

Effect of Ion Irradiation on the Mechanical Properties of High and Low Copper

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ABSTRACT

An investigation into the effects of proton beam exposure on high- and low-copper structural materials for nuclear reactors has been carried out. The aim of this work was to investigate the impact of proton energy irradiation on the damage of the materials. The damage parameter used in the evaluation was displacement per atom (dpa) in material as a function of proton energy. In addition, a TRIM code was used to identify the penetration depth in response to changes in proton energy. The effect of proton beam exposure on the irradiation induced hardening of the different copper levels was investigated by Vickers Hardness tests for microstructural changes examination. The proton beam incident energy was 3 MeV and the temperature was kept at approximately 30 °C. A 25 μm flat damage profile was achieved at 0.367 and 0.373 dpa for low and high copper samples, respectively. The hardness variation with depth and yield strength variation with dose (dpa) were also investigated. Based on the results, the study found that the hardness test for the high copper was higher than the low copper.

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INTRODUCTION

Ion irradiation experiments allow control of ion energy, dose, dose rate and temperature to obtain reproducible and specific results desirable for studying changes in irradiated micro-structure and property of a material in a shorter time [1-3]. However, the evolution of irradiated microstructure is dependent upon a combination of damage rate and irradiation temperature. Damage cannot always be reproduced in a relatively shorter time through increased displacement rate (dpa/s) alone, so temperature shift relations have to be created to allow for more correlation of one type of damage evolution from one irradiation environment to another using dose, dose rate and temperature [4]. One of the advantages of using proton beam is that parameters can be easily varied. Ion beams are also generally cheaper to produce a given dose compared to neutron sources.

Copper impurity has been found in older generation reactor pressure vessel (RPV) steels due

to the use of copper coated welding rods [5]. It was found that 0.3 and 0.05 wt% were used for high and low impurity levels of copper, respectively. A study by Zhang *et al.* [6] suggests that copper precipitates that are rich in other impurities (Ni, Al and Mn) formed under proton irradiation remain stable with further irradiation. Clusters of these precipitates inhibit the movement of dislocations under stress, which causes a net increase in hardness after irradiation. The mechanism of irradiation hardening is similar to that of aging except that proton radiation increases the free movement of vacancies, which encourages the diffusion of solutes. Another similarity to thermal aging is that under irradiation, instead of dissolving the precipitates coarsen. This phenomena cause the material to soften to some degree [6]. The conclusions that can be drawn from Zhang *et al.* [6] is that hardness increases with segregation of impurities in RPV steels, one of them is copper. Their binding to point defects in the matrix furthers the movement of these Cu-rich precipitate clusters. These clusters migrate to the areas of low energy such as grain boundaries, and sometimes they act as 'sinks' to which point defects

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continue to migrate. This process means that these sinks can also act as recombination sites, where interstitials and vacancies can combine and ‘recover’ the hardness of the material by some fraction. This process can occur during irradiation and promotes improvement of the mechanical properties of the material.

Further study conducted by Shibamoto *et al.* also suggests that hardening is caused by copper clustering in the matrix, and that nickel additions contribute to this mechanism. Their work agrees that Frenkel pairs increase hardening; however, they also suggest that after annealing above 400 °C, microvoids are no longer a cause of hardening, as they disappear above 200 °C [7].

Hardness is the capability of a substance to resist permanent shape change under applied strain. Historically, it has been defined as a material's ability to resist scratching from other materials. Many different methods for measuring hardness have been developed over the years, all of which have their own intricacies [8,9]. The Vickers hardness test has the advantage of very high versatility, for example, that it can be used on both the micro and macro scale [10,11]. The Vickers hardness test was the technique employed in this research. In this paper, TRIM is used to explore the irradiation effect on high and low copper. Moreover, this program is also useful in discovering the penetration depth of proton beams on high and low copper. By finding the penetration depth of protons, a suitable load for the Vickers hardness test can be selected.

MATERIALS AND METHODS

Microhardness for high and low copper samples was measured using a Vickers hardness machine. The indent was a diamond in the form of a square-based pyramid. The employed angle of the indent was 136 °. As generally known, hardness is a measure of force per area [12,13]. Hardness can be calculated by:

$$d = \sqrt{\frac{1.8544 * F}{HV}} \tag{1}$$

Additionally, it is possible to work out the indent depth:

$$h = \frac{d}{7.0006} \tag{2}$$

where *HV* is hardness value, *d* is diameter of the indentation, *F* is load and *h* is indenter depth.

The Stopping and Range of Ions in Matter (SRIM) and the Transport of Ions in Matter (TRIM), or SRIM-TRIM, is a Monte Carlo simulation code for the measurement of the stopping and range of ions into target matter by using a full quantum mechanical treatment of ion-atom collision (assuming a transferring atom as an ion, as well as all target atoms as an atom) [14]. Furthermore, this code can be used to gain information regarding the penetration depth of the ions into the surface of materials [15].

In this work, the type of TRIM Calculation used was the Ion Distribution and Quick Calculation of Damage, which is based on Kinchin-Pease model. The ion used was Hydrogen, which can perfectly simulate the effects of a proton. One target layer with 9 different elements was used as shown in Table 1. With the intention to simulate different compositions, the low and high copper ones, the atomic percent values of the elements were input. The simulation was performed for both compositions with ions of energies of 1, 3 and 9 MeV.

Table 1. Compositions of low and high copper (in at%).

| | Element | | | | | | | | |
|-------------|---------|------|-----|-----|------|-----|-----|-----|------|
| | Fe | C | Si | Mn | P | Cr | Mo | Ni | Cu |
| Low copper | 93.2 | 1.15 | 0.4 | 1.5 | 0.01 | 0.1 | 0.3 | 3.3 | 0.04 |
| High copper | 93.2 | 0.93 | 0.4 | 1.5 | 0.01 | 0.1 | 0.3 | 3.3 | 0.26 |

The Monte Carlo code TRIM was used to gain information about the displacement per atom into the surface of materials. Displacement per atom is a strong function to measure the amount of radiation damage in irradiated materials [16]. The ion used was hydrogen with ions of energies of 1, 3 and 9 MeV primary knock-on atom (PKA) into high and low copper for 5,000 incident ions

One sample of each composition was irradiated by 3 MeV protons. The beam current was 5 μA and the samples were irradiated for approximately 2 hours. The total charge applied was 36,000 μC. The temperature was controlled by cooled water and kept at approximately 30 °C. The beam area on each sample was 25 mm². An additional irradiation was performed with the same parameters except the temperature. The temperature was kept higher, at approximately 380 °C. Unfortunately, a contamination in the vacuum system caused a black layer on the surface, which invalidated the samples damaged at temperature. A Scanning Electron Microscope (SEM) test was done to identify the elements, but regrettably it was not possible to test these two specimens in the Vickers Hardness test.

Hardness tests were performed on the high and low copper samples irradiated at 30 °C as described before. It was not possible to continue with the hardness tests because the black layer on the surface had a brittle profile. The indents were causing cracks in the material so the result was not valid because Vickers Hardness test is not supposed to be performed in brittle surfaces. Figure 1 shows the photos of the invalid specimens.



Fig. 1. A contamination in the vacuum system has invalidated specimens irradiated at 380 °C.

Different loads were used in order to reach different indent depths in the sample. Three MeV protons can reach up to approximately 40 μm in the samples. Vickers Hardness test is able to measure hardness up to 25 μm, and a good simulation of neutron damage profile can be obtained. The indent depths for different loads were approximately 3, 8, 11 and 16 μm. Indents were 0.5 mm apart from each other to avoid interference in the measurements. Moreover, measurement started and ended far away from the edges of the specimens.

RESULTS AND DISCUSSION

One of the greatest advantages of using proton beam is the easy variation of parameters. Accelerators, such as the cyclotron used in this work, offer a huge variety of irradiation environments. It is important to express the displacements in terms of ion/cm² because this value can be easily changed for different irradiation conditions. The graphs for low copper compositions are shown in Fig. 2. The results were satisfactory as the dpa (displacement per atom) behaviour is clear. For higher energies, the dose at the surface decreases because they pass through the material easily, so the scattering cross section is lower. Moreover, the results were of the same pattern for high copper.

For the TRIM calculation of proton penetration in low copper, the proton penetration depth varies strongly with its energy, and no difference was observed between low and high

copper compositions. The penetration depth was approximately 7, 38 and 234 μm respective for 1, 3 and 9 MeV protons.

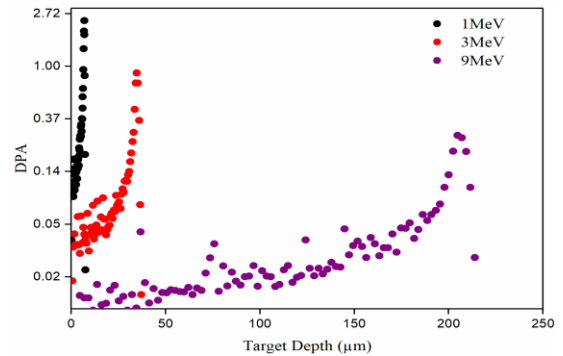


Fig. 2. Bragg peak in low copper, showing target depth change with increase displacement per atom at 1, 3 and 9 MeV proton.

Details of hardness results have made it possible to notice that it does not have the same number of points for each condition. This is because the work was held throughout the surface and the interface between non irradiated and irradiated sides was not exactly in the half of the row. Additionally, the indent depth was approximately the same for all specimens and conditions, so this value was also averaged in order to make comparisons easier. The study found that the influence of Copper in the specimen was noticeable. It was slighter than expected. The surface was considerably rough while doing the polishing was complex due to safety issues related to the residual activity of the samples. As shown in Fig. 3(c), the surface of the sample in what assumed to be the irradiated zones appears more dirty than the surface shown in Fig. 3(a) and (b).

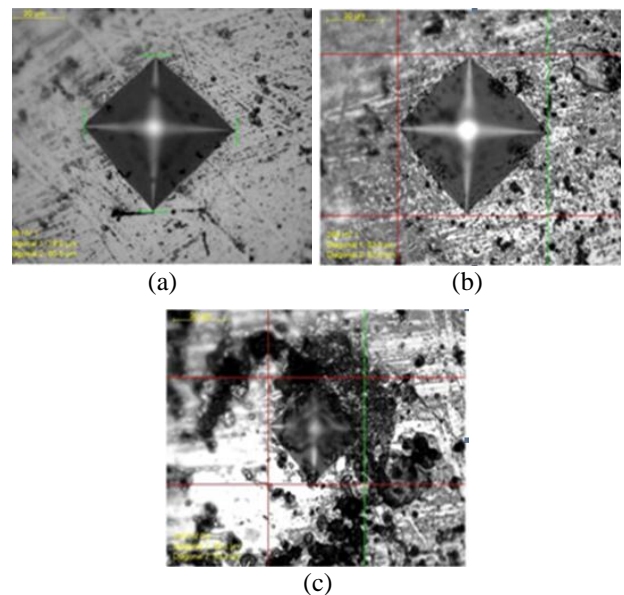


Fig. 3. Images from the high copper hardness analysis. Indent on the non-irradiated surface (a), Indent on the irradiated surface (b), Example of invalid indent (c).

Different depths on the material receive different damage accumulation due their different volumes. The deeper penetration, the greater dose is received. The hardness profile through sample thickness can be seen in Figs. 4 and 5. The values are averages for the non-irradiated and irradiated parts for both samples and the error bars express the standard deviation. Figures 4 and 5 show that irradiated values are slightly higher than non-irradiated values.

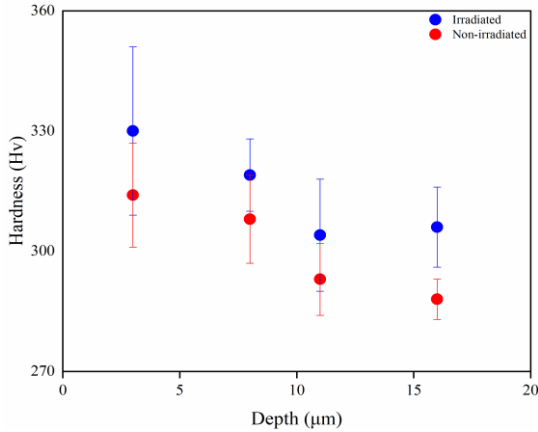


Fig. 4. Hardness versus depth for non-irradiated and irradiated low copper.

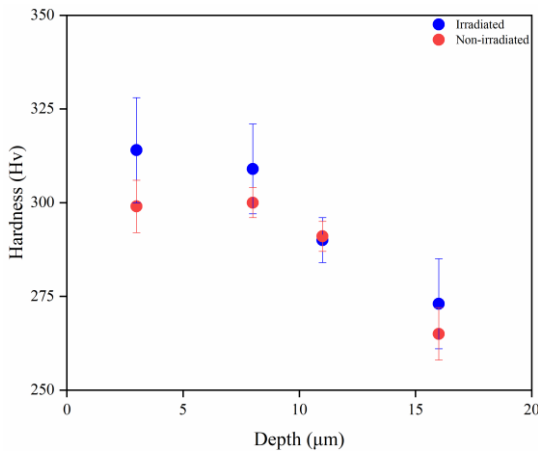


Fig. 5. Hardness versus depth for non-irradiated and irradiated high copper.

The profile was not the best expected because hardness, which is represented by depth, should increase with dose. However, it is noticeable that the standard deviation is large and ranging a great variation of values for both samples. In fact, the surfaces were not ideal to do hardness tests because they were rough.

The hardening result from proton irradiation is shown in terms of yield strength in Figs. 6 and 7. It was calculated from hardness results using the following relation [17,18]:

$$\Delta\sigma_y = 3.55 \Delta HV \quad (3)$$

where $\Delta\sigma_y$ is the increment in yield strength and ΔHV is the increment in hardness.

Was *et al.* [19] have conducted a calculation of the yield strength variation for protons and neutrons irradiations for two different types of austenitic steels and higher doses of 1-6 dpa. Their results are in excellent agreement, and the increase of yield strength with dose is clear for each experiment. The increase of yield strength with dose is anticipated. The errors bars in Figs. 6 and 7 represent the standard deviation for yield strength average values, and they cover a large variation of values.

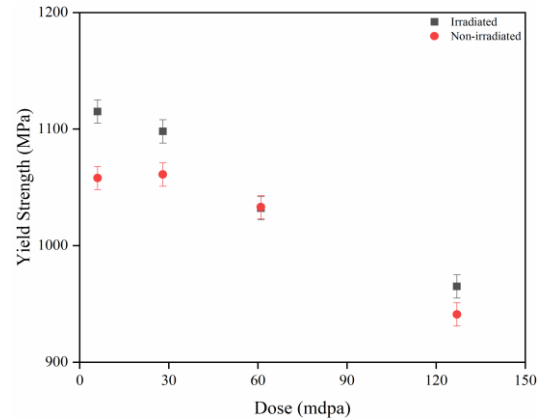


Fig. 6. Yield Strength versus Dose for non-irradiated and irradiated high copper.

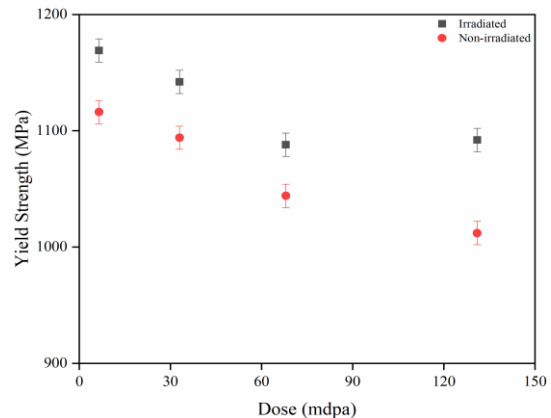


Fig. 7. Yield Strength versus Dose for non-irradiated and irradiated low copper

The yield strength variation through depth is not in great agreement with other experiments. However, the dose applied in this experiment was much lower ~150 mdpa. It is expected to have better results and cleaner profiles with higher doses. Additionally, the surfaces of the samples were not very smooth and shiny as expected for a hardness test.

CONCLUSION

Nano indentation experiments were performed to investigate the effect of ion irradiation on the hardness properties of high and low copper. Irradiated values for hardness were higher than non-irradiated ones as expected from radiation hardening. Furthermore, it was expected that high copper samples had a higher increase in hardness. This copper influence was not very perceptible in the results.

The hardness results in this work did not satisfy the predictions, where the values had great variation resulting in high standard deviations for the averages. The study found that the total dose in the first 25 microns of the flat damage profile were 0.367 dpa for low copper and 0.373 dpa for high copper samples. The hardness results appear to be clearer for higher doses, such as 1-6 dpa. Also, the surfaces were rough and not ideal for hardness tests.

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