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Influence of Microstructure on the Absorption of Tritium Into Gold-Plated 316 Stainless Steel

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Corresponding Author:	Matthew Sharpe University of Rochester Rochester, UNITED STATES
Corresponding Author Secondary Information:	
Corresponding Author's Institution:	University of Rochester
Corresponding Author E-Mail:	msha@lle.rochester.edu
Corresponding Author's Secondary Institution:	
First Author:	Matthew Sharpe
First Author Secondary Information:	
Order of Authors:	Matthew Sharpe
	W. T. Shmayda
Order of Authors Secondary Information:	
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Influence of Microstructure on the Absorption of Tritium Into Gold-Plated 316 Stainless Steel

M. Sharpe,[a]* W. T. Shmayda[a]

[a]*University of Rochester, Laboratory for Laser Energetics, Rochester, NY*

*E-mail: msha@lle.rochester.edu

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The interaction of tritium with metal surfaces is the initial step in the overall absorption of tritium by the substrate metal. As a result, limiting the adsorption of tritium to the surface may effectively reduce the quantity of tritium absorbed by a metal when it is in contact with tritium gas. To limit tritium adsorption, many tritium users electroplate gold onto the substrate metal. The gold layer is expected to reduce tritium adsorption, and subsequently absorption, by reducing water adsorption.

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KEYWORDS: tritium, gold, stainless steel, absorption

I. Introduction

Tritium absorption into stainless steels is a fundamental problem associated with tritium handling. Tritium will adsorb to the surfaces of steel, creating high localized

concentrations on the surface.[1,2] Additionally, the bulk steel substrate can absorb significant quantities of the isotope, creating a long-term radiation hazard. Finally, tritium gas interactions with stainless-steel surfaces provides a pathway for the accumulation of lighter hydrogen isotopes in the gas phase.[3] These lighter isotopes either cause an imbalance in the fusion fuel supply or poison the fusion reaction. To mitigate these concerns, many devices and gas-handling systems are gold plated.[4,5]

Gold plating offers an attractive solution to many of the aforementioned concerns. As a noble metal, gold does not readily form a surface oxide or a multilayer structure of water under standard atmospheric conditions,[6] as it does on stainless steel surfaces.[7] Further, hydrogen diffusion through gold is reported to be much slower than through stainless steel.[8] The combined gold properties of low surface interactions with slow diffusion have led to the assumption that any gold layer is sufficient for reducing tritium absorption and permeation.

In the current work, stainless-steel type 316 (SS316) samples were plated with gold by two different commercial vendors and one national laboratory. All gold layers were electrodeposited on the surface, with a nickel interface layer between the gold layer and steel substrate. The gold layers were analysed using a scanning electron microscope (SEM), a transmission electron microscope (TEM), x-ray photoelectron spectroscopy (XPS), and an interferometer for surface roughness. Gold-plated samples from each supplier were exposed to gaseous tritium. Absorbed tritium quantities were determined using thermal desorption.

II. Experimental Setups and Methods

To test the effectiveness of gold layers obtained from various vendors, a set of SS316 samples with dimensions of $5.1 \times 0.3 \times 1.8$ cm^3 and surface roughness Ra ~300 nm were manufactured by Torrovap Inc. and prepared according to procedures outlined previously.[9] Briefly, these samples were cut from a common plate and subsequently had 0.86 mm machined away to eliminate any surface inclusions arising from the manufacturing process and to expose the base metal lattice.

All electroplating was done by one of three suppliers; listed here as Vendor #1, Vendor #2, and NL for the two commercial vendors and one national laboratory, respectively. Prior to gold plating, the samples were mechanically polished to a mirror finish, with the final mean surface roughness being either 5 nm or 41.5 nm (Table I). Next, a nickel strike layer was electroplated to the SS316 substrate. Finally, gold was electroplated to the nickel layer. All gold plating conditions were considered proprietary and not shared with the authors. Vendors #1 and #2 followed a military specification for their gold layers: MIL-DTL-45204C. The types and hardness grades requested varied, as shown in Table I. The NL did not report a specification.

Prior to loading with tritium, all samples were sequentially washed in acetone, water, and isopropyl alcohol. This includes SS316 samples not plated with gold. After washing, the samples were exposed to deuterium–tritium gas at room temperature for approximately 24 h (Table I). The exposure cell allows for multiple samples to be exposed simultaneously while minimizing the contact each sample has with the walls of the exposure cell. After exposure, the samples were taken out and placed into individual storage containers under dry helium. The samples were loaded in several "batches." Each batch contained some

gold-plated samples, as well as non-plated SS316 samples for reference. For loading Batches 1–4, the samples were stored in individual cans. Batch 21 employed a newer storage rack design, which was constructed to be more leak tight. Each sample was kept in its storage container until retrieved for experimentation. To measure the total tritium absorbed in a sample, the sample was subjected to high-temperature thermal desorption (700°C/1 to 2 h).

III. Results

Upon receiving the gold-plated SS316 from each plater, one sample from each set was set aside for surface analysis. Each surface was viewed using a scanning electron microscope. Samples received from the NL and Vendor #1 were milled using a focused ion beam in the SEM to view a cross section of the gold layers.

III.A. Surface Analysis of NL Samples

Surfaces of the NL samples showed a large surface roughness, with valleys that may reach the substrate steel itself. Figure 1 shows a measurement of the surface roughness using atomic force microscopy (AFM) and a cross section of the gold layer, imaged using SEM. The AFM measurement shows peak to valley distances ≥100 nm. The SEM cross section shows the gold layer to be ~100 nm thick.

III.B. Surface Analysis of Vendor #1 Samples

Samples from Vendor #1 showed uniform surfaces, with sporadic pinholes ranging

from 1 μm to 100 nm. A cross section of the gold layer revealed voids in the gold layer averaging 450 nm in diameter [Fig. 2(a)]. The interfacial nickel layer appears to have no significant defects and is bonded well to the substrate SS316. While, the gold–nickel interface appears to have a gap, this may be a result of the ion beam milling.

III.C. Surface Analysis of Vendor #2 Samples

Surface analysis of the samples from Vendor #2 show dense gold layers with some microstructures. Figure 3 shows SEM images taken normal to the sample's surface for (a) hard gold and (b) hot gold. The hard gold sample showed microgranular features, while the hot gold showed a much smoother surface.

A small cross-sectional area of the hot gold sample was milled out of the surface with the focused ion beam and transported to the Transmission Electron Microscope (TEM) for further analysis (Fig. 4). The results show the nickel layer is bonded well to both the substrate steel and the gold coating. The gold layer contains micropores, with the largest features measured being less than 100 nm in size.

III.D. Results of Tritium Exposures

The effectiveness of the various gold-plated samples to reduce tritium absorption was tested by exposing the samples to DT gas and then measuring the total tritium absorbed using thermal desorption (Sec. II). Figure 5 shows a compilation of all the data collected from the various loading batches. These data show the tritium inventory in the gold-plated samples relative to the non-plated SS316 samples for the various suppliers. All but one of the coatings showed relative inventories greater than or equal to one. This indicates that

the quantity of tritium absorbed in the gold-plated samples is equal to or greater than that absorbed in the non-plated samples. Only the hot gold samples from Vendor #2 show a 25% reduction in tritium inventory, relative to the SS316 samples loaded at the same time.

IV. Discussion

The ineffectiveness of many of the gold layers to reducing tritium absorption is attributed to the structure of the gold layer. Pores, deep channels, and other defects may allow tritium to bypass the gold layer and directly enter the underlying nickel interfacial layer. Because tritium permeability through nickel is large relative to either gold or SS316, the nickel layer can act as a pump for any tritium bypassing or migrating through the gold layer. In the case of the NL samples, deep valleys were observed, which may indicate incomplete coverage of the substrate. The pinholes and large voids observed in the samples from Vendor #1 may artificially increase tritium solubility in the gold and/or offer an easier pathway for tritium to migrate through the layer. In contrast, the dense layers produced by Vendor #2 showed consistently lower inventories than Vendor #1 or the NL samples. Although the hot gold samples showed microporous layers, the voids were much smaller than those from Vendor #1. The smaller micropores may have lowered the tritium solubility in the gold layer, relative to the Vendor #1 samples.

V. Conclusions

The effectiveness of gold plating SS316 substrates to reducing tritium absorption was measured. It was found that the microstructure of the gold layer significantly impacts the absorption of tritium into the samples. Defects in the gold layer, such as pores and

channels, appear to influence the absorption of tritium. In contrast, a moderately dense layer obtained from Vendor #2 showed 25% reduction in tritium absorption, relative to non-plated SS316. Caution must be used when selecting a gold-plating method intended for tritium use.

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Figure captions

Fig. 1. (a) AFM results and (b) SEM cross section of gold surface on SS316 sample gold coated by NL.

Fig. 2. SEM cross section of gold and nickel layers on gold-plated SS316 provided by Vendor #1. (a) Zoomed in cross section of gold layer; (b) gold, nickel, and substrate SS316. Vertical streaking in (b) is a result of ion beam milling the surface.

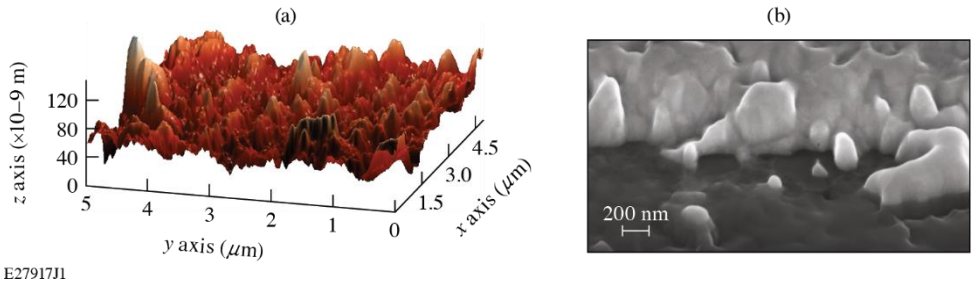
Fig. 3. SEM images of (a) hard and (b) hot gold samples. Images were taken normal to the plane of the sample.

Fig. 4. TEM cross section analysis of hot gold layers. (a) Gold, nickel, and substrate SS316, with atomic identification in the inset. (b) Zoomed in view of the gold layer.

Fig. 5. Comparison of tritium inventory in gold-plated samples and non-plated SS316 samples. Data shown here are relative to the tritium inventories in SS316 for each loading batch. Values less than unity indicate reduced absorbed in gold-plated samples.

Table I Summary of Gold Layers and Tritium Exposure Conditions

Batch	Δ*x* (*μ*m)		Ra (nm)	Suppliers	Specification	Loading pressure (kPa)	T %	Loading time (h)
	Au	**Ni**						
1	0.8, 1.7, 4.1, 8.3	6	41.5	Vendor #1	Type I, grade A	86.7	36.3	24
2	0.8, 4.1, 8.3	6	41.5	Vendor #1	Type I, grade A	72.4	56	24.25
3	1.7	6	41.5	Vendor #1	Type I, grade A	70.7	58	24
4	0.1, 1.7 100 nm	6	41.5	Vendor #1 NL	Type I, grade A (no reported spec.)	71.2	56	24
5	> 0.5	1.5	5	Vendor #2	Type II, grade D (hard) Type I/III, grade A (hot)	82.5	69.1	24



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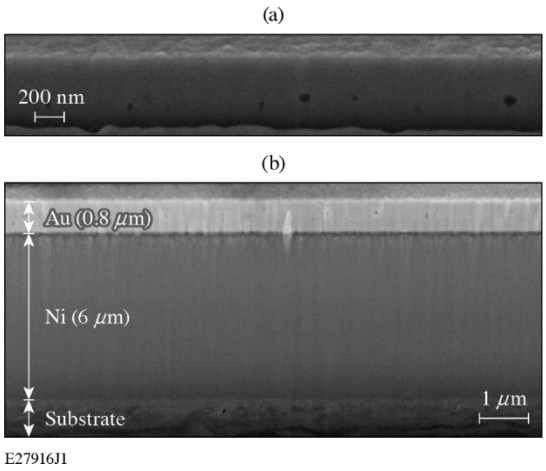

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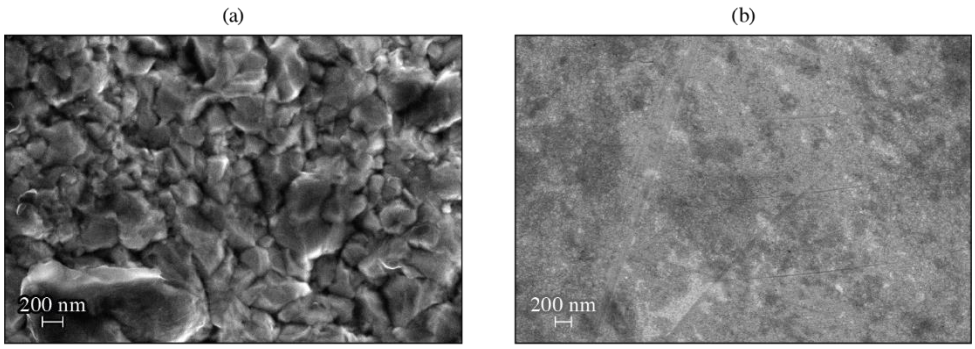

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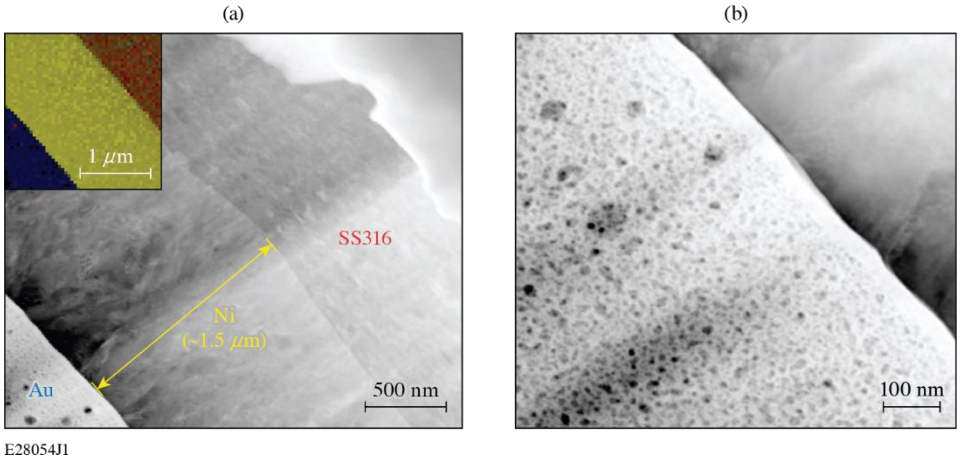

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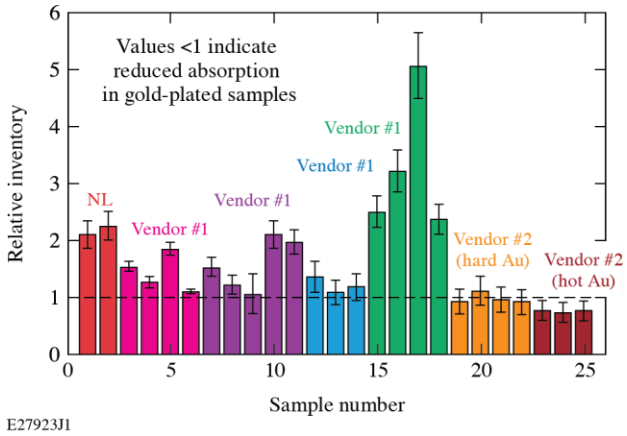

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