease in the electrical resistivity and CTE when the nickel content is increased from 40 to 50 wt %. Hence, SIM containing 40 wt % small nickel particles (1–5 μm) has a superior combination of electrical, mechanical and thermal expansion properties, so that it is technologically attractive for electronic packaging and other applications.

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