Evaluation of Irradiation Characteristics of ETU-10 Nuclear Graphite

Se-Hwan Chi[1], Hiroshi Okuda[2] and Jun Ohashi[2]
[1] *Global Institute for Nuclear Initiative Strategy (GINIS),*
302 Nutopia Bd., Yuseong-gu, Daejeon, 34166, Korea
[2] *FGM Unit, IBIDEN,*
1120-1, Suemori, Godo-cho, Anpachi-gun, Gifu pref.,503-2321, Japan
[1] *E-mail: shchi5301@gmail.com*

1. Introduction

Currently there are more than 50 small modular reactor (SMR) designs under development for different applications in all principal reactor lines: water cooled reactors, high temperature gas cooled reactors, liquid-metal, sodium and gas cooled reactors with fast neutron spectrum, and molten salt reactors. The key driving force behind the SMR developments are flexible power generation and increased applications, which offer better economic affordability [1].

Of these SMRs, 15-20 designs are high temperature gas-cooled reactors (HTGR) and molten salt reactors (MSR) with graphite moderation. In these reactors, graphite will be used for the construction of major core components including the fuel block and reflector.

In a graphite moderation reactor, the graphite core components are subjected to neutron irradiation under high temperature helium gas or salt environments, and subsequently manifest dimensional and properties changes. It is well known that these changes in dimension, physical and mechanical properties degrade the integrity of the graphite components, menacing the safety of the reactor.

Thus, for the reactor designer, selecting the graphite for the design and construction of a reactor is an important task, and requires information through which the irradiation characteristics of candidate grades can be compared.

In this study, to provide the graphite components designer with reference information for graphite selection, the irradiation characteristics of ETU-10 (Ibiden), a newly introduced nuclear graphite grade, were explored based on comparisons made between the reported irradiation test data for ETU-10, IG-110, and NBG-25.

2. Materials and irradiation condition

Table 1 and 2 provide a summary of the four different nuclear graphite reactor irradiation tests compared in this study and a short description of graphite grades compared in this study, respectively. Table 1 shows that the irradiation test data on ETU-10 and IG-110 were produced from the same reactor (HFIR), and the data irradiated at or near 600℃ were selected for comparison with each other from the HFIR (ETU-10), HFIR(IG-110, 1996), HFIR(IG-110, 2017), and HFR (INNOGRAPH) [2][3][4][5].

Table 1. Four nuclear graphite irradiation tests compared in this study.

Reactor	HFIR*			HFR**
Irr. Grade	ETU-10 [2]	IG-110 (1996) [3]	IG-110 (2017) [4]	INNOGRAPH [5]
Irr. Temp.	342~667℃	600℃	300 - 750℃***	750℃, 950℃
dpa	3~30	~25	5-30 dpa	4.1~ 25 (750℃) 15 (950℃)
Remark	598℃ and 667℃ irradiation data were used for comparison with HFIR (IG-110) and HFR (INNOGRAPH), respectively.		*** Target temperature. Data from 600℃ target temperature were used for comparison with IG-110 (1997) and ETU-10.	IG-110, NBG-10, -17, -18, -25, PCEA, PCIB, PPEA were irradiated in the INNOGRAPH project. Data from 750℃ Irradiation were used in this study.

*HFIR: High Flux Isotope Reactor in Oak Ridge National Laboratory (ORNL), USA. ** HFR: High Flux Reactor in Petten, The Netherlands

Table 2 shows that the ETU-10, IG-110, and NBG-25 have the same grain size and forming method. Table 3 compares the reported irradiation test data from the respective reactor irradiation test employed for comparison in this study. Based on this table, the dimensional change (DC), thermal conductivity (TC), elastic modulus (dynamic young's modulus) (EM), and coefficient of thermal expansion (CTE) data in common from each irradiation test were selected for comparison in this study.

Table 2. Characteristics of the nuclear graphite grades compared in the present study.

Grades	Manufacturer'	Cokes	Forming method	Grain size
ETU-10	Ibiden	pitch	iso-moulding	fine
IG-110	Toyo Tanso	petro-	iso-moulding	fine
NBG25	SGL	petro-	iso-moulding	fine

Table 3. Reported irradiation test data from each of the respective reactor irradiation tests compared in this study.

	HFIR* (ETU-10) [2]	HFIR (IG-110, 1996) [3]	HFIR (IG-110, 2017) [4]	HFR** (INNOGRAPH) [5]
Dimensional change	○	○	○	○
Thermal conductivity	○	○	○	○
Elastic modulus	○	○	○	○
Coefficient of thermal expansion	○	X	○	○
Electrical resistivity	X	○	X	X
Fracture strength	X	○	X	X
Four point flexure strength	X	X	○	X
Tensile strength	X	X	X	○
Creep	X	X	○	X

3. Comparison of reactor irradiation studies: HFIR (ETU-10), HFIR (IG-110, 1996), HFIR (IG-110, 2017) and HFR (INNOGRAPH) [2][3][4] [5].

3.1 Volume change (VC)

A comparison of the volume change behaviors with neutron irradiation, i.e., (△V/V₀, %)-dpa was conducted between the ETU-10 and IG-110 in Figure 1 by overlappig the HFIR (ETU-10) and HFIR (IG-110, 2017) data on the HFIR (IG-110, 1996) data, and between the HFIR (ETU-10) and HFR (INNOGRAPH) of 8 grades in Figure 2 by overlapping the HFIR (ETU-10) data on the HFR (INNOGRAPH) data, respectively.

In Figure 1, it is seen that, while HFIR (ETU-10) and HFIR (IG-110, 2017) show similar dimensional change behavior in turn-around and cross-over, large differences are observed between these two irradiations and HFIR (IG-110, 1996). Table 4 shows a summary of predicted turn-around dimensional change (contraction %), and cross-over dose (dpa) from Figure 1, where, it is seen that the maximum dimensional change (contraction) of ETU-10 (-4%) is smaller than IG-110 (-6.3 % and - 7 %).

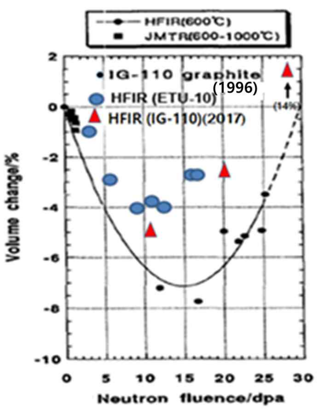

Figure 1. Comparison of volume change behaviors with neutron dose: HFIR (ETU-10), HFIR (IG-110, 1996), and HFIR (IG-110, 2017) [2][3][4].

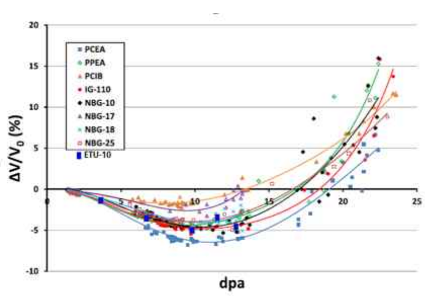

Figure 2. Comparison of the volume change behaviors with dose: Data from HFIR (ETU-10) were overlapped on the INNOGRAPH dimensional change data (HFR Innograph, Table 1) [2][5].

Table 4. Prediction of turn-around behavior and cross-over dose from Figure 1.

	Turn-around volume contraction(%) and dose (dpa)	Cross-over (dpa)	Irr. Temp (℃).
IG-110, 1996	-7, 15	30	600
IG-110, 2017	-5, 10	~20	667 *
ETU-10	-4, 10	20-25	598

*actual temperature

In Figure 2, it is worth noting that the ETU-10, IG-110 and NBG-25 with a fine grain and iso-molded grade showed similar turn-around behaviors at around 11-12 dpa and -5 % of volume contraction. However, while the high dose data are limited for ETU-10, some differences are predicted in the cross-over behaviors of the three compared grades. Compared to the PCEA with medium grain and extrusion molding, the volume contraction of these three fine grain and iso-molded grades appeared to show about 30% smaller contraction, i.e., -5% versus -7%, at a similar turn-around dose range, i.e., 10 dpa ~13 dpa.

3.2 Thermal conductivity (TC)

Three different TC values are being reported as an un-irradiated TC of IG-110: 120 W/m°K, 130 W/m°K and 160 W/mK. The present study regards 130 W/m°K as a reasonable TC value for IG-110 for comparison. The comparison of the irradiation-induced changes in the TC of HFIR (ETU-10) with those of HFIR (IG-110, 1996) and HFIR (IG-110, 2017) are shown in Table 5. Table 5 only shows a measurement for HFIR (IG-110, 2017). It is shown that both the un-irradiated and irradiated TC of HFIR (ETU-10) were smaller than those of HFIR (IG-110, 1996) and HFIR (IG-110, 2017) for 50% and 25%, respectively. Also, the HFIR (ETU-10) and HFIR (IG-110, 1996) showed a similar irradiation-induced decreasing rate of changes (%) in TC after irradiation. Related analyses showed that the TC of HFIR (ETU) was 16-24 W/mK for 12-20 dpa, and stayed at about 16 W/mK from about 20 dpa.

Table 5. Comparison of the irradiation-induced thermal conductivity change for HFIR (ETU-10), HFIR (IG-110, 1996), and HFIR (IG-110, 2017). Unit: W/m°K.

Dose (dpa)	HFIR (ETU-10)	HFIR (IG -110, 1996)	HFIR (IG -110, 2017)	Rate of Change (%)	
				ETU-10	IG-110 (1996)
Un-irr.	104	160	130	104	160
11.9	25.0	35.0	42.3	76.0	78.1
21.8	16.0	27.5		84.6	82.8
24.8	16.0	22.5		84.6	85.9

Figure 3 compares the irradiation induced changes in the TC of ETU-10 with those of HFR (INNOGRAPH) by overlapping the TC measurements from HFIR (ETU - 10) on the HFR (INNOGRAPH) TC measurements.

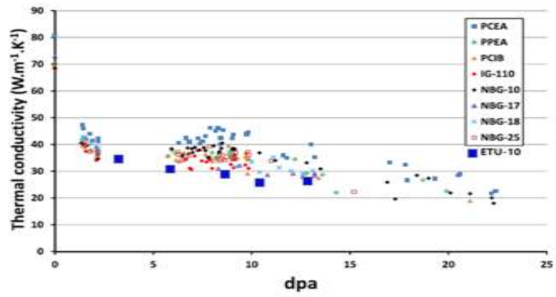

Figure 3. Irradiation-induced changes in TC of HFIR (ETU-10) and HFR (INNOGRAPH) [2][5].

It is seen that HFIR (ETU-10) and HFR (INNOGRAPH, IG-110, NBG-25) show a fast decrease in TC with irradiation (< 2-3 dpa) from the un-irradiated TC, and these three fine grain and iso-molding grades form a lower boundary in the HFR (INNOGRAPH) TC measurements.

3.3 Elastic modulus (EM)

To compare the irradiation-induced change in elastic modulus behaviors among HFIR (ETU-10), HFIR (IG-110, 1996), HFIR (IG-110, 2017), and HFR (INNOGRAPH, IG-110, NBG-25), the dynamic Young's modulus (DYM) measurement data from the HFIR (ETU-10) and HFIR (IG-110, 2017) were overlapped on the HFIR (IG-110, 1996) data (Figure 4) and data from the HFIR (ETU-10) were overlapped on the HFR (INNOGRAPH) DYM-dpa data (Figure 5).

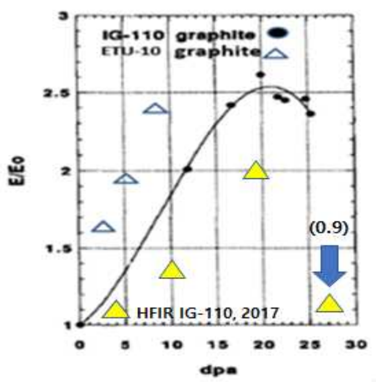

Figure 4. Comparison of irradiation-induced changes in DYM of HFIR (ETU-10), HFIR (IG-110, 2017), and HFIR (IG-110, 1996). Un-irradiated DYM of ETU-10 and IG-110 are 9.7-9.8 GPa and 9.1-10.2 GPa, respectively [2][3][4].

Figure 4 shows that the irradiation-induced change in DYM, i.e., E/Eo, of HFIR (ETU-10) was larger than those of HFIR (IG-110, 1996) and HFIR (IG-110, 2017) about 10% - 65% for 3-20 dpa. Here, the large difference in the irradiation-induced E/Eo behavior between the HFIR (IG-110, 1996) and HFIR (IG-110, 2017) is notable, since both results were obtained from the same grade (IG-110), same reactor (HFIR), same dose, at similar irradiation temperatures, Table 1.

Both grades showed a large increase in E/Eo after turn-around.

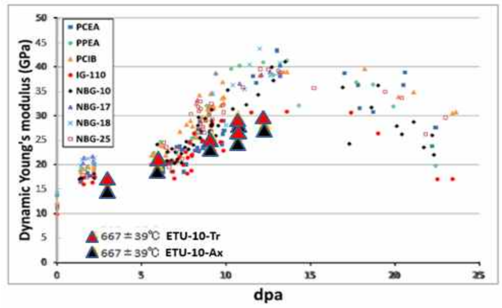

Figure 5. Comparison of DYM-dpa behaviors between HFIR (ETU-10) and HFR (INNOGRAPH). Here, HFIR (ETU-10) data are overlapped on HFR (INNOGRAPH) data for comparison [2][5].

Figure 5 shows that, with irradiation, both the DYM of HFIR (ETU-10) and the HFR (INNOGRAPH, IG-110, NBG-25) of fine grain and iso-molding were scattered along the lower boundary of the HFR (INNOGRAPH) measurements up to about 15 dpa.

3.4 Coefficient of thermal expansion (CTE)

The HFIR (ETU-10) was compared with HFIR (IG-110, 2017) since no CTE data were available from HFIR (IG-110, 1996). Figure 6 compares the CTE-dpa behaviors of HFIR (ETU-10) with that of HFIR (IG-110, 2017) by overlapping the CTE-dpa data from HFIR (IG-110, 2017) on the CTE-dpa data of HFIR (ETU-10). Figure 6 shows that the HFIR (ETU-10) data are scattered at around 2.8-3.2 x 10^{-6} K^{-1} and the HFIR (IG-110, 2017) shows a nearly constant CTE of 4.2 x 10^{-6} K^{-1} for 10 – 25 dpa. It may be concluded from Figure 6 that, while both grades show a similar CTE-dpa behavior up to about 15 dpa, HFIR (IG-110, 2017) appears to show a higher CTE value over HFIR (ETU-10) for 10-30 % up to 25 dpa from about 15 dpa.

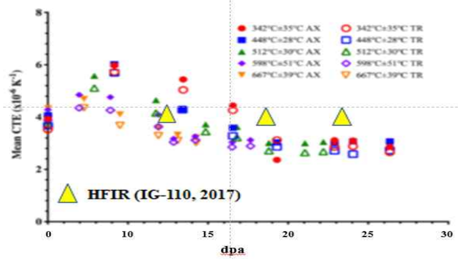

Figure 6. Comparison of CTE-dpa behaviors between HFIR (ETU-10) and HFIR (IG-II0, 2017). Here, HFIR (IG-110, 2017) data are overlapped on HFIR (ETU-10) data for comparison [2][4].

Figure 7 compares the CTE-dpa behaviors of the HFIR (ETU-10) and HFR (INNOGRAPH) by overlapping the HFIR (ETU-10) CTE-dpa data on the HFR (INNOGRAPH) CTE-dpa data. Figure 7 shows that HFIR (ETU-10) and HFR (INNOGRAPH, IG-110, NBG-25) show a rather similar CTE-dpa scattering behavior along the lower boundary of HTR (INNOGRAPH) CTE-dpa measurements.

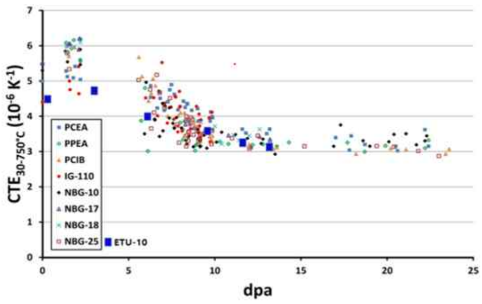

Figure 7. Comparison of CTE-dpa behaviors between the HFIR (ETU-10) and HTR (INNOGRAPH) [2][5]. Here, the HFIR (ETU-10) CTE-dpa data are overlapped on the HTR (INNOGRAPH) CTE-dpa data.

4. Discussion

The similar irradiation characteristics observed in the previous section on VC, TC, DYM, and CTE for ETU-10, IG-110, and NBG-25 of fine grain and iso-molding can be understood from their similarities in their microstructural characteristics. It is well known that several of the physical, thermal, and mechanical properties of graphite are determined largely by the coke particle size and forming method. Further, it is worth noting that all the grades compared in this study were already high quality nuclear graphite grades, basically satisfying all the mandatory material specifications in ASME Section III HHA-I.

However, some limited discussion can be offered to characterize the irradiation-induced property changes in the ETU-10 based on the comparison made with the limited irradiation data in Section 3.

ETU-10, IG-110, and NBG-25 of fine grain and iso-molding appeared to show a similar turn-around behavior. The comparison made with IG-110 with limited data on VC, however, tended to show that the VC was smaller for ETU-10, with a similar turn-around dpa.

For thermal conductivity, again the ETU-10, IG-110, and NBG-25 tended to show a similar changing behavior with irradiation, but, the limited data tended to show that the ETU-10 had a smaller TC, in both un-irradiated and irradiated conditions to IG-110. The smaller TC observed in ETU-10 may be attributed to the smaller crystallite and grain size in ETU-10 to IG-110. The observed similar irradiation-induced TC decreasing rate between the ETU-10 and IG-110 may imply a microstructural similarity between the grades in radiation defect thermal resistivity.

For DYM, ETU-10 appeared to show a larger increase to IG-110 after irradiation. It is known that the larger increase in DYM after irradiation has a close relationship with an increased sensitivity to thermal stress in the larger graphite components during operation.

For CTE, the ETU-10, IG-110, and NBG-25 tended to show a similar changing behavior up to about 15 dpa. However, limited data showed that IG-110 tended to show a slightly higher value to ETU-10 for 15~25 dpa.

5. Conclusions

Graphite has a major role in the graphite moderation reactor, as a structural component and core forming material. Thus, reliable reference information based on reactor irradiation testing is critically important for the design and construction of a graphite moderation reactor. To provide the graphite components designer with reference information for graphite selection, the irradiation characteristics of ETU-10 (Ibiden), a newly introduced nuclear graphite grade, were explored by comparing the dimensional change, thermal conductivity, elastic modulus (dynamic Young's modulus), and coefficient of thermal expansion of ETU-10, IG-110, and NBG-25. Limited data tended to show a smaller dimensional change with irradiation in ETU-10 to IG-110. While a comparison was made without considering the irradiation temperature effects, over all, all three grades of fine grain and iso-molding compared in the present study tended to show similar irradiation behavior for the properties examined, satisfying the ASME Section III Div. 5 HTR materials requirements (ASTM D 7219 and D 7301).

To reliably compare the irradiation characteristics of the grades of concern, data produced from the same irradiation conditions are needed.

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