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# Development of a non-destructive testing technique using ultrasonic wave for evaluation of irradiation embrittlement in nuclear materials

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### Abstract

To develop a non-destructive testing technique for evaluating embrittlement of irradiated materials, the correlation between ultrasonic characteristics and embrittlement was investigated from the results of the ultrasonic wave measurement and the Charpy impact test of irradiated specimens of commercial A533B-1 steel and welded material at the hot laboratory of the Japan Materials Testing Reactor (JMTR) in the Japan Atomic Energy Research Institute (JAERI). After irradiation at 523 or 563 K up to a fast neutron fluence of  $1 \times 10^{24}$  N/m<sup>2</sup> (E > 1 MeV), velocities of both shear and longitudinal waves in the irradiated specimen were lower than those in the unirradiated one. The decrease in the velocities may be caused by the reductions of the shear and Young's moduli in the irradiated specimen. The attenuation coefficient of the longitudinal wave in the irradiated specimens increased compared with unirradiated ones. With increasing the shift amount of the Charpy transition temperature at 41 J absorbed energy, the velocity and attenuation coefficient of the ultrasonic waves decreased and increased, respectively.

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# 1. Introduction

Since the non-destructive testing (NDT) technique is very essential to monitor the degradation of first wall materials in the nuclear fusion reactor and reactor pressure vessel (RPV) materials in the nuclear fission reactor under neutron irradiation during their opera-

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tion, the development of this technique is needed to maintain the integrity of materials used for structures and components such as the first wall and RPV. In particular, the surveillance testing [1], which is for monitoring the aged degradation and irradiation embrittlement of the RPV materials, is very important to predict the life of the nuclear power plant. However, the number of specimens for the surveillance testing will decrease if the plant life is extended [2], since the specimens applied to the surveillance testing cannot be reused. Therefore, the study on the non-destructive evaluation of the irradiation damage is carried out by T. Ishii et al. / Journal of Nuclear Materials 307-311 (2002) 240-244

thermoelectric power [7] as a substitution for the surveillance testing. The NDT technique to evaluate the irradiation embrittlement is suitable for the reuse of the surveillance testing specimens. On the other hand, some of these techniques can be applied directly to structures and components in the fusion reactor to detect their degradation caused by neutron irradiation. Furthermore, since the small specimen test technique is also essential for the recent study on fusion reactor materials [8], these NDT techniques will be suitable for applying to irradiated small specimens for evaluating the effect of neutron irradiation on the mechanical properties of these materials.

The present study was conducted to develop the NDT technique using an ultrasonic wave for characterizing the irradiation embrittlement of nuclear materials. The velocity and attenuation coefficient of the ultrasonic wave in the irradiated A533B-1 steel and welded material for the Charpy impact test were investigated on the basis of data obtained by the remote manipulation in the hot laboratory of the Japan Materials Testing Reactor (JMTR) of the Japan Atomic

Table 1

Chemical compositions of materials used in this test (wt%)

Energy Research Institute (JAERI). The changes in the velocity and attenuation coefficient of the ultrasonic waves propagating in the materials were observed after irradiation. The correlation between irradiation damage and ultrasonic characteristics of the irradiated materials was also discussed.

## 2. Experimental procedure

Chemical compositions of materials used in this test are given in Table 1. The commercial A533B-1 (ASTM A 533 Grade B Class 1) steel is a material for the RPV. The low P A533B-1 steel is a material melted in the laboratory to reduce the phosphorus content. The welded material has a chemical composition of the weld metal for the submerged arc welding. The configuration and dimensions of the Charpy impact specimen used in this study are in accordance with Japanese Industrial Standard (JIS) Z2202. The specimens were irradiated in the JMTR at about 523 or 563 K up to a fast neutron fluence of  $1 \times 10^{24}$  N/m<sup>2</sup> (E > 1 MeV). In order to prepare the specimens with the different embrittlement characteristics, some of the irradiated specimens were

Chemical compositions of materials used in this test (w(7))										
	С	Si	Mn	Р	S	Ni	Cr	Mo	Cu	
Commercial A533B-1 steel	0.220	0.31	1.36	0.010	0.012	0.58	0.130	0.52	0.140	_
Low P A533B-1 steel	0.210	0.28	1.36	0.003	0.010	0.60	0.110	0.51	0.160	
Welded material	0.076	0.23	1.46	0.016	0.010	0.66	0.037	0.47	0.089	



Fig. 1. Schematic diagram of experimental apparatus for ultrasonic wave measurement.

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annealed at 673 K for 5 min. Charpy impact tests of these specimens were performed complying with the specification of ASTM A 370 in the hot laboratory.

A schematic drawing of an experimental apparatus for the ultrasonic wave measurement is shown in Fig. 1. A clamping device with a probe was installed in the hot cell, and an ultrasonic testing instrument and a data processing unit were set up at an operation area of the hot laboratory. An ultrasonic probe fixed to the clamping device was contacted with the surface of the specimen by the remote manipulation to measure the propagation time and the pulse amplitude of the ultrasonic wave. Machine oil or glycerin was used as a coupling medium to propagate the ultrasonic wave from the probe to the specimen smoothly. A transducer of the probe was 6.3 mm in diameter. Five megahertz shear wave, and 10 and 15 MHz longitudinal waves were used in this experiment. The propagation time between the first and the second back wall echoes, and the pulse amplitude of these echoes were estimated from the pulse echo displayed on a CRT. The thickness of specimens was measured with accuracy of  $\pm 1 \ \mu m$  by a micrometer. The velocity and attenuation coefficient of both shear and longitudinal waves propagating in the specimen were calculated by the following equations [9,10]:

$$C = \frac{2T}{t_2 - t_1},$$
 (1)

$$\alpha = \frac{20\log_{10}(h_1/h_2)}{2T},\tag{2}$$

where *C* and  $\alpha$  are the velocity and the attenuation coefficient of the ultrasonic wave, respectively. *T* is the thickness of the Charpy impact specimen. *t* is the propagation time of the back wall echo. *h* is the pulse amplitude of the back wall one. The subscripts 1 and 2 denote the first and the second back wall echoes, respectively.

## 3. Results and discussion

Changes in velocities of the shear and longitudinal waves propagating in specimens that were unirradiated, irradiated, and annealed after irradiation are shown in Fig. 2. Velocities of both shear and longitudinal waves in the irradiated specimen were lower than that in the unirradiated one. The velocity of both waves in the irradiated specimens had a tendency to increase after annealing at 673 K for 5 min.

In general, the velocity of the shear wave,  $C_S$ , and the velocity of the longitudinal wave,  $C_D$ , are expressed by Eqs. (3) and (4) [9,10].

$$C_{\rm S} = \sqrt{\frac{\mu}{\rho}} = \sqrt{\frac{E}{\rho} \frac{1}{2(1+\sigma)}},\tag{3}$$



Fig. 2. Changes in velocities of shear and longitudinal waves.

$$C_{\rm D} = \sqrt{\frac{K + (4/3)\mu}{\rho}} = \sqrt{\frac{E}{\rho} \frac{(1-\sigma)}{(1+\sigma)(1-2\sigma)}},\tag{4}$$

where  $\mu$ , *E* and *K* are the shear, Young's and bulk moduli, respectively.  $\rho$  and  $\sigma$  are the density and Poisson's ratio, respectively. The density of the specimen made of commercial A533B-1 steel before and after irradiation was 2827.5 and 2821 kg/m<sup>3</sup>, respectively. The rate of the change in shear and Young's moduli calculated on the basis of Eqs. (3) and (4) is plotted against the change in the velocity of the shear wave in Fig. 3. The rates of the change in shear and Young's moduli of the irradiated specimen whose rate of the change in the density is -0.23% of the unirradiated specimen's density are -1.08and -0.98% of those of the unirradiated one, respectively. Therefore, it seems that the decreases in the velocity of both shear and longitudinal waves are related



Fig. 3. Rate of change in shear and Young's moduli as a function of change in velocity of shear wave.



Fig. 4. Attenuation coefficient of longitudinal wave propagating in both unirradiated and irradiated specimens of different materials.

to the reductions of the shear and Young's moduli in the irradiated specimen.

The attenuation coefficient of the longitudinal wave propagating in both unirradiated and irradiated specimens of different kinds of materials is shown in Fig. 4. The attenuation coefficient of the longitudinal wave in the irradiated specimens increased compared with unirradiated ones. In particular, the attenuation coefficient in the specimen irradiated at 523 K was larger than that in the specimen irradiated at 563 K. It may be related by the generation and aggregation of internal defects such as void and vacancy under neutron irradiation [9–11], that the pulse amplitude of the ultrasonic wave in irradiated materials becomes lower compared with that in the unirradiated ones.

The correlation between the velocity of 5 MHz shear wave and the shift amount of the Charpy transition temperature at 41 J absorbed energy is shown in Fig. 5. In all the material used in this study, the velocity decreases with increasing the shift amount of the transition temperature. Compared with the unirradiated specimen,



Fig. 5. Correlation between velocity of 5 MHz shear wave and shift amount of Charpy transition temperature at 41 J absorbed energy.



Fig. 6. Correlation between attenuation coefficient of 15 MHz longitudinal wave and shift amount of Charpy transition temperature at 41 J absorbed energy.

the decrease in the velocity was 23 m/s in the irradiated specimen of welded material whose shift amount of the transition temperature caused by neutron irradiation was 260 K. The amount of 23 m/s is corresponding to 0.7% of the velocity measured in the unirradiated specimen of the welded material. It was found that the velocity of 5 MHz shear wave depends on the degree of embrittlement in the materials used in this study.

The correlation between the attenuation coefficient of 15 MHz longitudinal wave and the shift amount of the Charpy transition temperature at 41 J absorbed energy is shown in Fig. 6. The attenuation coefficient increases with increasing the shift amount of the transition temperature. Compared with the unirradiated specimen, the increase in the attenuation coefficient was 0.05 dB/mm in the irradiated specimen of the commercial A533B-1 steel whose shift amount of the transition temperature caused by neutron irradiation at 523 K was 300 K. The amount of 0.05 dB/mm is corresponding to 50% of the attenuation coefficient measured in the unirradiated specimen of the commercial A533B-1 steel.

It was found that the NDT technique using the ultrasonic wave is applicable and indispensable for characterizing the neutron irradiation embrittlement of materials for the structures and components in the nuclear fusion and fission reactors.

### 4. Conclusions

Development of the NDT technique using the ultrasonic wave for characterizing the irradiation embrittlement of nuclear materials on the basis of the correlation between the shift amount of the Charpy transition temperature and the ultrasonic characteristics in the materials with different embrittlement characteristics was conducted to use this technique as a new method for evaluating the irradiation embrittlement of the RPV in the fission reactor and the first wall in the fusion reactor. The velocity and the attenuation coefficient of ultrasonic waves propagating in the Charpy impact specimens that were irradiated at 523 or 563 K up to a fast neutron fluence of  $1 \times 10^{24}$  N/m<sup>2</sup> (E > 1 MeV) were evaluated on the basis of experimental data obtained by post irradiation examinations in the hot laboratory. The main results obtained were as follows:

- Velocities of both shear and longitudinal waves in the irradiated specimen were lower than that in the unirradiated one.
- (2) The tendency that the attenuation coefficient of the longitudinal wave in the irradiated specimens increased compared with unirradiated ones in all the materials used in this study.
- (3) With increasing the shift amount of the Charpy transition temperature at 41 J absorbed energy, the velocity and attenuation coefficient of the ultrasonic wave decreased and increased, respectively.

The good correlation between the ultrasonic characteristic and the shift amount for evaluating the embrittlement of materials was observed in the experimental results for a preliminary examination. This NDT technique for evaluating irradiation embrittlement of materials quantitatively will be available for the practical fields by obtaining more data regarding the correlation between the ultrasonic characteristics and embrittlement ones for specimens, which have different irradiation embrittlement characteristics under the various kinds of irradiation test conditions, and by developing the technique for measuring propagation time and pulse amplitude of the ultrasonic wave with higher accuracy and resolution.

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# References

- Japanese Electric Association, Fracture Toughness Test Methods for Nuclear Power Plant Components, JEAC 4206-1991 (in Japanese).
- [2] Y. Nishiyama, K. Fukaya, K. Onizawa, M. Suzuki, T. Nakamura, S. Kaihara, A. Sato, K. Yoshida, ASTM STP-1329 (1998) 484.
- [3] H. Yoneyama, N. Ooka, Y. Futamura, T. Hirano, K. Yoshida, H. Kobayashi, ASME PVP-228 (1992) 57.
- [4] B.S. Viswanathan, D. Pachur, R.V. Nandedkar, ASTM STP-956 (1987) 369.
- [5] K. Ara, N. Ebine, N. Nakajima, J. Press. Vess. Technol. 118 (1996) 447.
- [6] I.E. Ukpong, A.D. Krawitz, D.F.R. Mildner, H.P. Leightly Jr., ASTM STP-956 (1987) 480.
- [7] J.F. Coste, S. Jumel, R. Borrelly, in: Proceedings of the 7th International Conference on Nuclear Engineering, Tokyo, Japan, 19–23 April 1999, JSME, Tokyo, 1999.
- [8] T. Ishii, M. Ohmi, J. Saito, T. Hoshiya, N. Ooka, S. Jitsukawa, M. Eto, J. Nucl. Mater. 283–287 (2000) 440.
- [9] Japan Society for the Promotion of Science (GAKU-SHIN), Ultrasonic Material Testing, The Nikkan Kogyo Shinbun Ltd., Tokyo, 1984 (in Japanese).
- [10] J. Krautkramer, H. Krautkramer, Ultrasonic Testing of Materials, Springer-Verlag, Berlin, 1983.
- [11] S. Ishino, Irradiation Damage, Tokyo University, Tokyo, 1979 (in Japanese).