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Letter to the Editor

Carbon composites reinforced by graphite grains

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Abstract

A new kind of composites – graphite grain/carbon (G/C) composites prepared by thermal gradient chemical vapor infiltration technology and elementary properties are investigated, and the microstructure evaluated. It is observed that pores among graphite grains are filled with pyrocarbon effectively, therefore, G/C composites with high density can be obtained. These composites offer the high temperature properties of carbon material, and excellent mechanical properties and isotropy.

1. Introduction

In recent years carbon-based composites have been used widely as plasma facing materials in fusion devices, fission reactor and high power accelerator due to their superior thermo-mechanical properties [1,2]. Traditional isothermal chemical vapor infiltration for preparing carbon/carbon (C/C) composites is complicated, time-consuming and costly. This severely impacts the application of C/C composites. Reinforced materials of three-dimensional or multi-dimensional carbon fiber weave are expensive. The popular preform is carbon fiber felt in which a continuous long fiber layer (carbon cloth without weft thread) and a random short fiber layer are laminated alternately and then needled to prepare the porous preforms. However, the strength is higher parallel than perpendicular to the fiber layer by this technology. The obvious disadvantages include anisotropy, complicated processing for preparation of preforms, high-cost preforms. Therefore, C/C composites with high performance and low cost prepared by novel experimental approaches are the focus of research [3].

Machining graphite creates powder and scrap, which not only waste resource, but also pollute the environment. Therefore, we develop new carbon materials – carbon composites reinforced by graphite grains (G/C composites) by the thermal gradient chemical vapor infiltration, which is low cost, high performance and isotropic. This kind material overcomes the shortcoming of anisotropic C/C composites with low strength perpendicular to the fiber layers.

2. Experimental

Reinforced materials, graphite grains with granularity of 300–640 μm, are used to prepare the preform, the size of which is 80 mm o.d. × 6 mm i.d. × 400 mm height with the apparent density about 0.6 g/cm^3. The carbon source gas used for the infiltration run is the natural gas (98% CH_4, 0.3% C_3H_8, 0.3% C_4H_{10}, 0.4% other hydrocarbons, 1% N_2), and molybdenum pole of 6 mm (diameter) × 400 mm (height) is used as the heater. Densification of porous graphite grains preform is carried out in a thermal gradient CVI furnace, which is cooled in the jacket by water. The detailed description of the preparation has been reported elsewhere [4,5].

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3. Results and discussion

3.1. The microstructures of composites

The micrograph of G/C composites shown in Fig. 1 is the microstructure of smooth laminar pyrocarbon consisting mainly of smooth surface, strong optical activity, and disk shape under the polarized light microscopy, and possess a very high degree of anisotropy. It can be seen from Fig. 2 that pyrocarbon is formed around graphite grains and graphite grains of different sizes are distributed among pyrocarbon. In G/C composites, the pores formed within graphite grains preform possess multi-entrance for precursor penetration, so graphite grains from different directions can be infiltrated with pyrocarbon, pores being completely filled. The density of G/C composites is 1.85 g/cm^3, and the density of graphite is 1.75 g/cm^3 which is similar to that of carbon fiber. On the other hand, the pores among fiber bundles are small because carbon fibers in C/C composites are overlapped and pores are tube-shaped which only possess two entrances for precursors such that pores are easily plugged resulting in low density of C/C composites.

From Fig. 3 we can see that graphite grains are wrapped by pyrocarbon in the form of layer and pyrocarbon distribute in different orientations due to the graphite grains. In C/C composites, the graphitic flat-layered structure crystallites of pyrocarbon are arranged on the surface of the carbon fiber, which grows around the carbon fiber in the form of ringed multilayer structure, in which six-membered ring carbon atom plane (0 0 2) is parallel to the direction of the fiber axis. However, G/C composites are anisotropic because pyrocarbon distributes in different orientations and graphite grains are used as reinforced particles.

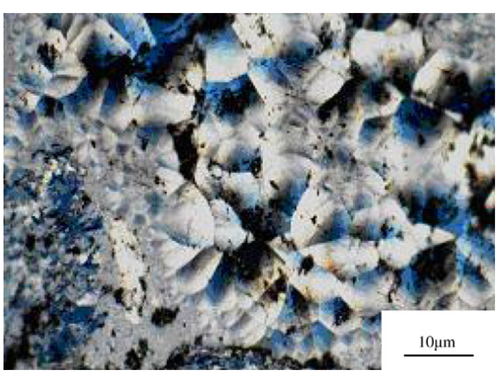

Fig. 1. Microstructurs of G/C composites under the optical microscopy.

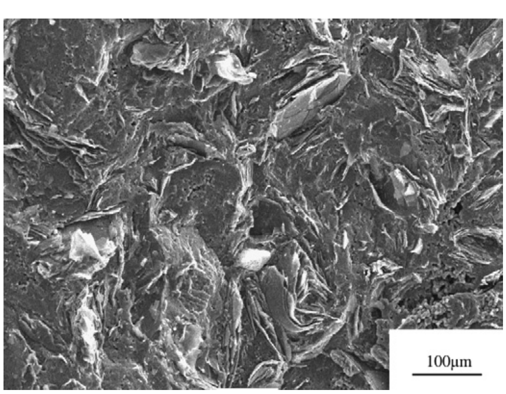

Fig. 2. SEM fracture morphology of G/C composites.

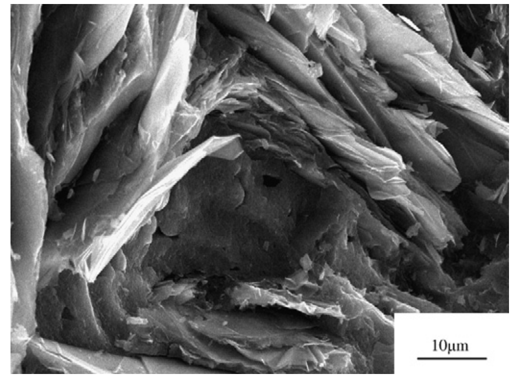

Fig. 3. SEM morphology of a graphite grain covered by pyrocarbon in G/C composites.

3.2. Mechanical property of G/C composites

The properties of graphite (Toyo Tanso grade IG 11), C/C composites reinforced by carbon fiber felt and G/C composites are listed in Table 1. The direction of flexural and compressive strength of C/C composites is perpendicular to the layer while the direction of shear strength and resistivity are parallel to the layer [4]. The strength of G/C composites is higher than that of graphite. The mechanical properties of turbostratic graphite planes structure in G/C composites are higher than those of hexagonal crystal in graphite. Slippage between graphite planes can be stopped by the adjacent graphite plane because disordered and angled graphite planes are overlapped and interlocked leading to high mechanical properties compared with graphite. However, flexural, shear strength of G/C composites are lower, and compressive strength is similar to that of C/C composites because load can not be passed effectively through graphite grains. In general, G/C composites are dimensionless materials, i.e. isotropic.

3.3. Resistivity of G/C composites

The results in Table 1 show that resistivity of G/C composites is higher than those of graphite and C/C

Table 1
Properties of pure graphite, C/C composites reinforced by carbon fiber felt and G/C composites

Materials	Density (g cm^{-3})	Flexural strength (MPa)	Compressive strength (MPa)	Shear strength (MPa)	Volume resistivity (μΩ m)
Graphite	1.77	39	78	–	11.0
C/C	1.75	80	120	20	82.7
G/C	1.85	50	121	18	148.4

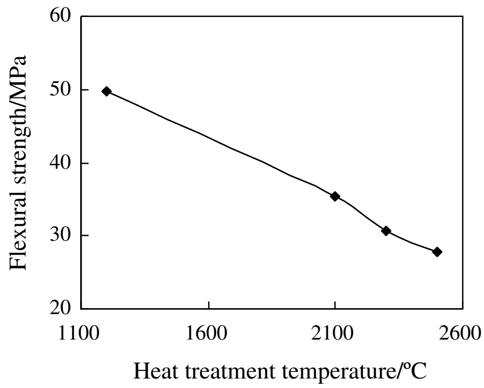

Fig. 4. Effects of heat treatment temperature on the bending strength of G/C composites.

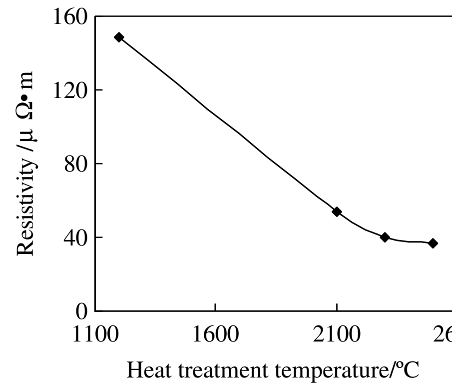

Fig. 5. Effects of heat treatment temperature on the resistivity of G/C composites.

composites. Electron cloud in the conjugated π band is electric current carrier. Therefore, for graphite hexagonal crystal, the crystal size is large, there are few scattering sites, so the resistivity is lowest. For C/C composites, the graphitic flat-layered structure crystallites of pyrocarbon are arranged on the surface of the carbon fiber, which grow around the carbon fiber in the form of ringed multilayer structure, in which six-membered ring carbon atom planes (002) are parallel to the direction of the fiber axis, there are relatively few intrinsic scattering sites, and the resistivity is low. However, for G/C composites, disorder structure contains many intrinsic scattering sites so electron transport is hindered and resistivity is high.

3.4. Effect of heat treatment on property of G/C composites

When G/C composites are in high temperature environment, the adjacent graphite layers are liable to sliding with thermal stress, resulting in transformation from turbostratic graphite planes structure to preferred orientation of pyrocarbon crystallites. Pores and fracture among crystallite are enlarged, which leads to the weak joining between pyrocarbon and graphite grains with increasing heat treatment temperature. Fig. 4 shows that the bending strength of the composites is reduced with increasing of the treatment temperature.

When the heat treatment temperature is increasing, the higher degree of graphitization results in the structure of G/C composites being much closer to that of graphite crystal. Improvements in crystallites result in reduced electrical resistivity. The experimental results show that the resistivity of the composites reduces with the increasing of heat treatment temperature as shown in Fig. 5.

4. Conclusions

Graphite grain/pyrocarbon (G/C) composites have been prepared by thermal gradient chemical vapor infiltration. It is shown that graphite grains are wrapped by pyrocarbon from all orientations, and pores between the graphite grains are effectively filled with pyrocarbon, The composites have a high density which not only has the high temperature properties of carbon, but also excellent mechanical properties and isotropy of composites reinforced by grains. Furthermore, its properties can be adjusted through heat treatment. The composites are suitable for use as heaters, electrodes, crucibles, first wall materials in nuclear fission and fusion reactors.

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