HIGHLY ORIENTED CARBON FILAMENTS FROM MESOPHASE PITCH FOR THERMAL MANAGEMENT APPLICATIONS

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Introduction

Mesophase pitch-based carbon fibres can have excellent thermal transport properties and thus may be ideal thermal management materials [1]. Amoco Performance Products Inc. recently developed a self-reinforced panel, ThermalGraph®, by pressing the as-spun mesophase pitch fibres before thermosetting and further heat treatment, which provides a new cost-effective method to produce carbon materials for thermal management. The high thermal conductivity of mesophase pitch-based carbon fibres is due to the extended graphitic crystalline structure [2]. The carbon fibres with a larger transverse area may allow the development of more extensive graphitic planes, thus improving the thermal transport properties. However, the diameters of carbon fibres are restricted by the time-consuming stabilisation process, therefore, carbon fibres with average diameters larger than 15 microns are rarely produced. There have been many attempts to produce carbon fibres with non-circular cross-sections such as trilobal, C-shaped and ribbon fibres [3,4]. The non-circular fibres can have bigger transverse area due to the shorter oxygen diffusion distance from the surface during stabilisation compared with round fibres with equivalent cross-sectional area, also they tend to have better mechanical properties [5]. The naphthalene derived mesophase pitch has high reactivity, fibres from which can be readily stabilised. This makes it a convenient precursor for the study. Here we report preliminary results from a study designed to develop high thermal conductivity graphitic materials. In this first stage we report the preparation of large diameter filaments of 15-50 microns diameters.

Experimental

The naphthalene derived mesophase pitch ARA24 (100% anisotropic content, softening point: 237 °C) was used to prepare the pitch filaments. The as-spun filaments were stabilised in air or oxygen, and the stabilisation process was evaluated using a thermobalance. The stabilised filaments were carbonised at 1300 °C or 2400 °C under inert atmosphere. The mechanical properties of carbon filaments were evaluated by single-filament tensile testing and 30 mm gauge lengths were used. The electrical resistivities of carbon filaments were determined using a standard four-point probe method and used to indicate the likely thermal conductivity.

Results and Discussion

The melt spinning process for the larger diameter filaments was easier than melt spinning of small diameter fibres (< 15 μm), with few threadline breakages. The stabilisation degree of the pitch filaments can be evaluated by the weight gain, which should be approximately between 8-10% to attain the infusibility during subsequent carbonisation. Fig. 1 shows the stabilisation weight gain of two sets of filaments with average diameters 27 μm, and 45 μm respectively. For the 45 μm filaments, the maximum weight gain obtained in air is about 5.7%, but reaches 9.7 % in oxygen atmosphere. Pitch filaments with diameters smaller than 30 μm can easily be stabilised in air, but those with diameters larger than 55 μm could not be sufficiently stabilised even in an oxygen atmosphere.
Fig. 2 shows the relationships between electrical resistivity and average diameters of carbon filaments at different heat treatment temperatures. The final heat treatment temperature strongly influences the electrical resistivity, increasing heat treatment temperature from 1300 °C to 2400 °C reduces the electrical resistivity by a factor of 3. It is interesting to note that the electrical resistivity decreases slightly with the increase of the diameters of the carbon filaments. This suggests that the thermal conductivity will also improve with increasing diameter because of the reverse relationship between electrical resistivity and thermal conductivity [6]. The general trends are consistent at two different heat treatment temperatures. It is believed that more planar graphitic structures are developed in the larger fibres. The SEM image (a) in Fig. 3 shows the typical gross sheath-core textures in the large diameter fibres, which was probably formed due to diffusion controlled stabilisation reactions. The central area of the filament can partially melt during carbonisation, leaving a hollow filament if the filament is not fully stabilised (Fig. 3b). The large filaments may have better transport properties, but not better mechanical properties as demonstrated in Fig. 4, because large filaments are prone to more defects and higher porosity. However, large diameter filaments of the order of 20 μm may provide a compromise; such fibres (1300°C carbonised) have a tensile strength of about 1 GPa, Young's modulus

approaching 180 GPa, and electrical resistivity of 8.7 μΩm. These results indicate that carbon filaments with significant larger cross-sections (say > 2000 square micrometers) with useful mechanical properties can not be realised by circular fibres. However, carbon ribbons may provide a better alternative, in terms of larger graphitic plane structures and mechanical properties. Results on carbon ribbons will be presented at the conference.

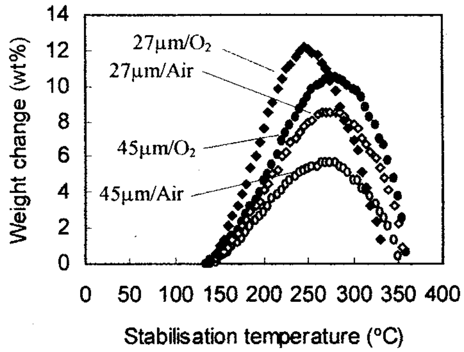

Fig. 1. The influence of filament diameters, oxidising atmosphere to the weight gain

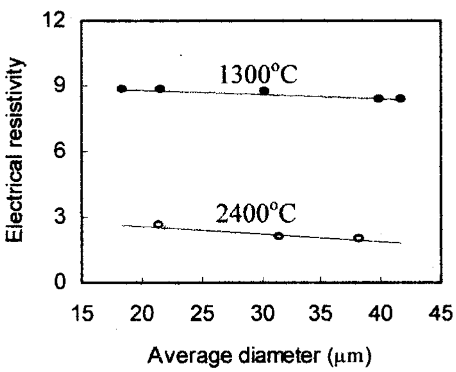

Fig. 2. Relationship between electrical resistivity (μΩm) and average diameter of carbon filament

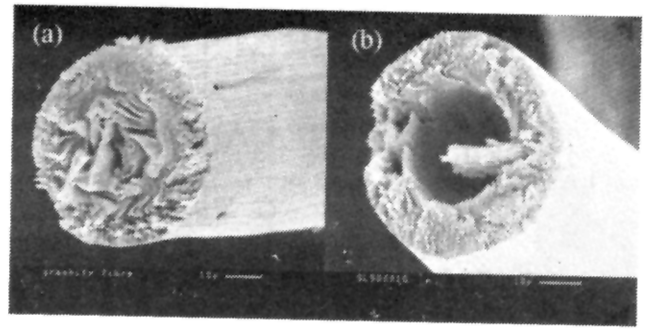

Fig. 3. SEM images of carbon filaments, (a) showing the typical sheath-core texture, (b) hollow filament

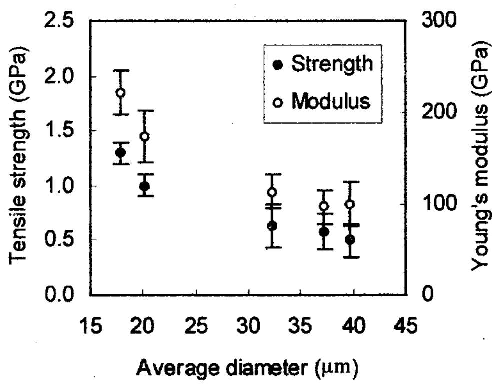

Fig. 4. Tensile properties as a function of average diameter of carbon filaments carbonised at 1300°C

References

1. Edie DD. The effect of processing on the structure and properties of carbon fibers. *Carbon* 1998; **36**(4): 345-362.
2. Adams PM, Katzman HA, Rellick GS and Stupian GW. Characterization of high thermal conductivity carbon fibers and a self-reinforced graphite panel. *Carbon* 1998; **36**(3): 233-245.
3. Edie DD, Hayes GJ, Rast HE and Fain CC. Processing of noncircular pitch-based carbon fibers. *High Temperatures-High Pressures* 1990; **22**: 289-298.
4. Mochida I, Yoon SH and Korai Y. Preparation and structure of mesophase pitch-based thin carbon tape. *J Mater Sci* 1993; **28**: 2135-2140.
5. Stoner EG, Edie DD and Kennedy JM. Mechanical properties of noncircular pitch-based carbon fibers. *High Temperatures-High Pressures* 1990; **22**: 299-308.
6. Lavin JG, Boyington DR, Lahijani J, Nysten B and Issi JP. The correlation of thermal conductivity with electrical resistivity in mesophase pitch-based carbon fiber. *Carbon* 1993; **31**(6): 1001-1002.