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Characterization the microstructure and defects of matrix graphite irradiated with Xe ions

H.X. Xu [a,*], J. Lin [a], J.J. Li [a], Z.Y. Zhu [a], G.L. Zeng [a], J.D. Liu [b], B.C. Gu [b], B. Liu [c]

[a] Shanghai Institute of Applied Physics (SINAP), Chinese Academy of Sciences (CAS), Shanghai 201800, China
[b] State Key Laboratory of Particle Detection and Electronics, University of Science and Technology of China (USTC), Hefei 230026, China
[c] Institute of Nuclear Energy and Technology (INET), Tsinghua University, Beijing 100084, China

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ABSTRACT

The matrix graphite of pebble fuel elements was irradiated with 1 MeV Xe ions at room temperature to fluences of 5.8×10^{14} ions/cm^2 and 2.9×10^{15} ions/cm^2, respectively. The microstructure and defects of matrix graphite samples were characterized by using scanning electron microscopy (SEM), Raman spectroscopy and slow positron beam techniques. The SEM result reveals that hundred-nanometer sized pores appear at the surface after irradiation and the density of pore increases with fluence. Raman results show that D peak (1350 cm^{-1}) and G peak (1580 cm^{-1}) are broadened after irradiation. In addition, the G peak position shifts from 1580 cm^{-1} to 1560 cm^{-1} with the linewidth increases from 21 cm^{-1} to 132 cm^{-1}, corresponding to the increase in bond-angle disorder as the matrix graphite transforms from microcrystalline to amorphous carbon(a-C). The slow positron beam study shows that the defects-trapped positron S parameter increases with fluence, suggesting that the vacancy-type defects concentration or size of open volume defects increases. The analysis of Raman and slow positron beam consistently conclude that the reason for the phase transition after irradiation is the increase in irradiation-induced vacancy defects accompanied by the overlap of disordered regions.

1. Introduction

Graphite has been widely used as nuclear materials, such as moderator and reflector in reactor cores, due to its beneficial performance of high temperature tolerance, high thermal conductivity, low coefficient of thermal expansion (L-CTE) and mechanical stability [1–3]. However, in the reactor core enviroment, graphite will suffer serious neutron irradiation which can cause significant changes in dimensions and physical properties such as, strength, thermal conductivity etc [4,5].

Tuinstra and Koenig [6] reported that polycrystalline graphite exhibited both D peak at 1355 cm^{-1} and G peak at 1580 cm^{-1} in the Raman spectrum, corresponding to phonon density of states and E_{2g}-mode lattice vibration, respectively. In addition, they found that the relative Raman intensity ratio of D peak to G peak ($R = I_D/I_G$) was inversely proportional to the crystalline size L_a of graphite ($R = 4.4\ nm/L_a$). Elman [7] studied the structural evolution of HOPG graphite implanted with various ions (Li^+, Be^+, B^+, C^+, P^+ and As^+) at 100 keV by using Raman scattering. Their results indicated that the critical fluence for inducing the transformation from microcrystalline to an amorphous carbon (a broad asymmetric Raman line at 1500 cm^{-1}) depending on the mass of ion. Nakamura, Kitajima [8] and Asari [9] studied the early stage of noble-gas ions (He^+, Ne^+, Ar^+, Kr^+ and Xe^+) irradiation on HOPG graphite prior to amorphization. They found that the peak intensity ratio R (I_D/I_G) was proportional to the square root of irradiation time due to the reduction of phonon correlation length (equal to a mean distance between vacancies) of graphite. However R saturated at about $t \sim 150s$ due to the creation of vacancy clusters. Recently, Yang [10] and Shi [11,12] investigated the thermal evolution of vacancy defects induced by C^+ and He^+ irradiation by using slow positron beam, and different thermal annealing stages were found for samples irradiated to low fluence and high fluence.

As known, the irradiation property of graphite is closely associated with their raw materials and preparation technology. One of the Thorium molten salt reactors (TMSR) design will utilize advanced spherical fuel elements of which the TRISO coated particles (CPs) [13,14] are embedded in graphite matrix. The raw materials and preparation technology of matrix graphite are different from traditional nuclear graphite, such as IG-110, ETU-10, NBG-10 and fine-grained isotropic nuclear graphite. Therefore, it is very necessary to characterize the changes in the microstructure and

* Corresponding author.
E-mail address: xuhongxia@sinap.ac.cn (H.X. Xu).

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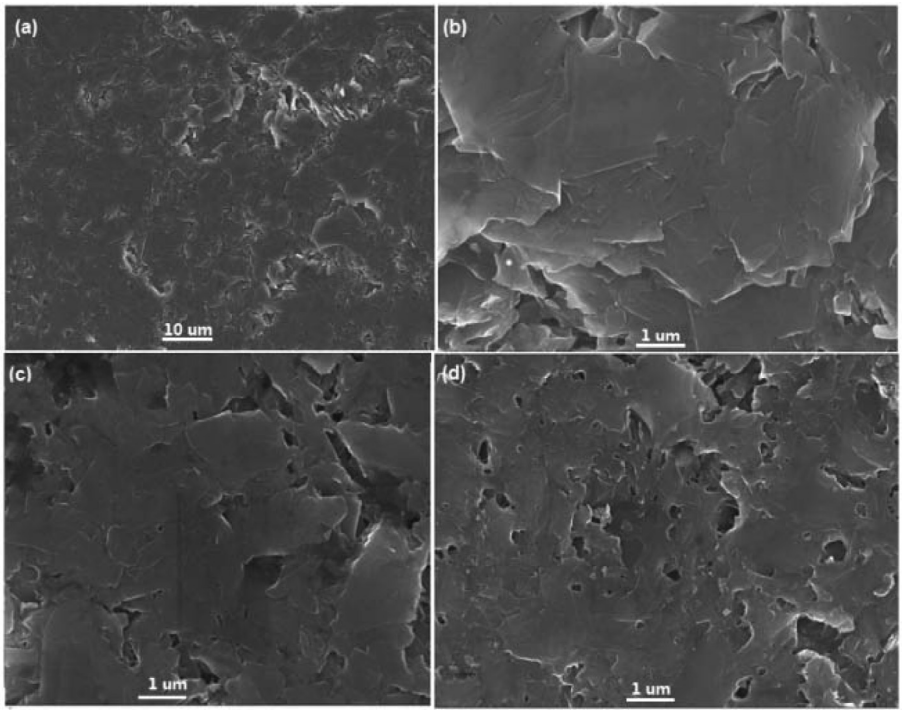

Fig. 1. Surface morphologies of matrix graphite before and after Xe ions irradiation. (a) and (b) are SEM images of virgin sample, (c) and (d) are SEM images of irradiated samples with Xe ions to fluences of 5.8×10^{14} ions/cm^2 and 2.9×10^{15} ions/cm^2, respectively.

defects of graphite matrix due to irradiation, which is also helpful in understanding the irradiation mechanism and to predict the radiation performance of matrix graphite.

In this work, matrix graphite of TMSR FEs was irradiated with 1 MeV Xe ions to various fluences (5.8×10^{14} ions/cm^2 and 2.9×10^{15} ions/cm^2) at room temperature. SEM, Raman spectroscopy and slow positron annihilation beam techniques were used to study the effects of Xe ions irradiation on microstructure, phase and defects of matrix graphite.

2. Experiments

2.1. Sample preparation and ion irradiation

Specimens used in this study were cut from matrix graphite spheres provided by Institute of Nuclear Energy and Technology (INET), Tsinghua University. Raw materials of matrix were natural-graphite, electro-graphite and phenol formaldehyde. They were firstly compressed to form a sphere with a diameter of 6 cm, followed by carbonization at 800 °C and purification at a temperature up to 1900 °C [13,14]. After heat treatments, the matrix graphite sphere has a density of 1.70–1.77 g/cm^3, a total porosity of approximate 20% and an anisotropic factor of 1.07.

For irradiation, specimens were cut into plates with a size of $0.8 \times 0.8 \times 1$ mm^3, then polished by hand with a flat surface. The surface morphology of as prepared specimen was shown in Fig. 1 (a) and (b) with different magnification. Ion irradiation experiments were performed with 1 MeV Xe ions at room temperature (RT) in a terminal of the 320 kV high-voltage experimental platform equipped with an electron cyclotron resonance (ECR) ion source in the Institute of Modern Physics, Lanzhou. Two specimens were irradiated to fluences of 5.8×10^{14} ions/cm^2 and 2.9×10^{15} ions/cm^2, respectively, corresponding to the maximum damage level of 1 DPA (displacements per atom) and 5 DPA, according to the TRIM calculation [15]. Details of the specimens and irradiation conditions were listed in Table 1.

Table 1
Details of the specimens, irradiation conditions and Raman results.

Specimen No.	1	2	3
Xe ion energy (MeV)	–	1.0	1.0
Ion fluence (ions/cm^2)	–	5.8×10^{14}	2.9×10^{15}
Damage peak (DPA)	–	1.0	5.0
D peak position (cm^{-1})	1350	1389	1388
D peak linewidth (cm^{-1})	38	258	228
G peak position (cm^{-1})	1580	1575	1560
G peak linewidth (cm^{-1})	21	113	132
I_D/I_G ratio	0.31	1.36	1.17

2.2. Characterization methods

Scanning electron microscope (SEM: MERLIN Compact) was performed to investigate the surface morphology of matrix graphite before and after irradiation.

Raman spectrum is a powerful and widely used tool to detect microstructure for carbon system due to the different bonding of carbon atoms [6–9]. Raman probes within an optical skin depth, which is appropriate for depth of ion irradiation-induced damage layer. Raman spectrum for specimens was measured with an Argon laser confocal Raman microscope (HORIBA XPloRA) using 532 nm wavelength at room temperature. The scanning range was from 1000 to 3500 cm^{-1} with a resolution of 1.0 cm^{-1}.

Positron annihilation spectroscopy (PAS) is a very sensitive tool to study the vacancy-type defects and phase transition. Especially the energy-tunable slow positron beam can be used for the study of depth profile of open structure defects (vacancy, cluster, voids and micro porosity) [16–18] induced by irradiation near surface or damage region. Since open structures often act as positron trapping sites with negative charge and low electrons density, they could trap positrons easily and then give an increased S parameter. The slow positron beam measurement was conducted in University of Science and Technology of China (USTC), to obtain the

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