Effect of Gamma-Irradiation on the Physical Properties of Nuclear Graphite

Gen-Chan Kim, Se-Hwan Chi, Jong-Hwa Chang
Korea Atomic Energy Research Institute, P.O.Box 105, Yuseong, Daejeon, Korea, 305-353, gchkim@kaeri.re.kr

1. Introduction

Gamma-radiation contributes a significant component of nuclear reactor radiation. Gamma-irradiation is used widely for the study of radiation effects in materials which are sensitive to electronic excitation, for instance, dielectrics and semiconductors. But, it is known that the electronic mechanisms of defect production are negligible in materials of high conductivity. Sometimes, such mechanisms are realized in the metals of high electronic energy loss [1]. However, it is reported that gamma-rays can induce atomic displacements in metals by compton effect [2,3]. In this work, preliminary results on the effects of gamma-irradiation from ^{60}Co isotope on the physical property of IG-110 nuclear graphite are presented.

2. Experimental

The IG-110 graphite specimens (10x10x2mm^3), packed in the glass ampouls in vacuum $\leq 10^{-2}$ Torr, were irradiated to $7\text{x}10^9$ R (flux: 765 R/s.) at $\leq 70^0$C. Changes in the microhardness due to irradiation were evaluated by the loading-unloading method using an ultra-microhardness tester DUH-200 (SHIMADZU). Test load was $9.8\text{x}10^{-3}$N. All the specimens for microhardness measurement were ultrasonically rinsed in acetone after mechanically polished with 0.05 μm alumina powder. Changes in the Raman spectrum were obtained using a LabRamHR Jobin-Yvon spectrometer of Ar-ion laser (λ = 514 nm). The parameters of Raman peaks were determined by Lorentzian fitting.

3. Results and discussion

3.1. *Evaluation of the recoil atom energy due to Compton electrons*

The average energy of ^{60}Co gamma-ray is about 1.25 MeV. The three main effects take place at interaction of gamma-rays with solid: photoelectric effect, Compton scattering and pair production [4]. The prevail mechanism of interaction of ^{60}Co gamma-ray in graphite is Compton scattering. As result the fast Compton electron and scattered gamma-quantum are created. The calculation shows that the maximal energy of Compton electrons reaches up to 1 MeV.

Compton electron can interact with graphite atoms by means of elastic impact. As result of elastic interaction a displaced carbon atom can be produced. We can estimate possibility of recoil atom production by next way. The energy passed to carbon atoms E_{max} (eV) from fast Compton electrons can be determined by the expression, taking into account the relativist effect of fast electrons [5]:

$$E_{\max} = \frac{2148 E_C \left(E_C + 1.0022\right)}{A} \tag{-1-}$$

where A is the atomic weight of carbon atom, $\mathbf{E_C}$ is the kinetic energy of electron (in MeV).

The energy necessary for displacing a carbon atom from the crystal lattice, i.e., E_d is about 25-35 eV [5,6]. If E_{max} is replaced by E_d = 25-35 eV, the minimum energy E_C of electrons which can displace the carbon atoms by the elastic interaction is calculated to be about **125-160 keV**. Then, from the the equation (1), the maximal energy passed by 1 MeV electrons to the recoil carbon atom is about **360 eV.** If compared this energy with the Ec of **125-160 keV**, it is clear that ^{60}Co gamma-rays could form new defects structure during irradiation.

3.2. *Hardness change due to gamma-irradiation*

Fig. 1 shows the dose dependence of the hardness change of IG-110 graphite after gamma-irradiation.

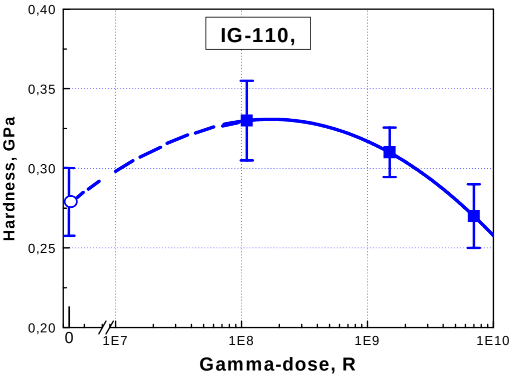


Fig. 1. The change of hardness of IG-110 graphite after ^{60}Co gamma-irradiation

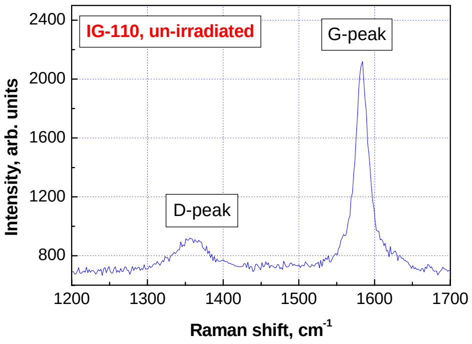
It is seen that, for the gamma dose of ~ $1\text{x}10^8$ R, the hardness show a maximum value of about 0.33 GPa. This value corresponds to about 17% increase from the unirradiated one. After the maximum hardness at about ~ $1\text{x}10^8$ R, it is seen that the hardness decreases with dose regaining the initial value after about $7\text{x}10^9$ R.

Irradiation-induced annealing of defects, for example, gamma heating, may be attributed to the present observation. From this observation, Gamma-irradiation is proved to ineffective

3.3. *Raman spectrum change due to gamma-irradiation*

To investigate the possible internal bonding structure change due to gamma irradiation, Raman spectroscopy was performed. The well-known two peaks were observed on the unirradiated specimen: D-peak (maximum at 1355 cm^{-1}) and G-peak (maximum at 1580 cm^{-1}) **(Fig. 2)**.

Fig. 2. Raman spectrum of un-irradiated graphite



The qualitative Raman spectrum pattern was not changed even after the maximum irradiation doses. However, the calculation of Raman parameters, such as the ratio of D peak intensity to G peak intensity (I_D/I_G), or the FWHM (full width at half maximum) of two peaks showed a noticeable change after different doses **(Table 1).**

Table 1. Raman parameters for un-irradiated and gamma-irradiated graphite.

Dose, R	I_D/I_G	FWHM D peak, cm^{-1}	FWHM G peak, cm^{-1}
0	0.15±0.01	48±2	21±0.2
$1.1x10^8$	0.21±0.01	53±3	23±0.3
$1.5x10^9$	0.14±0.01	46±3	20±0.2
$7.0x10^9$	0.16±0.01	47±3	21±0.3

Table 1 shows that a large change is observed in the intensity ratio (I_D/I_G) and the maximal change is observed at $1.1x10^8$ R.

It is evident from these results that gamma-irradiation changes the defect concentration (i.e., the ratio I_D/I_G). However, it is expected that the regularity of crystal lattice, i.e., crystallinity, is not changed practically since the change of FWHM of G peak is negligible.

It is known that the G peak is due to the stretch vibration of C-C bonds in graphite structure [7].

Taking into account the small penetration of laser beam (about 40-50 nm), it is understood that Raman test will only examine the surface layers of graphite. In contrast, the microhardness test examines about one micrometer layer from the graphite surface. In case of ^{60}Co gamma-irradiation, following discussions can be made as to the change of defect microstructure, thus, Raman spectrum change.

The energy spectrum of gamma-rays of majority nuclear reactors extends up to 10 MeV [2,3]. In this case, the maximal energy $\boldsymbol{E_{max}}$ to the recoil carbon atoms is calculated to be about 20 keV. Thus, the delievered energy is considerably larger than the displacement energy for carbon atoms in graphite (25-35 eV). From these discussions, it is clear that the gamma-rays of nuclear reactor can produce matrix defects by Compton effects of elastic interaction.

4. Conclusion

Compared to the dose, the Gamma-irradiation-induced microhardness change in nuclear graphite was negligible. From this observation, it is predicted that the expected Gamma-irradiation-induced damage of VHTR nuclear graphite components will not be so large during a reactor lifetime.

5. References

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