Accumulation of Radiation Damage and the Change of the Mechanical Properties of Nuclear Graphite due to Ion Irradiation

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1. Introduction

Graphite is main neutrons moderating and reflecting material for High Temperature Gas Reactor (HTGR). Therefore, the prediction of properties change of nuclear graphite is one of the important subjects of research. Unfortunately, the test experiments in nuclear reactor require the long time and such experiments are laborious and expensive. The ion irradiation is very useful instrument for radiation damage production up to highest doses and it allows shorten the evaluation test period considerably. The ion irradiation possesses many advantages such as the convenient operation of irradiation parameters, an absence of radioactivity of specimens. But the ion irradiation limits the use of many traditional experimental methods because radiation damage layer is very thin. Nevertheless, the ion accelerators are successfully using for the preliminary prediction of the radiation resistance of nuclear materials. In this paper, the method of determination of the hardness and Young's modulus of the ion irradiated nuclear graphite, the kinetics of radiation damage accumulation and the dose dependences of the mentioned mechanical properties are being described.

2. Experimental

2.1 The specimens preparation and irradiation conditions

The fine grained nuclear graphite IG-110 and IG-430 produced by Toyo Tanso Co., LTD (Japan) were selected for study. These graphite materials are differed by initial raw materials. Graphite IG-110 was synthesized from petroleum coke and IG-430 was synthesized from coal coke. Also, the average grain sizes in IG-110 and IG-430 are about 20 μm and 10 μm, accordingly. The kinetics of radiation damage accumulation was monitored by Raman scattering measurements in the wave range 1000-2000 cm^{-1}. The specimens for Raman specters measurements had size (3×3×2) mm^3. The measuring surface of specimens mechanically polished and then peeled by adhesive tape. The surface of specimens for hardness measurements was polished by 0.05 μm alumina powder in finally. All specimens were ultrasonically rinsed in acetone before irradiation. The specimens were irradiated in vacuum by 3 MeV C^+ ions at beam current density 0.83μA/cm^2. The temperature of specimens didn't exceed 60 ^{0}C. The peak damage range is placed at ~3.2 μm depths accordingly of TRIM Code calculation [1].

2.2 The method of hardness measurement of the ion irradiated specimens

The ultra-microhardness tester DUH-200 was used for hardness measurements. The loading-unloading method was used for evaluation of mechanical properties of graphite. The definition of hardness as $\boldsymbol{H = P/A}$ ($\boldsymbol{P}$ - the maximal load, $\boldsymbol{A}$ – projected area of impression) was used for the hardness determination [2]. Because the real diamond Vickers indenter has non-ideal form the special calibration experiment was conducted for correct determination of the area dependence from depth. Also, the size of plastic deformation zone for graphite was determined. This value exceeded 2.3-2.6 times of indenter penetration depth. The analysis of loading-unloading curves of different multi-layer structures such as thin soft layer-hard endless layer, soft thin layer-hard thin layer-thick hard layer showed that the maximum of hardness value must be observed in the ion irradiated graphite. At that, the maximum of hardness value is placing before of the peak damage range at depths 0.5 μm and smaller in dependence of the irradiation dose. On basis of this analysis the hardness and the effective elastic modulus values in the maximum on depth dependences were used as representative values for evaluation of the mechanical properties of the ion irradiated graphite. The effective elastic modulus value $\boldsymbol{E_r}$ was determined by formula $\boldsymbol{E_r = 0.179\ S/h_c}$ ($\boldsymbol{S}$-slope of unloading curve, $\boldsymbol{h_c}$ – the residual penetration depth of indenter).

3. Results and Discussion

3.1 Accumulation of radiation damage

The Raman specters of un-irradiated specimens include the two peaks at 1360 (D-peak) and 1580 cm^{-1} (G-peak), which due by defects in crystal lattice of graphite and stretch vibration of C-C bond accordingly.

The ratio of intensity of D peak to intensity of G peak, which corresponds to perfection order of graphite crystal lattice, is smaller for graphite IG-110 than IG-430. This is in accord with more density of IG-430 graphite and the larger average grain size in IG-110.

Fig. 1 shows dose dependence of change of the full width on the half maximum (FWHM) of D and G peaks.

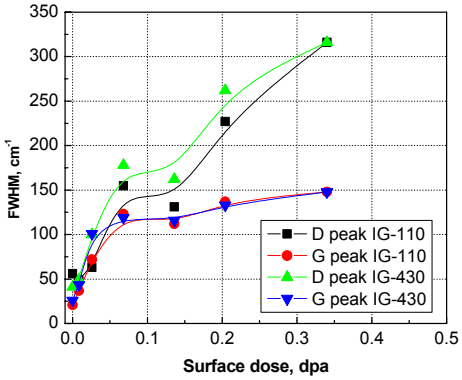

Fig.1. Dose dependences of the FWHM of D and G peaks at 3 MeV carbon ions irradiation

Dose dependences of ratio I_D/I_G have similar kind as dose dependences of FWHM of G peak in graphite. As is easy to see the two stages of damage accumulation are observed. In the beginning doses the FWHM of peaks increase sharply and then the saturation is observed. The values of FWHM increase again above the dose ~ 0.1 dpa. The analysis of the experimental data and comparison with known data of radiation damage accumulation in graphite at neutron irradiation show, that the first stage due by forming of point defects in graphite structure. The presence of saturation region and comparatively small dose of the saturation indicate on the limited volume of defect production source. It is possible, that these defects are being formed at defect places of lattice or near of impurities. The second stage of dose dependence was due by change of macrostructure predominantly. At that, the crystal structure of graphite subjected to considerable disordering right up to full transformation in amorphous state at the high irradiation doses. In particular, the decrease of the oxidation rate is observed at doses higher 0.1 dpa in the irradiated graphite IG-110 [3].

3.2 The hardness and the effective elastic modulus of the ion irradiated graphite

The relative changes of hardness and modulus were measured up to peak dose about 18 dpa (fig. 2). The hardness of graphite is increased in beginning doses, then the increase is being reduced and the following growth of the hardness is observed after dose about 10 dpa. In contrast to the hardness change the beginning growth of the effective elastic modulus is accompanied with following stabilization of the values at doses higher than 2-4 dpa. Starting from these differences of the hardness change and the elastic properties it can suppose that the changes of the plastic properties of the irradiated graphite are more sensible to the changes of macrostructure of graphite.

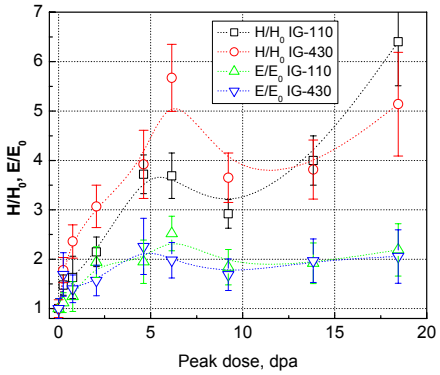

Fig.2. Relative change of hardness and the effective elastic modulus of graphite irradiated by 3 MeV carbon ions.

The elastic properties of the irradiated graphite are more determined by changes of defect structure of graphite lattice. It is necessary to note, that the relative increase of hardness is more in graphite IG-430. The similar behavior is observed for FWHM of D peak in Raman specters of IG-430 (fig. 1). Most probably this is connected with more small average grain size of crystallites and accelerated change of macrostructure.

4. Conclusion

1. The method of determination of the hardness and the effective elastic modulus of the ion irradiated graphite was being developed.
2. The accumulation kinetics of radiation damage at the ion irradiation has two stages in the range up to 0.4 dpa which due by forming of defects in graphite structure and the change of macrostructure.
3. The effective elastic modulus of graphite fast increases at beginning irradiation doses and then the values are being stabilized. The hardness of graphite increases in all dose range up to ~18 dpa.

REFERENCES

1. J.F. Ziegler, J.P. Biersack and U. Littmark. The Stopping and Range of Ions in Solids, Pergamon, 1985.
2. M. F. Doerner, W. D. Nix, A Method for interpreting the data from depth-sensing indentaion instrument. J. Mater. Res., Vol.1, p. 601, 1986.
3. Se-Hwan Chi, Gen-Chan Kim, Oxidation Behavior of Carbon Ion Irradiated IG-110 Nuclear Graphite, Proc. of KNS-2004, Fall Meeting.