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Effect of shot peening and post-peening heat treatments on the microstructure, the residual stresses and hardness, corrosion and deuterium uptake resistance of Zr-2.5Nb pressure tube material

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ABSTRACT

The effect of shot peening and post-peening heat treatment on the microstructure, the residual stresses, hardness, corrosion and deuterium uptake behaviour of Zr-2.5Nb pressure tube material has been evaluated. Shot peening produced a heavily cold-worked microstructure, with the original grain structure completely obliterated by the shot-peening process. Shot peening also caused a change in the texture of this material. Heat treating shot-peened specimens at temperatures from 670 to 770 K caused recrystallization of the cold-worked surface layers and breakup of the grain-boundary β-phase network. The recrystallization is accompanied by precipitation within the α-Zr grains of fine uniformly distributed Nb-rich β-phase particles and the reduction in the Nb supersaturation of the α-Zr phase from $\simeq$1.5 to $\simeq$0.5 at.% Nb. Shot peening produced an improvement in the corrosion and deuterium uptake resistance of Zr-2.5Nb pressure tube material. The improved corrosion and deuterium uptake resistance was attributed to the intense cold work and to microstructural changes produced by shot peening and post-peening heat treatments.

INTRODUCTION

Zirconium alloy (Zr-2.5Nb) pressure tubes in CANDU (CANada Deuterium Uranium) reactors contain the fuel bundles and the pressurized heavy-water coolant, and operate at $\simeq$570 K. The pressure tubes pick up deuterium as a result of corrosion by the heavy-water coolant during reactor operation. This deuterium pickup is a concern because, if the terminal solid solubility (TSS) limit for hydrogen isotopes in the Zr-2.5Nb alloy at reactor operating temperatures is exceeded within the lifetime of the pressure tube, delayed hydride cracking (DHC) may occur.

The occurrence of DHC may be reduced or eliminated by cold working the surface of the pressure tube to reduce the rate of corrosion and the related deuterium uptake. Cold working Zr-Nb-based alloys has been found to improve its corrosion resistance e.g., Bertea et al. [1]. Improvement in the corrosion resistance and the reduction in the hydrogen pickup rate depended on the extent of cold reduction. Increasing the amount of cold reduction from 0 to ≃80% resulted in a twofold improvement in the corrosion and hydrogen pickup resistance of Zr-2.5Nb-based alloys, e.g., Bertea et al. [1]. By inducing large amounts of cold work in the surface layers, shot peening of pressure tubes should produce similar reductions in corrosion and deuterium pickup rates without affecting the bulk properties.

The present investigation studied the effect of shot peening and post-peening heat treatment on the microstructure, corrosion, and deuterium uptake resistance of Zr-2.5Nb pressure tube specimens. The microstructure and properties of surface layers of shot peened, and shot peened and heat-treated Zr-2.5Nb pressure tube specimens were characterized. The effect of shot peening and post-peening heat treatment on aqueous corrosion and deuterium pickup behaviour of Zr-2.5Nb pressure tube material was also studied.

EXPERIMENTAL PROCEDURES

Shot peening of cold-drawn (30%) Zr-2.5Nb pressure tube specimens was carried out using commercial equipment and procedures. The specimens were shot peened with a coverage greater than 100% at Almen intensities of N5, A8, A11, A16 and C7. Coupons, 25.4 mm x 12.5 mm x 4 mm, were shot peened on all six faces for corrosion and deuterium uptake measurements. Some of the specimens and coupons were heat-treated at 670 K for 24 h, 750 K for 6 h or 770 K for 3 or 24 h to investigate the effect of post-peening heat treatments on the microstructure, the corrosion, and the deuterium uptake. The shot-peened and shot-peened and heat-treated corrosion coupons were electropolished or chemically polished before being corrosion-tested at 570 K or 625 K in pressurized light or heavy water. As-received and ground (400 grit) Zr-2.5Nb pressure tube coupons were also corrosion-tested at 570 K for comparison. The corrosion behaviour of these specimens was characterized by weight-gain measurements (gravimetry) or measurements of oxide thickness on the axial-transverse plane of the corrosion coupon using a Fourier transform infrared reflection (FTIR) interferometer, e.g., Ramasubramanian and Ling [2]. The microstructure of shot-peened and shot-peened and heat-treated Zr-2.5Nb pressure tube specimens was characterized by transmission electron microscopy (TEM). Energy dispersive X-ray (EDX) microanalysis was used to determine the Nb concentration in α-Zr grains after heat treatment and post-peening heat treatment. The texture of the surface-treated layers was measured by the inverse pole figure technique using X-ray diffraction. The X-ray diffraction technique was also used to measure the residual stresses induced by the shot peening process. The residual stresses were measured at the surface of the shot-peened specimens (peened at Almen intensities A11, A16 and C7) and at several depths from the peened surface after chemically removing layers of metal and examining the X-ray peaks from (21$\bar{3}$1) and (11$\bar{2}$4) planes, each of which had been calibrated for stress measurement, e.g., Winegar [3]. Thus, profiles showing a variation of residual stress with depth from the surface have been obtained. The hardness of the surface layers of the shot-peened specimens was characterized using microhardness tests.

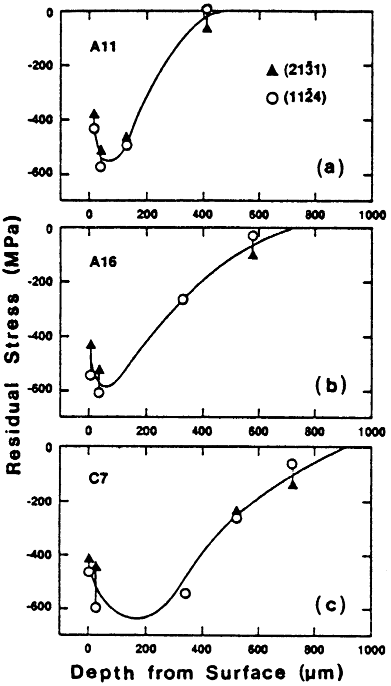


Fig. 1 Residual stress distribution in Zr-2.5Nb pressure tube specimens shot peened at Almen intensities (a) A11, (b) A16, (c) C7.

RESULTS

Figure 1 shows the variation with depth from the peened surface of the induced residual stresses in Zr-2.5Nb pressure tube specimens shot peened at A11, A16 and C7. The maximum compressive residual stresses were approximately 560, 587 and 633 MPa for Almen intensities A11, A16 and C7 respectively. These results showed that increasing the shot-peening intensity slightly increases the induced maximum compressive residual stresses. The depth of the compressive residual stresses, in contrast, significantly increases from approximately 400 to 900 μm as the peening intensity increases from Almen intensities A11 to C7. The effect of shot-peening intensity on the depth of the cold-worked layers, as determined by microhardness measurements, is shown in Figure 2 for three different Almen intensities: A8, A11 and C7. The depth of cold-worked layers increases as the peening intensity increases from A8 to C7, similar to the variation of compressive stresses with depth from surface (Figure 1). However, the depth of cold-worked layers is shallower than that of the compressive residual stresses. Shot peening produced an approximately 30% increase in hardness, compared with the as-received material.

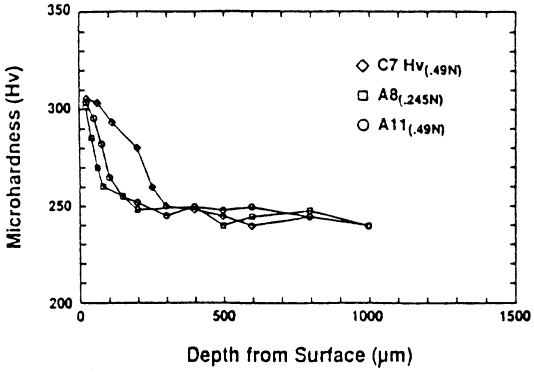
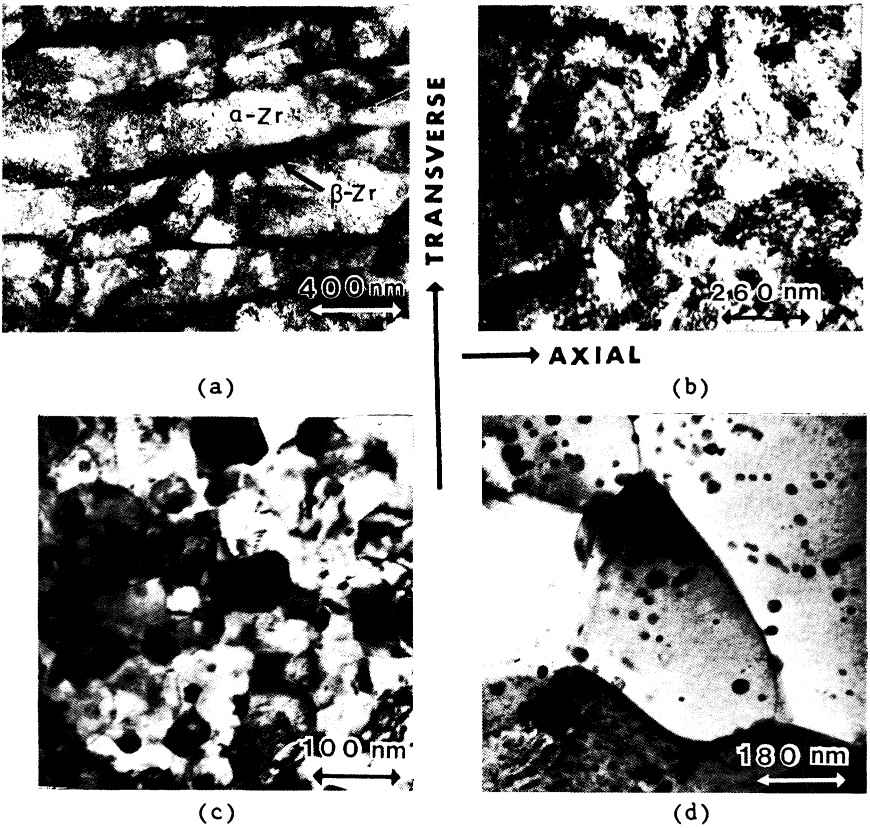


Fig. 2 Microhardness profiles of shot-peened Zr-2.5Nb pressure tube specimens obtained using loads of 25 g (0.245 N) and 50 g (0.49 N).



(a)
(b)
(c)
(d)

Fig. 3. TEM micrographs showing the microstructure of Zr-2.5Nb pressure tube specimens (a) as-received, (b) shot-peened, (c) shot-peened and heat-treated at 770 K for 3 h, (d) shot-peened and heat-treated at 770 K for 24 h.

The TEM examination of surface layers of shot-peened Zr-2.5Nb specimens revealed a heavily cold-worked structure with the original grain structure (Figure 3(a)) completely obliterated by the shot-peening process (Figure 3(b)). Heat treatment of the shot-peened specimens at 670 K for 24 h produced a partial recrystallization of the cold-worked structure with evidence of breakup of the Nb-rich β-phase grain boundary network in the bulk. Heat treating the shot-peened specimens at 750 K for 6 h or 770 K for 3 to 24 h resulted in a complete recrystallization of the shot-peened structure (Figure 3(c) and 3(d)). Recrystallized α-Zr grains with average grain size ranging from 20 nm to 2 μm in size were observed in Figures 3(c) and 3(d), along with a uniform dispersion of fine Nb-rich (> 60 at.% Nb) β-phase particles. The size of the recrystallized α-Zr grains and the density and size of the precipitates were found to depend on the heat-treatment temperature, the annealing time and the depth from the shot-peened surface. For a given annealing temperature and time, the finest α-Zr grains were observed in the near-surface layers. EDX microanalysis showed that the Nb concentration in the α-Zr phase was reduced from between 1.3 and 1.5 at.% to a concentration of 0.5 at.% Nb, whereas no significant reduction in Nb concentration in the α-phase of the as-received material took place after heat treatment at 770 K for up to 72 h. Crystallographic textures of the shot-peened surface layers measured by the inverse pole figure technique showed that the peening increased the average resolved fraction (F_R) of basal plane normals in the radial direction from 0.31 to 0.44. Heat treating shot-peened Zr-2.5Nb pressure tube material at 670 K for 72 h did not produce a significant change in texture, whereas a slight decrease in F_R occurred after heat treatment at 750 K for 6 h. Heat treating shot-peened specimens at 770 K for 24 h resulted in a texture with $F_R = 0.33$.

The effect of various surface treatments on the corrosion behaviour (characterized by weight-gain measurements) of Zr-2.5Nb pressure tube specimens is shown in Figure 4. The shot-peened and electropolished specimens exhibit 20 to 30% lower weight gain than ground and electropolished Zr-2.5Nb pressure tube specimens. Figure 4 also shows that the weight gains of the as-received, and the as-received and ground specimens are 1.5 to 2 times higher than that of shot-peened or electropolished specimens. The effect of shot peening and post-peening heat treatments on the thickness of the oxide films grown on Zr-2.5Nb pressure tube specimens after exposure for 17 and 41 d in pressurized water with pH of 10.5 is shown in Table 1. The experimental data in Table 1 represent an average of 3 measurements. These data also show that shot peening improves the corrosion resistance of Zr-2.5Nb pressure tube specimens. Heat treating at 770 K for 24 h produced a 10% improvement in the corrosion resistance of as-received specimens and 30 to 50% improvement in the corrosion resistance of shot-peened specimens. The results of deuterium uptake measurements on duplicate pickled specimens of as-received shot-peened, shot-peened and heat-treated Zr-2.5Nb pressure tube material, exposed for 268 d at 625 K in pressurized D_2O with

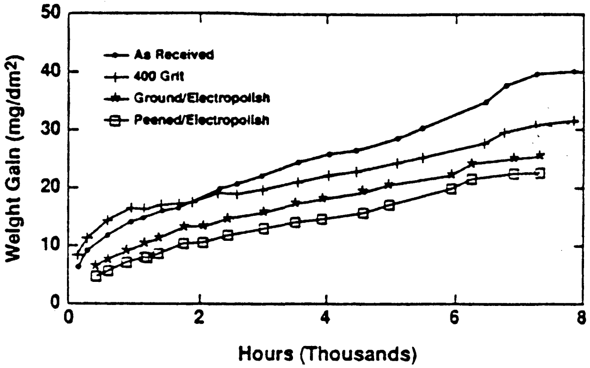


Fig. 4. Effect of different surface treatments on corrosion behaviour of Zr-2.5Nb pressure tube specimens at 570 K in pressurized water with pH of 10.5.

TABLE 1
Short-term Corrosion Behaviour of Zr-2.5Nb Pressure Tube Specimens Exposed at 570 K for 17 and 41 d in Pressurized Water with pH of 10.5

Material Conditions	Oxide Thickness(μm) After 17 d	41 d
As-received (cold drawn)	1.20	1.50
As-received, ground and electropolished	0.90	1.15
Shot-peened and electropolished	0.70	0.90
As-received, heat-treated at 770 K for 24 h, electropolished	0.80	1.05
Shot-peened, heat-treated at 670 K for 72 h, electropolished	0.60	0.75
Shot-peened, heat-treated at 770 K for 24 h, electropolished	0.50	0.66

TABLE 2
Deuterium Uptake Behaviour of As-received and Shot-peened Zr-2.5Nb Pressure Tube Specimens Exposed at 625 K for 268 d in Pressurized D_2O with pH of 10.5

Material Conditions	Deuterium Uptake at.% (ppm)	Oxide Thickness (μm)
As-received (cold drawn), pickled	0.055 (11.7)	5.6
Shot-peened, pickled	0.027 (5.8)	4.1
Shot-peened, heat-treated at 770 K for 24 h, pickled	0.025 (5.1)	2.9

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pH ≃ 10.5, are shown in Table 2. These results show that shot peening has reduced the deuterium uptake of Zr-2.5Nb pressure tube specimens by a factor of two, even though FTIR oxide thickness measurements showed only ≃30% improvement in the oxidation resistance.

DISCUSSION

The results of this study indicate that the depth of induced cold-work in shot-peened Zr-2.5Nb materials is significantly lower than the depth of compressive residual stresses. The reason why the depth of the compressive residual stresses is significantly higher than that of the cold-worked layers is not understood. TEM examinations showed that shot peening has produced sufficient plastic deformation to obliterate the grain structure of the as-received Zr-2.5Nb pressure tube material by mechanically breaking up the β-phase grain boundary networks. Shot-peened surface layers were found to recrystallize readily at temperatures as low as 670 K, whereas in unpeened as-received material, with ≃30% cold work, recrystallization did not occur until the monotectoid temperature (880 K) was reached. The temperature at which recrystallization begins is a function of the amount of cold work, the purity of the metal and time. In general, for a given composition and annealing time, the recrystallization temperature decreases with increasing amount of cold work. The minimum recrystallization temperature of a zirconium alloy containing 2.4 wt.% hafnium, 0.02 wt.% oxygen and 0.07 wt.% iron and with 96% cold work is 670 K, e.g., Kaufmann and Magel [4]. Thus, the propensity of shot-peened specimens to recrystallize at such low temperatures suggests that shot peening has introduced a very high amount of cold work in the surface layers. This cold work is estimated to range between 90 to 95%, e.g., Kaufmann and Magel [4], DeSalvo and Zignani [5], Douglass [6], on the basis of the recrystallization behaviour of the shot-peened specimens. The extremely fine recrystallized grains, 20 to 100 nm in size shown in Figure 3(c), are consistent with this estimated high amount of cold work produced by the shot-peening process.

The texture produced in as-shot-peened specimens could improve the DHC behaviour of shot-peened pressure tubes, since most grains would be oriented with their basal plane normals perpendicular to the hoop stress, a condition which generally produces the maximum DHC resistance, e.g., Coleman et al. [7]. The change in texture on heat treating for 24 h at temperatures as low as 770 K, was unexpected. In a previous work, Cheadle and Ells [8] reported that no change in texture took place when Zr-2.5Nb pressure tube material was annealed for 100 h at 770 K. In zirconium, recrystallization does not significantly change the texture, e.g., Douglass [6].

From EDX analyses, which revealed a significant reduction in Nb concentration in solid solution, we conclude that some of the uniformly distributed Nb-rich particles in Figure 3(d) had precipitated from solid solution during heat treatment. Air cooling from the extrusion temperature (1090 to 1130 K) to room temperature during the manufacturing of the pressure tube produces some Nb supersaturation of the α-Zr phase. The very large cold work produced by the shot peening is believed to be the main contributing factor to the precipitation because no significant reduction in Nb concentration was measured in the as-received material (non-shot-peened) after heat treatment at 770 K for up to 72 h. Previous work, e.g., Lesurf [9], showed that cold working a quenched Zr-2.5Nb alloy prior to heat treatment at 770 K for 24 h enhanced the precipitation from supersaturated solid solution. The large deformation induced by the shot peening introduced into the material a very high number of dislocations and vacancies. Since dislocations act as precipitation nuclei and diffusion is promoted by vacancies, e.g., Christian [10], precipitation and redistribution of Nb-rich particles occurred faster in shot-peened material than in as-received material. Thus, shot peening has accelerated the kinetics of recrystallization, precipitation and redistribution of Nb-rich particles.

The improved corrosion and deuterium uptake resistance of shot-peened and chemically-polished or electropolished Zr-2.5Nb pressure tube specimens, compared with unpeened and chemically-polished Zr-2.5Nb pressure tube specimens, is attributed to the intense cold work produced by the shot-peening process. The beneficial effect of the intense cold work on the corrosion behaviour of Zr-2.5Nb pressure tube material can be explained by the work hardening of a surface layer by the shot-peening process. Assuming that thermally grown oxide films on zirconium alloys are stabilized by the compressive growth stress, which should be directly proportional to the strength of the underlying material, Cox [11] predicted that an increase in strength of the alloy should result in less cracking in the oxide and thereby make it more protective. Measurements of mechanical properties of oxide layers and scanning electron microscopy (SEM) examinations, e.g., Amouzouvi and Clegg [12] showed that shot-peened specimens developed fewer cracks than unpeened specimens, hence confirming the above hypothesis. Another possible beneficial effect of the intense cold work is the complete obliteration of the original β-phase grain-boundary network, by shot peening. The β-phase was previously shown to exhibit a higher oxidation rate than the α-Zr phase, e.g., Warr et al. [13] possibly because of the higher rate of oxygen diffusion in this phase than in the α-phase. The local difference in oxidation rate between the β- and α-Zr phases was found to result in development of holes and pores in the oxide film, rendering the oxide film non-uniform and less protective, e.g., Warr et al. [13]. Breaking up the grain-boundary β-phase network by shot peening or heat treatment produced an oxide film, which is more uniform, less cracked and more protective. Additional improvement in corrosion resistance was achieved by heat treatment of shot-peened Zr-2.5Nb specimens. Such additional improvement was ascribed to the breakup of the grain-boundary β-phase network, e.g., Urbanic et al. [14], and to the reduction of Nb concentration in the α-phase from a supersaturated solid solution due to precipitation of Nb-rich β-phase particles, e.g., Lesurf [9], Urbanic et al. [15]. Shirvington [16] suggests that a reduction of Nb in solid solution would result in a lower concentration of Nb in the oxide, which would decrease the electronic and ionic conductivity of the oxide, hence producing a lower oxide growth rate.

CONCLUSIONS

Shot peening Zr-2.5Nb pressure tube material produced a heavily cold-worked surface layers in which the original grain structure was completely obliterated. Shot peening also changed the texture of the surface layers of the Zr-2.5Nb pressure tube material. Heat treating shot-peened Zr-2.5Nb pressure tube material caused recrystallization of the cold-worked surface layer and precipitation of Nb-rich β-phase particles from the Nb supersaturated α-Zr phase. Shot peening improved the corrosion and deuterium uptake resistance of Zr-2.5Nb pressure tube. This improvement is attributed to strain-hardening and to microstructural changes produced by the shot peening and post-peening heat treatments.

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