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Homogenous and highly isotropic graphite produced from mesocarbon microbeads

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A B S T R A C T

Mesocarbon microbeads (MCMBs) are attractive precursors for high-density, high-strength polycrystalline graphite due to their self-sintering capability. They have the shape of a standard sphere and stack in a completely random orientation during compaction, thereby leading to an isotropic texture. In this paper, MCMB-based graphite shows better isotropic property than commercial nuclear graphite, as demonstrated by a coefficient of thermal expansion-based isotropy ratio of 1.00. It was found that variations in apparent density within block are approximately 2–5% in graphitized artifacts. The variation of flexural strength within block is small, revealing excellent homogeneity of MCMB-based graphite. Density distribution measurement indicates that artifacts produced in the same batch also have low variation from block to block. Furthermore, the flexural strength of this material is greatly influenced by heat treatment temperature and mean particle size. The fracture mechanism is also shown by a detail examination of fracture surfaces. These results may help to gain a better understanding of MCMB-based graphite.

1. Introduction

Polycrystalline graphite is essential in modern technologies. For example, ultra high power graphite electrodes produced from needle coke are used for electric arc furnace steelmaking; graphite blocks are used as cathodes in aluminum-electrolysis cells; iso-molded graphite can be used as heater and crucible for single crystal silicon growth; in addition, isotropic nuclear graphite serves as moderator and reflector in high-temperature gas-cooled reactor (HTR). For the HTR-pebble module (HTR-PM), which is the first commercial HTR in China, each module contains more than 500 tons of graphite components.

Highly isotropic property of polycrystalline graphite is required for nuclear applications for two reasons. Firstly, isotropic graphite shows much longer irradiation lifetime than

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anisotropic graphite due to better dimensional stability. Secondly, anisotropic dimensional change may cause distortion or mismatch of graphite components, leading, for example, to interference with control rod operation or coolant leakage. The isotropy ratio is usually defined by the ratio of the against-grain (AG)/with-grain (WG) values of the coefficient of thermal expansion (CTE) [1]. Graphite in which the CTE-based isotropy ratio is in the range from 1.00 to 1.10 is named isotropic graphite; graphite in which the isotropy ratio based on the CTE is between 1.10 and 1.15 is named near-isotropic graphite; and graphite in which the isotropy ratio based on the CTE is greater than 1.10 is named anisotropic graphite [1,2].

Conventional polycrystalline graphite is manufactured by the process of bonding coke fillers with coal-tar pitch by carbonizing and graphitizing the artifact. The selection of the filler materials essentially dominates the properties of the final product. By careful selection of fillers and manufacturing settings, CTE values in the region of 1.05 can be achieved, as shown in Table 1. Gilsocarbon which has an isotropy ratio of 1.03 has been proven to be one of the most isotropic graphite grades ever produced [3]. This graphite is produced from coke obtained from naturally occurring pitch found in Utah in the USA. The structure of Gilsonite coke is made of spherical particles of about 1 mm in diameter, and shows an onion-like texture, i.e. a concentrically arrangement of graphite basal planes.

It is well known that mesocarbon microbeads (MCMBs) are self-sintering carbon powder precursors. This raw material is able to be compacted and to become strongly bonded via ‘sintering’ in the absence of a binder. The MCMB-based graphite shows high strength, fine microstructure, and a marked inhibition of liquid fluoride-salt infiltration [9]. Due to their completely spherical shape, MCMB particles spontaneously stack in random orientation upon compaction, thereby forming an isotropic texture in spite of the anisotropic nature of MCMB itself. Nevertheless, the isotropic property of this graphite is rarely mentioned by previous studies.

Therefore, the primary objective of this work is to show the processing of MCMB to highly isotropic graphite. Linear thermal expansion testing was used to evaluate the isotropy behavior of MCMB-based graphite. The product is considered

Table 1 – CTE values and isotropy ratios of some commercial nuclear graphites.

	CTE (10^{-6} K^{-1})	Isotropy ratio	References
Gilsocarbon	//4.99, ⊥4.84* (20–120 °C)	1.03	[4]
H-451	//4.00, ⊥4.55	1.14	[5]
ATR-2E	//4.4, ⊥4.9	1.11	[6]
ASR-1SR	//4.70, ⊥4.87	1.04	[6]
PCEA	//4.0, ⊥4.24	1.06	[7]
IG-110	4.5 (350–450 °C)	–	Catalog value
	//4.03, ⊥4.16 (20–500 °C)	<1.05	[8]
IG-430	4.8		Catalog value
NBG-17	//4.5, ⊥4.6	<1.05	Catalog value
NBG-18	//4.5, ⊥4.7	<1.05	Catalog value
NBG-25	//3.9, ⊥4.3	<1.10	Catalog value

* //, means WG; ⊥, means AG.

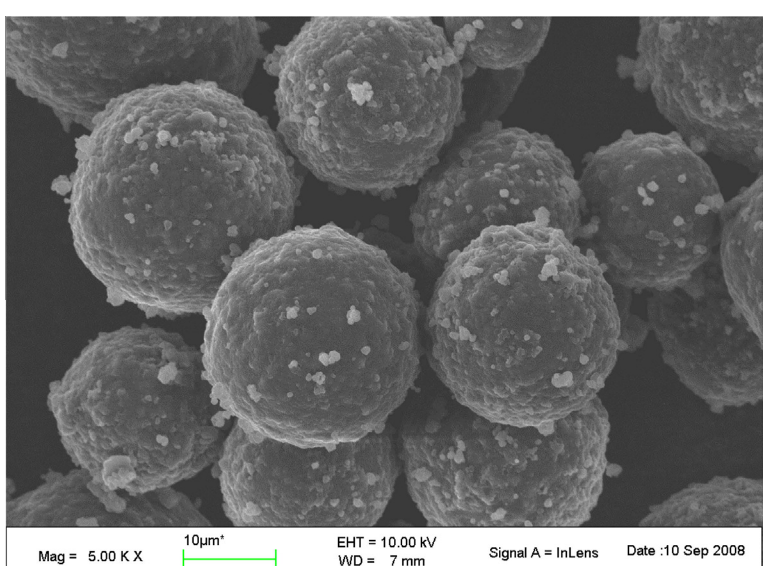

Fig. 1 – Scanning electron microscope image of MCMB green powder. (A color version of this figure can be viewed online.)