

THE INFLUENCE OF ACTIVE ELEMENT ADDITIONS FOR THE INTERFACE BETWEEN CARBON FIBRE- COPPER COMPOSITES

Katarzyna Pietrzak

Carbon fibre reinforced copper matrix composites have been produced to provide materials with high thermal conductivity for use in semiconductors (diodes and thyristors) and electrical contacts. The problem of poor wettability of the carbon fibres by the copper matrix was overcome by the addition of zirconium or chromium, which react with the carbon fibres to form carbides. These carbides promote wetting by molten copper and lead to the formation of composites with good interfacial bond strength characteristics, as well as favourable electrical properties.

INTRODUCTION

Copper matrix composites have been widely used when very high thermal and electrical conductivity are required in conjunction with high strength and thermal resistance over wide ranges of temperature. Carbon fibres are characterized by both very good conductivity and outstanding mechanical properties. It appears therefore that carbon fibre-copper composites should have optimum properties from this point of view. Typical applications might be in aerospace components where high strength or stiffness is required together with the ability to withstand high thermal loading [1-2]. Both the physical and mechanical properties of this type of composite are strongly influenced by the properties of the individual constituents of the composite, and also by the microstructure and properties of the products in the reaction zone at the fibre-matrix interface. The matrix and its reinforcing fibres must exhibit both chemical and mechanical compatibility. Two of the main problems to be overcome are the limited wettability of fibres by many metals and the formation of undesirable phases at the fibre-matrix interface caused by inter-diffusion. In the case of carbon fibre-copper composites, the wetting angle is about 160° , and therefore the wettability of the carbon fibres by pure copper is very low. This problem

Instytut Technologii Materiałów Elektronicznych
ul. Wólczyńska 133, 01-919 Warszawa

may be solved in either one of two ways. Firstly, the fibres can be coated with a different metal that is readily wetted by copper, or, secondly, the copper matrix material can be admixed with active additions that will form desirable compounds through reaction with the carbon fibres. Ideally, these compounds should minimize thermal or elastic mismatches at the fibre-matrix interface and hence minimize interfacial stresses, and they should not change the properties of the copper matrix itself. In order to control the interface reactions, the concentrations of the admixed alloying elements in the copper matrix should not be too high and the reactions should be thermodynamically moderate.

INVESTIGATIONS

Carbon fibres (produced by the Institute of Carbon Fibres) with the symbol SAF D 67/a 12000 WW2 were used in these experiments. The main characteristics of the fibres are shown in Table 1 below.

Table 1. Characteristics of carbon fibres.

Nominal quantity	12 000
Cross-section of filament	round
Diameter of filament	7 μm
Breaking strength	3,6 GPa
Young's module	215 GPa
Elongation	1,67 %
Density	1,73 g/cm ³

The fibres were ultrasonically cleaned in acetone for 5-10 minutes. They were then covered with nickel or copper, by brush-plating, before being cut into lengths of 1-2 mm. By protecting the fibres in this way by a 1 μm thick metallic layer, by brush-plating, the strain to failure increases by a factor of two compared to un-plated fibres.

The matrix materials used in this study included a) pure copper powder, b) copper powder with 0.4% Cr and 0.1% Zr, c) copper powder with 18% Cr, and d) copper powder with 18% Zr. All the powders were produced by the Institute of Electronic Materials Technology, and they had a mean particle size of approximately 120 μm .

Composites were prepared by mixing the carbon fibres with one of the matrix powders, including both the pure copper powder and each of the admixed powders, to achieve a uniform distribution. The composite powders were consolidated by volumetric bonding, using a diffusion bonding instrument. The conditions used for diffusion bonding were the temperature range of 750-1050°C, time of 15-60 minutes, pressure of 30-60MPa, and vacuum 2.66×10^{-3} to 2.66×10^{-4} Pa [4]. [Fig.1]

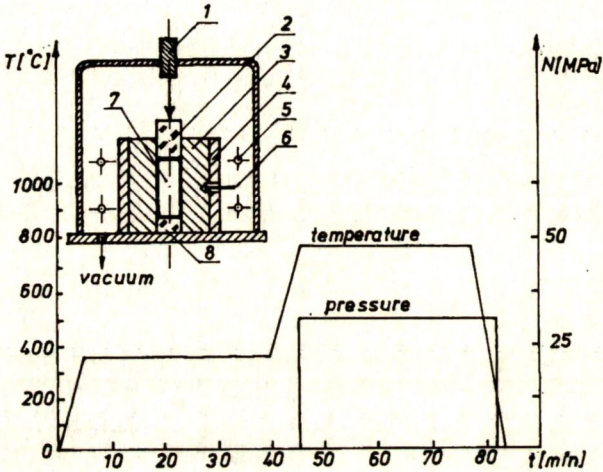


Fig. 1. Cycle of volumetric bonding and scheme of the instrument.

1-pressure, 2-ceramic plunger, 3-graphite mould, 4-clamping ring, 5- heating inductor, 6-thermocouple, 7-sample, 8-ceramic base.

It should be noted that the hold period at 300-350°C is to allow the composite powder and graphite mould to out-gas adequately before pressure is applied.

RESULTS AND DISCUSSION

The microstructure of a typical carbon fibre-copper composite is shown in Fig. 2.

From the analysis made of the thermodynamic conditions of the volumetric bonding process of the composite it was found that when copper powder is admixed with active element additions such as Cr or Zr, the carbides of these elements should form. The X-ray diffraction patterns obtained from the compacted composite specimens confirmed this prediction. The X-ray diffraction patterns were recorded using a Phillips PW-1840 X-ray diffractor equipped with a solid state detector and an APD 1877 automated computer identification system.

X-ray diffraction patterns were recorded in the 2 θ range from 3..60° using

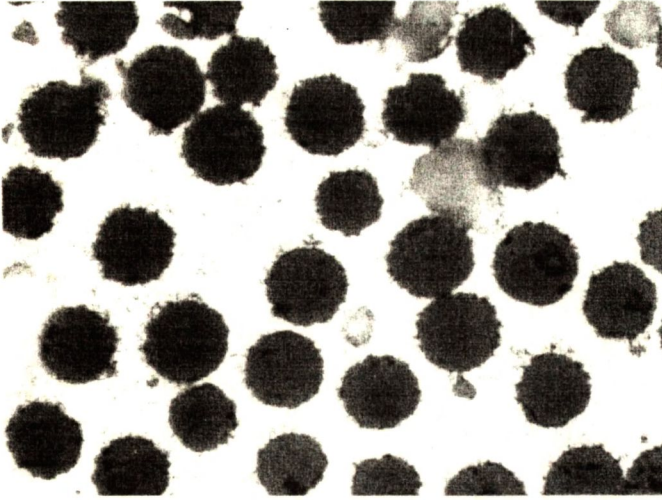


Fig. 2. Optical micrograph of a copper-carbon fibres composite (copper-copper coated carbon fibres).

filtered K_{α} radiation, a tube voltage of 40 kV, filament current of 45 mA, and a time constant of 1s.

For the composite specimen consisting of un-plated carbon fibres in a copper-18% chromium matrix, the presence of a Cr_2C_3 phase was clearly revealed by X-ray diffraction, see Fig. 3. Also, tests to detect the presence of unreacted free elements confirmed this result, and showed a concentration of chromium at the fibre-matrix interface, corresponding to the carbide, and a depletion of chromium in the matrix away from the fibres. In the case of the composite containing uncoated carbon fibres and a copper-18% Zr matrix, the X-ray diffraction data revealed the presence of ZrC together with a unstoichiometric compound of $ZrC_{0.7}$, see Fig. 4.

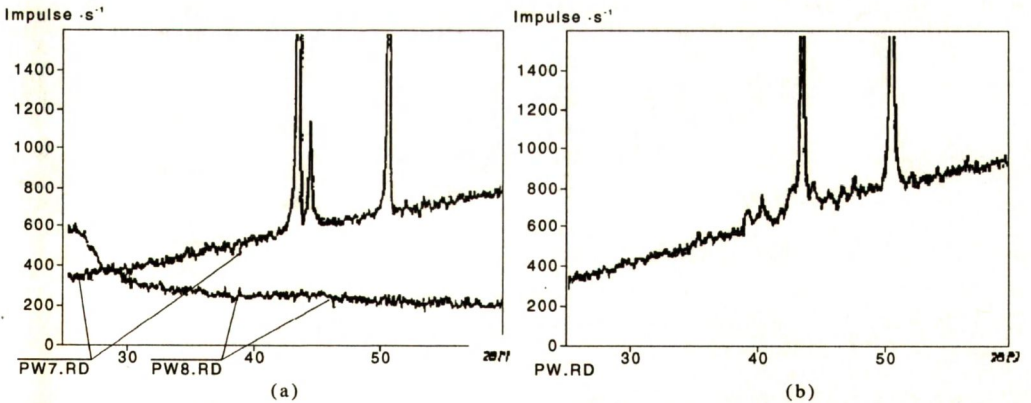


Fig. 3. X-ray diffractometer. (a) initial components 82%Cu+18%Cr and carbon fibre, (b) new phase - composite Cu18Cr-carbon fibre.

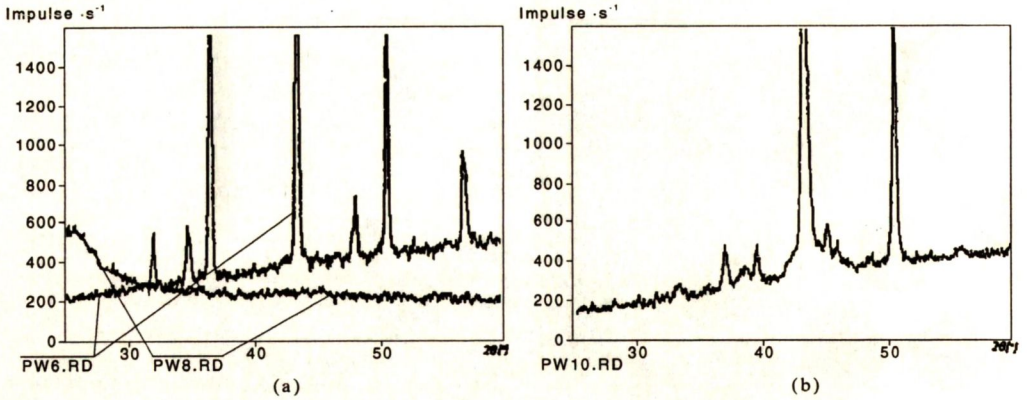


Fig. 4. X-ray diffractometer. (a) initial components 82%Cu+18%Zr and carbon fibre, (b) new phase - composite Cu18Zr-carbon fibre.

Scanning electron microscopy, together with electron microprobe analysis, confirmed that zirconium was present in higher concentrations in the interface region adjacent to the reinforcing carbon fibres, as shown in Fig. 5.

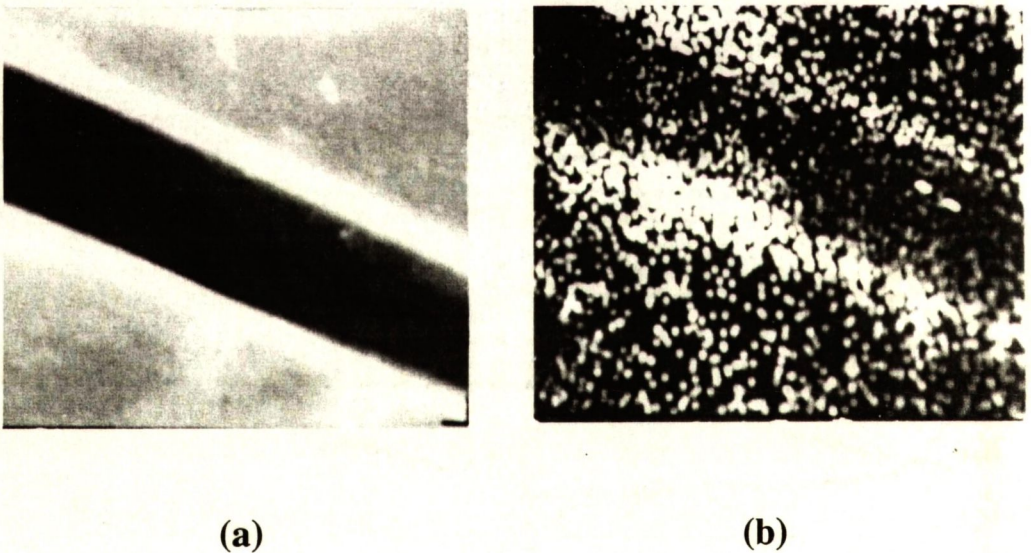


Fig. 5. SEM micrograph and microprobe map of the zirconium of carbon fibre-CU18Zr composite. (a) SEM micrograph of selected area, (b) electron microprobe map of the same area.

The presence of both copper and chromium, or copper and zirconium together in the same fibre may depend on the method of preparation of the composites, the substantial differences in the hardnesses of the different matrix elements, and their diffusion characteristics. Chromium carbides were not observed to form, for example, when the carbon fibre was plated with pure copper prior to hot pressing with the copper-18%Cr matrix. In this case inter-diffusion between the chromium and the copper plating was insufficient to allow Cr_2C_3 to form at the fibre-matrix interface. The method of fabrication of the composite, and especially the presence or absence of a copper or nickel plating on the carbon fibres prior to hot pressing with the alloy matrix, also had a marked influence on the hardness of the final composite, as shown in Table 2. Also, as expected, composites containing larger volume fractions of carbon fibre and those containing carbide forming elements were the hardest of the specimens produced, as shown in Table 2.

Table 2. Hardness of composites carbon fibre-copper. (Conditions of receiving $T=870^\circ\text{C}$, $t=60$ min., $p=2.66 \times 10^{-3}$ Pa).

Composite contents	Q-ty of carbon fibres in % vol.	Hardness HB [kG/mm ²]
Carbon fibre uncovered-Cu0,4Cr0,1Zr	10	44,31
Carbon fibre uncovered-Cu0,4Cr0,1Zr	30	63,67
Carbon fibre uncovered-pure Cu	10	50,02
Carbon fibre uncovered-Cu18Cr	30	113,00
Carbon fibre covered with Cu-Cu18Cr	30	44,80
Carbon fibre covered with Cu-Cu18Zr	30	101,00
Carbon fibre covered with Ni-pure Cu	10	55,60
Carbon fibre covered with Cu-pure Cu	10	49,07

CONCLUSION

Based upon the results obtained in this study the following conclusions may be reached:

1. The lack of wettability of carbon fibres may be overcome by admixing copper with active, carbide forming elements.
2. The carbides formed at the carbon fibre-copper matrix interface determine the physical characteristics of the interface and the mechanical properties of the composite.

3. Knowing the optimum thermodynamic conditions prevailing during the formation of the composite, the compositions and form of the initial constituents, one may forecast the mechanical properties of the composite. This has been demonstrated in the present study with regard to hardness.
4. Although not demonstrated in this study, similar predictions are likely to be possible in the case of thermal and electrical properties.

The present study was not intended to determine the optimum processing conditions but only to examine the interlayers formed at the carbon fibre-copper matrix interface.

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СОДЕРЖАНИЕ

ВЛИЯНИЕ АКТИВНЫХ ЭЛЕМЕНТОВ НА ПЕРЕХОДНУЮ ЗОНУ В КОМПОЗИТАХ УГЛЕРОДНОЕ ВОЛОКНО-МЕДЬ

Композиты углеродное волокно - медь являются перспективными материалами для производства диодов, тиристоров и электрических контактов благодаря их очень хорошим коэффициентам тепло- и электропроводности. Основная проблема получения этих композитов - плохое смачивание углеродного волокна через медь. Показан один из путей её решения - добавление активных элементов: циркония и хрома, которые взаимодействуют с углеродом до карбидов. Полученные карбиды отвечают за микроструктуру переходной зоны, а следовательно за механические и электрические свойства композита.