EFFECT OF HEAT TREATMENTS ON THE RELIEF OF RESIDUAL STRESSES AND RECRYSTALLIZATION BEHAVIOUR OF SHOT-PEENED ZR-2.5NB PRESSURE TUBE MATERIAL

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ABSTRACT

The effect of heat treatment on stress relief and recrystallization behaviour of shot-peened Zr-2.5Nb pressure tube material was investigated to understand the mechanisms involved in the significant improvement in long-term corrosion and deuterium uptake resistance of this material after shot peening. Shot-peened Zr-2.5Nb material was found to recrystallize at temperatures as low as 400°C after 24 h stress relief. Residual stress measurements by X-ray diffraction technique show that the residual stress relief at 300°C is fast, and the stress relief is nearly completed in less than 5 h. It was concluded that the improvement in corrosion and deuterium uptake resistance in shot-peened Zr-2.5Nb material is not due to the presence of compressive residual stresses. The microstructural changes due to shot peening and post-peening heat treatment (the destruction and decomposition of the β-phase networks, and the fine and uniform dispersion of this phase) are responsible for the improvement in the corrosion and deuterium uptake resistance of this material.

KEYWORDS

Shot peening, corrosion, deuterium uptake, residual stress, heat treatment, stress relief, recrystallized grains, zirconium alloy

INTRODUCTION

Shot peening is being considered for use for CANDU® reactor Zr-2.5Nb pressure tubes to improve the corrosion and deuterium uptake resistance of the tubes. Previous studies have shown that shot peening produces a significant improvement in the corrosion resistance of these tubes material in long-term (≈ 700 days) out-reactor tests (1,2). A twofold increase in the deuterium uptake resistance was produced by shot peening Zr-2.5Nb material (1).

Shot peening has also been found to reduce by 25% the fraction of hydrogen absorbed in steel (3). The benefit of shot peening for steels is thought to be due to compressive surface stresses (3). If this is also the reason in the Zr-2.5Nb material, the benefit may only be short term because the residual stresses will be relieved during prolonged exposure to the reactor operating temperatures (260-300°C). Thus, we have determined whether the improvement in the deuterium uptake resistance produced by shot peening is due to the presence of compressive residual stresses, as suspected in steel.

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We have undertaken a systematic study of the effect of the temperature on the residual stresses, and of the microstructures produced by shot peening, and post-peening heat treatments in Zr-2.5Nb pressure tube material. We have evaluated the temperature dependence of the stress relief of the shot-peened Zr-2.5Nb material for temperatures from 25°C up to 500°C for 24 h and different stress relief conditions as outlined below:

(a) Stress relief at 300°C (typical reactor operating temperature) for times ranging from 5 to 50 h.
(b) Stress relief at 400°C for 24 h, which simulates the typical stress relief treatment of pressure tubes prior to installation in a CANDU reactor.
(c) Stress relief at 500°C for 3 h and 24 h to determine the extent of microstructural changes that a shot-peened Zr-2.5Nb pressure tube would undergo during a high temperature stress relief. The high temperature stress relief is used in an alternative pressure tube production route, Task Group 3 (TG3) Route 1 (4).

EXPERIMENTAL PROCEDURES

Specimens, 30 mm long (longitudinal), 12 mm wide (transverse), 4 mm thick (radial), cut from a standard CANDU reactor Zr-2.5Nb pressure tube (≈ 2.5 at.% Nb, ≈ 0.6 at.% O, and Zirconium balance) were shot peened on all six surfaces to Almen intensity A15 with 200% coverage. About 150 μm of heavily cold-worked surface layers were produced by the shot-peening process. The shot-peened specimens were individually encapsulated in a quartz tube, evacuated and sealed under 13.2 kPa of argon and then stress relieved as outlined above.

The residual stresses in as-shot-peened and the stress-relieved specimens were evaluated on the concave surface in the hoop direction, using a Rigaku D/Max-2BX X-ray diffractometer operated at 40 kV. The residual stresses were determined by the $\sin^2\psi$ (5) method, at $\psi = 0, 10, 20$ and 35°, and the percentage of stress relief defined as (6):

$$\Delta\sigma/\sigma_0 = [(\sigma_0-\sigma_r)/\sigma_0] \times 100 \qquad (1)$$

where σ_0 represents the initial residual stress in the as-shot-peened specimen, and σ_r is the residual stress in the stress-relieved specimen.

The microstructures of the surface layers of the shot-peened specimens that were heat treated at 350°C, 400°C for 24 h and at 500°C for 3 h and 24 h, were examined in transmission electron microscopy (TEM), and were compared with those of the peened and unpeened material. The TEM specimens were thinned to electron transparency using the following techniques. Slices about 200 μm thick were cut parallel to the shot-peened surface using a low-speed diamond saw. These slices containing the shot-peened layers were ground on the unpeened surface to about 125-μm thickness using 600-grit abrasive paper and then electropolished on one side (the unpeened surface) to about 5-μm thickness in a solution of 6 vol.% ethyl alcohol, which was continuously stirred and maintained at -60°C. Discs 3 mm in diameter were punched from the electropolished slices, and were electropolished from the unpeened side until perforation occurred at the shot-peened surface. Thus, microstructures of the shot-peened surface layers could be examined very close (a few microns) to the shot-peened surface.

RESULTS

Effect of temperature on the stress relief behaviour

The effects of temperature on the percentage of stress relief after 24 h, and for an initial compressive residual stress, $\sigma_0 = 300$ MPa (as-shot-peened material), is shown in Fig. 1. There are three temperature regimes for the relief of residual stresses of shot-peened Zr-2.5Nb pressure tube material. Below about 100°C, no significant stress relief takes place. Between 200 and 350°C, the amount of stress relief markedly increases with increasing temperature. Above 350°C, the compressive residual stress is completely relieved after 24 h.

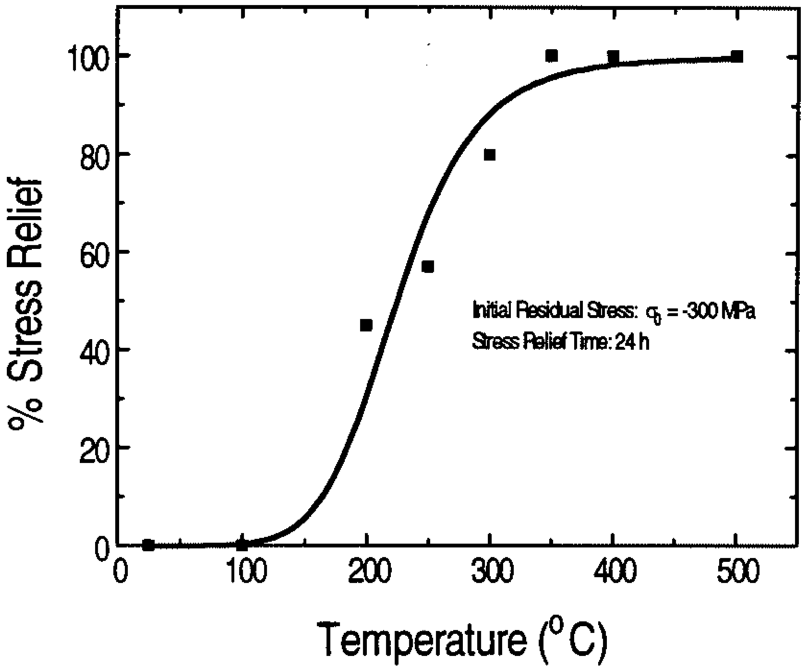


Figure 1: Effect of temperature on the percentage stress relief of shot-peened Zr-2.5Nb pressure tube material, for 24 h stress relief and an initial compressive residual stress of 300 MPa.

Residual stress relief kinetics

The variation with time of the percentage of stress relief at 300°C, for an initial compressive residual stress of 300 MPa is shown in Fig. 2. The stress relief is very fast. Over 85% of the residual stress is relieved in the first 5 h, and no significant stress relief occurs after this initial 5 h. The stress relief saturates with no further stress relief up to 50 h.

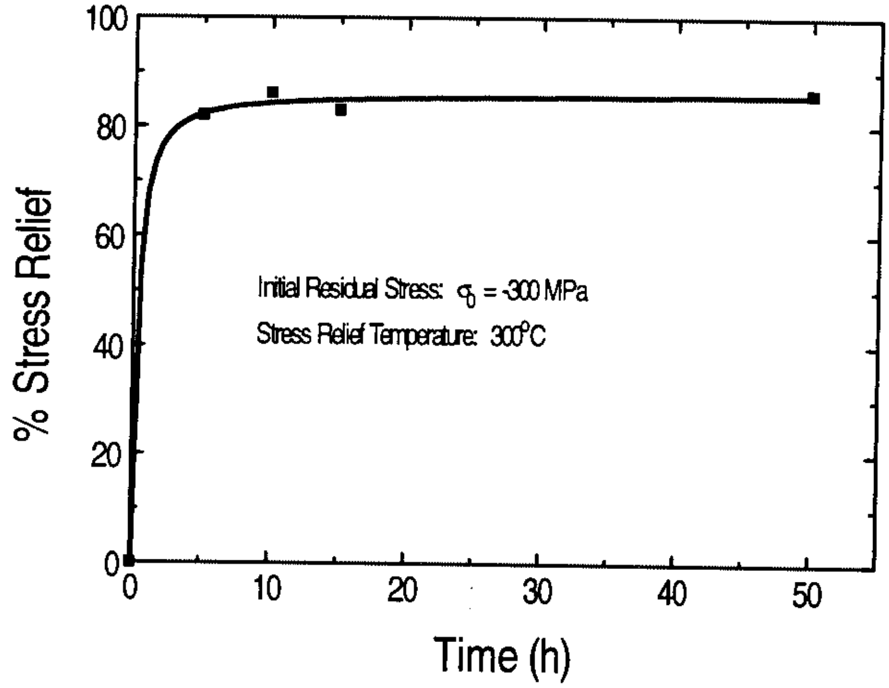
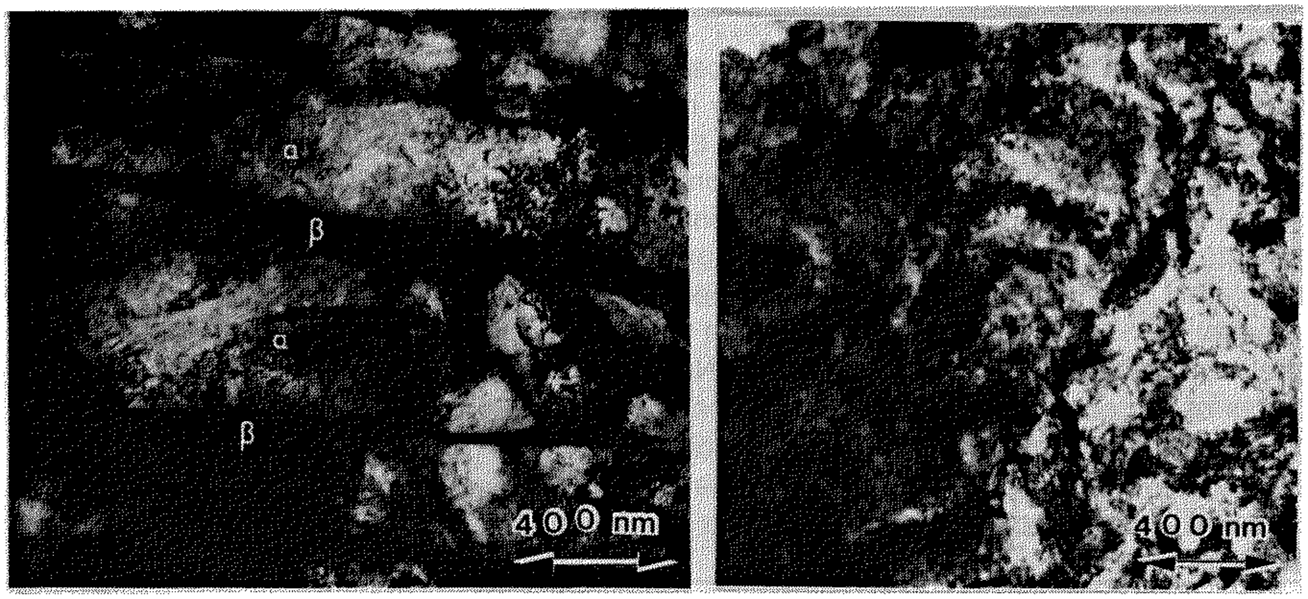


Figure 2: Variation with time of the percentage residual stress relief at 300°C for an initial compressive residual stress of 300 MPa.

Microstructural examinations and recrystallization behaviour

The pressure tube material has an initial microstructure that consists of elongated α-zirconium grains surrounded by a network of a continuous β-phase (Fig. 3(a)). Fig. 3(b) shows the microstructure of the surface layers of Zr-2.5Nb pressure tube material after shot peening. Shot peening has heavily cold worked the surface layer and has completely obliterated the original grain structure. Heat treating shot-peened Zr-2.5Nb pressure tube material at 350°C for 24 h did not produce any recrystallization. The microstructure of a shot-peened Zr-2.5Nb pressure tube material that has been stress-relieved for 24 h at 400°C is shown in Fig. 4. This figure shows a partially recrystallized structure. Thus in shot-peened Zr-2.5Nb material, recrystallization occurs at temperatures as low as 400°C after 24 h heat treatment. In contrast, in unpeened Zr-2.5Nb pressure tube material with about 30% cold work, no recrystallization was observed after 24 h heat treatment at 500°C.

The recrystallization process in shot-peened material is very fast at 500°C and was complete in 3 h (Fig. 5(a)). The recrystallization is accompanied by the formation of β-phase particles that are uniformly distributed within the recrystallized grains. Growth of the recrystallized α-zirconium grains and of the β-phase particles occurred after 24 h stress relief at 500° C (Fig. 5(b)).



(a) (b)

Figure 3: Transmission electron micrographs showing (a) elongated α-zirconium grains surrounded by a network of continuous β-phase in as-received (30% cold drawn) Zr-2.5Nb material and (b) heavily cold-worked structure in shot-peened Zr-2.5Nb material.

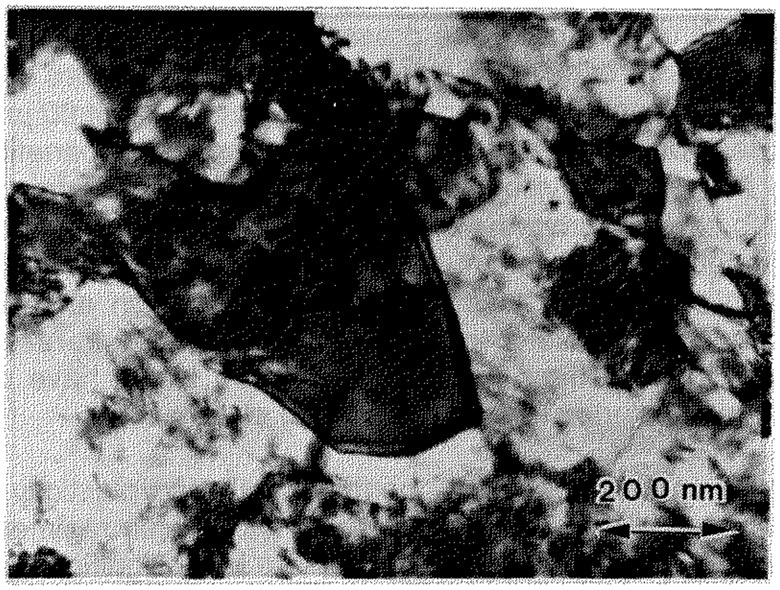


Figure 4: Transmission electron micrograph showing partially recrystallized grains in shot-peened Zr-2.5Nb material after 24 h stress relief at 400°C.

DISCUSSION OF RESULTS

The benefit of shot peening could not be due to the effect of the residual stresses because the residual stresses are quickly relieved. The stress relief at 300 and 350°C was found to be fast, with about 85% of the residual stress removed within the initial 5 h at 300°C, and the residual stress completely relieved at 350°C and 400°C after 24 h. Thus, the prefilming treatment in an autoclave (400°C for 24 h) that the specimens underwent prior to long-term corrosion testing at 300 and 350°C would have completely relieved the compressive residual stresses produced by the shot-peening process. These observations indicate that the improvement in corrosion and deuterium uptake resistance of shot-peened Zr-2.5Nb pressure tube material is not due to the effects of the compressive residual stresses, as suggested for steel. In previous work (1), the drastic microstructural changes produced by shot peening or post-peening heat treatment were suggested as one of the possible reasons for the improvement in corrosion and deuterium uptake resistance due to shot peening.

The microstructure of Zr-2.5Nb pressure tube material, particularly the grain-boundary β-phase network plays a crucial role in the corrosion and deuterium uptake behaviour of Zr-2.5Nb pressure tube material. It has recently been shown that the presence of a continuous β-phase network at the grain boundary significantly decreases the corrosion resistance of Zr-2.5Nb pressure tube material (7). Thus, shot peening, or post-peening heat treatment that breaks up or decomposes the grain boundary β-phase network should improve the corrosion and deuterium uptake resistance of Zr-2.5Nb pressure tube material (8). The results of the present study would also indicate that the observed improvement in corrosion and deuterium uptake resistance due to shot peening should persist at the reactor operating temperatures, as long as shot-peened layers are present on the surface of the pressure tube (if shot-peened layers are not consumed by the corrosion reaction). Since the maximum thickness of oxide formed on pressure tube during the service life of the pressure tube is about 60 μm, which is much less than the thickness of the peened layers, we can expect the benefit of shot peening to last the 30 to 40 years service life of the pressure tube. Irradiation-enhanced precipitation of β-Nb particles from the α-phase during reactor operation is also known to affect the corrosion and deuterium pickup behaviour of Zr-2.5Nb pressure tube material. The effect of such a precipitation on the corrosion and deuterium uptake behaviour of shot-peened Zr-2.5Nb material is not known. In-reactor tests are in progress to determine what effect in-reactor irradiation would have on the corrosion and deuterium pickup resistance of the shot-peened material.

The mechanisms of stress relief involve thermally activated plasticity processes such as creep. The results of the residual stress relief show three regimes of temperature dependence. At low temperature, below 100°C, there is no significant stress relief after 24 h heat treatment. Between 100 and 350°C, the stress relief process shows a strong temperature dependence, and may be controlled by thermally activated plasticity processes such as dislocation glide, climb, and cross slip observed during a recovery process (9) or creep. The saturation in the percentage stress relief in this temperature regime would indicate that the maximum annealing time of 50 h is not sufficiently long for the stress-relief process to be completed. The stress relief appears to proceed at

(a) (b)

Figure 5: Transmission electron micrographs showing (a) fine equiaxed grains in shot-peened Zr-2.5Nb material stress relieved at 500°C for 3 h and (b) large recrystallized grains with uniformly distributed β-phase particles in shot-peened Zr-2.5Nb material stress relieved at 500°C for 24 h.

very low rate after the initial rapid increase. Above 350°C, the stress-relief process is completed in 24 h, because the temperature is sufficiently high for the stress-relief process to proceed quickly. Recrystallization starts at a temperature as low as 400°C. The relatively low recrystallization temperature of the shot-peened pressure tube material can be explained by the intense cold working produced by the shot-peening process (1), which has introduced in the material large numbers of lattice defects such as dislocations. The presence of large numbers of dislocations would facilitate the formation of recrystallization nuclei at relatively low temperatures. The recrystallization temperature is known to decrease as the amount of prior deformation increases (10).

CONCLUSIONS

The residual stress relief in shot-peened Zr-2.5Nb pressure tube specimens is found to be very fast at 300°C. Nearly 85% of the residual stress is removed in the initial 5 h at 300°C. The stress relief is completed at 350°C, and recrystallization starts at about 400°C after 24 h stress relief. The improvement in corrosion and deuterium uptake resistance of Zr-2.5Nb pressure tube material observed in long-term tests at 300 and 350°C is not due to the residual stresses produced by the shot-peening process. Microstructural changes produced by shot-peening or post-peening heat treatment are believed to be responsible for the improvement in corrosion and deuterium uptake resistance. Thus, the benefit of shot peening is expected to last at the CANDU reactor operating temperatures.

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REFERENCES

1. Amouzouvi, K.F., Clegg, L.J., Styles, R.C., Winegar, J.E., in Computer Methods and Experimental Measurements for Surface Treatment Effects, Computational Mechanics Publications, Southampton, UK, Aliabadi, M.H. and Brebbia, C.A., Editors, 145-154, 1993.
2. Amouzouvi, K.F., Mechanical Properties of Thin Zirconia Corrosion Films Thermally Grown on Zr-2.5Nb Materials, Scripta Met. et. Mater., 30, 1139-1143, 1994.
3. Wilde, B.E. and Shimada, T., Surface Modification: A Potential New Approach to Combating Hydrogen Induced Fracture in Steel, Scripta Met., 22, 551-556, 1988.
4. Fleck, R.G., Price, E.G., and Cheadle, B.A., in Proc. of the 6th International Symposium on Zirconium in the Nuclear Industry, ASTM STP 824, 88-105, 1984.
5. Klug, H.P. and Alexander, L.E., Editors, X-ray Diffraction Procedures for Polycrystalline and Amorphous Materials, John Wiley & Sons, New York, USA, 1974.
6. Fox, A., in Residual Stress and Stress Relaxation, Sagamore Army Materials Research Conference Proceedings, Kula, E. and Weiss, V., Editors, Plenum Press, New York, USA, 181-203, 1982.
7. Ding, Y. and Northwood, D.O., SEM Examination of the Oxide-Metal Interface Formed during Aqueous Corrosion of a Zr-2.5Nb Alloy. Journal of Materials Sciences 27, 1045-1052, 1992.
8. Urbanic, V.F., Warr, B.D., Manolescu, A., Chow, C.K. and Shanahan, M., in Zirconium in the Nuclear Industry, Proceedings of the Eighth International Symposium, ASTM STP 1023, 20-35, 1989.
9. Reed-Hill, R.E., Physical Metallurgy Principles, D. Van Nostrand Company, Inc., Princeton, USA, 1964.
10. Douglass, D.L., The Metallurgy of Zirconium, International Atomic Energy Agency, Supplement 1971, Vienna, 1971.