Effects of Neutron Irradiation on Mechanical Properties of Heat-Treated Zr-2.5Nb Pressure Tube in Prototype Advanced Thermal Reactor FUGEN

K.Ohara, Y.Abe, S.Ukai and M.H.Koike

O-arai Engineering Center,
Power Reactor and Nuclear Fuel Development Corporation (PNC)
4002, Narita, O-arai, Ibaraki, 311-13, Japan

ABSTRACT

The heat-treated Zr-2.5Nb is a pressure tube alloy utilized in prototype advanced thermal reactor Fugen. Fugen surveillance specimens as well as three kinds of modified HT Zr-2.5Nb, i.e. as received, 200 ppm hydrogen enriched and radially hydride reorientated specimens, were prepared and irradiated in the special fuel assemblies of Fugen up to neutron exposure of 7×10^{25} n/m^2 (E$>$ 1MeV) at around 560 K. A series of post irradiation examination including tensile test, bending fracture test, compact tension test and metallographies have been extensively conducted to investigate the effects of neutron irradiation on mechanical property changes of pressure tube.

Tensile strength of pressure tube increases within the initial stage of 1×10^{25} n/m^2, and thereafter takes saturation around 1000 MPa and 800 MPa at room temperature and at 573 K, respectively. The fracture toughness of the modified HT Zr-2.5Nb pressure tube is improved at both unirradiated and irradiated conditions. The fracture toughness is very sensitive to the orientation relationship between hydride platelets and crack extension axis. The radially reorientated hydride specimens degrades in fracture toughness, but satisfy the Fugen design value. Based on present results, the integrity of the Fugen pressure tube was appropriately demonstrated for 30 years operation.

1. INTRODUCTION

The prototype advanced thermal reactor, Fugen (power output 165 MWe), is a heavy-water-moderated, boiling-light-water-cooled, pressure-tube-type reactor, and has been commercially operating since March 1979. The pressure tubes used in the Fugen are made of heat-treated (HT) Zr-2.5Nb with high strength and low absorption rate of neutrons, which contain fuel assemblies and boiling light water coolant at about 550 K under an operating pressure of 7 MPa.

In order to characterize the material property changes of the pressure tubes under operation of the Fugen, surveillance testing has been continuously conducting using specimens cut from the actual pressure tubes. Those surveillance specimens were assembled inside of the special fuel assemblies from the initial stage of the operation. Up to the present, surveillance specimens had been taken out three times in 1984[1], 1989[2] and 1995 for post-irradiation examination.

PNC had also tried to improve the mechanical properties of pressure tubes by mean of modifying the heat-treatment at the final stage of Zr-2.5Nb tubes, which was formed at the domestic fabrication vendor from the billets purchased from the United State.

One of the main concerns in designing the pressure tube so as to prevent unstable fractures during operation involves the effects of the concentration and orientation of the zirconium hydrides on the fracture toughness of the pressure tube. Zirconium alloys pick up hydrogen under reactor operating condition, and circumferential hydride platelets have a tendency to form in the radial - axial plane perpendicular to the hoop stress direction. These concentrated and radial hydrides cause the pressure tube material to significantly decrease in fracture toughness[3][4]. To investigate such individual effects, Zr-2.5Nb pressure tubes modified with heat-treatment in PNC had been irradiated in special fuel assemblies of Fugen, including materials with enriched hydrogen content and reoriented hydrides in radial direction.

These specimens had been transported from Fugen site to the Materials Monitoring Facility in the O-arai Engineering Center, and post irradiation examination had been conducted. In this paper, results of tests concerning the Fugen pressure tube surveillance specimens as well as modified alloys containing enriched hydrogen and reoriented hydrides are described.

2. MATERIALS

Fugen pressure tube is in dimension of 118 mm inner diameter and 4.3 mm thickness. Its chemical composition and heat-treatment condition are shown in Table 1. HT Zr-2.5%Nb contains hydrogen of 18 ppm and about 1,000 ppm oxygen. They are water-quenched after a final solution treatment at a temperature of about 1160 K, cold-drawn in 5 to 15 %, and aged 24 h at 773 K. For the pressure tube surveillance test of Fugen, tensile specimens, bending specimens, burst specimens, corrosion specimens and hydrogen analysis specimens were cut and prepared from the actual Fugen pressure tubes.

The chemical composition and heat-treatment condition of the Zr-2.5Nb pressure tubes modified with heat-treatment is also shown in Table 1. The final solution treatment was taken to be slightly lower of 1143 K in order to improve the fracture toughness of pressure tube. The other specification of modified alloy is the same as that of Fugen pressure tube. This Zr-2.5Nb alloy was formed in pressure tube at the domestic fabrication vendor. To investigate the effect of hydrogen on the mechanical properties of modified pressure tube, hydrogen was charged about 200 ppm. Some of the pressure tube specimens were added the tensile stress of 150 MPa in hoop direction under thermal heat cycles of 543 K to RT at the 30 K/h to reorient the hydrides in radial direction of a thickness of the pressure tube. In the latter, the tubes were cut into half sections and flattened by using a continuous reverse bending technique. The flattened plates were then stress-relieved at 723 K for 15 h. **Figure 1** represents three types of hydride morphology of the as-received, hydrogen enriched of 200 ppm and hydrides reoriented specimens, which are optical micrographics at the transverse cross section of the pressure tube. The tensile and compact tension specimens were cut so as to arrange the tensile axis in parallel with the hoop direction of the pressure tube, which corresponds to the most severe condition from the viewpoint of the hydrides reorientation. The schematic diagram of the test specimens to be irradiated in the Fugen is represented in **Figure 2**, which includes tensile test specimens, bending and compact tension specimens for fracture toughness evaluation.

3. IRRADIATION

The surveillance examination of the Fugen pressure tubes is conducted in accordance with the stipulations as established by the Ministry of International Trade and Industry of Japan in the Ministerial Ordinance 62 and Notification 501 as well as in accordance with the Surveillance Examination Procedure of Nuclear Reactor Structural Material of JEAC 4201 of the Electrical Technical Regulation of the Japan Electrical Society. The type of surveillance specimens and the time when such surveillance specimens to be taken out are established with consideration of the characteristics of the Fugen advanced thermal reactor.

The surveillance test specimens as well as modified pressure tube specimens were contained in the capsules. The nine capsules containing such test specimens can be inserted into the fuel bundle center of the special fuel assembly. There are four special fuel assemblies loaded in the rotational symmetry at the intermediate portion of the Fugen reactor core, which is shown in **Figure 3**, and each special fuel assembly is removed from the core at two to four year intervals for a total of eight times during the reactor life.

The irradiation condition of the surveillance test specimens and modified pressure tube specimens, which were post-irradiation examined and presented in this paper, are summarized in **Table 2**. The surveillance test specimens had been irradiated from the initial to the 18 cycle of the Fugen, which corresponds to the 3,173 days. The neutron exposure level attains to 7.0×10^{25} n/m^2 (E>1MeV). As for the modified pressure tube specimens, irradiation had been started at the 11 cycle and continued up to the 20 cycle of Fugen, which results in 1,693 days operation and neutron exposure of 3.7×10^{25} n/m^2 (E>1MeV).

4. RESULTS AND DISCUSSION

4.1 Tensile Properties

The tensile tests are conducted in accordance with ASTM E-8-79a for RT and ASTM E21 for 573 K, respectively. The strain rate was set to be 8×10^{-5}/s up to 0.2 % yield point, and changed to 8×10^{-4}/s until occurrence of rupture.

Results of tensile test conducted at room temperature and 573 K are shown in **Figure 4**. These tests were carried out at the tensile stress in parallel with the hoop direction for the specimens cut from both Fugen surveillance and modified HT Zr-2.5Nb pressure tubes. As compared with un-irradiated specimens, tensile strength increases with neutron fluence and saturates at certain level within the neutron fluence of about 1×10^{25} n/m^2 (E>1MeV), which can be caused by the irradiation hardening due to the accumulation of point defects, i.e. frank loop and vacancy cluster formation. All of these results of irradiated pressure tubes demonstrate that one thirds of minimum tensile strength at both RT and 573 K clearly satisfy the Fugen design value of Sm.

HT Zr-2.5Nb modified heat-treatment exhibits similar tensile strength to that of Fugen actual surveillance specimens at both RT and 573 K. From the comparison of as-received specimens with 200 ppm hydrogen enriched specimens, both specimens show almost same strength level. Those results suggest that hydrides orientation in parallel with circumferential direction of pressure tube, same as stress axis, does not degrade the tensile strength. On the other hand, radially reorientated hydrides specimens have a tendency to be slightly lower strength, which cab be induced by morphology change of hydride platelets perpendicular to the stress axis.

Figure 5 shows the rupture elongation of tensile tests for the as-received, 200 ppm hydrogen enriched and hydride reorientated specimens at room temperature. The rupture elongation decreases with neutron fluence, corresponding to the irradiation hardening shown in **Figure 4**. As for the radially reorientated hydride specimens, there is slightly lower rupture elongation compared with as-received and hydrogen enriched specimens.

4.2 Fracture Toughness

Fracture toughness values were obtained by means of the bending fracture tests for surveillance test specimens as well as compact tension tests for modified HT Zr-2.5Nb specimens. The bending fracture test specimen, shown in **Figure 2**, is set with both ends on the jig inclined at 14.5 degree from the load axis to avoid out-of-plane bending at the notch area. Prior to performing the bending fracture test, the specimen is pre-cracked by fatigue. The conditions for pre-cracking and loading rate are according to ASTM Test Method for Plane-Strain Fracture Toughness of Metallic Materials (E 399-90). The equation by Brown [5] for the three-point bending of flat plate having a crack on one side is used to calculate the stress intensity factor.K, for the bending specimens.

The compact tension tests are conducted according to the ASTM E 399 and JSME S001. As shown in **Figure 2**, pre-cracking in the compact tension specimen is introduced by fatigue with frequency of 10 Hz up to 1 mm depth by loading of 3.5 to 60 Kg and subsequently 0.5 mm depth by loading of 4.5 Kg to 40 Kg. The compact tension test was performed at constant loading rate of 200 Kg/min, and corresponding strain was monitored with the clip gages. For the compact tension specimen, the following equation was used to calculate the stress intensity factor:

$$K = \{P/(B \times W^{1/2})\} \times f$$

$$f = (2+\xi)\times(0.886+4.64\xi-13.32\xi^2+14.72\xi^3-5.6\xi^4)/(1-\xi)^{3/2}$$

where
P = applied load
B = thickness
W = width
$\xi = a/W$
a = initial crack length

Fracture toughness Kc was calculated from the maximum applied load by substituting Pmax for P in above equation.

Fracture toughness values Kc obtained from the bending fracture test and compact tension test are shown in **Figure 6** at room temperature for as-received, hydrogen enriched and hydride reorientated specimens. The fracture toughness values of as-received specimens show the opposite tendency to tensile strength as shown in **Figure 4**, and decrease with neutron fluence, which is in agreement with CANDU reactors data for cold-worked Zr-2.5Nb pressure tubes[6]. Because the hydrogen pickup is very small during the initial period of irradiation, the initial decrease of fracture toughness is considered to be caused by accumulation of defect clusters due to neutron irradiation. Fracture toughness at 573 K were also obtained, and it was demonstrated that fracture toughness values at operating temperature is higher than at RT.

It is to be noticed from **Figure 6** that fracture toughness of as-received specimens is improved in the modified HT Zr-2.5Nb at both unirradiated and irradiated conditions, compared with Fugen surveillance specimens. Fracture toughness decreases in 200 ppm hydrogen enriched specimens, although the data are scattered. In the hydride reorientated specimens, fracture toughness degrades even at the unirradiated condition. These results suggest that fracture toughness is very sensitive to the orientation relationship between hydride platelets and crack extension axis, as shown in **Figure 1**. The hydride reorientated specimens showed low fracture toughness values, which is caused by the excessive amount of hydrogen of 200 ppm. Considering that the actual pressure tube utilized in Fugen 30 years operation could contain adequately low hydrogen content as compared with 200 ppm, it can be said that fracture toughness obtained was confirmed to satisfy the Fugen design value.

Using the fracture toughness obtained for the Fugen surveillance specimens at RT and 573 K, the critical crack length (CCL) was estimated based on the Newman's equation[7], which represents the correlation between stress and CCL for certain fracture toughness value. **Table 3** summarizes results of calculation, in which design stress of 179.5 MPa was utilized. The estimated minimum CCL are 32 mm at 573 K and 16 mm at RT, respectively. On the other hand, the postulated initial flaw with 5.0 mm length was estimated to extend to 6.30 mm at the Fugen life time of 30 years operation on the basis of design analyses. Therefore, it can be said that there still exists sufficiently allowable safety margin of 5 for unstable fracture at operation condition of Fugen pressure tube.

5. CONCLUSION

Based on the post-irradiation examination of Fugen surveillance specimens as well as modified HT Zr-2.5Nb pressure tube specimens, the following results were obtained:

1. The tensile strength increases initially and saturates with neutron fluence by the irradiation hardening. For the tensile stress in parallel with the hoop direction of the pressure tube, the radially reorientated hydride specimens have a tendency to be slightly lower strength and lower rupture elongation, which are induced by the hydride platelets orientation perpendicular to the stress axis. The irradiated pressure tube satisfies the Fugen design limit in tensile strength.

2. The fracture toughness of the modified HT Zr-2.5Nb pressure tube is improved at both unirradiated and irradiated conditions, as compared with Fugen surveillance specimens. The fracture toughness is very sensitive to the orientation relationship between hydride platelets and crack extension axis. The radially reorientated hydride specimens degrades in fracture toughness, but satisfy the Fugen design value. Based on present results, the integrity of the Fugen pressure tube was demonstrated for 30 years operation.

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Table 1 Specification of Fugen Pressure Tube and Modified Pressure tube

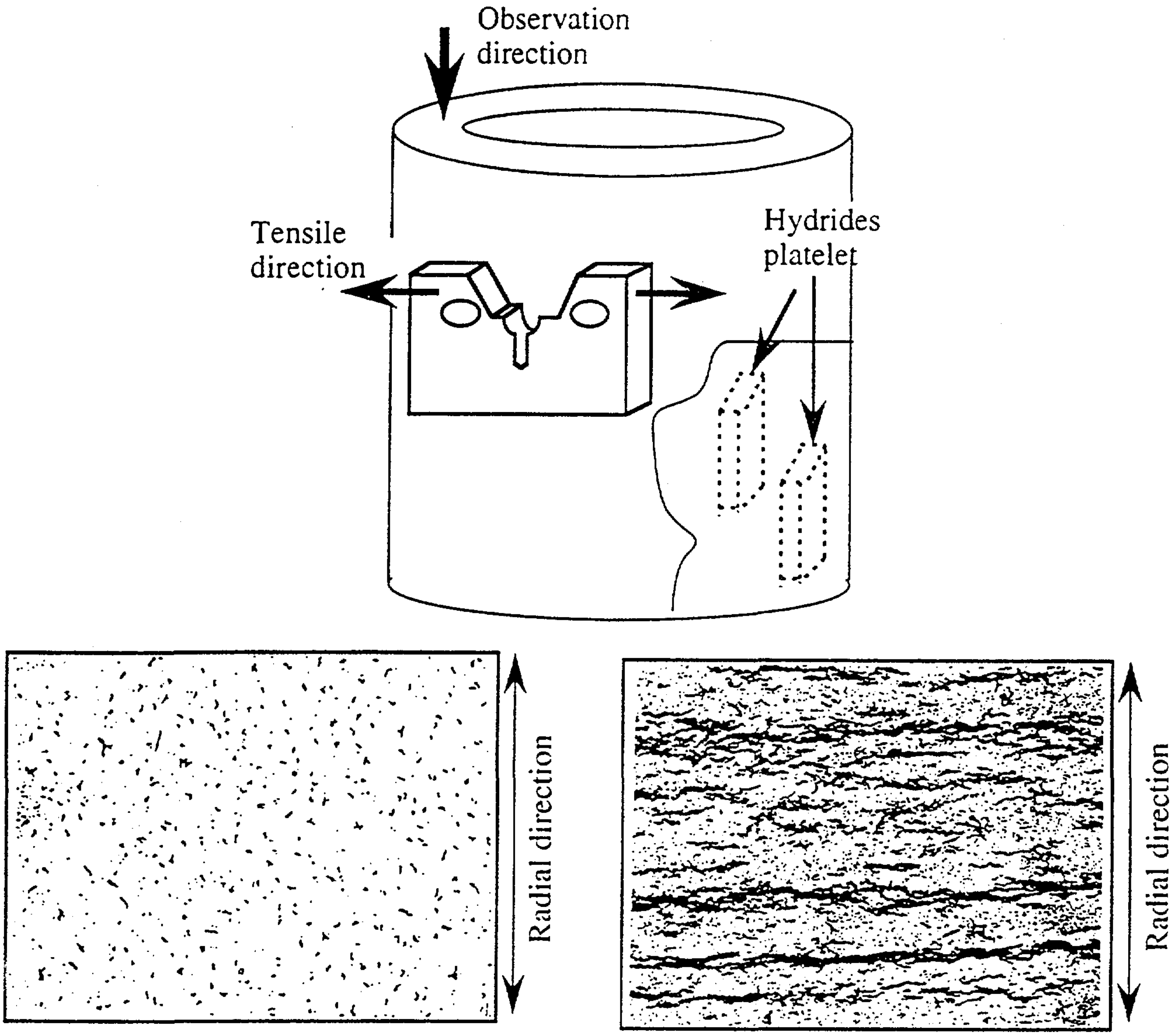
Item	Fugen Pressure Tube	Modified Pressure Tube
Chemical Composition	Nb; 2.40 - 2.80 wt% O ; 900 - 1300 ppm Zr ; ballance	Same
Annealing Quench	1160 K ± 15 K Water Quench below 323 K	1143 K ± 15 K Water Quench below 323 K
Cold Work	5 - 15 %	Same
Aging Treatment	773 K ± 15 K 24 h in Vacuum	Same

Table 2 Irradiation Condition of the Fugen Surveillance and Modified Pressure tube

	Fugen Surveillance	Modified Zr-2.5Nb
Fugen Cycle	1 - 18 cycle	11 - 20 cycle
Duration	3173 days	1693 days
Neutron Fluence	$7.0\text{x}10^{25}$ (n/m^2, E>1MeV)	$3.7\text{x}10^{25}$ (n/m^2, E>1MeV)
Irradiation Temperature	560 K	560 K

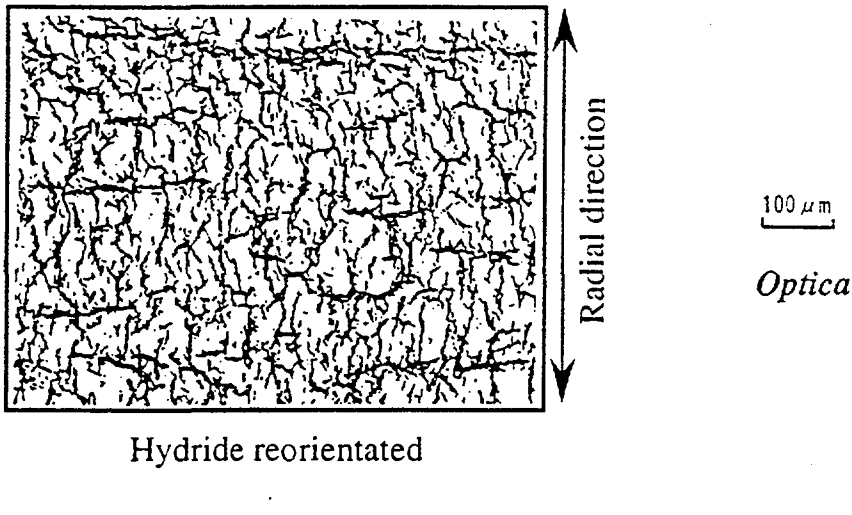
Table 3 Estimation of Critical Crack Length (CCL) and Safety Margin for Unstable Fracture

	Fracture Toughness (MPa$\sqrt{m}$)	Critical Crack Length (mm)	Safety Margin	Fugen Design
at RT				
Minimum	37.5	16	2.5	Initial Flaw Length 5.0 mm
Average	43.5	20	3.1	↓
at 573 K				
Minimum	65.7	32	5.0	6.3 mm after 30 Years Operation
Average	67.6	33	5.2	



As-received

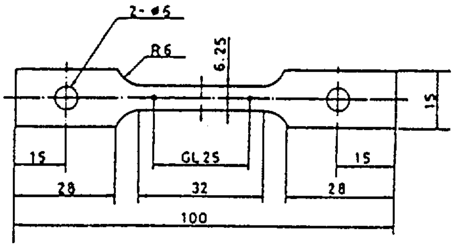
200 ppm Hydrogen enriched



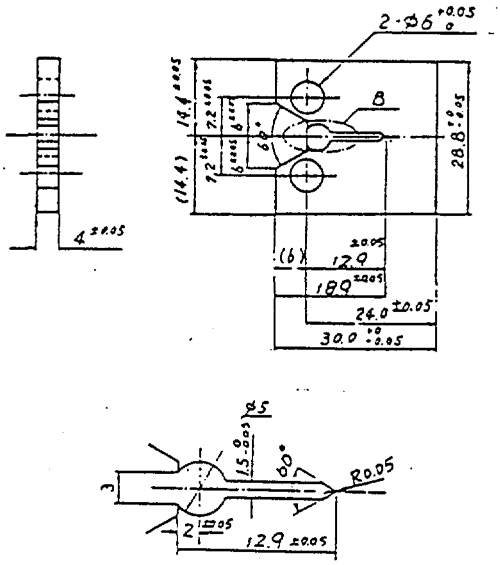
Optical micrographies

Hydride reorientated

Figure 1 Geometrical Relationship between Hydrides Orientation and Tensile and Compact tension Tests

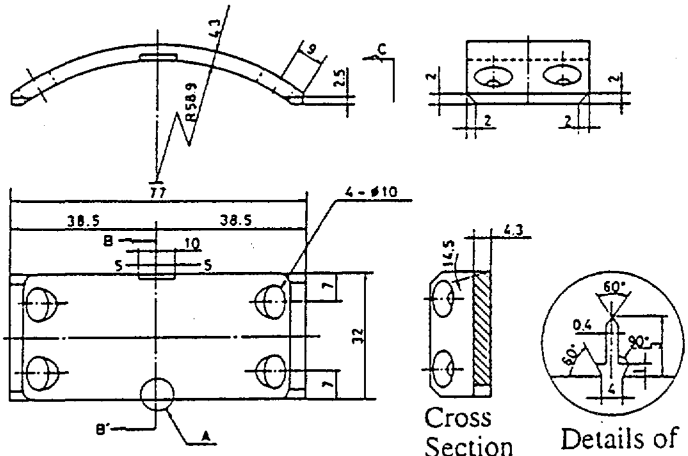


Tesile Test Specimen



Detail of B Part

Compact Tension Test Specimen

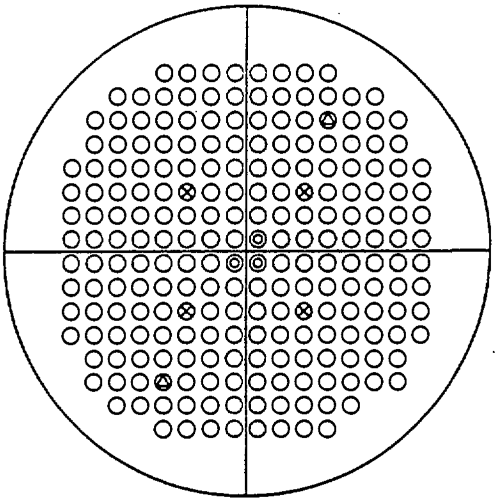


Cross Section of B-B'

Details of A Part

Bending Specimen

Figure 2 Test Specimens (unit:mm)



Symbol	Description	Quantity
○	Standard Fuel Assembly	215
⊗	Special Fuel Assembly	4
◎	36-rod Fuel Assembly	3
◬	R&D Fuel Assembly	2

Figure 3 Special Fuel Loading Position in the Fugen Core (1989)

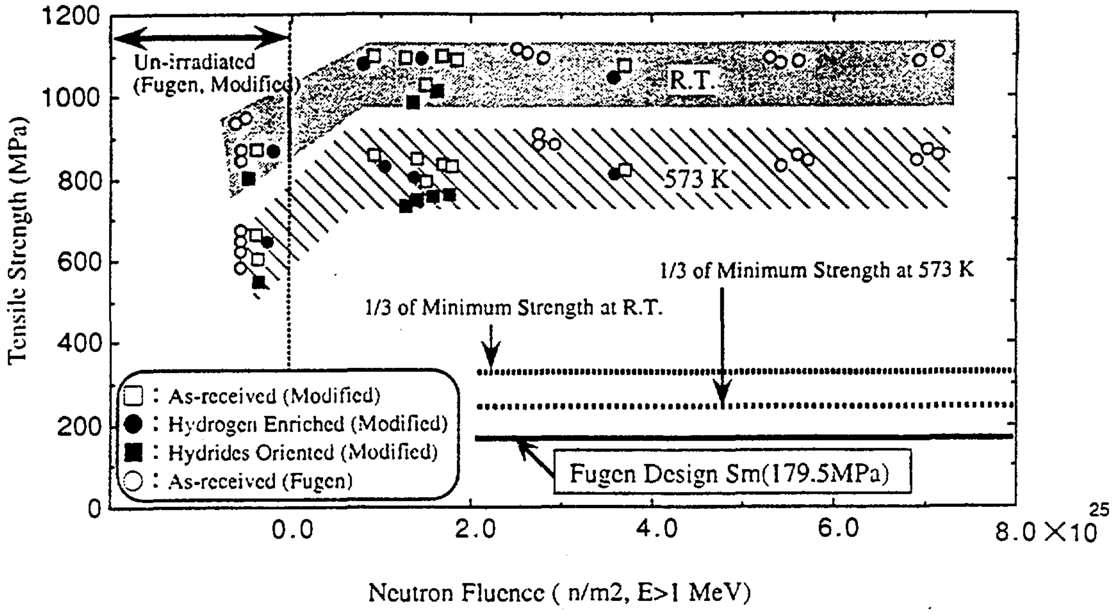


Figure 4 Tensile Strength at Room Temperature and 573 K as a Function of Neutron Fluence

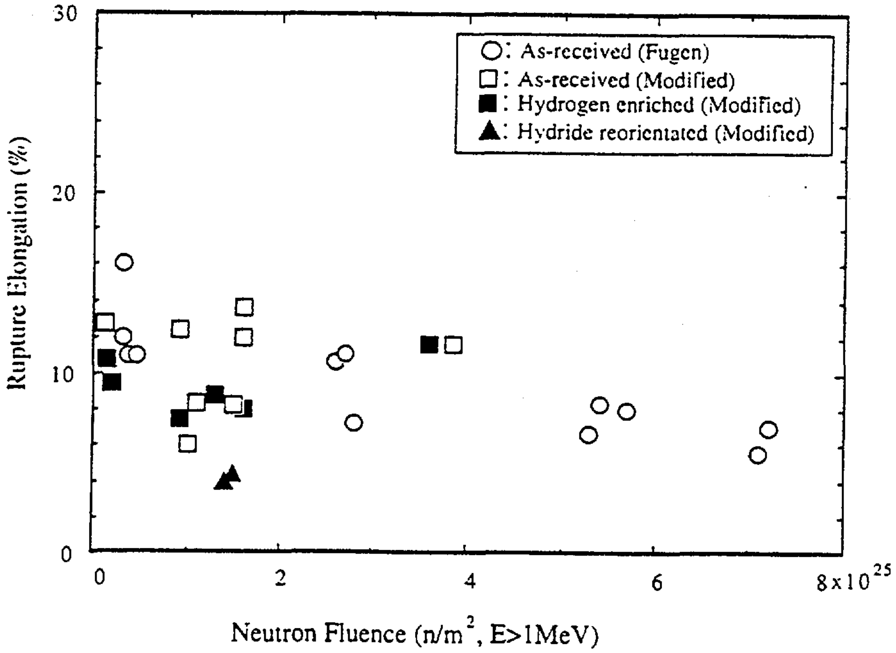


Figure 5 Tensile Rupture Elongation at Room Temperature as a Function of Neutron Fluence

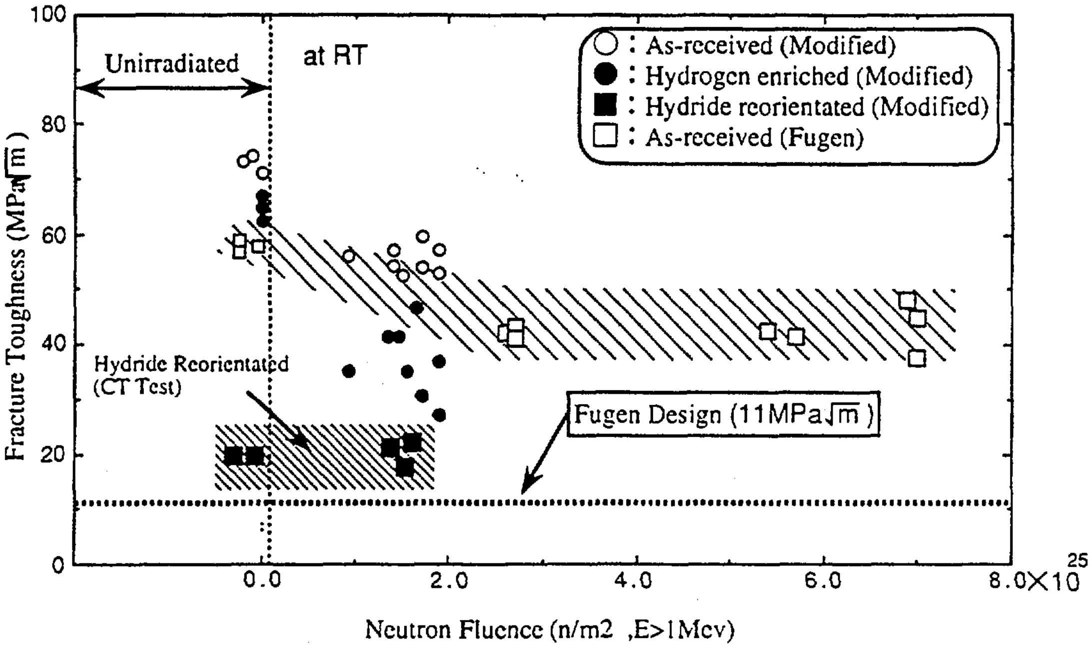


Figure 6 FractureToughness at Room Temperature
as a Function of Neutron Fluence