Distribution of Tritium in the Near Surface of Stainless-Steel 316

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The interaction and subsequent retention of tritium in stainless steel impacts many activities and procedures at LLE, ranging from protium contamination of the DT fuel supply to decontamination procedures for items fielded in OMEGA. The adsorption of tritium onto the surface of stainless steel represents the first step in the overall retention and permeation of tritium through stainless steel. Understanding the distribution of tritium in the surface region is necessary in determining how tritium responds to an exposure of tritium gas. This knowledge can help determine the effectiveness of barrier materials deposited on stainless-steel surfaces and various surface treatments designed to reduce tritium absorption. Measurement of the near-surface concentrations that develop after storage help quantify the classical observation of tritium migration to steel surfaces after surface decontamination.[1]

There are three regions of interest regarding tritium retention in stainless steel: the surface, the near surface, and the bulk metal. Each region has different chemistry and structures, causing various quantities of tritium to bind in each region. The bulk metal is comprised of a regular lattice of metal atoms, where tritium binds in the interstitial spaces between the metal atoms. The "near surface" represents the transition between the regular metal lattice and the native metal oxide. The surface of stainless steel contains several monolayers of water molecules, which are bound to the metal oxide by a hydroxide layer.

Measuring the tritium concentrations in each region requires three different techniques, each specifically designed to probe the tritium content in that region. Tritium bound to the steel's surface was measured by immersion in a zinc chloride ($ZnCl_2$) solution. This technique has been effective at removing surface-bound tritium without etching into the metal oxide.[2,3] The tritium concentrations in the near surface were measured by dissolving the metal in diluted mixtures of hydrochloric and nitric acid.[4,5] The residual tritium was determined by heating the samples to elevated temperatures.

The migration of tritium from the bulk to the surface was measured by removing surface tritium with an $ZnCl_2$ wash and an acid etched after storing the sample for several days. This removal and storage sequence was repeated several times to monitor how much tritium had migrated to the surface. Such an experiment mimics the effectiveness of decontamination procedures used to clean steel surfaces exposed to tritium gas.

The total tritium contained in the stainless-steel 316 (SS316) samples was 180±20 MBq (4.9±0.5 mCi). The results are shown in Fig. 1 as a function of the storage time for each sample. Six different sets of SS316 samples are included in this figure. Each set was charged with tritium on a different date. These data show negligible, if any, loss of tritium over the 70-day storage period in a dry environment. Two samples contained significantly more tritium than observed on average. This apparent higher activity is most likely a result of cross contamination during sampling.

The combination of the $ZnCl_2$ wash and acid etching provides the distribution of tritium on the surface and in the near surface (<10 μm). The concentration profiles determined from these measurements are shown in Fig. 2. Nine different samples were used. Each sample was stored from 33 to 233 days between charging and measurement. The concentration profiles show that the adsorbed water layers contain large concentrations of tritium. These concentrations decrease by a factor of 10^7 over an ~5-μm
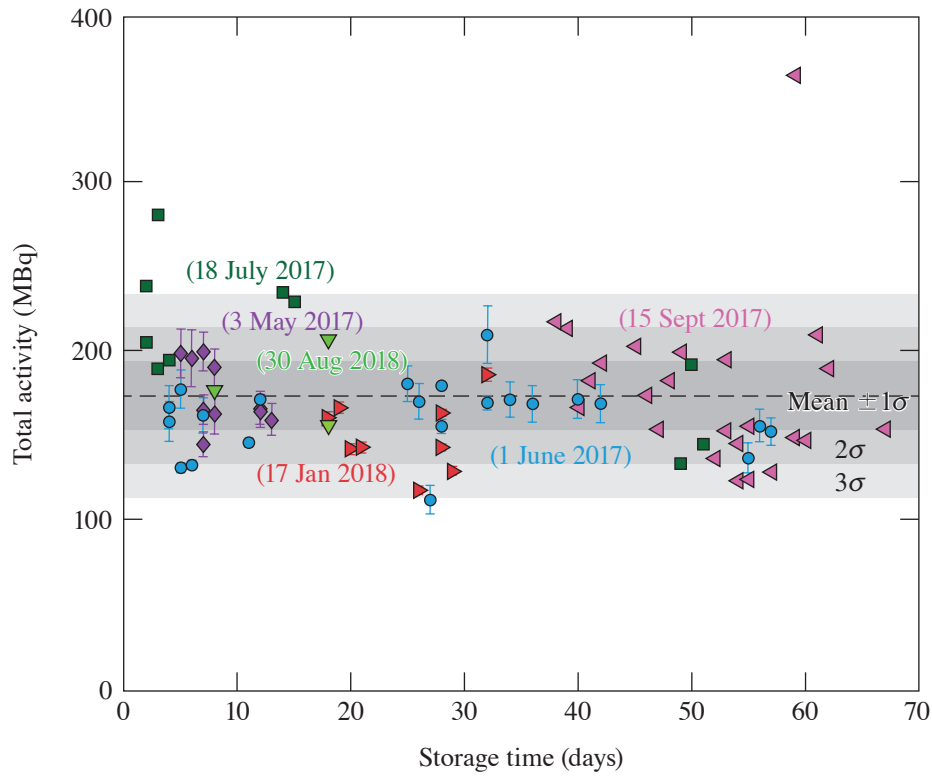
Figure 1
Summary of total tritium inventories from six different sets of samples. Each set was charged with tritium on a different date.
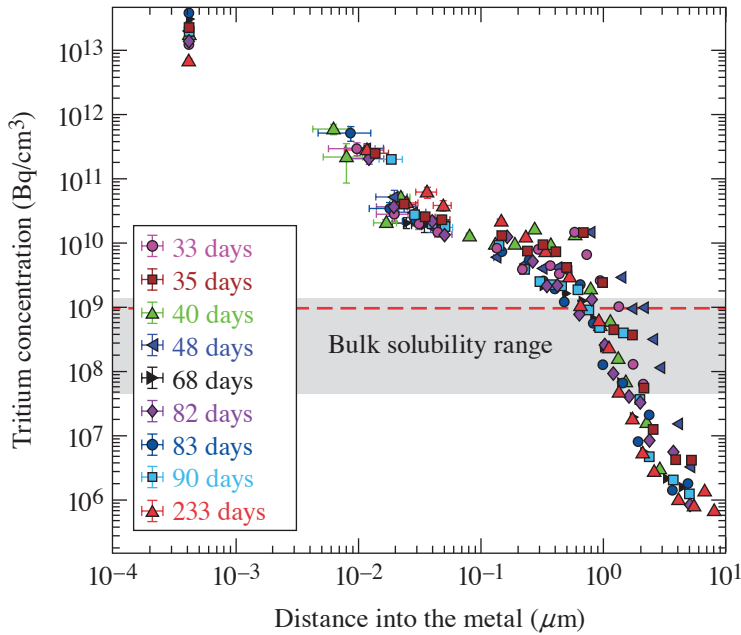
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Figure 2
Tritium concentration profiles in the near surface of SS316, as measured using a ZnCl$_2$ wash, followed by acid etching. Shown in the figure are the results of nine different samples, stored from 33 to 233 days prior to measurement.

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depth into the metal. Figure 2 also shows the mean and range of hydrogen isotope solubilities in SS316 at 25°C from higher temperature data reported in the literature.[6]

The measured concentration profiles in the near surface of SS316 indicate several things: First, ~30% of the surface is occupied by tritium. Second, the distribution of tritium in the near surface does not change during the 33 to 233 days of storage. This is striking since tritium migration is expected, given the observed concentration gradient and the diffusivity of tritium in SS316 reported in the literature. Tritium appears to be in a quasi-equilibrium state. Finally, comparing the measured concentrations with the reported solubility of tritium in SS316 shows the measured concentrations at depths less than ~1 μm exceed the reported solubilities. These higher concentrations are attributed to the storage capacity of the metal oxide.

Thermal desorption, acid etching, and $ZnCl_2$ wash measurements show the distribution of tritium to be fixed over the measured time period with 40±20% residing in the bulk metal, 42±9% in the near surface, and 21±5% in the adsorbed water.

Tritium migration from the SS316 surface was measured for several samples. Figure 3 presents activity as a function of storage interval, where each interval was between 2 and 4 days. Tritium migrating to the surface decreases after each successive cleaning and storage period, suggesting a depletion of tritium in the near-surface region and an inability to resupply the surface. Perturbing the quasi-equilibrium concentration profiles (shown in Fig. 2) by removing surface tritium concentrations cause some of the tritium to migrate back to the surface from the underlying subsurface in order to re-establish the equilibrium between the surface and the subsurface.
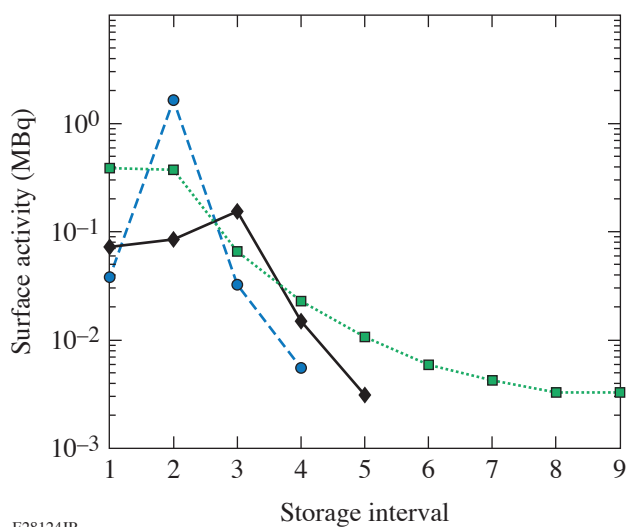

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Figure 3

Surface activity for three SS316 samples, measured after the surfaces were cleaned using acid etching and then stored for 2 to 4 days.

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