- 9. N. H. VAN MAAREN, G. M. SCHAEFFER, and F. K. LOTGERING, *Phys. Letts.* 25A (1967) 238.
- 10. E. BUCHER and J. L. STAUDENMANN, cited by F. Hulliger, Structure and Bonding Vol. IV (Springer-Verlag, Heidelberg 1968), Vol. IV, 83.
- 11. E. REVOLINSKY, G. A. SPIERING, and o. J. BEERNTSEN, *J. Phys. Chem. Solids* **26** (1965) 1029.
- 12. H. FRÖHLICH, *Phys. Letts.* **26A** (1968) 169; *Proc. Phys. Soc.* Ser. 2 (1968) 544; H. FRÖHLICH and T. K. MITRA, *Proc. Phys. Soc.* Set. 2 (1968) 548.
- 13. S. FURUSETH, K. SELTE, and A. KJEKSHUS, *Acta Chem. Scand.* 19 (1965) 95.

*Work done at Cyanamid European Research Institute, Geneva.

14. P. JENSEN, A. KJEKSHUS, and T. SKANSEN, *ibid*, 20 (1966) 403.

Amended version received 27 August and accepted 22 September 1971

K. ANDRES

G. W. HULL, JUN. *Bell Telephone Laboratories Murray Hill, New Jersey 07974, USA* F. HULLIGER* *Laboratorium fiir Festk6rperphysik ETH CH-8049 Ziirich, Switzerland*

Continuously Cast Aluminium-Carbon Fibre Composites and their Tensile Properties

There is a current interest in reinforcing aluminum with carbon fibres [1-3] to produce a low density composite material with high tensile strength and modulus parallel to the carbon fibre axes. Major difficulties include the lack of wetting between aluminium and the fibres and also the formation of aluminium carbide Al_4C_3 at temperatures as low as 550° C [4]. We have studied the influence of prior outgassing of the fibres and further, the use of a coating to promote wetting.

The experimental equipment [5] which was developed for the study, is shown in fig. 1. It is based on a semi-continuous casting principle: a carbon fibre tow is drawn through a long die located at the bottom of a crucible containing the aluminium melt. The liquid metal permeates the tow and solidifies in the die to produce a continuous wire. The method has the advantage of a short contact time between melt and tow and also allows an easy and rapid choice of melt temperature. The drawn wires were made from Courtaulds Grafil A or HT fibres and 99.99 pure aluminum to produce wires normally 0.5 mm in diameter and 300 mm long. Fibre content was varied between 5 and 40 vol $\%$ (fig. 2b).

A first series of carbon fibre tows was outgassed by direct Joule heating at 1000° C in a

Figure I Wire-drawing apparatus.

vacuum of 5×10^{-7} torr for 2 h. No wetting of these fibres was found to occur below about 1100° C. Above this temperature, wetting occurred but was accompanied and probably caused by the formation of aluminum carbide (fig. 2a) with a consequent loss of tensile properties and a concurrent proneness to decomposition in atmospheric humidity.

A second series of experiments was carried out using tows in which the fibres had been individually coated with 1 μ m of nickel. The mechanical

9 1972 Chapman and Hall Ltd.

Figure 2 (a) Micrograph showing the formation of $\mathsf{Al}_4\mathsf{C}_3$ around outgassed fibres heated in AI at 1150°C (\times 415). (b) Cross-sections of two composite wires with fibre concentrations of 8 and 40%. Diameter of composite wires 0.5 mm. (c) Scanning electron micrograph of the fracture surface of a composite wire $(x 625)$.

properties of these fibres have been discussed elsewhere [6]. The nickel coating dissolved in the aluminium to form the compound $Al₃Ni$ whilst the fibres were wetted by the melt. (It might be remarked that the addition of massive nickel directly to the melt did not induce such wetting.) Typical micrographs are shown in fig. 2b. In contrast to the outgassed fibres, the coated fibres formed very little carbide with the melt temperature set at 660 to 700 $^{\circ}$ C and with drawing speeds in the range 5 to 100 mm/min. In view of the corrosion susceptibility of the aluminium carbide, an attempt was made to obviate its formation completely by adding up to 1% titanium to the melt with the object of producing stable titanium carbide instead of aluminium carbide as the former is thermodynamically more favoured.

The tensile properties of composites made with nickel coated fibres are given in fig. 3, and no improvement with the titanium addition is evident. No difference was detected between wires made from HT or A fibres, the ultimate tensile strength of both wires and fibres [7] being quite similar. It should be added that whilst there is a wide scatter between different wires, the properties along the length of a given wire were remarkably uniform. Amongst the causes for the variation were the existence of small voids in the matrix (fig. 2b) and a lack of alignment of the fibres.

The "law of mixtures" curve which is included in fig. 3 is based on the average properties of a batch of carbon fibres (see for example ref. [6]). The range of tensile properties of the fibres extends well below the average value and a second (dashed) curve, based on the lower bound, is included in fig. 3. Both curves were calculated for pure A1 and uncoated carbon fibres. In reality, as the fibre volume fraction grows, the proportion of the Al_aNi -phase increases, modifies the tensile properties of the matrix and must finally embrittle it. Under these conditions, as soon as a weak fibre breaks, a crack forms and leads to catastrophic failure. This is a likely process, as can be seen from the line-up of many experimental points close to the dashed curve of fig. 3. Furthermore, the fractograph of a specimen containing 16 vol $\%$ carbon fibres reveals areas where brittle fracture has taken place (fig. 2c). In the absence of nickel, the matrix would be more capable of absorbing the crack energy with a consequent improvement in tensile properties.

Figure 3 Ultimate tensile strength of C-fibre/aluminium composites made with nickel coated fibres as a function of fibre volume fraction. \bigcirc : specimens made from a pure aluminium melt. $+$: specimens made with Ti addition to the melt. Each datum point represents the average value of several specimens taken from a singlewire.Continuous line: Law of mixtures based on $\sigma_c = 200 \text{ kg/mm}^2$. Dashed line: Law of mixtures based on $\sigma_c = 130 \text{ kg/mm}^2$.

In conclusion, a semi-continuous method of producing aluminium-carbon fibre composite materials has been developed. The mechanical properties of the composites reach the lower bound predicted by the law of mixtures. This limit is thought to be related to the wide strength range of the carbon fibre tow leading to tensile failure when the weakest fibres break in the presence of the embrittling Al_aNi -phase.

Acknowledgements

The authors are indebted to Dr D. J. Rowcliffe for many fruitful discussions. They also wish to thank Messrs H. Beutler and M. Villat of Sulzer Brothers, Winterthur, for their collaboration.

References

- 1. A. W. H. MORRIS, Intern. Conf. on Carbon Fibres, Paper 17 (The Plastics Institute, London, 1971).
- 2. K. E. SAEGER, *Aluminium* 46 (1970) 681.
- $3. R. T. PEPPER, J. N. UPP, R. C. ROSSI, and E. G.$ KENDALL, *Met. Trans.* 2 (1971) 117.
- 4. G. BLANKENBURGS, J. *of the Australian Inst. of Metals* 14 (1969) 236.
- 5. E. DE LAMOTTE and n. R. KILLIAS, Proceedings of the 1970 Constance Conference on Composite Materials *(Deut. Gesellsch. f. Metallk.)* Dr Riederer Verlag (Stuttgart, 1971).
- 6. A. J. PERRY, E. D. LAMOTTE, K. PHILLIPS, J . *Mater. Sci.* 5 (1970) 945.
- 7. A. J. PERRY, K. PHILLIPS, and E. DE LAMOTTE, *Fibre Sci. and Tech.* **3** (1971) 317.

Received 13 September and accepted 29 September 1971

E. DE LAMOTTE K. PHILLIPS[†] A. J. PERRY H. R. KILLIAS* *Brown Boveri Research Center 5401 Baden, Switzerland*

*Present address: Landis & Gyr AG, 6301 Zug, Switzerland.]'Present address : Department of Materials Science, University of Sussex, Brighton UK.

A Comment on "A Decade of Quenching from the Melt" by T. R, Anantharaman and C. Suryanarayana (J. Mater. Sci 6 (1971) 1111-1135),

The authors have produced an interesting and useful review, particularly in respect of their tabulation of information on the structure and properties of a large number of alloys in various metastable conditions induced by quenching from the liquid state. While being among the first to recognise the importance of cooling-rate variations, both within and among specimens, in accounting for variations in structure and properties [1], the authors accept uncritically the proposal of Matyja *et al* [2] that measurements of local dendrite arm spacing can be used to estimate cooling-rates of samples. They reject, on the other hand, another proposal [3] that eutectic interphase spacing be used for this purpose.They support this rejection by a number of incorrect statements and specious arguments. These will be dealt with first, followed by a consideration of the basis of the dendrite spacing method preferred by the authors.

Firstly, they state (p. 1116) that the coolingrates of 7 \times 10⁵ and 3 \times 10⁴ K.sec⁻¹ derived for two particular conditions in reference 3, are "at least two or three orders of magnitude less than the *usual* cooling-rates associated with the gun technique" (my italics). The higher value is in fact a factor of only three less than that quoted by the authors only two paragraphs earlier as being typical of their own previous experiments [1] on Al-Ge alloys with the gun technique. On the other hand, the authors, not without some justification, imply that all direct measurements of cooling-rates in splat-cooling are unreliable,

yet, as we shall see, one such measurement provides the crucial support for the dendrite arm spacing correlation they endorse so enthusiastically. This particular direct measurement by Predecki *et al.* [4] gave 1.5 to 3×10^7 K.sec⁻¹ for A1 splats cooled on a composite Ni/Ag substrate. The smaller splat thickness obtained in that study would indeed be expected to give a higher cooling-rate than in reference 3, except in the unlikely event of thermal contact being poorer. If we follow Predecki *et al* in assuming Newtonian cooling applied in their experiment, the heat-transfer coefficient they derived is identical within experimental error to that derived in reference 3 for a considerably thicker splat. Even though there is some uncertainty about the significance of directly measured cooling-rates, they can hardly have been overestimates and there is no better alternative at present than to accept them provisionally at their face value. Their consistency with the indirect results of reference 3 is, at least temporarily, reassuring.

Secondly, the authors state incorrectly that a foil thickness of 30 μ m was *assumed* in reference 3. This value was in fact directly measured by optical microscopy on the actual local foil crosssections used for the eutectic spacing measurements. Foil thickness should always be indicated in reports of work on splat-cooling because of its known importance in affecting cooling-rate. Among others, the authors fail to do this in their previous report $[1]$ of the effect of cooling-rate on metastable phase formation in A1-Ge alloys, although this information might have lent support to their cooling-rates derived entirely from measured dendrite spacings.

Thirdly, the authors state that the use of equilibrium eutectic phase proportions and