

ULTRASONIC CHARACTERISTIC ANALYSIS OF THE REACTOR PRESSURE VESSEL MATERIALS DEGRADATION DUE TO NEUTRON IRRADIATION

KeeOk Chang, SamLai Lee, Chulmoo Sim, ByoungChul Kim
Korea Atomic Energy Research Institute, 150 Dukjindong,
Yusonggu, Taejon, KOREA
kochang@kaeri.re.kr

Abstract

Ultrasonic signal analysis from specimens have been performed in order to understand the ultrasonic characteristics of reactor vessel materials by neutron irradiation. Ultrasonic test parameters including amplitude and attenuation were analyzed for different frequencies and they were correlated with the quantity of material degradation due to neutron fluence. The results showed that the peak amplitude difference in the frequency domain for the front and back signals from the specimen increased as the amount of neutron fluence increased in a base metal, while such a consistent relationship was not shown in the welded metal. A similar relationship was observed in the attenuation analysis of the frequency domain. These phenomena indicate that the ultrasonic parameters might have some correlation with the amount of neutron irradiation for a specimen and the possibility that a nondestructive evaluation method could be used to predict the effect due to the fluence of neutron irradiation in nuclear reactor vessel materials.

Introduction

The core beltline region in a reactor vessel is subjected to neutron irradiation at both high temperatures and pressure as the reactor starts operation. The neutrons produced from nuclear fuel have high levels of energy and cause collisions with the material lattice structure and thus the materials experience an increase in strength and hardness and a decrease in ductility and toughness through this collision. Thus, the life prediction of an operating reactor vessel can be obtained by correlating the test results from mechanical test specimens inserted in the reactor and the neutron fluence, and therefore, research for irradiation embrittlement has been actively performed since the embrittlement phenomena by neutrons directly affects the safe operation of a plant as well as the reactor's life span[1-3]. Up to now, research related to embrittlement by neutrons has been performed on specimens irradiated in a research reactor using a higher level of energy within a short period of time compared to a commercial nuclear power plant, and the effect due to embrittlement will be considerably different from the accumulated dose effect for such a long period of irradiation. Although a sufficient number of mechanical test specimens have to be available in order to accurately evaluate the

embrittlement effect related to reactor operation, a limited number of specimens have to be inserted into the reactor during construction due to space limitations and thus ample information is not available for maintaining the integrity and making decisions on life extensions. Thus, USNRC recommended the development of various nondestructive evaluation methods to compensate for the current integrity evaluation practice[4]. Not much research work using NDE methods has been performed for irradiated materials due to the contamination problem. This work covers material characterization by ultrasonics for reactor materials in the form of Charpy impact specimens made of Mn-Mo-Ni low alloy steel withdrawn from a reactor vessel during a reactor outage.

Theoretical Background

Reactor vessels in a nuclear power plant are irradiated by fast neutrons under high temperatures as well as a high pressure and thus both irradiation hardening and embrittlement effects occur at the same time due to severe irradiation damage. Although irradiation hardening itself may show a positive effect from a metallurgical point of view, the life of the vessel can be shortened by the accompanying decrease of ductility due to irradiation. The low alloy steel used for reactor vessels shows a ductile-brittle transition temperature in Charpy impact testing and this phenomenon occurs due to various defects in the material and especially, irradiation hardening occurs by clusters of impurities rather than a single impurity. Embrittlement by neutron irradiation is affected by various factors such as chemical composition, microstructure and neutron fluences, etc. In order to improve the strength and stiffness, various chemical compositions are added and some remain after manufacturing. One of the elements, Cu, deepens the irradiation embrittlement phenomenon[5]. Therefore, many researchers have tried to measure the degree of embrittlement from copper-rich-precipitates by various methods. More sophisticated and reliable methods are expected to be developed in order to assist such microscopic analyses. Apart from such analysis, the structural integrity of the reactor vessel can be evaluated nondestructively. One of the methods is ultrasonic testing widely used to detect minor cracks within the material, as well as measuring the mechanical properties. Since the ultrasonic beam

is a stress wave, evaluation of the material properties is done using the fact that velocity and energy vary when energy passes through the material and it also depend on the density, grain size, dislocation and precipitates which may be affected by neutron irradiation and thus it becomes possible to correlate the interrelationship between the attenuation, velocity and neutron fluence. Some research has been performed using ultrasonic characteristics such as Rayleigh wave criticality, electromagnetic acoustic effects, magnetostrictive effects and ultrasonic nonlinearity[6-8]. However, up to now, research for the evaluation of irradiation embrittlement using ultrasonics has not been sufficiently performed due to the limitation of specimen availability[9].

Experiment

The specimens used in the present study were obtained from the reactor pressure vessel surveillance program of a commercial reactor both the unirradiated condition and following irradiation to two different fluences ($2.2 \times 10^{19} \text{ n/cm}^2$, $3.8 \times 10^{19} \text{ n/cm}^2$, $E > 1.0 \text{ MeV}$, 288). These fluences correspond to 7 and 10 years of exposure at the surveillance capsule location in the reactor, respectively. In order to identify the change of ultrasonic characterization due to neutron irradiation, an ultrasonic pulse echo data acquisition system was built. A JSR ultrasonics board was used as a pulser and receiver and a Compuscope265 board from Gage Applied Science as an A/D board. An Aerotech transducer 5.0MHz with a diameter of 6mm was used. The dimension of the specimen was so small in depth that a delay line was attached in order to reduce the effect of the near zone. The ultrasonic signal was acquired under the same condition since the purpose of this experiment was to

Table 1. Chemical composition(wt%) and heat Treatment of specimens

Base	
C: 0.16 Si: 0.17 Mn: 1.28 P: 0.006 S: 0.005 Mo: 0.48 Ni:	
0.71 Cr: 0.17 Cu: 0.06 Cl:0.011 Co: 0.012 Fe: Bal	
871°C 4h, austenizing, water quenching	
663°C 4h, tempering, air cooling	
595°C 6h, stress relieving, air cooling	
Weld	
C: 0.06 Si: 0.37 Mn: 1.68 P: 0.007 S: 0.008 Mo: 0.55 Ni:	
0.58 Cr: 0.03 Cu: 0.046 Cl:0.004 Co: 0.011 Fe: Bal	
595°C 6h, post-weld stress relief	

measure ultrasonic attenuation by a direct contact method and thus a constant weight was applied upon the transducer in order to maintain a constant

contact pressure constantly, although it does not greatly affect the attenuation measurement. Since the irradiated specimen is highly contaminated by radioactive materials, measurement was performed using a lead shielded facility as shown in Fig. 1. The chemical composition and heat treatment conditions of the specimens are given in table 1.

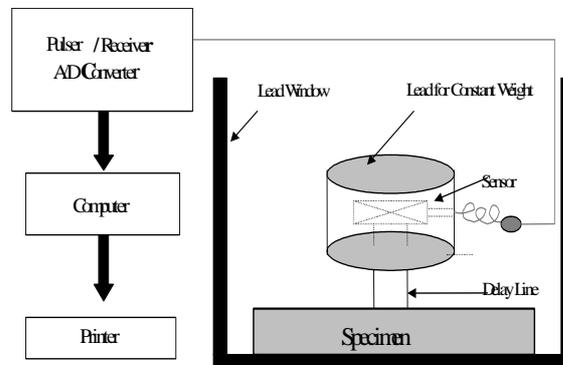


Figure 1. Experimental set-up for acquiring ultrasonic signals

Result and Discussion

Ultrasonic signal acquisition and analysis for the Charpy impact specimen was performed. Figure 2 shows a typical ultrasonic signal acquired from the specimen by a transducer. Signals (1) and (2) represent the front and back surface of the delay line and (3) and (4) are the signals from the front and back surface of the specimen.

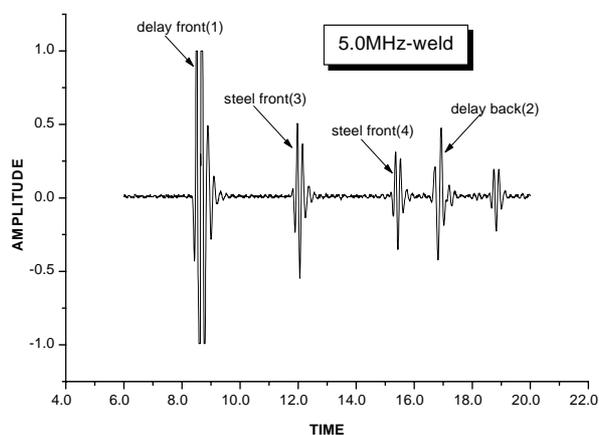


Figure 2. Typical ultrasonic signal acquired from the weld specimen

Each signal acquired from the back surface of the specimen was analyzed by the fast Fourier transform, the phase and velocity of the signal was calculated[10] and their changes were then observed depending on the specimens as well as the amount of neutron fluence. An ultrasonic evaluation was conducted for the signals acquired from the Charpy impact test specimens made of the same material representing

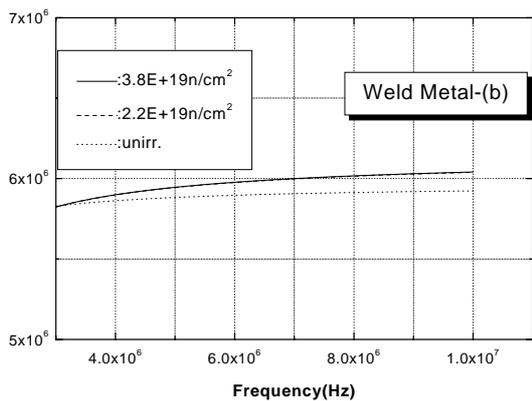
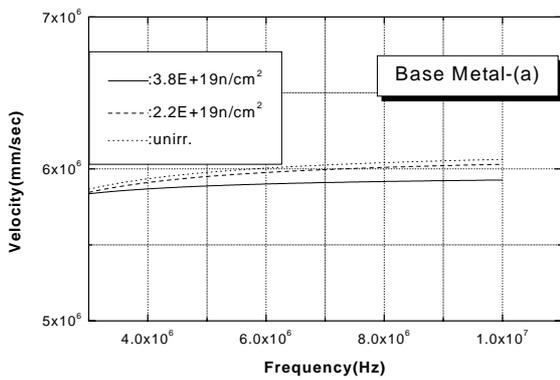


Figure 3. Relationship between ultrasonic velocity and frequency for base and weld metal

base and welded metal from the reactor vessel. The evaluation was performed for an irradiated specimen as well as an unirradiated specimen and the relationships between the phase velocity versus frequencies are shown in Fig. 3. One can notice that the velocity decreases while the fluence increases in the base metal as shown in Fig.3(a). Such results may be interpreted from the effect generated by a microstructural defect and it may be considered to be consistent with previous work where ultrasonic propagation could be deterred by the generation of various defects such as point defects and defect clusters, etc., within the material due to various environmental reasons[11]. The velocity difference was not noticeable between the 2nd and 3rd extracted specimens since the difference in the amount of each fluence was not so great compared to the result that was appreciably seen in the unirradiated specimen. However, an inverse phenomenon was observed in weld metal shown in Fig.3(b) compared to the base metal, which was very strange from a metallurgical point of view. Let us presume that certain factors played an important role in this experiment. Such a phenomenon may be understood

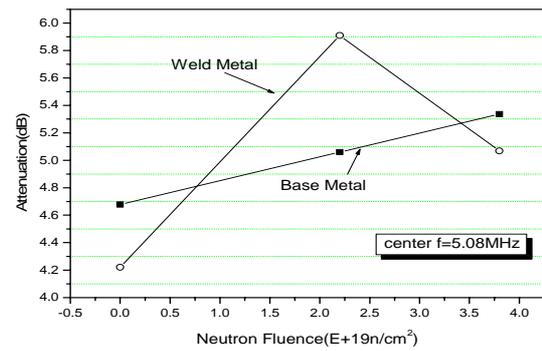


Figure 4. Relationship between attenuation and neutron fluence for base and weld metal

as having occurred due to the compositional characteristics of the welded specimen. Figure 4 shows the relationship between the attenuation and neutron fluence for the base and welded metal. The attenuation change due to irradiation is similar to the velocity effect of figure 3. And also an inverse phenomenon was observed in the welded metal compared to the base metal. Such a discrepancy was verified by the test parameters including amplitude and attenuation which were analyzed for the different frequencies and were correlated with the quantity of material degradation due to neutron fluence.

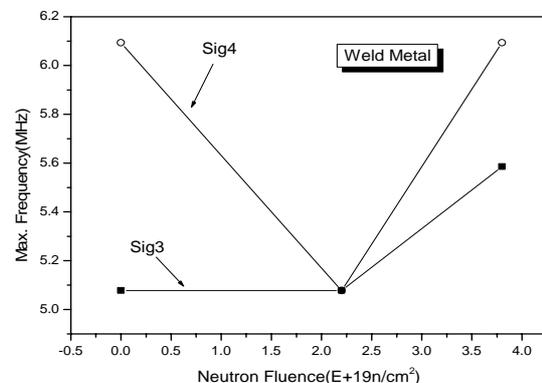
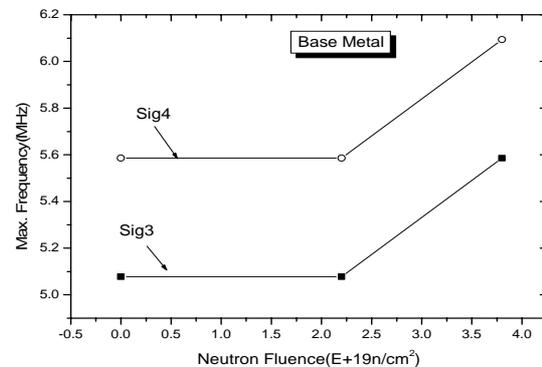


Figure 5. Relationship between different frequencies and neutron fluence

Figure 5 shows that the peak frequency in the frequency domain for the front and back signals from the specimen increased as the amount of neutron fluence increased in the base metal, while such a consistent relationship was not shown in the welded metal. A similar relationship has been observed in the attenuation analysis of the frequency domain. These phenomena indicate that the ultrasonic parameters might have some correlations with the amount of neutron irradiation for a specimen and the possibility that a nondestructive evaluation method could be used to predict the effect due to the fluence of neutron irradiation in nuclear reactor vessel materials. Looking at these results, one may presume that the metallurgical complexities of the welded metal must have had an affect on the ultrasonic measurement and the effects showed similar results while measuring the ultrasonic velocity.

Conclusions

Ultrasonic evaluation for Charpy impact test specimens, in order to evaluate the integrity of the reactor vessel, were performed and the results are as follows:

The ultrasonic peak frequency in the frequency domain increases as the fluence increases in the base metal, while such a consistent relationship was not shown in the welded metal. Such a discrepancy was also verified by the test parameters including velocity and attenuation which were analyzed for the different frequencies and were correlated with the quantity of material degradation due to neutron fluence. This indicates that the metallurgical complexities of the welded metal must have had an affect on the ultrasonic measurement and the effects showed similar results while analyzing the ultrasonic attenuation. Therefore, a parametric analysis by ultrasonics may be useful for identifying the material state related to neutron irradiation embrittlement and can be considered useful for supplementing the current technology for the evaluation and prediction of a reactor vessel's integrity if further research to identify such an inconsistent phenomenon can be performed.

References

- [1] T. J. Griesbach and W. L. Server " Management of Neutron Radiation Embrittlement: A view from the United States," in *Radiation Embrittlement of Nuclear Reactor Pressure Vessel Steels: An International review(Fourth Volume)* -1990, ASTM STP 1170, Philadelphia, 1993, Griesbach, pp.30-39.
- [2] W. M. de Villier and de H. J. Klerk, *Int. J. Pressure Vessel & Piping* **50**, 1992, pp.215-230.
- [3] K. Popp and U. Bergmann, et. al., "Irradiation and Annealing Behavior of 15Kh2MFA Reactor pressure Vessel Steel," in *Radiation Embrittlement of Nuclear Reactor Pressure Vessel Steels: An International review(Fourth Volume)*-1990, ASTM STP 1170, Philadelphia, 1993, pp.344-368.
- [4] S. Jackson "The direction and trend in US nuclear regulatory policy," in *The first annual KAIF/KNS Meeting*, Seoul, 1996.
- [5] A. Astafev, et. al., *Atomnaya Energiya* **42**, 187-193(1977).
- [6] G. A. Alers and Ray Santoyo, "Shear Wave Velocity Response to Copper Precipitate and Neutron Irradiation," in *Workshop on Nondestructive characterization of embrittlement in reactor pressure vessel steels*, National Institute of Standards and Technology, United States Department of Commerce, 1999. Chapter19.
- [7] B. Igarashi and G. A. Alers, "Ultrasonic measurement of Dynamic Magnetostriction," in *ibid*, Chapter23.
- [8] D. C. Hurley, D. Balzar and P. T Purtscher, "Ultrasonic Nonlinearity Parameter in Precipitate-hardened Steels," in *ibid*, Chapter16.
- [9] A. L. Hiser Jr and R. E. Green Jr, "Evaluation of Ultrasonic Attenuation Measurements for Estimating Irradiation Embrittlement of RPV Steels," in *ibid*, Chapter18.
- [10] H. Jeong and D. K. Hsu, *Ultrasonics* **33**, 195-203(1995).
- [11] Y. B. Cohen and A. K Mal, "Ultrasonic Inspection," in *Metals handbook 17, Nondestructive Evaluation and Quality Control*, ASM International, Metals Park, 1989, pp.231-277.