Exploring the Effect of Irradiation Time on the Damping of Surface Acoustic Waves

by

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Abstract

Finding relationships between material structure and properties is a key discipline of materials science. Many methods have been used to examine material properties, including transient grating spectroscopy (TGS). These properties were used to probe several scales of structure, and TGS has been applied in this way to solve several challenges. This thesis explores the details of the TGS signal itself, and how the property of irradiation time affects different qualities of the signal. In particular, the acoustic damping parameter is scrutinized in relation to the irradiation time of the samples from Byron Nuclear Generating Station. The trend was found to be similar to that of other material properties when compared with irradiation time, denoting the creation and subsequent clustering of defects. Further research with higher granularity into the relationship is suggested in order to further the development of nondestructive evaluation techniques.

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1 Introduction

One of the core principles of materials science is to understand the structure of materials and how that affects its properties. Being able to monitor and control these properties has a vast array of uses, such as doping semiconductors, controlling polymers in plastics, doping ceramics, and working metal to behave in different ways. There are many methods to go about characterizing materials, all with different affects on the sample used and degrees of precision.

In some cases, it is perfectly fine to take a small sample of a material and do whatever tests are needed. Some tests, such as Charpy impact tests, destroy a sample; a pendulum is dropped onto a notched sample of a material and the energy transferred to the material is inferred from the change in height of part of the pendulum. Some other techniques change a material's structure, such as calorimetry studies. During one, the sample is heated slowly to record the amount of power required to do so. This anneals the sample, so its structure is changed from what it was before.

There are some cases where these methods are not optimal. One example is reactor pressure vessels, where there is a finite number of samples that can be taken. When the pressure vessel is built, it has some number of tabs of metal known as "inspection coupons" placed inside the vessel that are made of the same material as the rest of it. When the state of the metal needs to be inspected, a coupon can be taken out and tested. These tests (such as Charpy impact tests) are destructive, so that particular coupon is not a good candidate for future testing. While there are many inspection coupons built into a reactor pressure vessel, some US nuclear power plants are getting on in years and are running low on them. For further testing, old coupons have to be reinserted and used or steel has to be taken from the bulk pressure vessel. This is obviously detrimental to the long term health of the pressure vessel.

The health of reactor pressure vessels is key to the continued service of the nuclear power plant it is located at. Nuclear power provides a sizable portion of many countries' power supplies, and in order to operate, all parts of a reactor must pass safety inspection. In order to pass safety inspection, there must be a way to test that each element operates correctly and will continue to do so. If some element does not pass, especially a reactor pressure vessel, there are two outcomes: either losing power output, accompanied by a loss of electricity to the grid or time of experiments depending on the reactor's purpose; or risking a failure and resulting accident, which will also bring power loss as well as put the surrounding area at risk. In order to prevent that, finding ways to sustainably gauge RPV health is an important goal to pursue.

A possible way of finding such sustainable methods is through studying nondestructive evaluation techniques. An example of this is transient grating spectroscopy (TGS), which uses laser pulses to create waves of thermal expansion which interfere and cause a standing acoustic wave on the surface of a material. When analyzed, the signal has several properties that correlate with the material's properties, such as thermal diffusivity. It is possible that other traits of the signal are influenced by the material's structure. If such a relationship could be identified and characterized, it would enable those inspecting reactor pressure vessels to use a nondestructive method of evaluation. This would extend the length of time that the RPV could be reliably evaluated for safety, which would be very beneficial for aging nuclear power plants. Additionally, more points of reference with respect to safety inspections provides more information for better evaluation of the condition of the pressure vessel itself, contributing to an overall increase in nuclear safety.

2 Background

2.1 Aging in RPVs

One of the primary mechanism through which irradiation aids degradation of steel is through neutron embrittlement. This happens when neutrons hit the atoms in a material and cause microscopic movement and defects, which build up over time and can move and cluster when exposed to heat, as happens in the annealing process. This, paired with the water and pressure that reactor pressure vessels are constantly exposed to, can cause cracking. These conditions also cause corrosion, which can be aided by irradiation as well. Precipitates form in places in the steel and can compromise the structural integrity of the RPV.

Previous work of tracking precipitate growth in reactor pressure vessels has shown an overarching pattern. The steel starts clean, with only the defects found in any unirradiated steel. At first, very tiny defects are distributed evenly throughout the metal. Over time, these defects cluster into smaller clusters, and eventually larger clusters. The larger clusters leave the surrounding metal in a similar state to pre-irradiation and may be annealed out entirely. Several depictions of this process in steel can be found in Almirall et. al.'s publication on Mn-Ni-Si precipitates in RPVs [1]. This process of damage and recovery is reflected when properties that depend on material structure are probed. There are a number of examples of this, one of which is the frequency of a generated acoustic wave when related to displacements per atom (dpa). This can be seen depicted in Figure 4 of Almousa et. al. [2]. The acoustic wave travels more easily through homogeneous material, so in their samples with more void swelling, the frequency changed more around mid-level doses. Similar patterns can be seen with a different method of magnetic investigation of RPV steel, where a U-shaped curve can be observed as a function of neutron fluence in Figure 7 of Kobayashi et. al [3]. Yet another example can be seen in Figure 3 of Mamivand et. al., with the number density of copper precipitates in RPV steel increasing rapidly and then gradually decreasing over irradiation time [4].

The current exact procedures for evaluating reactor pressure vessels varies by the regulatory body in each country. However, they all have in common the assessment of structural integrity by testing the steel's resistance against strength and fracture [5]. Frequently this test is destructive, testing the inspection coupon's fracture mechanics by fracturing it. These inspections are done periodically over the RPV's licensed life. When they reach the end of their license, it is sometimes the case that the license can be extended for an additional several decades. This time is approaching for a large number of the world's nuclear power reactors, as well over half of them have been in operation for over 30 years [5].

2.2 Transient Grating Spectroscopy

Transient grating spectroscopy (TGS) is a material evaluation technique that is nondestructive and, somewhat uniquely, does not involve contact with said material. Two pulsed laser excitation beams are overlapped and create an interference pattern that forms a periodic intensity pattern. The absorption of the light causes thermal expansion, which results in a periodic variation in height on the material and two surface acoustic waves (SAWs) propagating in opposite directions. The technique monitors the resultant grating and SAW decay using a probe laser beam diffracted off the grating and a reference beam reflected from the material. The signal itself is a measure of intensity of said laser over time in nanoseconds [6]. Vibrations propagate best through a medium when they are unobstructed by foreign objects, so it can be inferred that defects in a sample that TGS is done on can cause the signal to decay more quickly in a more heterogeneous material.

TGS has potential in both exploring thermal and elastic properties in materials as well as inferring other information from said properties, such as changes in a material's microstructure. Of course, before TGS there has been frequent forays into evaluating material properties with other nondestructive methods.

2.3 Wave Energy Dissipation

In the past, most experiments using the dissipation of SAWs have looked at either the elastic properties of their samples or the growth of precipitates, or how these properties relate to each other. As far back as 1959 experiments were being conducted, such as Folweiler and Brotzen using contact ultrasonics to characterize the Young's modulus of aluminum [7]. In the 90's Li and Szpunar also used ultrasonics to probe elastic properties [8], and since then several others have used SAWs to investigate elastic constants and stiffness tensors in pure metals [9], nickel superalloy [10], and austenitic steel [11].

The monitoring of growth of precipitates in materials with SAWs has been examined at length with Rayleigh nonlinear ultrasonics. They connect the change of a nonlinearity term, typically denoted as β, to infer the corresponding change in precipitates. This has been done in several types of steel, such as P92 steel [12], 17-4PH stainless steel [13][14], and steel from reactor pressure vessels [15], as well as a titanium alloy [16]. This technique has also been used to characterize stress corrosion cracking more specifically [17], as it is a phenomenon caused in part by the growth of precipitates.

One more novel way found of inducing SAWs to examine material properties is through optics, as Verstraeten et. al. did, where the hardness of his steel samples was related to the wave amplitude of the signal [18].

Finally, TGS has been used by several people to examine a variety of properties. Dennett has used it for monitoring thermomechanical properties [19], finding elastic constants in aluminum and copper [20], and monitoring swelling evolution in nickel alloys [21]. Almousa has also used TGS to investigate swelling, although this time in irradiated steels using the frequency of the signal [2]. Ferry used it to monitor microstructural evolution in niobium via thermal diffusivity [22], and found a similar trend to that discussed in section 2.1.

3 Methods

3.1 TGS Data

The samples of RPV steel came from Byron Nuclear Power Plant. They were irradiated in surveillance capsules in the Byron Unit 2 PWR. There are four, with irradiation times of 1.2, 4.7, 8.6, and 20 effective full power years. Their specimen IDs are YT2, YT8, YT10, and YT13 respectively. The data for these samples was taken on 3/27/2023, at 19 different spots on each of the samples, with a grating spacing of 6.40μm.

As explained in section 2.2, TGS works by pulsing a laser to create a temperature grating, producing SAWs. The grating is probed by a laser along with a reference laser to record intensity over time. The setup used to take data from these samples is identical to the one used by Dennett et. al. to take data from various metal alloys at the Sandia Ion Beam Laboratory [19], as it was relocated to an MIT lab.

3.2 Analytical Methods

The script used to fit the TGS signals to an equation was developed by C.A. Dennett [23]. Mathematically speaking, it uses an iterative procedure, the Levenberg-Marquardt nonlinear least squares method. The code goes through several aspects of the signal and fits terms to it one at a time with inital guesses for each. It first fits the amplitude of the signal, then figures out the phase of the sinusoid aspect. The final part of the fit is the physical parameters of α and β. It determines these by starting with an estimate of α and then using expressions that relate α and β to iterate through values of them until they converge.

Inputs

The script requires inputs of the positive and negative phases of the signal measured by the TGS setup, as well as the size of the grating used for that batch of runs. The data taken from the RPV steel had a grating spacing of 6.40μm. The script also requires an arbitrary integer between 1 and 4 to start its fit from, and has an option to do a baseline subtraction of background noise. The data for a baseline subtraction was only available for and thus performed on the YT2 and YT10 RPV samples.

Outputs

The script outputs all of the parameters for expressing the fit of the TGS signal. This fit is of the form

$$I_P(t) = A[\operatorname{erfc}(q\sqrt{\alpha t}) - \frac{\beta}{\sqrt{t}}\exp\Big(-q^2\alpha t\Big) + B\sin(2\pi f t + \theta)\exp\Big(-\frac{t}{\tau}\Big) + C] \tag{1}$$

where $I_P(t)$ is the intensity of the phase grating signal, A, B, and C are amplitude constants, f is the frequency of the acoustic oscillation, θ is the acoustic phase, τ is the acoustic decay constant, q is the constant grating wave vector defined as $q = \frac{2\pi}{\Lambda}$,

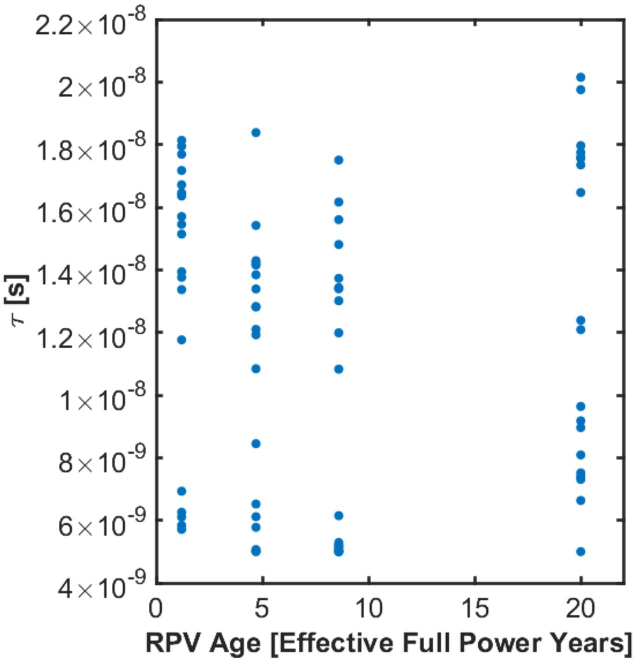

Figure 1: RPV Age vs. τ.

β is the ratio of diffraction contributions from reflectivity to displacement, and α is the isotropic thermal diffusivity [23].

Physically speaking, these variables are rather straightforward. The features of the base sinusoid are expressed in f and θ, the frequency and phase of the oscillation. q is not an output of the script, but is calculated from the grating spacing input. τ represents how fast the amplitude of the sinusoid decays. α is an expression of rate of heat transfer. β expresses how much of the signal's diffraction came from the reflectivity of the sample as compared to the displacement of the surface of the sample. Finally, A, B, and C have no physical meaning, acting as fitting constants.

4 Results

The raw values of τ are plotted against RPV age in Figure 1. It is distributed somewhat bimodally, with groups falling somewhat above and below 1×10^{-8} s, from here on referred to as the top and bottom groups respectively. The data was analyzed both in these bimodal groups and together as an aggregate group.

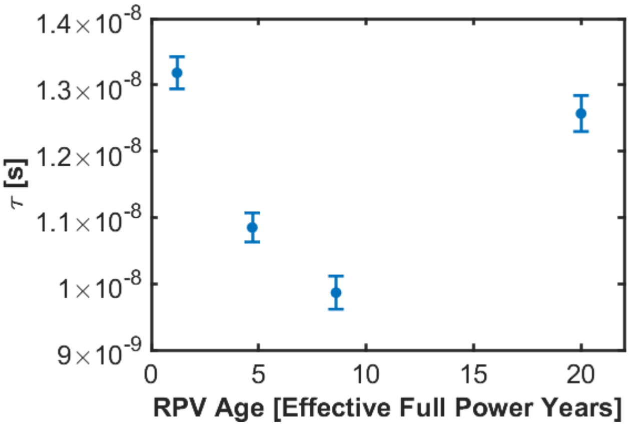

Figure 2: τ over RPV age, aggregate group

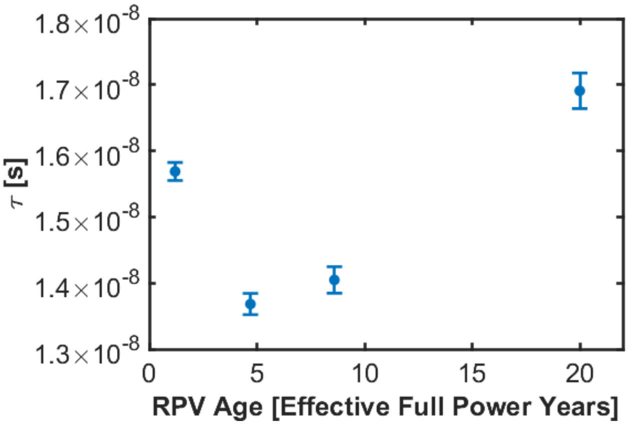

Figure 3: τ over RPV age, top group

All three groups displayed similar behavior, with τ decreasing over the first decade or so of the pressure vessel's life and then increasing as it became older.

The plots of the top (Fig. 3) and bottom (Fig. 4) groups differ slightly, with τ increasing between 4.7 and 8.6 years in the top group and decreasing over that time interval in the bottom. Additionally, they both differ from the aggregate data (Fig. 2) in that their values of τ at 1.2 years are less than those at 20, while the reverse is true for the aggregate data.

Additionally, histograms of the values of τ were made for each RPV age to illustrate the bimodal distribution and to check if they adhered to Gaussian statistics. They can be seen in Figures 5, 6, 7, and 8. They were only somewhat normally distributed; standard error was used on the plots of τ over time regardless.

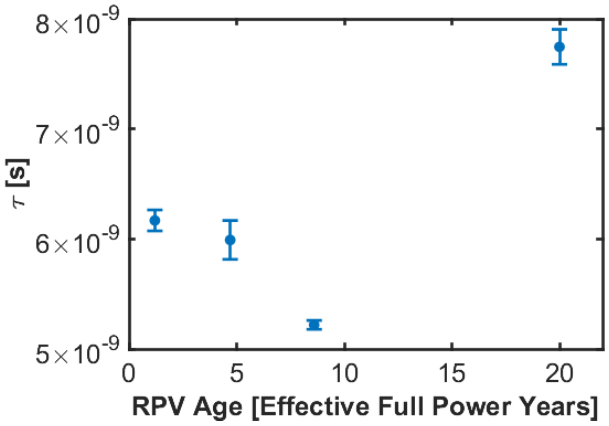

Figure 4: τ over RPV age, bottom group

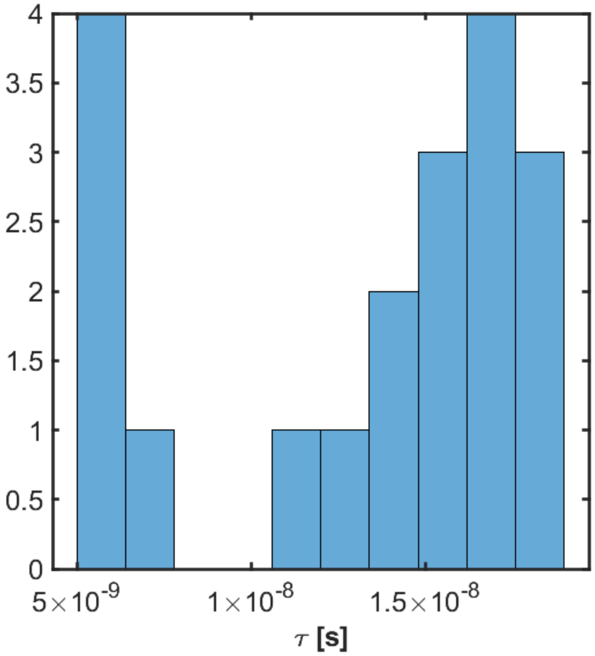

Figure 5: Values of τ for 1.2 year old sample

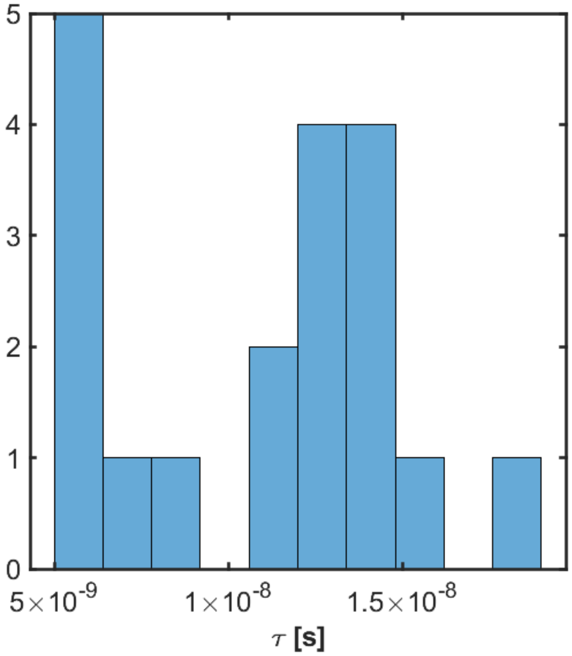

Figure 6: Values of τ for 4.7 year old sample

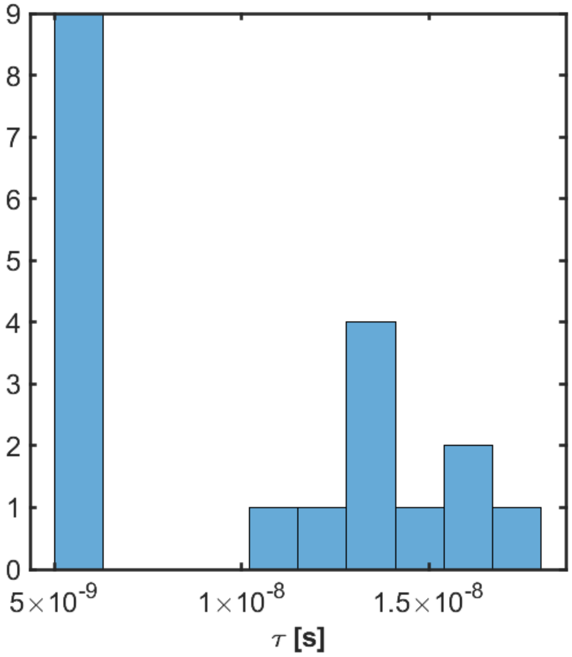

Figure 7: Values of τ for 8.6 year old sample

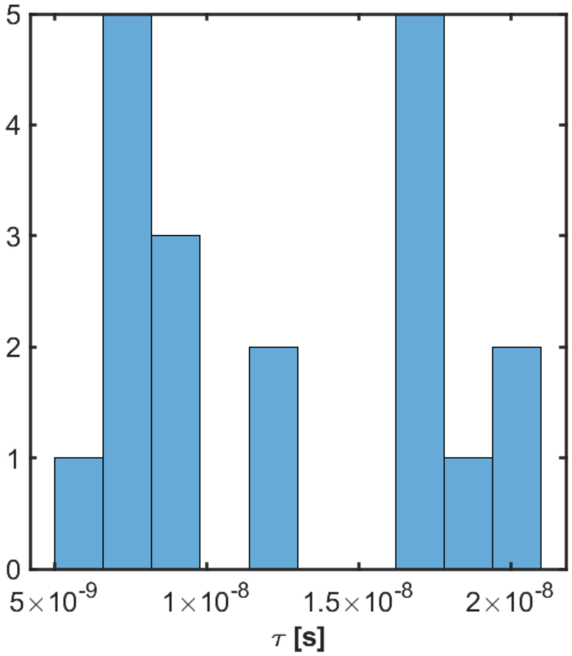

Figure 8: Values of τ for 20 year old sample

5 Discussion

The data displays the same trends as other forays into examining material properties as a function of irradiation time. A lower value for the acoustic damping parameter implies more damping, so it can be seen that there is an initial increase in damping in the first several years of an RPV's service followed by a decrease over time as it ages, going back to a similar state as it was at the beginning of its life. This is in line with the damage and recovery mechanism discussed; initially defects are tiny and spread out, making the steel more heterogeneous. Over time the defects start to cluster, keeping some level of heterogeneity. More time brings more clustering and some defects annealing out, leaving the steel with more swathes of defect-free material. Eventually the defects form large clusters, with the steel having a similar structure as it had in the beginning, but with some pockets of damage. This leaves more material for the SAWs to propagate unperturbed, resulting in less acoustic damping. The same goes for the middle stage; many scattered defects makes it more difficult for the SAWs to propagate, so the damping is thus higher.

The precise shape of the trend across different studies is not consistent. In general the peak is somewhat soon after irradiation begins, but it is hard to draw solid conclusions from a small number of studies investigating very different things.

It is possible that bimodal distribution of the data comes from there being two main types of material in the samples: the steel and the precipitates. Regardless of whether this is the case, to characterize the steel as a whole the data should be taken as an aggregate.

The statistically significant trend found for this sample warrants further investigation into using τ as an indirect way of assessing RPV health. An experiment with data from a greater number of pressure vessel ages would give greater resolution to the damage and recovery trend, and taking a greater number of data points from each sample would facilitate investigation of the apparent bimodal distribution of data, should that be a useful course of investigation. Additionally, it is possible that other parameters in the fit of the TGS data have some correlation with RPV age, so a short study there could also be beneficial.

6 Conclusion

When studying materials science, determining methods of monitoring material properties is an important area of study. Finding nondestructive methods is useful in a number of situations, such as when only a limited number of samples can be taken from the source. TGS offers such a method, and its output signal can and has been used to identify material properties such as elastic constants and to monitor swelling and precipitate growth. In scrutinizing the signal in the specific case of data from RPV samples, RPV age and the acoustic damping parameter were found to have a relationship that follows the pattern found in other studies comparing irradiation and various material properties. The pattern denotes the creation and subsequent clustering of defects, changing how SAWs propagate through the steel. This suggests that the ADP could be used in future efforts to develop a nondestructive evaluation technique using TGS, as well as in monitoring precipitate growth. Further study with a higher resolution of RPV ages would help to establish a more detailed view of precipitate growth over an RPV's lifetime. While there is still much work to be done in finding a way to relate TGS data to material properties that is as effective as destructive tests that directly measure strength, more avenues to explore makes it easier to get a full picture of how the data can be used.

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