X-Ray Diffraction Measurements of Ion-Irradiated Graphite

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1. Introduction

In high temperature gas cooled reactors, neutron irradiation causes dimensional changes and reduction of the crystallite size of nuclear grade graphite, which are determined from shift and broadening of X-ray diffraction peak, respectively [1-3]. For the accurate measurement, internal standard method is generally used to remove instrumental error and unexpected shift and broadening of diffraction peak caused by deep penetration depth of x-ray into the specimen. However, ion-irradiation experiment is performed with bulk specimen, and X-ray penetrates too deeply through damaged layer by ion-irradiation. Therefore, this study investigates the method of the X-ray measurement for thin damaged layer of ion irradiated graphite by standard method.

2. Experimental

2.1 Test Specimens and Equipment

Powder X-ray diffractometer (XRD) was used to make a comparison between internal and external standard method. IG-11 graphite powder was sieved less than 45 ㎛. Silicon powder of high purity (99.999%) below 25 ㎛ was used for standard material. Sample for internal standard method was a mixture of graphite with 20 mass% silicon, and graphite for external standard method was measured after the measurement of the silicon sample.

Proton irradiated IG-110 graphite was bulk specimen (10 x 10 x 1 mm^3), which was measured by thin-film XRD with external standard method.

2.2 Test Conditions

X-ray source is $Cu_{K\alpha}$. K_β was filtered with graphite monochromater. Accelerating voltage and current is 40kV and 45mA for powder XRD and 30kV and 60mA for thin film XRD, respectively. Profile of Thin-film XRD was obtained by step scanning with 0.01 internal, 2-theta axis to scan, 2° incident angle and 2 s dwelling time. Profile of powder XRD was obtained by continuous scanning with 0.25/min and theta/2-theta axis at 24°~29°, 75°~80° scanning angle(2θ).

2.3 Pattern Processing of X-ray Diffraction

Diffraction peak was corrected by intensity correction factor, $FCT=LPAf_c^2$, where Lorentz(L), polarization(P), absorption(A) and atomic scattering factors(f_c) because of more intensive peak at low diffraction angle. A little smoothing was performed due to intensive and clear peak. After removing background and $K\alpha_2$, profile was fitted by Pseudo-Voigt fitting function [4].

2.4 Measurement of X-Ray Penetration Depth

Bulk sample was polished up to 60, 80, 120 and 160 ㎛ to measure the damaging depth of graphite by 3 MeV proton irradiation. And then the X-ray measurement was performed with putting polished graphite on Ag film.

3. Results and Discussions

Figure 1 shows X-ray diffraction peak of IG-11 graphite measured by Internal and external standard method.

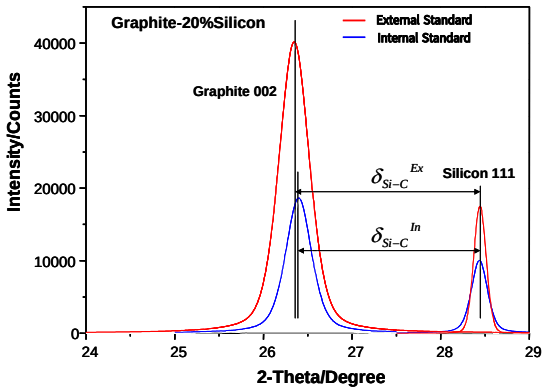

Figure 1. X-ray diffraction peak of IG-11 graphite by internal and external standard method.

The corrected diffraction angle of graphite is
For 002 peak, 2θc =2θsi - δsi-c (1)
For 110 peak, 2θc =2θsi + δsi-c
Crystallite size can be calculated as
For the 002 reflection, Lc=9.1/β (2)
For the 110 reflection, Lc=11.3/β
Where β is true FWHM.

However, FWHM strongly depends on the peak intensity. Diffraction peak by external standard method is more intensive than that by internal standard method as shown in Figure 1, because the intensity of X-ray diffraction peak depends on weight fraction of graphite and silicon. Therefore, correction by normalization was performed.

The results of X-ray measurement by internal and external standard method are shown in Table 1.

Table 1. Crystallite size and lattice parameter of IG-11 graphite by internal and external standard method

	Internal Standard Method		External Standard Method	
	c-direc.	a-direc.	c-direc.	a-direc.
Crystallite size(nm)	51.996	99.036	40.837	81.381
Lattice parameter(Å)	3.3654	1.2299	3.3715	1.2301

Large difference between the crystallite sizes may be caused by different absorption coefficient between silicon and graphite. And a little shift of the diffraction peak may result from residual stress after polishing.

The results of X-ray diffraction measurement of 3 MeV, 3 μA/4hr proton-irradiated IG-110 graphite by external standard method are shown in Table 2.

Table 2. Crystallite size and lattice parameter of proton irradiated IG-110 graphite

	Unirradiated IG-110 graphite		Irradiated IG-110 graphite	
	c-direc.	a-direc.	c-direc.	a-direc.
Crystallite size(nm)	43.409	98.099	44.915	102.21
Lattice parameter(Å)	3.3682	1.2310	3.3710	1.2310

The thickness of proton irradiated layer is just about 80 μm. Thus, X-ray penetration depth must be less than the thickness of damaged layer. Theoretically, X-ray penetration depth, x, can be obtained as follows:

$$G_x = 1 - \exp[-2\mu x / \sin\theta] \quad (5)$$

where G_x is the fraction of the total diffracted intensity contributed by a surface layer of depth x (0.95). θ the incident angle (2°) and μ the absorption coefficient ($7.50cm^{-1}$) [5]. From eq. (5), penetration depth is about 70 μm. However, measured penetration depth of IG-110 graphite was the deeper (above 160 μm) than calculated one because of many pores within graphite matrix (Figure 2).

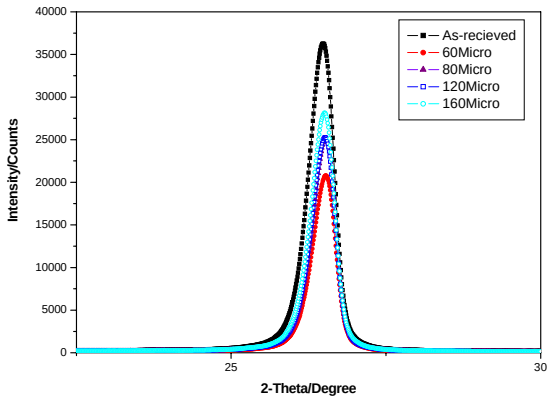

Figure 2. X-ray diffraction peak of IG-110 graphite of different thickness for the measurement of X-ray penetration depth

4. Summary

There are some differences as a result of comparison between internal and external standard method. Thin-film XRD was used to measure the thin damaged layer by proton irradiation. Experiment was performed by external standard method to measure bulk sample accurately. A little changes of crystallite size and lattice parameter by small dose were observed. X-ray penetrates too deeply above damaged layer of graphite despite of small X-ray incident angle.

Acknowledgments

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