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Partitioning of tritium between surface and bulk of 316 stainless steel at room temperature

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ABSTRACT

The distribution of tritium between the near surface and the bulk of 316 stainless steel has been measured using two independent techniques: pulsed-plasma exposures and a zinc-chloride wash. Between 17% and 20% of the total inventory absorbed into a stainless-steel sample during a 24-h exposure to DT gas at room temperature resides in the water layers present on the metal surface. Redistribution of tritium between the surface and the bulk of stainless steel, if it occurs, is very slow. Tritium does not appear to enter into the bulk at a rate defined solely by lattice diffusivity.

1. Introduction

The interaction of tritium with the native oxides on the surface of stainless steel is the first step toward adsorption and absorption of tritium into the bulk stainless-steel lattice. Understanding this fundamental interaction is necessary for the development of surfaces that minimize tritium absorption and tritium permeation through piping materials in nuclear reactors, which mitigate the radiological hazards associated with tritium contamination of metal tools and structures. Much work has been done to measure the distribution of tritium within stainless-steel samples [1–7] and to test the influence of different surface modifications on the absorption and permeation of tritium through stainless steel [8–15]. Several conclusions can already be drawn from this body of work. Firstly, the surface comprising < 15-nm and near surface comprising < 0.1-µm layers of tritium-loaded stainless-steel samples contain relatively large fractions of the total tritium inventory [4]. These large surface concentrations are likely caused by tritium dissolved within the ubiquitous adsorbed water layers on stainless-steel surfaces [16–19]. Secondly, the relatively large surface concentrations are strongly influenced by the condition of the metal surface [19]. Finally, modifying the metal surface can significantly alter the permeation [8,9,12,14] and absorption [13,15] of tritium into the substrate metal, an effect likely caused by a local equilibrium established between the tritium concentrations in the adsorbed water layers and the bulk metal lattice [20].

Here, we present new data about the tritium partition between the adsorbed water layers and the bulk metal lattice. This distribution was measured using two different techniques: a low-temperature pulsed plasma [20] and an aqueous zinc chloride method adapted from Tanaka et al. [21]. Both methods remove surface-adsorbed tritium. Thermal desorption was also used to measure tritium dissolved in the bulk metal lattice. The pulsed-plasma and zinc-chloride methods allow for the removal of the adsorbed water layers without etching into the substrate metal lattice. The pulsed-plasma method accomplishes this by bombarding the surface with energetic ions. The zinc-chloride method removes the adsorbed water layers by binding the zinc-chloride complex to the oxygen atoms in the hydroxyl layer. This hydroxyl layer is directly bound to the underlying native metal oxide that forms naturally on exposure to ambient air. By binding the zinc-chloride complex to the surface, the multilayer structure of adsorbed water is expected to be liberated into the solution along with any tritium contained therein. Both surface-removal techniques allow for the measurement of tritium adsorbed solely within the adsorbed water layers and at a finer resolution than has been reported to date.

2. Experimental setups and procedures

Samples of dimensions $5.1 \times 1.9 \times 0.3\,\text{cm}^3$ were cut from a common plate of stainless steel type 316. The total geometric surface area of each sample is $23.5\,\text{cm}^2$. An ~0.86-mm-deep surface layer was machined away to eliminate any surface inclusions produced in the manufacturing process and to expose the base metal lattice. The machining process produces surface striations along the machining path axis, which adds to the overall surface roughness as discussed elsewhere [15].

Two surface treatments were used to probe the relationship between

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surface finish and tritium uptake compared to unmodified surfaces. Samples that did not receive any pre-treatments served as benchmarks and were labelled “as received.” Ten as-received samples were baked for 2 h in dry helium at 200 °C in a dry box to remove physisorbed water from the surface. These samples were labelled “as-received baked.” Another subset of as-received samples was mechanically polished using 3-μm MetaDi Mono Suspension diamond paste with a low-nap cloth wheel, followed by final polish with 0.3-μm α-alumina provided by a commercial vendor. These samples are labelled as “polished.” The surface roughness of the samples was measured with a Zygo NEXview interferometer. The average surface roughness for an as-received sample was 0.27 ± 0.06 μm. Polished samples showed an average surface roughness of 0.02 ± 0.01 μm.

All samples were placed in an ultrasonic bath and subjected to three washes: first with acetone to degrease the surfaces, then with isopropyl alcohol, and finally with de-ionized water to remove any chemical residue. The samples were then transferred to a glovebox and stored under dry helium at a dew point of −65 °C. After drying in the glovebox, the samples were loaded with tritium by exposure to a 1-atm mixture of DT gas with an isotopic ratio comprising 70% tritium and 30% deuterium. The tritium partial pressure was 0.54 atm. The samples were soaked in the DT gas mixture for 24 h at room temperature. Under these exposure conditions the diffusivity of tritium through stainless steel is 3.76 × 10–16 m^2/s at room temperature [20] and the tritium concentration within the bulk is not expected to be at equilibrium. After this loading, each sample was placed in an individual pod of a storage rack and sealed under dry helium to provide leak-tight storage. The storage racks were placed in a transfer box filled with a dry nitrogen atmosphere to minimize air exposure during the transfer from the storage rack to the experimental setup. All samples were stored in the racks until retrieved for an experiment.

The samples were subjected to one of three treatments: thermal desorption, plasma irradiation, or zinc-chloride washes. In the first treatment, a sample was subjected to temperature-programmed thermal desorption (TPD) to release and measure the total quantity of tritium retained by the metal sample. Tritium release was measured by scintillation counting of the activity collected with bubblers using a Perkin Elmer Tri-Carb 2910 TR liquid scintillation counter. In the second treatment, a sample was exposed to a room-temperature, Tonks–Langmuir, pulsed, radio-frequency–driven plasma to release and measure the quantity of tritium bound on and in the near-surface. Tritium release was measured with an in-line ionization chamber. The pulse duration was 2 s followed by a 20-min dwell between each pulse. After the pulsed-plasma treatment, the sample was placed in the thermal desorption setup to measure the residual tritium in the bulk of the metal. Both of these techniques are described in detail elsewhere [20,22]; however, some improvements have been installed recently. The carrier gas used in the TPD procedure has been upgraded to ultrahigh-purity argon and a load-lock system is used to transfer samples from storage into the pulsed-plasma chamber to minimize exposing samples to air during the transfer.

In the third treatment, samples were washed in an aqueous zinc-chloride solution to remove adsorbed tritium following a method adapted from Tanaka et al. [21]. In this case, a sample was removed from storage under argon and soaked in a beaker containing 25 mL of 4 M ammonium chloride, 12.5 mL of 0.4 M zinc chloride, and 12.5 mL of de-ionized water with the pH adjusted to 7 using concentrated ammonium hydroxide. The solution was stirred for 5 min. Afterward, the sample was removed, dried with filter paper, and placed in the thermal desorption setup to measure the residual tritium in the bulk of the metal. The activity of the tritium removed by the zinc-chloride mixture and by the drying filter paper was measured using liquid scintillation counting to give the total quantity of surface tritium removed in the wash.

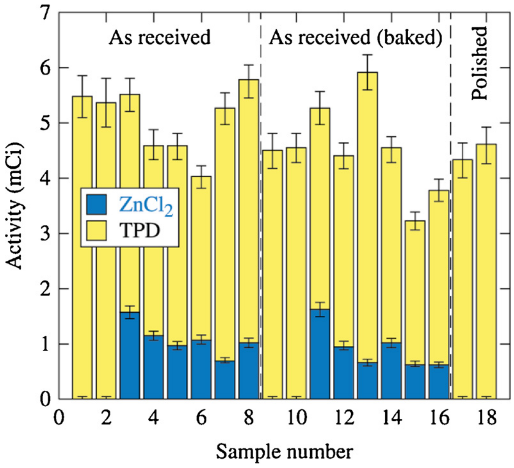

Fig. 1. The total tritium inventory for as received, as-received baked, and polished stainless-steel samples. Tritium fractionation between the surface and bulk for as-received and as-received baked samples using the ZnCl$_2$ treatment method.

3. Results and discussion

To determine the tritium removal efficiencies of the pulsed-plasma and zinc-chloride methods, a series of six samples were treated with each method. Following the pulsed-plasma or the zinc-chloride treatment, each sample was heated to 700 °C in the thermal desorption facility to measure the residual tritium in the sample bulk. Surface activities collected from the two sample sets are shown in Figs. 1–3. The first set of measurements focused on determining the total tritium inventory and its fractionation between surface and bulk. Samples were subjected to either thermal desorption or to the zinc-chloride treatment followed by thermal desorption. In the second set of measurements the efficacy of removing surface tritium by pulsed-plasma or by zinc-chloride washes was compared. Samples were washed in the zinc-chloride solution or exposed to the pulsed-plasma and then heated in the thermal desorption facility. Figs. 1–3 compare the quantities of tritium removed from the surface and from the bulk for the as received, baked as received, and the polished cases.

The data collected using the first series of samples are shown in Figs. 1 and 2. These data were collected using a set of as-received samples, a set of baked as-received samples, and a set of mechanically polished samples. In this series, each sample was subjected to either thermal desorption alone or to the combined procedure of zinc-chloride

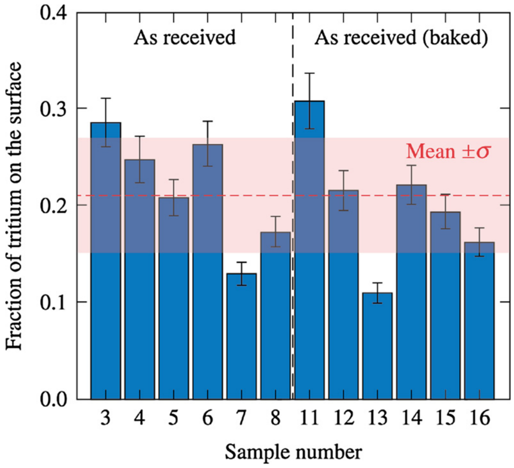

Fig. 2. The relative fraction of the total tritium inventory residing on the surface of as-received and as-received baked stainless-steel samples. The dashed line provides the mean fraction without differentiating between the two treatments. The shaded band illustrates one standard deviation about the mean.

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