CONTROL OF THERMAL PROPERTIES OF CC COMPOSITES BY OPTIMISATION OF STRUCTURE

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Introduction

The thermal conductivity of CC composites is structure sensitive and is thus a complex function of the composite architecture, the constituent volume fractions, texture and crystallinity, and the spatial arrangement of voids and other defects. Although thermal properties of CC composites frequently govern their application, there are not many reports dealing with the systematic correlation of these properties with structure [1]. In this paper, a number of model unidirectional CC composites are described in which the development of matrix texture is both systematically enhanced by thermal treatment and disrupted by the addition of particulate coke fillers. The effects of these processing variables on measured values of thermal conductivity are described and related to the composite structure.

Experimental

Two series of unidirectional (1D) CC composite panels, of thickness 10 mm, were fabricated in-house by a sequence of wet-winding, lay-up, moulding and carbonisation (at 1000 °C). The fibres were untreated high modulus PAN-based Grafil HM-370 fibres. A solution of petroleum pitch (Aerocarb 75) and tetrahydrofuran (THF) was used in the wet-winding of the 'HMP' composite series. The 'HMPC' composite series was prepared using a slurry prepared by mixing a more dilute pitch-THF solution with finely divided coke filler particles (prepared from Aerocarb 75). Composites in each series were densified by means of three cycles of liquid pitch impregnation and carbonisation. Selected composites were then heat treated at temperatures ranging up to 2400 °C.

Table 1. Composite bulk density and constituent volume fractions (fibre, matrix, void).

Composite	Density	Average volume fraction		
(HTT °C)	(g.cm⁻³)	V_f	V_m	V_v
HMP1000	1.62	}0.48	0.38	0.14
HMP2400	1.58	}		
HMPC1000	1.68	}	0.06	
HMPC1500	1.70	}0.47	(C matrix)	0.13
HMPC2000	1.69	}	0.34	
HMPC2400	1.68	}	(C filler)	

Thermal diffusivity measurements were made over the temperature range 25 to 1000 °C in directions both parallel and perpendicular to the fibre axis. Measured diffusivity data were converted to values of thermal conductivity using values of bulk density (Table 1) and computed values of specific heat [2].

Results and Discussion

Figures 1 and 2 illustrate the temperature dependence of thermal conductivity of the pitch-based composites HMP1000 and HMP2400, parallel to the fibre direction. Also plotted is the thermal conductivity of the HM fibre in the axial direction. As the fibre remained structurally unchanged by heat treatment at 2400 °C, the observed difference in thermal conductivity of composites HMP1000 and HMP2400 was wholly attributed to an increase in thermal conductivity of the matrix. Given the simple geometry of the 1D composite, heat flow in the composite parallel to the fibres may be assumed to be the sum of the products $V_f.K_f$ and $V_m.K_m$, where K_f and K_m are the values of thermal conductivity of the fibre and matrix, respectively. Approximate values for the matrix thermal conductivity in the direction parallel to the fibres were calculated using this simple relationship (Figures 1 and 2). The effect of heat treatment on the matrix thermal conductivity (at ambient temperatures) was to increase its value from ~ 30 W.m⁻¹.K⁻¹ to over 250 W.m⁻¹.K⁻¹. The origin of this increase in thermal conductivity was an improvement in the crystallinity and orientation of the matrix associated with the development of a graphitic matrix 'sheath' around each fibre. Figure 3 illustrates that the values of thermal conductivity of the HMP1000 and HMP2400 composites were considerably lower in the direction perpendicular to the fibre axis. Of particular interest was the effect on thermal conductivity of small additions of filler particles. Coke particles were added to the primary matrix in order to modify the development of the structure during moulding and subsequent processing. Their effect was to refine the composite structure and to partly disrupt the development of the matrix sheath structures. Additions of the coke particles were found to reduce the anisotropy ratio of thermal conductivity parallel and perpendicular to the fibre direction from ~11:1 (HMP2400) to ~6:1 (HMPC2400). This reflected a 15 % decrease in thermal conductivity parallel to the fibre direction, together with a 57 %

increase in thermal conductivity perpendicular to the fibre direction. The absolute magnitude of the thermal conductivity is determined by both the crystallite size and the orientation of the layer planes [3]. As the carbon matrix and filler materials in the HMPC composites had the same origin and thermal treatment history, the observed changes in thermal conductivity were attributed to disruption of the orientation of the layer planes.

Conclusions

On graphitisation, the development of matrix texture significantly increased the thermal conductivity in the fibre direction; this effect has been quantified. The addition of coke filler particles partially disrupted the matrix orientation and resulted in increased values of transverse thermal conductivity. Curve-fitting routines based on established theories, which relate the thermal conductivity to crystallite size and layer-plane orientation, may be applied in the analysis of the measured data [4]. Further work will seek to deconvolute the fibre and matrix contributions towards thermal conductivity in the direction transverse to the fibre axis.

References

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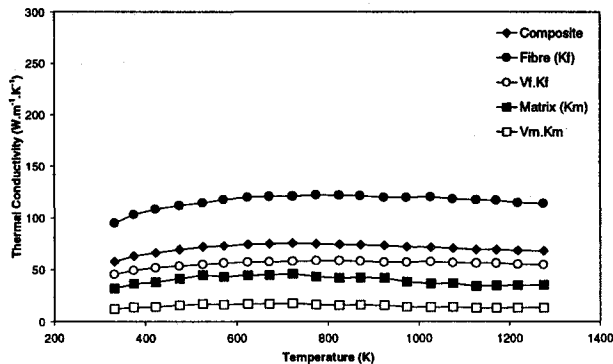
Figure 1. Thermal conductivity of HM carbon fibre and HMP CC composites (HTT 1000 °C; measured parallel to fibre axis); thermal conductivity of matrix calculated using mixture rule.
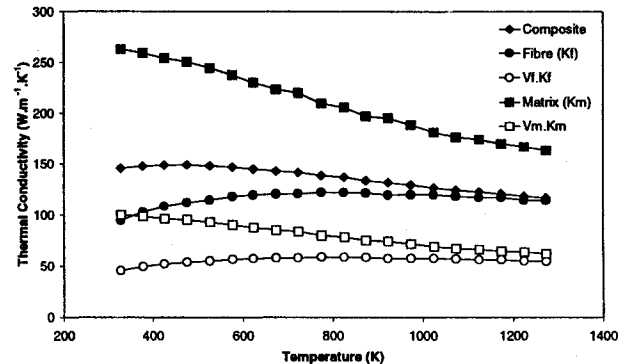

Figure 2. Thermal conductivity of HM carbon fibre and HMP CC composites (HTT 2400 °C; measured parallel to fibre axis); thermal conductivity of matrix calculated using mixture rule.
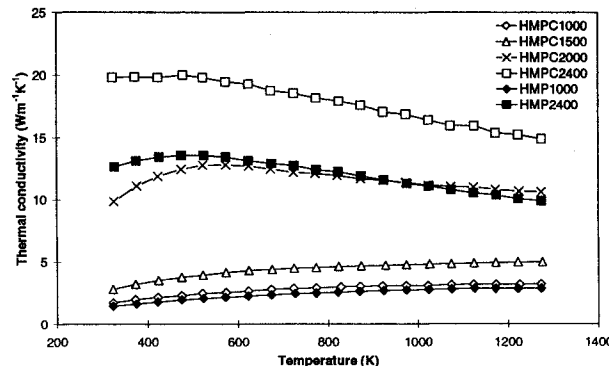

Figure 3. Thermal conductivity of HMP and HMPC CC composites (measured perpendicular to fibre axis).

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