

NEUTRON DAMAGE IN REACTOR PRESSURE-VESSEL STEEL EXAMINED WITH POSITRON ANNIHILATION LIFETIME SPECTROSCOPY

Stephen E. Cumblidge, Arthur T. Motta, and Gary L. Catchen
Nuclear Engineering Dept., The Pennsylvania State University, University Park, PA, USA

ABSTRACT

We have used positron annihilation lifetime spectroscopy to study the development of damage and annealing behavior of neutron-irradiated reactor pressure-vessel steels. We irradiated samples of ASTM A508 nuclear reactor pressure-vessel steel to fast neutron fluences of up to 10^{17} n/cm², and we examined these samples using positron annihilation lifetime spectroscopy (PALS) to study the effects of neutron damage in the steels on positron lifetimes. Non-irradiated samples show two positron lifetimes: a 110 ps component corresponding to annihilations in the bulk material, and a 165 ps lifetime corresponding to annihilations in dislocation defects. The irradiated samples show an additional lifetime component of 300 ps in the PAL spectra and an increase in the proportion of annihilations with a 165 ps lifetime, suggesting that vacancies and vacancy clusters are present in the material after room temperature irradiation. The samples were then annealed to temperatures ranging from 210° C to 450° C. The positron lifetimes introduced by neutron damage disappear after annealing the samples at 280° C.

INTRODUCTION

Reactor pressure-vessel steel embrittlement is one of the most difficult and serious problems facing nuclear utilities around the world. Decades of neutron irradiation alter the microstructure of the reactor pressure-vessel steel, causing it to become harder and less ductile. The embrittling mechanism is associated with the formation of nano-sized defect clusters such as depleted zones, small Cu-rich precipitates, and interstitial clusters which are invisible to TEM, but which may be visible to positron annihilation lifetime spectroscopy. By providing a means to measure and characterize the damage, positron annihilation lifetime spectroscopy can help identify the degree of embrittlement in the reactor pressure-vessel.

The examination of radiation damage by positron annihilation lifetime spectroscopy is based on the principle that defects created by irradiation trap positrons and measurably increase their lifetimes. PALS has been extensively used to study damage in metals [2]. A different positron lifetime is associated with each type of defect [1]. Because most reactors operate at around 280°-300° C, it is important to know the effects of annealing the irradiated steel at these temperatures. Positron annihilation lifetime measurement can potentially give insight into the neutron damage annealing kinetics, which are relevant to assessing the effects of proposed pressure-vessel annealing schedules. In this paper we describe the effects of irradiation on samples of reactor pressure-vessel steel and the effects of annealing these samples to temperatures from 210° to 450° C.

EXPERIMENTAL METHODS

ASTM A508 reactor pressure-vessel steel was obtained from the Westinghouse Electric Corporation. The steel was treated at 615° C for 30 hours prior to irradiation, to simulate a post-weld heat treatment. Samples were cut to 1 cm² x 1 mm thick using a wire saw. This created a cold-worked region that had to be removed, because the damage in this cold worked region interferes with accurately measuring the positron annihilation lifetime spectra. The steel was etched in nitric and hydrochloric acid to remove these artifacts.

In these experiments we used a fast-slow coincidence positron annihilation lifetime spectrometer. Our system uses BaF₂ detectors, and has a resolution of 280 ps, measured using the FWHM of a ⁶⁰Co spectrum. The samples were irradiated using Penn State's TRIGA reactor to fluences of 10¹⁷ n/cm² and 7x10¹⁶ n/cm². The positron spectra were deconvoluted using the PATFIT computer program package [3] furnished by Risø Laboratory. We used the program RESOLUTION to determine the resolution function of the spectrometer, and the POSITRONFIT program to separate the lifetime information from the spectrum. The positrons source consisted of ²²NaCl encapsulated between two Kapton foils. The corresponding effects of the annihilations in the foils and in the salt were evaluated and removed from each spectrum. The temperature in the room was controlled to (± 2° C) to minimize electronics drift. A three-lifetime model was used to represent the spectrum:

$$R(t) = I_1 e^{-t/\tau_1} + I_2 e^{-t/\tau_2} + I_3 e^{-t/\tau_3} \quad (1)$$

where $R(t)$ is the number of annihilations, I_i are the percentages of annihilations with lifetime i , and the τ_i are the positron lifetimes in different traps. Clearly, in a real material, there are more than three lifetimes corresponding to various microstructural features. Thus, the use of three lifetimes is an approximation, that achieves a balance between the need to accurately model the material and the need to obtain unique, consistent sets of parameters from the fitting. Moreover the physical significance of the lifetimes must be considered when the effects of high fluences are involved. We present lifetime assignments that pertain to the case of low-fluence, low-defect concentration samples.

The lifetimes used in this model were determined by the following controlled experiments. The first positron lifetime was determined by examining pure iron (99.5%), after annealing at 1200°C for 2 hours. We found the spectrum to be well fit in this case by a single lifetime of 110±1 ps, in good agreement with previous studies in the literature [4]. Thus the first lifetime corresponds to annihilations in bulk Fe. The second positron lifetime, which we associate with positrons annihilating in dislocation defects, was determined by examining cold-worked iron. The well-annealed iron was cold-worked by rolling, which introduced a positron lifetime of 162 ps, also in good agreement

with previous work [5] in which a positron lifetime of 165 ps for dislocations is found. We used 165 ± 2 ps for the second lifetime in the three-lifetime model. The third lifetime, associated with vacancy clusters, was found by examining reactor pressure-vessel steel samples irradiated at room temperature to neutron fluences of 10^{17} n/cm². Samples showed an additional positron lifetime of 300 ± 3 ps, which corresponds well with the positron lifetimes attributed by Brauer [6] to small vacancy clusters.

RESULTS

After room temperature neutron irradiation, the reactor pressure-vessel steel material shows an increase in average positron lifetime and shows a percentage of annihilations occurring at larger lifetimes (300 ps). The results of the deconvolution of the positron spectra using the three-lifetime model are shown in figure 1, where we plot the percentage of annihilations with each of the three lifetimes against neutron fluence.

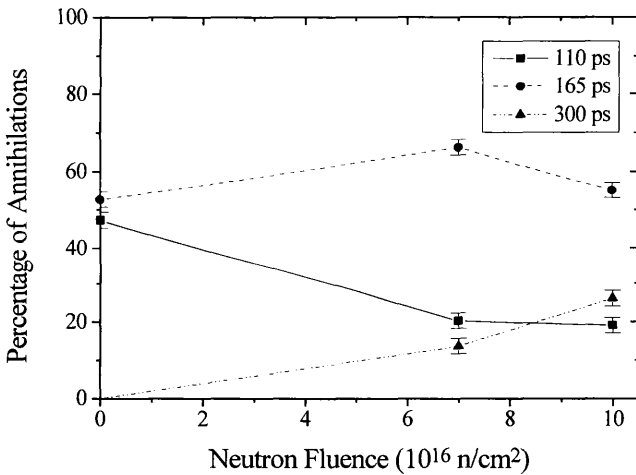


Figure 1: Percentage of annihilations with different lifetimes versus neutron fluence measured on reactor pressure-vessel steel samples.

As the neutron fluence increases, the percentage of positrons annihilating with a lifetime of 300 ps also increases. Irradiation creates vacancies which at room temperature may be immobile and whose concentration therefore steadily increases with fluence. Although the 300 ps lifetime represents all the contributions from clusters of different sizes, the spectra were very well fit using a three-lifetime model. In the irradiated pressure-vessel steel samples the number of positrons annihilating with a lifetime of 165 ps increases at a fluence of 7×10^{16} n/cm². The ratio of the percentage of positrons annihilating with a

lifetime of 165 ps to the percentage with a lifetime of 110 ps is higher than the ratio in the non-irradiated material. This increase is likely caused by the formation of vacancies under neutron irradiation.

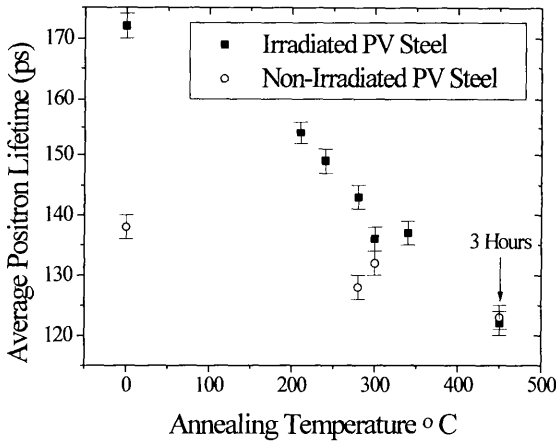


Figure 2: Average positron lifetime vs. 24-hour isochronal annealing temperature for reactor pressure-vessel steel samples irradiated to a fluence of 7×10^{16} n/cm².

Figure 2 shows the average positron lifetime observed after the material irradiated to a fluence of 7×10^{16} n/cm² was subjected to different annealing schedules. The average positron lifetime decreases as the annealing temperature increases. The average positron lifetime after 24-h annealing of the irradiated and non-irradiated reactor pressure-vessel steels converge at 300°C. Figure 3 shows the results of the analysis using the three-lifetime model for samples subjected to the different annealing schedules. The percentage of positrons annihilating with a lifetime of 300 ps goes to zero at 280°C. This effect is consistent with the disappearance of the vacancy clusters at this temperature. The percentage of positrons annihilating at 110 ps and 165 ps goes to 50% at these temperatures. This is close to the results of fitting the spectrum for non-irradiated reactor pressure-vessel steel. Thus, both the lifetimes and the intensities of irradiated material are similar to those of non-irradiated material after annealing.

Studies are under way on reactor pressure-vessel steel samples that were irradiated to 1.5×10^{17} n/cm² at 300°C. Preliminary results show that the average positron lifetime in the high-temperature-irradiated material increases from 138 ps for non-irradiated steel samples to 149 ps for the samples irradiated to 1.5×10^{17} n/cm² at 300°C. The complete results of the experiments performed on the high-temperature-irradiated material will be presented in a later paper.

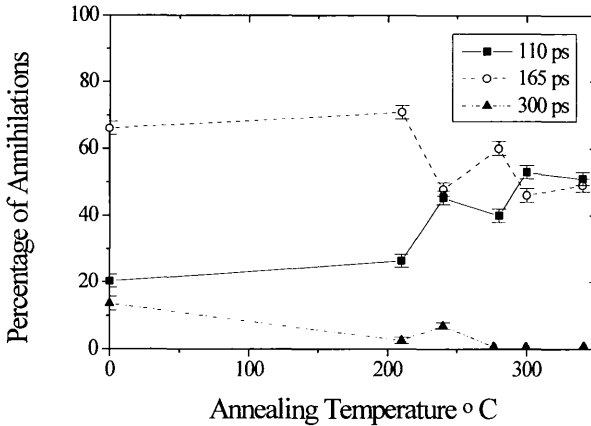


Figure 3: The percentage of annihilations for each positron lifetime observed after 24-hour isochronal annealing of reactor pressure-vessel steel irradiated to $7 \times 10^{16} \text{ n/cm}^2$.

DISCUSSION

Using the three-lifetime model, we can discern the effects of radiation-produced vacancies on the measured lifetimes, even though the lifetimes associated with single vacancies, 175 ps [7] and those associated with dislocations, 165 ps, are not easily resolvable. On irradiated samples we would expect the 165-ps percentage (representing dislocations) to the 110-ps percentage (representing the bulk crystal) either to decrease or to change very little, because irradiation effects would increase the percentage of annihilations that occur in vacancy clusters. Instead, after irradiation to a fluence of $7 \times 10^{16} \text{ n/cm}^2$, the 165-ps percentage increases and the 110-ps percentage decreases. This behavior likely arises, because the fitting procedure combines the 175-ps and 165-ps lifetimes, which represent annihilations in dislocations and vacancy clusters, respectively. Annealing the samples at 300°C brings these ratios close to the pre-irradiation value. This suggests that annealing sharply reduces the concentration of vacancies.

The formation of vacancy clusters is evident, as the 300 ps lifetime corresponds well to the results of other work done in this field. At the low fluences that these samples were irradiated to, the intensity of the 300 ps lifetime component increases with fluence. Because the concentration of vacancy clusters in low-fluence materials increases with fluence, this result suggests that the increase of the 300 ps lifetime component follows the increase in the concentration of vacancy clusters. The 300 ps lifetime likely results from the contribution of clusters of several sizes with an average overall lifetime of 300 ps.

The samples lose all detectable indications of having been irradiated after annealing at 300° C: the PAL spectra for the 300° C annealed samples and the as-received samples are nearly identical. The annealing effects of irradiating at 300° C would obviously not prevent embrittlement caused by mechanisms such as radiation-enhanced precipitate formation in the steels, or by the formation of dislocation loops in the material. The 450° C annealing removes all detectable neutron damage and reduces the intensity of the 165 ps lifetime (relative to the non-irradiated material) as well. This is interesting, as the material had previously been heat-treated at 615° C for 30 hours before irradiation. One would not expect heat treatment under 615° C to further anneal the sample.

CONCLUSIONS

We have measured positron-lifetimes in non-irradiated and irradiated reactor pressure-vessel steel samples up to fluences of 10^{17} n/cm². The measured average positron lifetimes increase with the neutron fluence. These results suggest that vacancies and vacancy clusters form in the irradiated reactor pressure-vessel samples. The lifetimes associated with the defect formation disappeared after the material was annealed at elevated temperatures. Annealing samples irradiated to fluences of 7×10^{16} n/cm² at 300° C for 24 hours removes the detectable of damage from the material from the point of view of positron annihilation lifetime spectroscopy.

ACKNOWLEDGMENTS

We thank Paula Freyer, James Williams and Rick Rishel of the Westinghouse Electric Corporation for furnishing the samples and for their interest in the research, Chris Davis for assistance in performing the experiments, and Morton Eldrup for useful advice on the use of PATFIT. We acknowledge the expert technical help we received at the Breazeale reactor. The FERMI Utilities Consortium, (which includes GPU Nuclear, PSE&G, PP&L, PECO and Westinghouse Electric Corporation) has funded this research. We thank the FERMI members for their continued interest and support of this work.

REFERENCES

- [1] Y.C. Jean, Positron and Positronium Chemistry, World Scientific, Singapore (1990)
- [2] S. Berko and H. Weisenburg, *Physical Review B* 154 (2) (1967) 249
- [3] P. Kirkegaard, M. Eldrup, O. E. Mogensen, and N. J. Pedersen, *Comput. Phys. Commun.* 23 (1981) 307
- [4] H. E. Birringer, R. Wurschum, R. Birringer, and H. Gleiter, *Phys.Rev.B*, 38 (14) (1998) 37
- [5] G. Brauer, L. Liszakay, B. Molnar, and R. Krause, *Nuc. Eng. Des.* 127 (1991) 47-68
- [6] G. Brauer, *Journal de Physique IV* (5) Jan. (1995) C1 143
- [7] P. J. Schultz and C. L. Snead Jr., *Metallurgical Transactions A* (1990) 21