Improving the electrical properties of structural epoxy resin adhesives

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The role played by metallic fillers in improving the electrical conductivity of structural epoxy resin adhesives has been investigated by adding powdered filler materials having different particle sizes in different weight percentages. Fibrous fillers, effectively oriented as well as randomly chopped, have also been studied. Results showed that the randomly chopped fibres are superior in improving the electrical conductivity of structural epoxy resin adhesives. However, powdered fillers are better at enhancing the mechanical strength of these adhesives.

Key words: adhesives; electrical resistivity; mechanical strength; aluminium powder; copper wires; copper network

Adhesives are used in the electrical and electronics industry in a variety of ways. These range from holding microcircuits to bonding coils in mammoth electrical generators[1]. In addition to mechanical fastening[2-7], adhesives are required in electrical applications to seal and protect substrates, and conduct or insulate heat and electricity. Since adhesives normally function as good insulators, the objective of the present work was to improve their electrical conductivity.

Fillers which are added to epoxy resin to reduce cost, reduce shrinkage and lower the coefficient of thermal expansion are expected to contribute to both the thermal and the electrical conductivities. The role played by the filler particle size, shape and orientation should be considered. In general, the smaller the particle size, the more easy the filler will be to incorporate and the less tendency for it to settle. The loading volume of the filler affects the handling characteristics, particle wetting and the ultimate properties of the adhesive[8]. Although most fillers reduce the impact resistance[8,9], fibrous fillers were found to preserve the adhesive impact strength. Random chopped fibres were found to be not quite as effective as laid-up continuous fibres, which can be oriented to maximize their contribution to mechanical strength[9].

In the present work, filler materials of a wide range of particle size, including random chopped fibres as well as oriented fibres, were mixed with adhesives at different weight percentages. Their effect on electrical resistivity and mechanical strength was investigated and is reported in this paper.

Experimental details

Materials

The adhesive used throughout the present work was a room-temperature cure, two-part epoxy resin with a mix ratio of 100 parts by weight or volume of resin to 40 parts by weight or volume of hardener. The adhesive had a pot life of 30 min at 23°C and a cure cycle of 24-28 h at 24°C.

Nine filler materials having different particle sizes were used in the present work (see Fig. 1). These fillers can be classified into three groups:

1) *powders* — graphite (5-10 μm), iron (147 μm), cast iron (~100 μm), aluminium (~160 μm), bronze (~100 μm), nickel (~45 μm) and copper (~45 μm);
2) *fibres* — chopped copper wires having a diameter of 0.077 mm and lengths ranging from 0.04-0.08 mm;
3) *network* — brass (60/40) having a mesh number of 0.123.

Test specimens

The dimensions of the test specimens used for measuring electrical resistivity were 9 × 6 × 13 mm. A total of 81 specimens — nine fillers at nine different weight percentages — was prepared and tested.

The mechanical strength of the different mixtures was assessed using a single lap specimen according to British Standard BS 5350[11]. The configuration and dimensions of this specimen are shown in Fig. 2. The adherends were cut from low carbon steel sheets. After

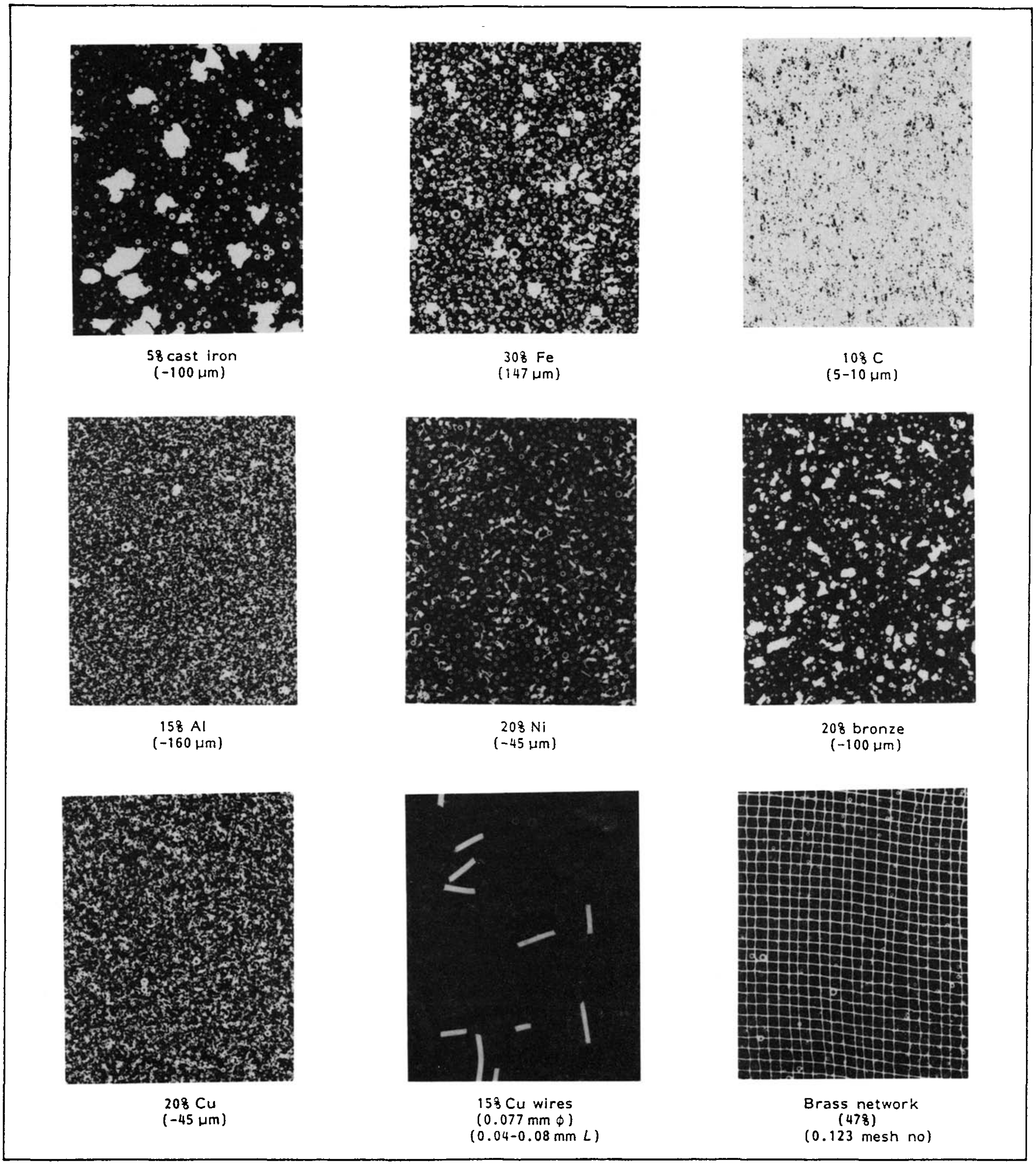

Fig. 1 Photographs showing the effect of filler addition on the structure of the epoxy resin adhesive (percentages are by weight). Dark field, ×13

cutting they were subjected to a stress relief anneal at 650°C for 30 minutes and then abraded using emery papers of different grades down to 400. After the physical treatment the adherends were subjected to a chemical treatment in acetone.

Adhesive bonding

The procedure followed throughout the bonding stage of specimen manufacture was as follows:

1) melting of the adhesive during processing to ensure the wetting of surfaces and to promote contact;
2) removal of unwanted adhesive components such as solvents and water; and
3) application of pressure to the joint during cure to maintain the integrity of the assembly.

Results

Effect of fillers on mechanical strength

Fig. 3 shows the effect of adding different weight percentages of filler on the shear strength of the structural epoxy resin adhesive. It can be seen that the

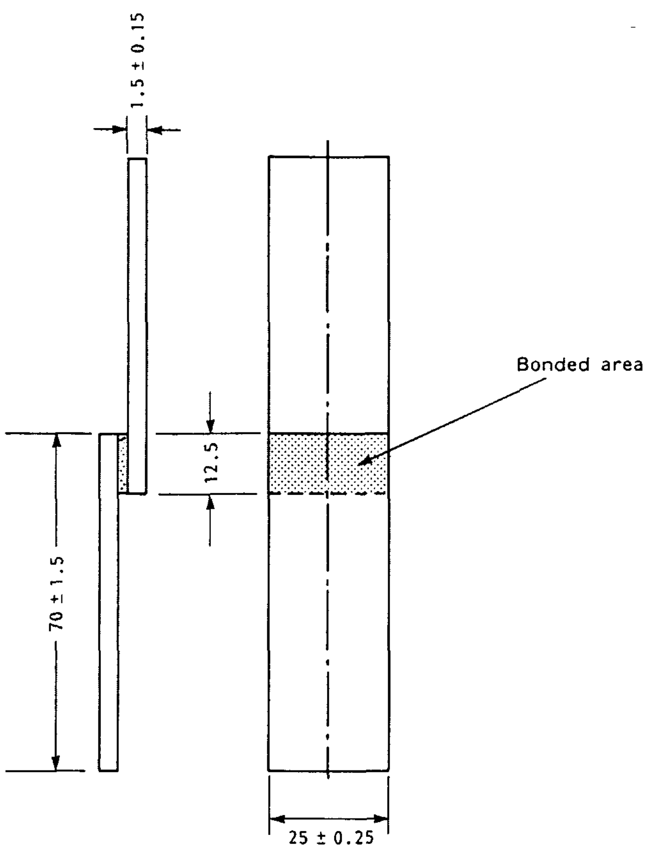

Fig. 2 Configuration and dimensions (in mm) of single lap joint (BS 5350)

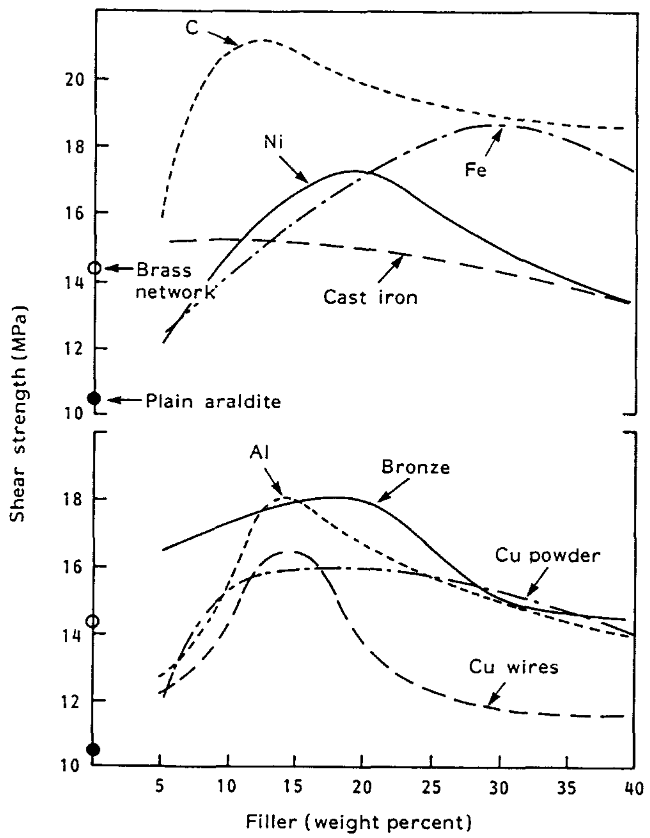

Fig. 3 Effect of filler addition on the shear strength of epoxy resin adhesive

Table 1. Effect of working temperature on shear strength of adhesive formulation

	Shear strength (MPa)			
	25°C	50°C	75°C	100°C
Plain Araldite 2004	14.70	11.86	9.636	7.40

Data from Reference 10

Fillers at 200 phr	Shear strength (psi)			
	23°C	75°C	90°C	105°C
Aluminium powder	2970	-	1470	1390
Carbon black	2000	555	980	980
Zinc dust	2510	600	300	225

Data from Reference 12

shear strength increases with weight percentage of filler up to a maximum value, after which the shear strength decreases. The characteristics of the strength/filler percentage curve (i.e., maximum shear strength value and the critical filler percentage) mainly depend on the filler characteristics (grain size, shape and orientation). Although these factors interact with one another, the following can be concluded from Figs 1 and 3.

1) The finer the filler particle size, the higher the value of maximum shear strength and the lower the filler percentage at which the maximum is attained (graphite powder is finer than copper, nickel and aluminium while iron powder is finer than cast iron).
2) The randomly oriented filler particles strengthen the epoxy resin matrix more effectively than oriented ones (bronze powder versus copper wires and both compared with the brass network).
3) The brass network markedly increases the shear strength of the epoxy resin adhesive although its weight percent is rather high (47%).
4) The peak in the graphite curve suggests the formation of carbon chains at 10% concentration, after which the carbon tends to form aggregates.

Effect of fillers on electrical resistivity

Electrical volume resistivity (ohm cm) was taken as an index for assessing the contribution of fillers to the electrical conductivity of adhesives. Due to the fact that the shear strength of unfilled and filled adhesives appreciably decreases as the testing temperature increases (see Table 1), the mixtures giving the maximum shear strength shown in Fig. 3 were chosen for the electrical resistivity specimens. The measured values of electrical resistivity are presented in Table 2 and Fig. 4. From these data the following conclusions can be drawn.

1) At room temperature, conducting fillers (bronze, nickel and iron) decrease the electrical resistivity of the unfilled epoxy resin adhesive to 54%, 22% and 19%, respectively, of its original value. Good conductors (copper, aluminium and graphite)

Table 2. Electrical resistivity v. temperature of the adhesive without and with a filler*

Temperature (°C)	ρ(ohm cm × 10^{11})						
	—	Bronze 20%	Ni 20%	Fe 30%	Cu‡ 20%	Al 15%	C 10%
31	137.4	73.53	30.73	26.51	4.29	3.82	2.56
33.5	124.9	68.7	25.02	21.8	4.19	2.91	2.04
36	113.5	64.6	14.71	15.9	4.05	2.03	1.66
38.5	85.5	56.6	8.34	10.89	3.95	1.34	1.21
41	72.5	37.2	4.74	6.53	3.81	0.79	0.91
43.5	47.4	14.96	1.97	3.19	3.00	0.50	0.67
46	28.7	5.25	1.27	2.07	1.98	0.33	0.45
48.5	16.1	1.53	0.92	1.19	1.34	0.22	0.35
51.5	9.2	0.65	0.52	0.744	0.91	0.14	0.25
53.5	4.6	0.36	0.32	0.42	0.56	0.093	0.17
65.5	0.39	0.12	0.05	0.042	0.08	0.015	0.03
77.5	0.21	0.08	0.01	0.007	0.013	0.003	0.006
90.5	0.14	0.053	0.0044	0.005†	0.0013	0.001	0.002
98.5	0.10	0.050	0.0035	—	0.0010	0.0005	0.001

*The filler concentration is that of the maximum shear strength of the tested bonded joint
†Measured at 87.5°C
‡As a powder

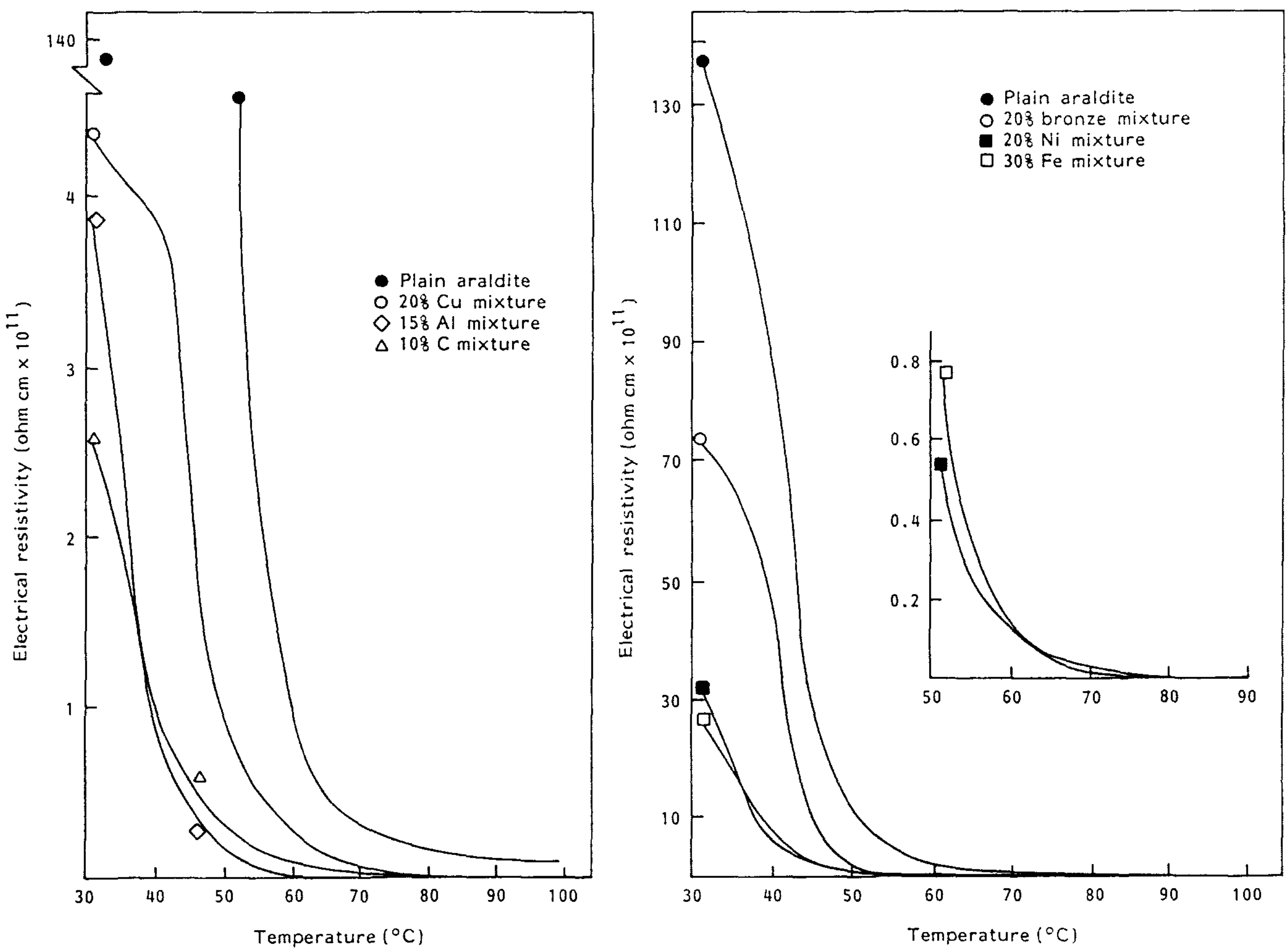

Fig. 4 Effect of filler on the electrical resistivity/temperature curve of filled adhesives

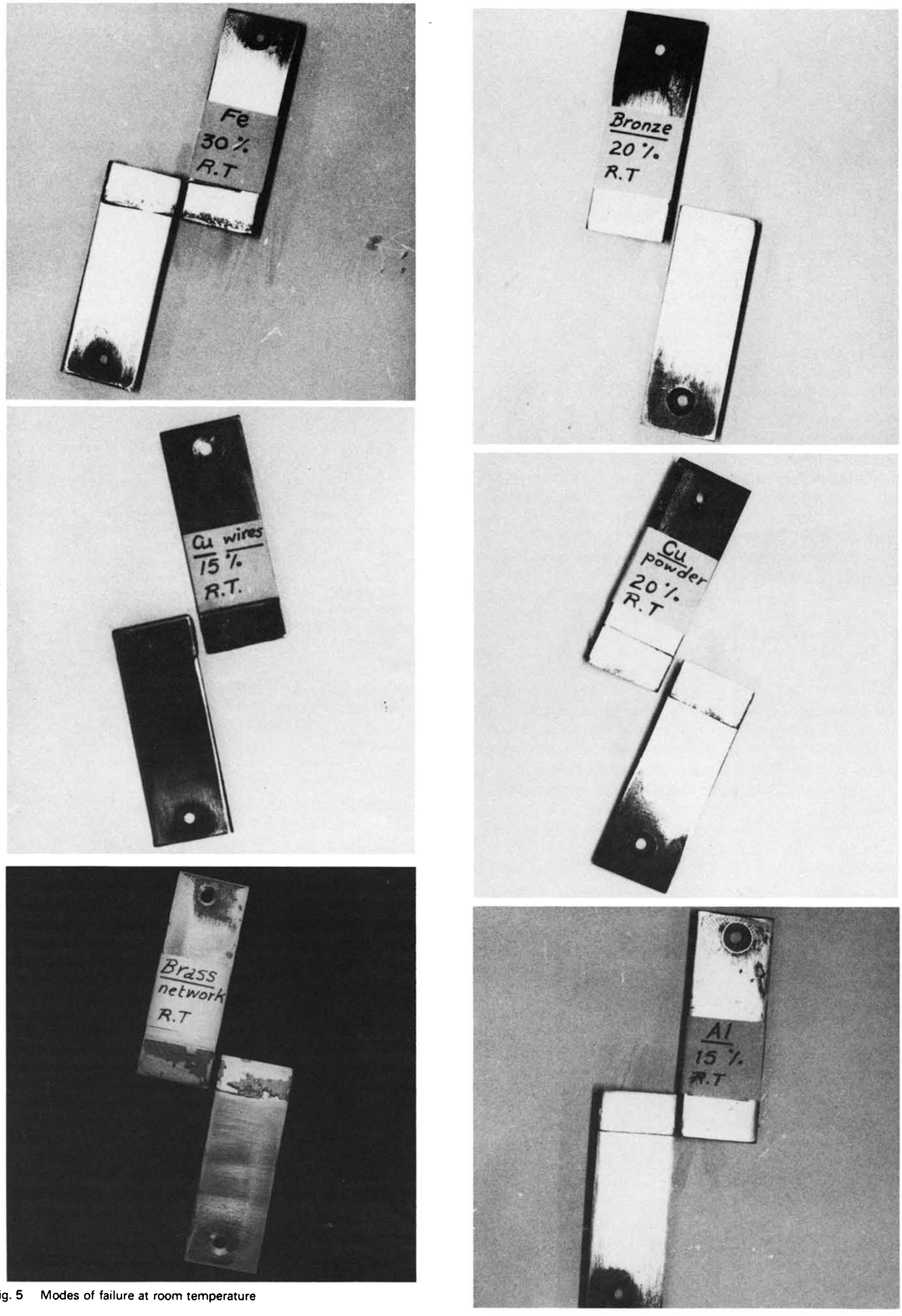

Fig. 5 Modes of failure at room temperature

resulted in a decrease of the electrical resistivity of the unfilled epoxy to 0.031%, 0.028% and 0.919%, respectively, of its original value.

2) The electrical resistivity temperature spectrum of Fig. 4 can be divided into two stages. In the first (room temperature up to 50°C), the electrical conductivity of the filled epoxy resin adhesive increases to several times the value for the unfilled epoxy resin, for all filler materials. The second stage (50–100°C) is characterized by an asymptotic increase of conductivity to a plateau, with the aluminium-filled adhesive being found to have the greatest conductivity throughout this stage.
3) Specimens containing copper wires had a pronounced high electrical conductivity (only 250 ohm cm) at room temperature. This might be related to the random orientation of the copper wires which established much higher contact.

Effect of fillers on mode of failure

Fig. 5 shows the modes of failure of the epoxy resin adhesive reinforced with the critical weight percentage of filler, where a combination of cohesive and adhesive modes of failure appears. Nearly half the area of each adherend is lacking adhesive; this area is observed to be greater in those specimens containing more or less than the critical percentage of filler. Close visual inspection revealed fracture debris around copper wires and cast iron particles.

The mode of failure of the unfilled epoxy resin adhesive was difficult to photograph due to the higher transparency of the layer.

Conclusions

1) Effective improvements in the electrical conductivity of structural epoxy resin adhesives can be achieved when fillers (powder) are mixed with the adhesive. Additions of 10% graphite, 15% aluminium or 20% copper powder by weight markedly improve electrical conductivity.
2) Fibrous filler in the form of copper wires proved to be greatly superior in improving electrical conductivity.
3) The shear strengths of the epoxy resin adhesives attained a maximum at some critical filler weight percentage (10% graphite, 30% iron and 15% aluminium).

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