Nonlinear Ultrasound to Monitor Radiation Damage in Structural Steel

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ABSTRACT

This work presents how the nonlinear ultrasonic technique of second harmonic generation can be used to monitor damage typical of nuclear reactor structural steel material. Second harmonic generation occurs when an ultrasonic wave interacts with microstructural features that create a nonlinear medium for the propagating ultrasonic wave. This phenomenon is measured by the acoustic nonlinearity parameter. Radiation damage causes microstructural evolution such as changes in dislocation density and the formation of precipitates, both of which have been shown to give rise to changes in the acoustic nonlinearity parameter. Previous work has shown that nonlinear ultrasonic techniques are sensitive to radiation damage, specifically that increases of radiation dose are detectable by changes in the acoustic nonlinearity parameter. For these measurements to be robust, alignment, clamping, and mounting of ultrasonic transducers to a sample must be simple, accurate, and repeatable. Nonlinear ultrasonic measurements were run on two types of nuclear reactor steel samples that were previously irradiated in the Rheinsberg power reactor to two fluence levels, up to 10^{20} n/cm^2 (E > 1MeV), through a previous study by the IAEA. More extensive experiments were run on unirradiated standard Charpy samples to test repeatability of the measurements using the fixture and to isolate measurement variations such as surface roughness and clamping force effects.

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INTRODUCTION

Nonlinear ultrasonic (NLU) techniques are a powerful nondestructive evaluation tool that can characterize precursors to macroscopic damage in metallic materials. It has recently been shown that NLU methods can detect changes in the microstructure of reactor pressure vessel steels due to irradiation [1]. This was done by monitoring the acoustic nonlinearity parameter, β, over increasing levels of neutron fluence. Irradiation causes voids and point defects to form [2], causes changes in dislocation density [2-4], and formation of precipitates [3], all of which have been shown to give rise to acoustic nonlinearity [5-7]. NLU methods have further been shown to monitor microstructural evolution in fatigue prior to macroscopic cracking [8, 9], creep [10], and cold work [11] for example.

When an ultrasonic wave propagates through a nonlinear material, a second harmonic wave is generated. It has been shown that for longitudinal waves, the acoustic nonlinearity parameter, in terms of this generated second harmonic, is expressed as [12]

$$\beta = \frac{8A_2}{A_1^2 x \kappa^2} \tag{1}$$

where A_1 is the amplitude of the first harmonic wave, A_2 is the amplitude of the second harmonic wave, x is the propagation distance of the ultrasonic wave, and κ is the wavenumber of the first harmonic wave. Measurements of the acoustic nonlinearity parameter for a given material sample typically track the first and second harmonic wave amplitudes while varying a controllable parameter in Eq. (1); in typical measurements with longitudinal waves, the input amplitude of the first harmonic wave is varied [9].

Multiple factors can influence the measurement of the acoustic nonlinearity parameter. Contact nonlinearities, for example from surface roughness and contact forces, effectively increase the measured acoustic nonlinearity parameter, plus lead to variation among samples if inconsistent [13]. In this work, measurement variation due to the experimental fixture, surface roughness of the samples, and variations in sample thickness (and thus clamping force) are investigated. Results of acoustic nonlinearity over fluence level are adjusted accordingly to account for these effects.

EXPERIMENTAL

Material Samples

Two types of nuclear reactor pressure vessel steels were investigated – ASTM standard A533B Cl.1 (IAEA reference material code ‘JRQ’) and A508 Cl.3 (IAEA reference material code ‘JFL’). These samples were part of a previous unrelated IAEA study, and material property details can be found in the literature [14, 15]. Samples were standard Charpy-V geometry of 10mm x 10mm x 55mm, and were irradiated at the Rheinsberg power reactor to two dose levels, up to a neutron fluence of 10^{20} n/cm^2 (E > 1 MeV), at a coolant temperature of 255°C [16]. For the study on relating the acoustic nonlinearity parameter to irradiation damage, unirradiated samples were machine polished with abrasive paper up to 600 grit, and irradiated samples were machine polished with abrasive paper up to 240 grit. Variation in sample thickness along the wave propagation direction (10mm direction) ranged from 0.02 mm to 0.13 mm for each set of materials at each fluence level. Sample specifications are summarized in Table I. Note that this work also utilized a separate set of unirradiated Charpy-V samples with as-is (unpolished) surface conditions, for studying surface roughness effects and clamping force influences on measurement results.

Table I. JRQ and JFL sample specifications.

Material	Fluence, n/cm^2 (E > 1 MeV)	Surface condition	Thickness variation in samples
JRQ	Unirradiated	600 grit	± 0.13 mm
JRQ	54.85 x 10^{18}	240 grit	± 0.05 mm
JRQ	98.18 x 10^{18}	240 grit	± 0.02 mm
JFL	Unirradiated	600 grit	± 0.02 mm
JFL	51.21 x 10^{18}	240 grit	± 0.04 mm
JFL	86.98 x 10^{18}	240 grit	± 0.02 mm

NLU Experiments

A laboratory measurement of acoustic nonlinearity using bulk ultrasonic waves is as follows; refer to Figure 1 (left). Transducers are mounted on opposite sides of a material sample with a small amount of oil coupling for efficient acoustic transmission. A high-power amplifier excites the transmitting transducer with a tone burst signal at or around its center frequency. The signal propagates through the material sample where the second harmonic wave is generated. The other transducer, operating at twice the center frequency of the transmitting transducer, receives the time signal consisting of both the first and second harmonic wave. The signal is transferred to an oscilloscope for viewing and then to a computer for post-processing to extract amplitude information, i.e. A_1 and A_2 in Eq. (1), by taking a Fast Fourier Transform of the time signal. This process is repeated for input amplitudes of 40%-90% of the amplifier output (roughly 734 V_{pp} with transducer loading). Taking a linear fit of A_2 and A_1^2 gives the acoustic nonlinearity parameter, β, as demonstrated in a representative measurement in Figure 1 (right). Note that for measurements of radioactive material, sufficient shielding surrounding samples is necessary to isolate equipment and operator from radiation exposure. The experimental setup allows for sufficient shielding and arrangement of the measurement apparatus in shielded area.

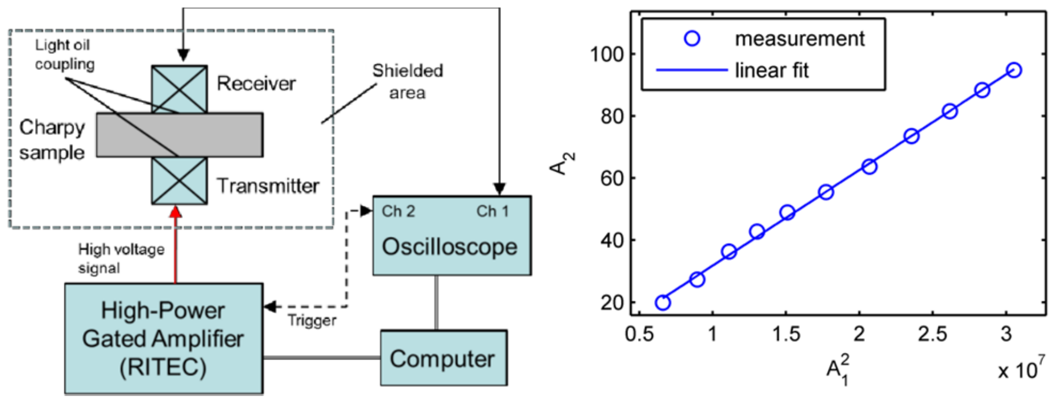

Figure 1. Laboratory schematic for nonlinear ultrasonic measurement (left), and representative measurement, showing linear growth of A_2 compared to A_1^2 (right). The slope of the linear fit is equal to β, the acoustic nonlinearity parameter.

NLU Measurement Fixture Design

In order to make measurements of the acoustic nonlinearity parameter robust, the experimental measurement fixture must allow for accurate and repeatable measurements. The fixture must align both transducers with a high degree of precision, must clamp the transducersto the surface of the material sample with a consistent contact pressure, and must minimize setup time since sensitive samples – radioactive material – were handled. These design points ensure accuracy of the extracted first and second harmonic amplitudes that make up the measured acoustic nonlinearity. To make nonlinear ultrasonic measurements more accessible, the fixture must have a simple and intuitive setup. The proposed fixture design for measurements on irradiated samples enabled quick set up of an experiment to make a nonlinear ultrasonic measurement.

The main functions of the fixture are to clamp the sample, and self-align and clamp the transducers in a repeatable manner. All parts are mounted to a plate and are shown in an exploded view in Figure 2 (left). The sample (a) is placed on fixed plate (c) by pulling back the movable plate (d) and placing sample in between this plate and the fixed plate (b). The fixed plate (c) is L-shaped to make sample mounting quick and easy while ensuring NLU measurements are taken at the same location on each sample. The movable plate (d) is spring- loaded (e) and clamps the sample in place once released, while also allowing for easy removal of the sample. The spring mechanism works by mounting the springs (e) on alignment rods (f) in between the moveable plate (d) and another fixed plate (g). Transducers (h) are housed in notches in plates (b) and (d) on opposite sides of the sample (a) that align transducers in the vertical center of the sample. Toggle clamps (i) are used to clamp transducers onto sample, while also allowing for a user-defined clamping force. The clamps used have a maximum holding force of 445 N. By changing the length of the toggle clamp, measurements can be made on samples with different thicknesses. The alignment rods (f) enable an automatic alignment of all plates. Setscrews (j) mounted on plates (b) and (d) provide accurate horizontal alignment of the transducers. The photograph in Figure 2 (right) shows the fully assembled fixture with mounted sample and transducers.

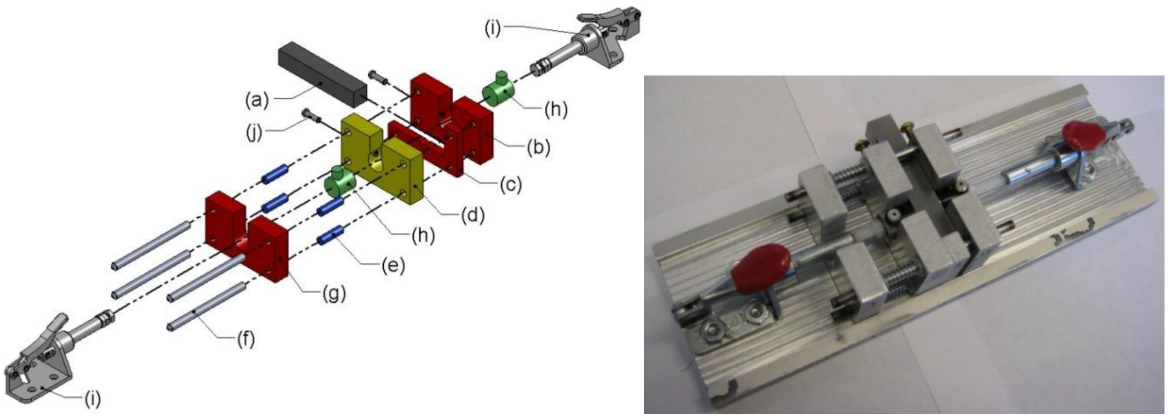

Figure 2. Exploded view of fixture design (left), and photograph of fixture with mounted sample and transducers. (right).

RESULTS AND DISCUSSIONS

Previous work showed that β increased from unirradiated to medium fluence, and then either leveled off (JRQ material) or decreased (JFL material) from medium to high fluence for reactor pressure steels [1]. In these results, β was measured in three different samples at each fluence level, for two different materials. The acoustic nonlinearity parameter varied by 8-30% among samples of same material and fluence level. These results are shown in Figure 3, along with an adjusted set of data that accounts for surface roughness effects as described in sections below. Note that this adjustment does not change the trend of the results but provides a more quantitatively accurate representation of β over fluence level.

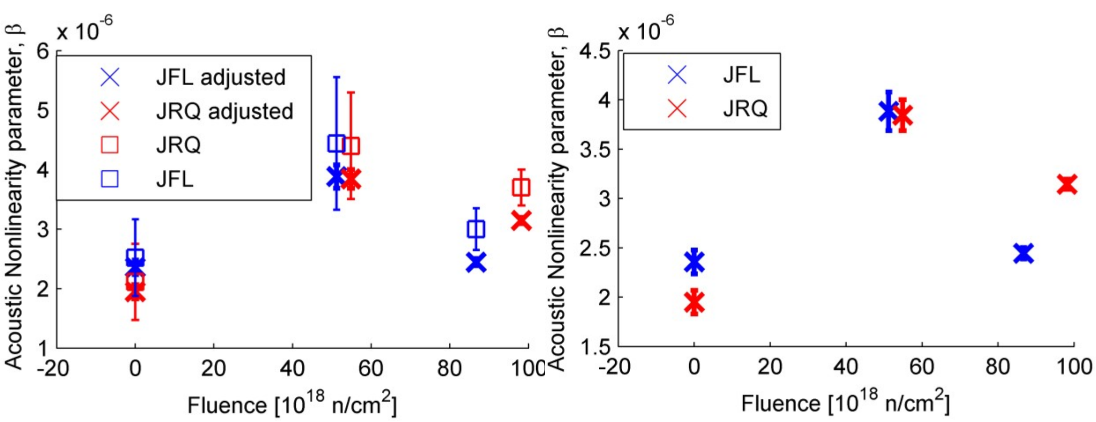

Figure 3. Acoustic nonlinearity parameter measured over fluence level for unadjusted results[1] and results adjusted to account for surface roughness effects (left), and adjusted results shown only for clarity (right).

Variation from Measurement Fixture

The fixture was used to measure the acoustic nonlinearity parameter in steel samples with two different radiation doses [1]. The fixture was transported to a laboratory in Dresden, Germany for these measurements, and experiments were set up by staff certified to handle the radioactive samples. Thus, these experiments provided a good means of evaluating the performance of the fixture. It was found that there was a maximum of 5% variation between different measurements on the same sample, which is small compared to variation due to microstructural variations and surface roughness that cause up to 30% variation in these measurements. Thus, the fixture provides repeatable measurements.

Surface Roughness Effects

Surface roughness of samples caused both an increase in β and a variation in β. To quantitatively investigate how surface roughness increased β, nonlinear ultrasonic measurements were taken on unirradiated samples at decreasing levels of surface roughness. As-is samples were polished up to 800 grit, and β was measured after each polishing increment. Since unirradiated samples were polished to a finer grit than irradiated samples and thus irradiated samples had a higher surface roughness, the β measured in irradiated samples should be larger than the actual value. The evolution of β over increasing levels of polish (and thus decreasing levels of surface roughness) is shown in Figure 4. These results show that β varies inversely with level of surface polish. With these results, β measurements for samples polished to different grit levels

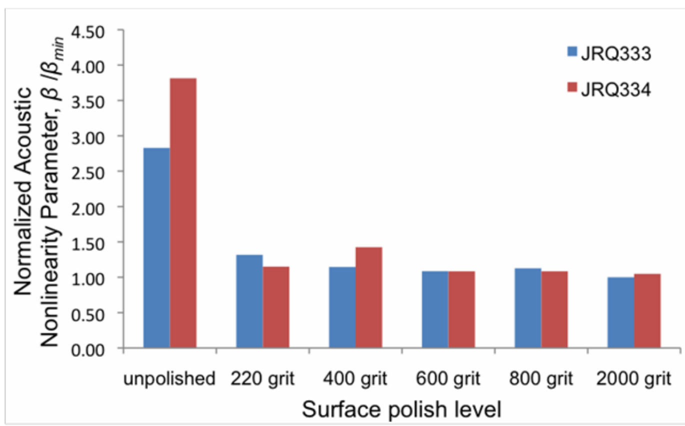

Figure 4. Dependence of acoustic nonlinearity parameter on surface polish level for two JRQ samples.

can be compared by accounting for the increase in β caused by surface roughness at that polish level. In this way, results of β over irradiation fluence level were adjusted, as shown in Figure 3.

To quantitatively investigate effects of variation from sample surface roughness, nonlinear ultrasonic measurements were taken on ten undamaged steel samples that were previously machine-polished to 600 grit. Samples were then hand polished to 2000 grit with a small amount of oil lubricant, and measurements were repeated. The variation in measurements among samples dropped to 3.27% for JFL and 4.57% for JRQ. Data for variation on acoustic nonlinearity before and after polish are given in Table II. The results for β measurements for all ten samples are also shown in Figure 5 (left), with upper and lower bounds of the β measurements indicated for each surface condition. Note that these results are normalized to the average value for each material and polish level.

Table II. Variation in β before and after surface polish to 2000 grit.

	***β* variation**		**Thickness variation**	
Material	**Before polish**	**After polish**	**Before polish**	**After polish**
JFL	15%	3.27%	0.144%	0.14%
JRQ	15.79%	4.57%	1.10%	1.12%

Clamping Force Variations

Due to the design of the fixture, there is a greater clamping force on thicker samples and less clamping force on thinner samples. This can easily be adjusted by the operator to obtain exactly the same clamping force for different measurements, but for sensitive samples such as irradiated samples, small adjustments for slight thickness variations can be time-consuming. To isolate and quantitatively evaluate effects of thickness variation among samples on β measurements, measurements were made on one sample while the length of one toggle clamp in the fixture was incremented (to simulate samples of different thicknesses). Measurements of β over changes in toggle clamp length are shown in Figure 5 (right), and are referenced to the length of the toggle clamp used in actual measurements. The upper and lower bounds for the toggle clamp length were selected as the points just before the toggle clamps could not fully close since the force was too great and just after the toggle clamps did not reach the transducers to clamp them to the

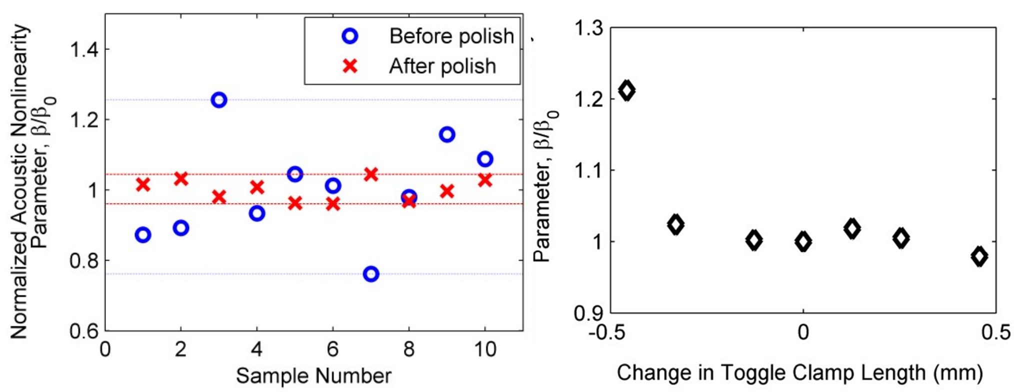

Figure 5. Normalized acoustic nonlinearity parameter before polish (polished to 600 grit) and after polish to 2000 grit (left). Variation in normalized acoustic nonlinearity parameter with change in toggle clamp length (right).

sample. Generally, acoustic nonlinearity varies inversely with increasing clamping pressure and levels off to a constant value, and this trend is generally consistent with previous experimental studies on how contact pressure influences the acoustic nonlinearity parameter [13]. Variations in β are within 5% for toggle clamp length (and thus sample thickness) variation of 0.8mm. To accommodate samples with more variation in thickness, toggle clamp length can be manipulated to provide the same clamping force. However, samples in this study varied up to only 0.13mm in thickness, so variations in clamping force had only a small effect (up to 5% variation) in measured acoustic nonlinearity.

CONCLUSIONS

This work shows how the acoustic nonlinearity parameter, β, can be used to monitor irradiation damage in reactor pressure vessel steels. The acoustic nonlinearity parameter is a measure of the second harmonic wave that is generated by interactions with microstructural features as an ultrasonic wave propagates through material. Microstructural evolution such as changes in dislocation density, point defects, and precipitates give rise to this second harmonic wave, and it is known that irradiation causes these microstructural changes [2-4]. It has previously been shown that the acoustic nonlinearity parameter is sensitive to increasing levels of neutron irradiation [1]. Further experiments were run on unirradiated samples to characterize the variations in clamping force and surface roughness that influenced the measurements on irradiated samples. It was found that after adjusting the acoustic nonlinearity results for these variations, the trend in β over fluence level remained the same, and the majority of the variation in the measurements was due to surface roughness variations.

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REFERENCES

1. K. H. Matlack, J. J. Wall, J.-Y. Kim, J. Qu, L. J. Jacobs, and H.-W. Viehrig, *Journal of Applied Physics,* vol. 111, pp. 054911-1 - 054911-3, 2012.
2. E. Meslin, M. Lambrecht, M. Hernandez-Mayoral, F. Bergner, L. Malerba, P. Pareige, B. Radiguet, A. Barbu, D. Gomez-Briceno, A. Ulbricht, and A. Almazouzi, *Journal of Nuclear Materials,* vol. 406, pp. 73-83, 2010.
3. J. Kocik, E. Keilova, J. Cizek, and I. Prochazka, *Journal of Nuclear Materials,* vol. 303, pp. 52-64, 2002.
4. W. G. Wolfer and B. B. Glasgow, *Acta Metall.,* vol. 33, pp. 1997-2004, 1985.
5. J. H. Cantrell, *Proceedings of the Royal Society of London, Series A (Mathematical, Physical and Engineering Sciences),* vol. 460, pp. 757-80, 2004.
6. W. D. Cash and W. Cai, *Journal of Applied Physics,* vol. 109, 2011.
7. J. H. Cantrell and W. T. Yost, *Applied Physics Letters,* vol. 77, pp. 1952-1954, 2000.
8. J. H. Cantrell and W. T. Yost, *International Journal of Fatigue,* vol. 23, pp. S487-S490, 2001.
9. J.-Y. Kim, L. J. Jacobs, J. Qu, and J. W. Littles, *Journal of the Acoustical Society of America,* vol. 120, pp. 1266-73, 2006.
10. S. Baby, B. N. Kowmudi, C. M. Omprakash, D. V. V. Satyanarayana, K. Balasubramaniam, and V. Kumar, *Scripta Materialia,* vol. 59, pp. 818-821, 2008.
11. A. Viswanath, B. P. C. Rao, S. Mahadevan, P. Parameswaran, T. Jayakumar, and B. Raj, *Journal of Materials Processing Technology,* vol. 211, pp. 538-544, 2011.
12. A. Hikata, B. B. Chick, and C. Elbaum, *Journal of Applied Physics,* vol. 36, pp. 229-236, 1965.
13. O. Buck, W. L. Morris, and J. M. Richardson, *Applied Physics Letters,* vol. 33, pp. 371-373, 1978.
14. IAEA, "Reference Manual on the IAEA JRQ Correlation Monitor Steel For Irradiation Damage Studies," Vienna2001.
15. A. Ulbricht, J. Bohmert, and H.-W. Viehrig, *Journal of ASTM International,* vol. 2, pp. 301-314, 2005.
16. C. Zurbuchen, H. W. Viehrig, and F. P. Weiss, *Nuclear Engineering and Design,* vol. 239, pp. 1246-53, 2009.