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Development of non-destructive evaluation methods for degradation of HTGR graphite components

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ABSTRACT

To develop the non-destructive evaluation method for degradation of HTGR graphite components, the applicability of the micro-indentation method to detect residual stress was studied. The fine-grained isotropic graphites IG-110 and IG-430, the candidates for the VHTR, were used. The following results were obtained:

(1) The residual stress in a graphite block at the HTTR in-core condition was analyzed. It was suggested that, for the components in the VHTR which would be used at much severer condition, the development of lifetime extension methods is an important subject.
(2) The micro-indentation behavior at stress free condition was investigated with some indenters. The spherical indenter R0.5 mm was selected to detect the specimen surface condition sensitively. The indentation load of 5 and 10 N was selected to avoid the pop-up effect in the loading process.
(3) The relationship between the average value of normalized indentation depth and compressive stress of the specimen was expressed by an empirical formula. It would be possible to evaluate the residual stress by the indentation behaviour. It is necessary to assess the variation of data with statistic method and it is the subject of future study.

1. Introduction

Graphite materials are used for reactor internals of high temperature gas-cooled reactors (HTGR), which provides high temperature coolant helium gas at about 950 °C at the reactor outlet. It is possible to utilize this high temperature helium gas not only for power generation but also as a process heat source for a hydrogen production system. The very high temperature reactor (VHTR), a HTGR, is a promising candidate for the Generation IV nuclear energy system [1]. For research and development (R&D) on the VHTR, international collaboration is carried out through the Generation IV international forum [1]. The R&D on graphite is one of the important subjects for the VHTR. The graphite components are degraded by neutron irradiation at high temperatures. They show creep behavior above 500 °C under neutron irradiation as well as irradiation-induced dimensional change. These effects cause stress in the components and are a crucial factor in the component's lifetime [2].

The high temperature engineering test reactor (HTTR), having the maximum thermal power of 30 MW, operated by the Japan Atomic Energy Agency (JAEA) is the first HTGR in Japan [3,4]. JAEA carries out R&D for the VHTR based on the technologies established through the HTTR construction and operation. The HTTR uses a pin-in block type fuel element which is a hexagonal graphite block, 360 mm across the flats and 580 mm in height. The fine-grained isotropic graphite IG-110 (Toyo Tanso Co.) is used for the core components. The residual stress in the components, caused by heat and neutron irradiation, is evaluated by visco-elastic analysis with VIENUS code. For the components in the HTTR, the lifetime is determined by the comparison of stress analysis result and the allowable stress limit of IG-110 which is set as 1/3 of the specified minimum ultimate strength, Su, for the core components. Since the in-core graphite components of the VHTR would be used at severer condition than that of the HTTR, it is important to assess their structural integrity during the component lifetime which is expected to be extended.

It would be possible to prolong the allowable component lifetime if we can measure the residual stress directly, as stress analysis is a major factor in component lifetime regulation and safety margin. The present authors are trying to develop the non-destructive evaluation technique for the degradation of mechanical properties of graphite components by using a micro-indentation method [5–8]. Although Ishihara and Oku experimentally showed the possibility of this method to detect the tensile stress condition of IG-11 graphite with the Vickers indenter [9], it is necessary to

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evaluate the applicability of this method for compressive stress conditions. In this study, the applicability of the micro-indentation method is discussed and experimental results are shown.

2. Analysis

Since the details of the VHTR in-core condition are not fixed yet, we use the HTTR in-core condition for the stress analysis on the graphite block. The cumulated stress in the graphite block by the reactor operation is calculated by VIENUS code which is a FEM code with visco-elastic analysis [10]. Fig. 1 shows the shape of pin-in block type graphite fuel block used in the HTTR and Fig. 2 shows the created mesh for the analysis, which is the model for the upper surface of the block. The cumulated stress by the reactor operation is analyzed for the HTTR in-core condition with reactor outlet temperature of 850 and 950 °C. The subjected fast neutron fluence is set as 1.0×10^{25} n/m^2.

3. Experimental

3.1. Graphite specimens

Two grades of fine-grained isotropic graphites manufactured by isostatic pressing, IG-110 and IG-430 graphites (Toyo Tanso Co.), are used in this study. Their typical material properties are shown in Table 1. IG-110 is used for the in-core graphite components in the HTTR and it is made from petroleum coke. IG-430 has about 20–40% higher strength than IG-110 and it is an advanced grade for the in-core components of the VHTR. It is made from coal tar pitch coke. Both are candidate graphite grades for the in-core graphite components of the VHTR. For the micro-indentation test, specimens with $5 \times 5 \times 10$ and $10 \times 15 \times 20$ mm^3 were prepared for compressive condition. Flat plate type specimens were used for tensile conditions, with the gauge being 15×10 and 15×15 mm^2 for IG-110, and 10×5 and 10×10 mm^2 for IG-430.

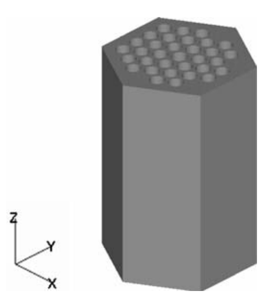

Fig. 1. In-core graphite block for analysis.

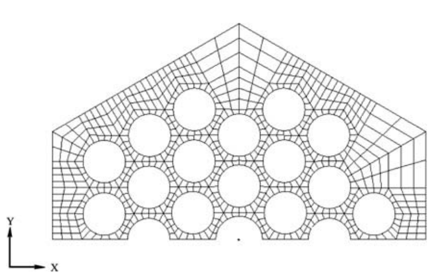

Fig. 2. FEM mesh for residual stress analysis with VIENUS code.

Table 1
Typical material properties of IG-110 and IG-430 graphites

Material	IG-110	IG-430
Bulk density (mg/m^3)	1.78	1.82
Tensile strength (MPa)	25.3	37.2
Compressive strength (MPa)	76.8	90.2
Young's modulus (GPa)	8.3	10.8
Poisson's ratio	0.14	–
S_{ut} value for tensile (MPa)	19.4	–
S_{uc} value for compressive (MPa)	61.4	–

S_u: specified minimum ultimate strength is determined from statistical treatment of strength data such that the survival probability is 99% at a confidence level of 95%. S_{ut}: S_u value for tensile, S_{uc}: S_u value for compressive.

3.2. Micro-indentation test

The micro-indentation behaviors on graphite materials are measured by a testing machine (SHIMADZU Co.) shown in Fig. 3. The residual compressive or tensile strains on the graphite specimens are simulated by applying compressive or tensile loads on the specimen in the horizontal direction as shown in Fig. 4, and the micro-indentation behaviors are examined by the indenter in the vertical direction. The strain is measured by the strain gauge attached on the side surface of the specimen. Five types of indenters, spherical shape with 0.5, 1.0 and 2.0 mm in radius, Vickers and Knoop, are used to examine the indentation behaviour on the indenter shape dependency. The relationship between indentation load and indentation depth is continuously recorded during the indentation test. The maximum indentation load is set as a parameter in the range from 5 to 150 N.

4. Results and discussion

4.1. Residual stress analysis

The analytical results of residual stress on the graphite block are shown in Figs. 5 and 6 for the operation mode of reactor outlet temperature of 850 and 950 °C respectively. We can see that the residual stress is in the range from −4 (compressive) to +4 (tensile) MPa for the 850 °C and from −6 to +6 MPa for the 950 °C operation. It is obvious that the severer operational condition, the larger cumulative residual stress is. The Su value for the IG-110, as shown in Table 1, is 19.4 and 61.4 MPa for compressive and tensile strength, respectively. The limit of the cumulate stress is determined as 1/3 Su for the core components in the HTTR. Although there is some margin between the limit and residual stress for the HTTR condition, it is necessary to develop lifetime extension methods for the components in the VHTR which would be used at much severer condition than in the HTTR.

It is thought that there would be two possible solutions for this issue. One is to use the stronger graphite, e.g. IG-430 rather than IG-110. The other method is to measure the residual stress directly by experimental method, because the residual stress analysis includes some safety margin for the uncertainties of in-core condition at reactor operation. If we can measure the residual stress directly, it is possible to reduce the safety margin and to extend the lifetime. We tried to develop the measurement method with the micro-indentation technique.

4.2. Micro-indentation behavior on indentation load and indenter shape

Micro-indentation behavior on IG-110 and IG-430 graphites at stress free condition was studied with each indenter [8]. Fig. 7 expresses the typical relationship between the indentation load and

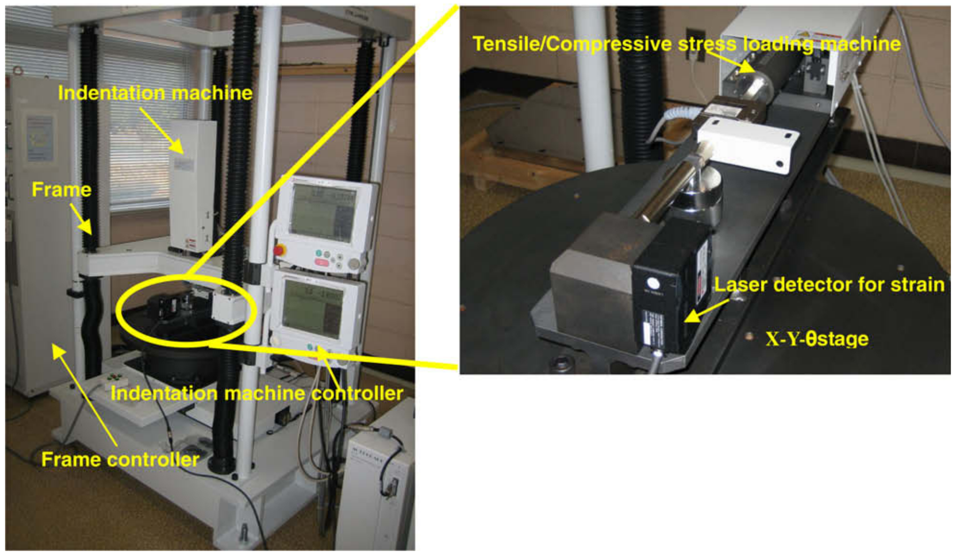

Fig. 3. Micro-indentation testing machine for graphite stressed condition.

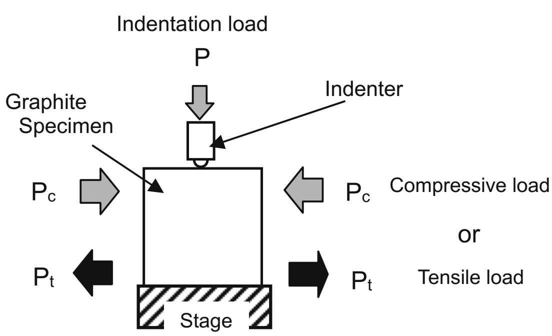

Fig. 4. Micro-indentation test method for graphite at tensile and compressive stress condition.

depth during the test. The maximum indentation load is set as 50 N. At the maximum load point, the Vickers indenter shows the largest depth and the spherical indenter R0.5 mm shows the second. Since the indentation behavior depends on the surface condition of the specimen, it is thought that these indenters are sensitive enough to detect the specimen.

Fig. 8 shows the experimental results of the relationship between the maximum indentation depth and the maximum indentation load. It is seen that the depth almost linearly increases with the load for each indenter. The experimental data of the Vickers indenter contains a larger variation than that of the other indenters. The spherical indenter R0.5 mm has the largest depth, except for the Vickers indenter.

The depth of maximum indentation load with 150 N for IG-110, Fig. 8(a), seems to be small compared to the linear extrapolation from the data with small indentation load. The surface conditions of IG-110 after the indentation test with the spherical indenter R0.5 mm are shown in Fig. 9. For the maximum indentation load with 150 N, the circular imprint is quite clear compared to that at the load with 50 N. Fig. 10 expresses the continuous relationship between the indentation load and indentation depth with R0.5 mm indenter for each maximum indentation load. The obtained curves show similar shape except the maximum load of 150 N which expresses the pop-up at loading process. It is suggested that the circular imprint shown in Fig. 9 was made at the loading process. It is thought that the low indentation load should be selected to avoid the pop-up effect. From this point, we selected the maximum indentation load of 5 and 10 N with the spherical indenter R0.5 mm for the stress condition measurements.

4.3. Application to stress condition

We are studying the applicability of the micro-indentation method for graphite materials [6–8]. The applicability for the stressed condition was examined.

(1) Compressive stress condition

The indentation behavior under the compressive stress condition was examined. The maximum indentation depth for compres-

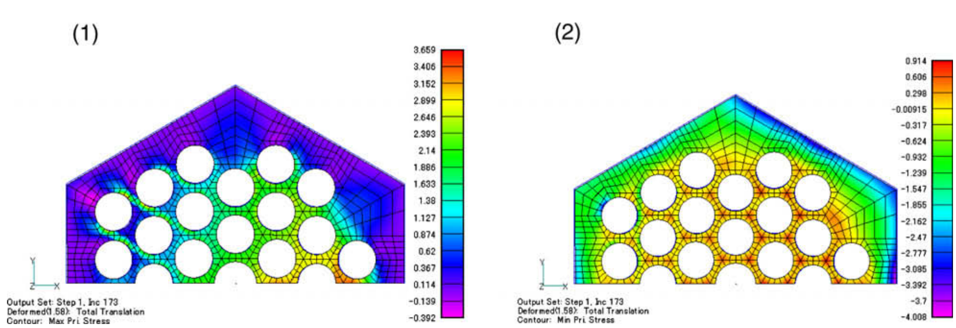

Fig. 5. Analytical result of residual stress on graphite block surface at the operation mode of reactor outlet temperature of 850 °C; (1) maximum principal stress, (2) minimum principal stress.

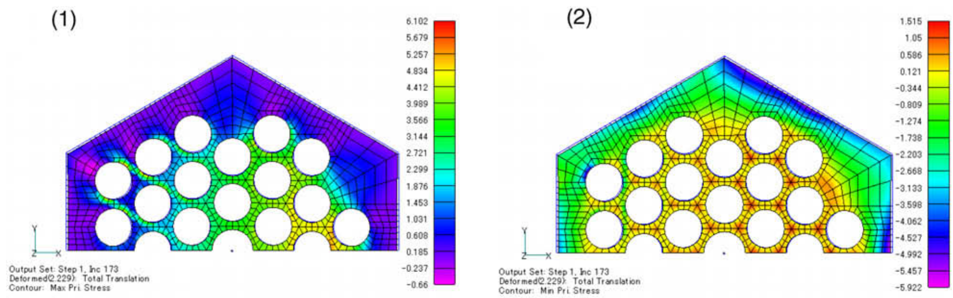

Fig. 6. Analytical result of residual stress on graphite block surface at the operation mode of reactor outlet temperature of 950 °C; (1) maximum principal stress, (2) minimum principal stress.

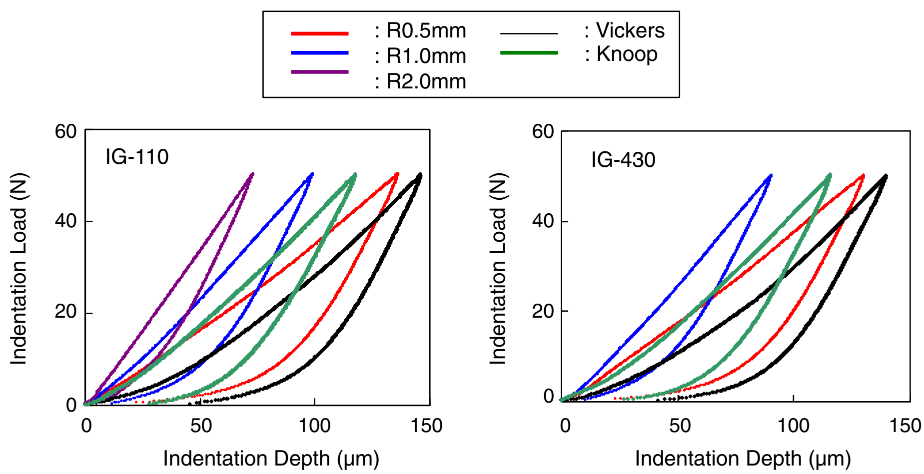

Fig. 7. Typical micro-indentation behavior for each indenter with maximum indentation load of 50 N.

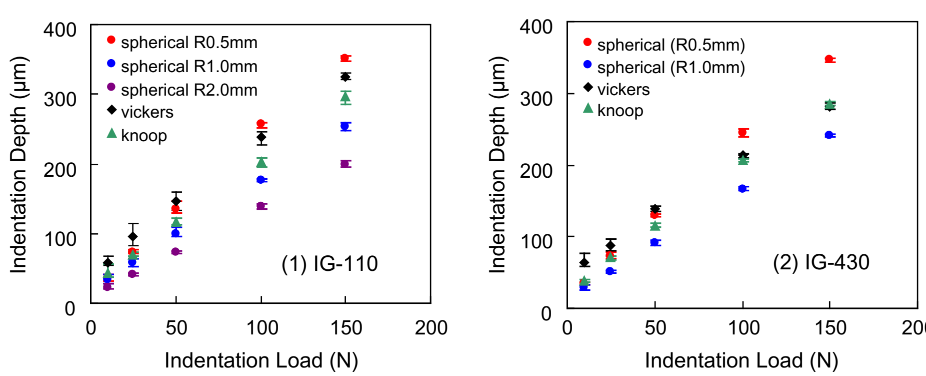

Fig. 8. Maximum indentation depth for each indenter in relation with maximum indentation load; (1) for IG-110, (2) for IG-430.

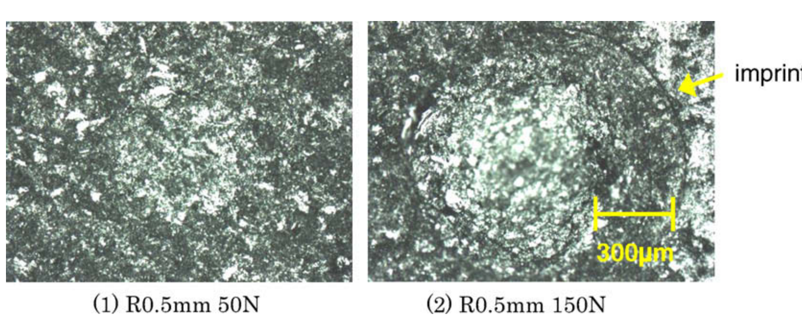

Fig. 9. Surface condition of IG-110 after micro-indentation test with maximum indentation load of (1) 50 and (2) 150 N.

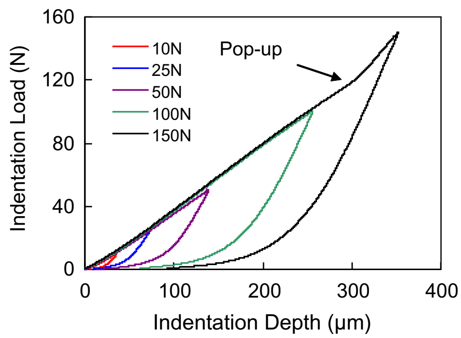

Fig. 10. Micro-indentation behavior for IG-110 for each maximum indentation load case.

sive stress condition of IG-110 was plotted in Fig. 11 for the maximum indentation load of 5 N (1) and 10 N (2). The compressive stress test was performed in the range of the specified minimum ultimate strength of IG-110, 61.4 MPa. The depth data is normalized by the average value at the data of non-stressed condition. The relationship between the normalized depth δ and compressive stress σ (MPa) is expressed by an empirical formula as:

$$5\text{N}: \quad \delta = -1 \times 10^{-6}\sigma^3 + 1 \times 10^{-4}\sigma^2 - 5.9 \times 10^{-3}\sigma + 1 \quad (0 \leqslant \sigma \leqslant 30)$$

$$10\text{N}: \quad \delta = -3 \times 10^{-7}\sigma^3 + 5 \times 10^{-5}\sigma^2 - 3.3 \times 10^{-3}\sigma + 1 \quad (0 \leqslant \sigma \leqslant 61.5) \tag{1}$$

Although we can see the variation of the data, the mean depth with the load of 5 N is decreased with increasing the compressive stress in Fig. 11 (1). From the result of Eq. (1), it seems to be possible to measure the compressive stress by the micro-indentation test. On the other hand, this trend is not followed with the load of 10 N in Fig. 11 (2). It is probable that the resistance in the graphite specimen against the indentation load is not large enough compared to the indentation load of 10 N. In this case, it would be difficult to detect the stress condition by micro-indentation characteristics. It is necessary to assess the variation of the test data with statistic methods to specify the stress condition. This will be investigated in a future study.

(2) Tensile stress condition

The indentation behavior to the tensile stress condition was also examined. The maximum indentation depth for tensile stress condition of IG-110 was plotted in Fig. 12 for the maximum load of 5 N (1) and 10 N (2). The tensile stress test was performed in the range of the specified minimum ultimate strength of IG-110, 19.4 MPa. The depth data is also normalized by the same manner as described above. For cases (1) and (2), it is difficult to see an obvious relationship between the depth and stress, unlike the obvious relationship seen in the compressive stress condition. It is probable that the resistance in the graphite specimen against the indentation load was reduced by the tensile stress, and it would reduce the sensitivity of the indentation behaviour on the specimen condition. In the tensile condition, it would be necessary to use the other indentation condition, e.g. other indenters and large indentation load. It is also necessary to use statistic method for the data evaluation. These are also subjects for future investigation.

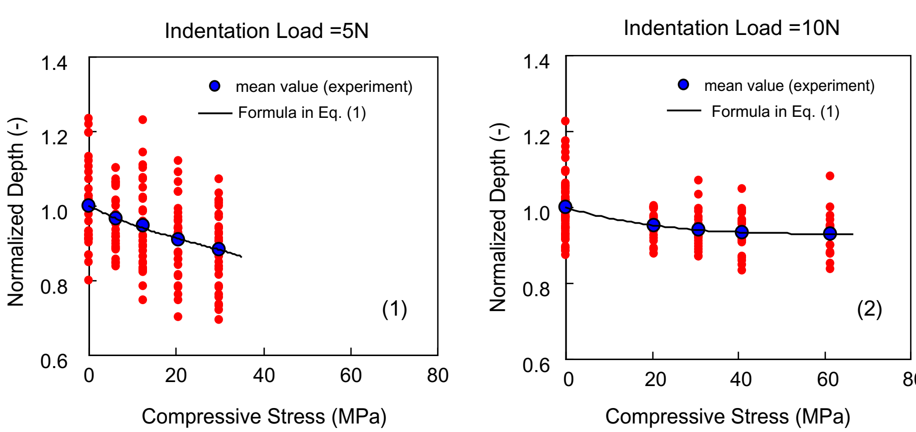

Fig. 11. Change of maximum indentation depth for IG-110 in relation with compressive stress with maximum indentation load of (1) 5 and (2) 10 N.

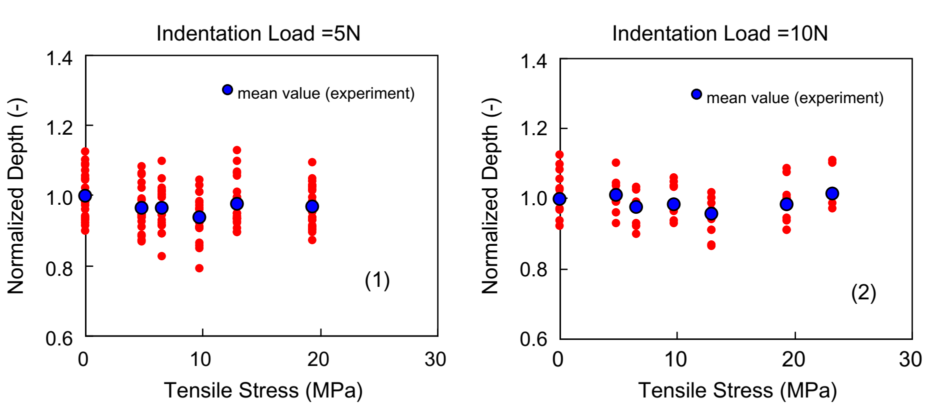

Fig. 12. Change of maximum indentation depth for IG-110 in relation with tensile stress with maximum indentation load of (1) 5 and (2) 10 N.

5. Conclusions

To assess the degradation of graphite components in the HTGRs by non-destructive methods, the applicability of the micro-indentation method or the residual stress evaluation was investigated. The fine-grained isotropic graphites IG-110 and IG-430, the candidates for the VHTR, were used. The following results were obtained.

(1) The analysis with VIENUS code in the graphite block at the HTTR condition showed that the residual stress caused by fast neutron fluence of 1.0×10^{25} n/m^2 is in the range of −6 (compressive) to +6 (tensile) MPa for the 950 °C operation. Although the tensile stress is lower than 1/3 Su value of IG-110, it is important to develop lifetime extension methods for the components in the VHTR, which would be used at much severer condition in the HTTR.
(2) Micro-indentation behavior at the stress free condition was investigated with different indenters. It was thought that the Vickers indenter and the spherical indenter R0.5 mm are sensitive enough to detect the specimen surface condition. The R0.5 mm indenter was selected to reduce the variation in the data, and indentation loads of 5 and 10 N were selected to avoid the pop-up effect in the loading process.
(3) The indentation behavior at the stressed condition was also examined. For the compressive condition, the relationship between the average value of normalized depth and compressive stress is expressed by an empirical formula. On the other hand, for the tensile condition, it is difficult to see the obvious relationship between depth and stress. It should be possible to evaluate the residual stress by the indentation behaviour. It is necessary to assess the variation of the test data with statistic methods and with other indentation conditions. This is the subject for future studies.

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