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Development of an environmentally friendly protective coating for the depleted uranium–0.75 wt.% titanium alloy

Part II: Coating formation and evaluation

Donald F. Roeper^{a,*}, Devicharan Chidambaram^{b,1}, Clive R. Clayton^a, Gary P. Halada^a

^a Department of Materials Science and Engineering, Stony Brook University, Stony Brook, NY 11794-2275, USA

^b Department of Environmental Sciences, Brookhaven National Laboratory, Upton, NY 11973-5000, USA

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Abstract

Molybdenum oxide based coatings have been formed on the surface of the depleted uranium–0.75 wt.% titanium alloy. Surface activation prior to coating formation has been examined using fluorides and concentrated nitric acid. The electrochemical characteristics of the coating formation processes were studied using open circuit potential measurements. Residual fluoride from the activation process has been found to interfere with coating formation and surface activation by nitric acid yields a relatively thinner but more robust coating.

The corrosion protection characteristics of the coatings were evaluated by potentiodynamic polarization testing in quiescent 0.05 M sodium chloride. The coatings have been studied using scanning electron microscopy, energy dispersive spectroscopy and optical microscopy.

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1. Introduction

Depleted uranium (DU) has both military and commercial applications but its potential use is limited due to its corrosion properties. The corrosion of uranium can release airborne particles due to spalling of the oxide layer [1]. Inhalation and ingestion are dangerous routes of exposure to uranium particles [2,3]. While radiation is still a concern for DU alloys, they have been largely depleted of ²³⁴U and ²³⁵U, which are the more highly radioactive isotopes of uranium than the ²³⁸U isotope [4]. In order to prevent corrosion of the DU–0.75 wt.% Ti alloy, we are developing a protective chemical conversion coating with more environmentally friendly [5] chemicals than are currently used in the chromate conversion coatings (CCC) which are applied to other metals and alloys.

Previous work in this laboratory [6,7] has shown that electrochemical activation of the alloy in an acidic sodium molybdate solution can cause the deposition of a molybdenum oxide coating on the DU alloy. The alloy was anodically polarized in the acidic sodium molybdate solution and this resulted in the formation of a thick blue coating. However, the coating was loosely adhered and easily rinsed off. A thin rudimentary coating can be formed by simple immersion in a sodium molybdate and nitric acid solution but the coating is non-uniform and pitting occurs during the process [6]. A complex chemical bath that can both activate the surface and deposit a protective coating in one step is desirable for a fast and inexpensive process. CCC is an example of such a coating formed in a complex bath; however, CCCs are being phased out in some applications because of the carcinogenic nature of hexavalent chromium [8–10]. Our earlier work [6] has also shown that certain fluorides can be used as a surface activator for the DU alloy. This study examines the use of fluorides in the development of a molybdate-based coating on the DU alloy. In addition, a surface pretreatment and activation by a

* Corresponding author. Tel.: +1 6316328068; fax: +1 6316328052.

E-mail address: droeper@notes.cc.sunysb.edu (D.F. Roeper).

¹ Goldhaber Distinguished Fellow.

strong acid bath is examined and compared to the fluoride pretreatment.

2. Experimental

2.1. Sample preparation

The samples, 2.5 cm diameter discs cut from a DU–0.75 wt.% Ti alloy penetrator (0.75 wt.% Ti, less than 0.2 wt.% ^{235}U and ~ 0.0008 wt.% ^{234}U , balance is ^{238}U with various trace impurities), were polished in an oxygen-free contained environment as described earlier [6] then degreased with isopropanol and rinsed with deionized water prior to receiving any coating treatments.

2.2. Surface morphology and chemistry

The morphology and surface chemistry of the coated depleted uranium titanium alloy samples were studied using the scanning electron microscope (Model 1550, LEO Electron Microscopy Inc., NY, USA). The SEM uses a Schottky Field Emission Gun. The ultra-high resolution (2.1 nm at 1 kV) SEM was also equipped with an energy dispersive X-ray detector (EDAX) with a polymer window and a retractable Robinson backscattering detector.

2.3. Optical microscopy

The surfaces of the coated DU alloy samples were observed using an optical microscope supplied by MEIJI that was interfaced with a Nikon digital camera (model DXM 1200) capable of a resolution of 3840 pixel \times 3072 pixel (12 million pixel resolution). All images were obtained at high resolution and sensitivity.

2.4. Electrochemical cell and instrumentation

A corrosion flat cell that was built in-house was used in these studies. A platinum coated niobium mesh was used as the counter electrode. All potentials were measured with respect to a saturated calomel electrode (SCE), which was used as the reference electrode. Electrochemical tests were performed using a PC4/FAS1 model potentiostat supplied by Gamry Instruments Inc. The dc corrosion techniques software, DC 105 (also supplied by Gamry Instruments Inc.) was used to control the potentiostat and analyze the data. More details about the system can be found elsewhere [11].

2.5. Electrochemistry of the depleted uranium–0.75 wt.% titanium alloy

The samples were exposed to the coating treatment solutions and the open circuit potential (OCP) was monitored. A magnetic stirrer was employed during open circuit potential

measurements in order to minimize any stagnant layer formation. Deionized water was used to prepare the solutions and the solutions were not purged during the experiments. Bullock et al. [12] suggested that an oxide film formed under oxidizing conditions would have better corrosion properties than a film formed under non-oxidizing conditions. The following reagent grade chemicals were used in the coating treatment baths: $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$, NaF, K_2ZrF_6 , K_2SiF_6 and HNO_3 . The pH of the solutions was in the range of approximately 1.2–2.0, with the exception of the 50% nitric acid solution. This pH range places uranium in a location on its Pourbaix [13] diagram analogous to aluminum during chromate conversion coating formation.

After drying for a minimum of 48 h, the coated samples were potentiodynamically polarized in 0.05 M NaCl at a scan rate of 1 mV/s from approximately 250 mV below the OCP to 250 mV above it. Several samples were allowed to age in air under ambient conditions for several weeks prior to testing. To act as a control, OCP measurements and potentiodynamic polarization scans were performed on uncoated, polished DU alloy samples in 0.05 M NaCl.

3. Results and discussion

3.1. Coating formation

The open circuit potentials of three fluorides previously tested, NaF, K_2ZrF_6 and K_2SiF_6 , indicated that they should all work well as surface activators [6,14,15]. Each of these fluorides displayed similar OCP and potentiodynamic polarization curves when exposed to DU in 0.1 M nitric acid solutions [6]. In this paper, we will focus on the behavior of K_2ZrF_6 as an activating agent, which is one of the fluoride constituents of the CCC treatment bath. However, each of these three fluorides were tested and they all displayed similar behavior when used in equivalent coating experiments. The concentration of the chemical inducing the maximum change in the OCP from the previous work [6] was considered to be the optimal concentration of that chemical to be used in these studies.

The most simple coating treatment bath would consist of an activator, (F^-), and a coating formation component, (MoO_4^{2-}), together in a single solution. This chemical bath consisted of 0.0025 M K_2ZrF_6 and 0.025 M $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$ in 0.1 M nitric acid. Immersion of the DU alloy into this simple bath for up to 1 h forms a very loosely adhered precipitate on the surface of the alloy. The OCP for the DU alloy immersed in this bath is shown in Fig. 1. The OCP is initially at a higher potential but quickly drops to a much lower potential showing the surface to be in a more active state. It is likely that the presence of the fluoride ion interferes with the ability of the molybdate to bond well with the DU surface and causes the precipitation of a molybdate salt instead of a chemically bonded coating. The salt that formed on the surface is easily rinsed off by water.

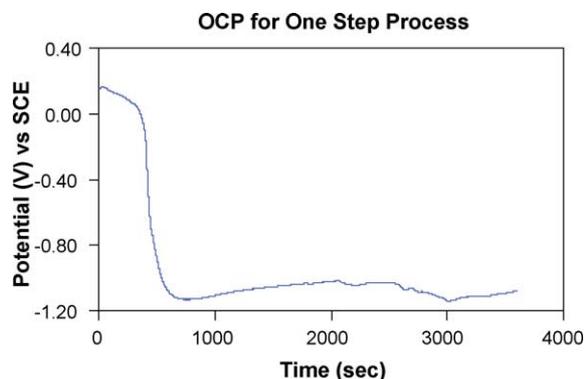


Fig. 1. A single bath treatment containing both an activator, 0.0025 M K_2ZrF_6 , and a protective species, 0.025 M Na_2MoO_4 , in 0.1 M nitric acid did not form a protective coating.

The electrochemistry of the single bath process was examined in greater detail by adding the chemicals separately to the bath. As can be seen in Fig. 2, when the fluoride is added at $t = 100$ s, there is a steep drop in potential, indicating the activation effect. After 5 min of pretreatment in the fluoride solution, the molybdate was introduced. Adding the molybdate to this solution raised the potential, but it was a short-lived effect. The potential quickly drops back down almost to the initial active potential. The fluoride appeared to prevent any chemically bonded coating formation on the surface and instead there was a dark blue precipitate that easily rinsed off.

Since the presence of fluoride ions in the treatment solution seems to inhibit coating formation, a two-step process was investigated. In the first step, the DU alloy was immersed in a solution of 0.0025 M K_2ZrF_6 in 0.1 M nitric acid. This caused the characteristic steep drop in potential, indicating surface activation. After 5 min of activating the surface, the K_2ZrF_6 solution was drained out of the electrochemical cell and in the second step, the 0.025 M sodium molybdate

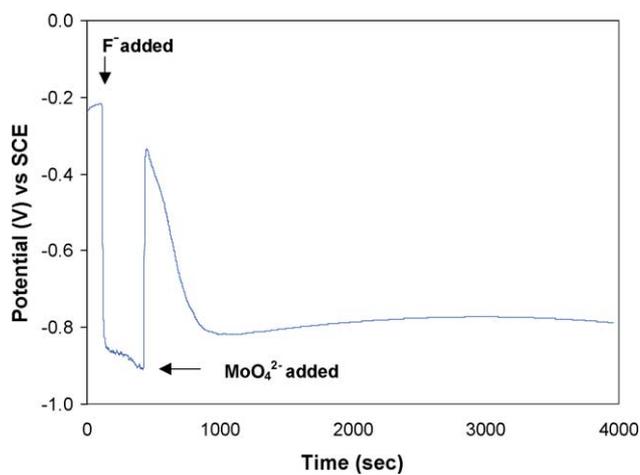


Fig. 2. In a single treatment bath, the addition of fluoride at $t = 100$ s caused a steep drop in potential. Adding the molybdate raised the potential but it quickly dropped back down.

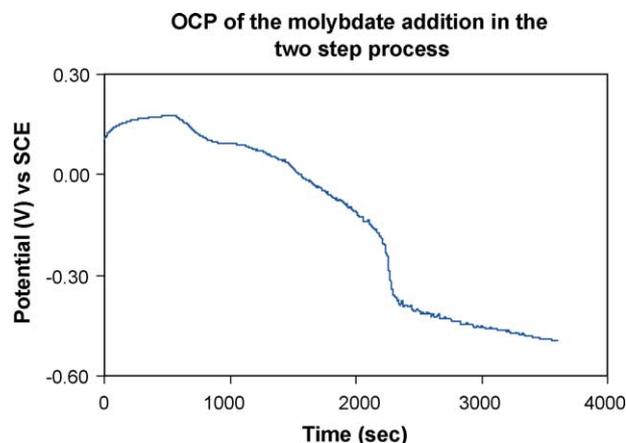


Fig. 3. OCP vs. time behavior for the DU alloy immersed in 0.025 M Na_2MoO_4 and 0.1 M HNO_3 solution the after sample had been pre-treated in the 0.0025 M K_2ZrF_6 and 0.1 M HNO_3 bath. Removing the fluoride before adding the molybdate raised the potential to a peak at +0.175 V vs. SCE. It then began dropping to -0.5 V vs. SCE over an hour.

solution was added. Fig. 3 shows the effect of the second step. There was a rise in potential to approximately +175 mV versus SCE with the addition of the molybdate. Removing the K_2ZrF_6 solution before adding the molybdate enabled the surface to grow a thick coating as seen in Fig. 4. The surface cracking of molybdate coatings has been shown before [16–18] and is similar to the coating pattern that is seen in CCC coatings on aluminium [19]. However, the outermost layers of this coating are loosely adhered and can be removed by a damp paper towel, leaving a thin underlayer. Also, the potential is seen to drop down again to close to -500 mV versus SCE over the course of an hour. The potential is ennobled for only about 10 min before it begins to drop. A subsequent sample was treated in a similar manner but removed from the solution and rinsed with D.I. water after 8 min of treatment, before the potential could significantly drop and the OCP is shown in Fig. 5. The coating appears to have better coverage and displays somewhat less cracking and is shown in Fig. 6.

The presence of fluoride ions appears to interfere with the formation of a molybdate coating but even after the cell is drained and replaced with a molybdate solution without any fluoride, the potential still drops over time. This is likely caused by residual fluoride ions absorbed on the surfaces of the electrochemical cell as well as the surface of the DU alloy. Another sample was prepared in a similar manner but the cell and the surface of the DU alloy was flushed with 0.1 M nitric acid after the fluoride solution was drained. Fig. 7 shows the OCP illustrating the steps involved. At $t = 100$ s, the fluoride is added and there is the characteristic steep drop in potential. After 5 min of fluoride exposure, the cell is drained and flushed with 0.1 M nitric acid. Fresh 0.1 M HNO_3 is added which causes a small rise in potential and with the addition of the molybdate, the potential rises to almost +200 mV versus SCE and remains ennobled for one hour without any decline. This sample, shown in Fig. 8, has the previously seen cracked pattern but was more uniform than the previous coatings. The

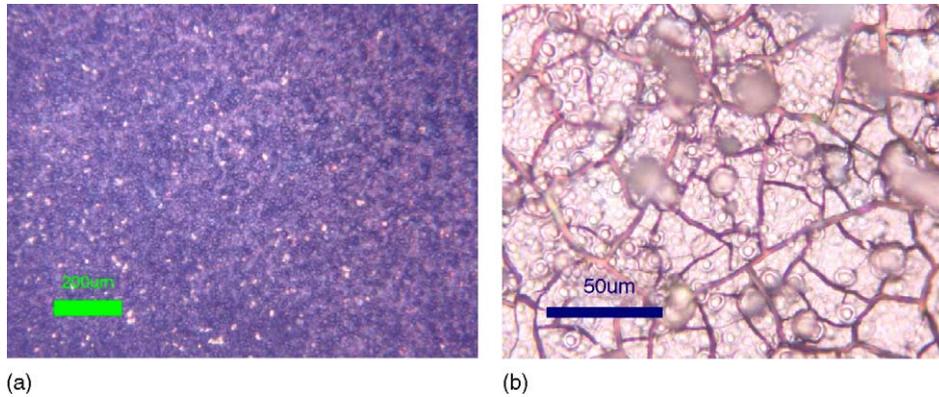


Fig. 4. Digital micrograph of the sample after the OCP measurement shown in Fig. 3. (a) Removing the fluoride before adding the molybdate enabled the surface to grow a blue coating. (b) The cracked pattern is similar in appearance to CCC coatings.

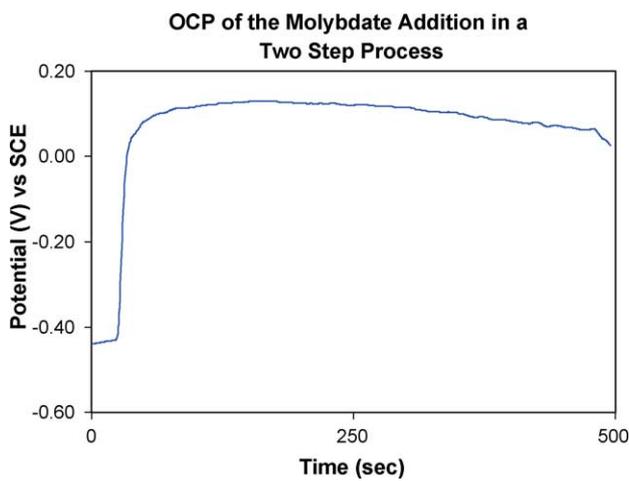


Fig. 5. OCP vs. time behavior for the DU alloy immersed in 0.025 M Na_2MoO_4 and 0.1 M HNO_3 solution the after sample had been pre-treated in the 0.0025 M K_2ZrF_6 and 0.1 M HNO_3 bath. Removing the fluoride before adding the molybdate raised the potential but the sample was removed after 8 min, before the potential dropped below 0 V vs. SCE.

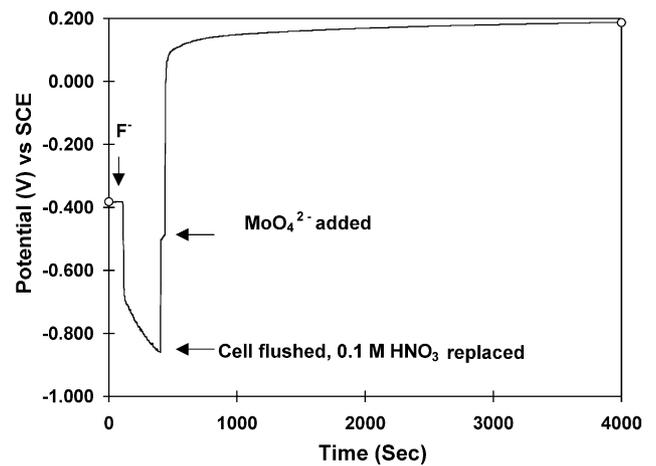


Fig. 7. Two-step process. The fluoride activates the surface causing a steep drop in potential and is followed by a nitric acid flush. The addition of the molybdate elevates the potential into the positive region. Flushing the cell with nitric acid to remove any remaining fluoride allowed the reaction to proceed for 60 min at a positive potential.

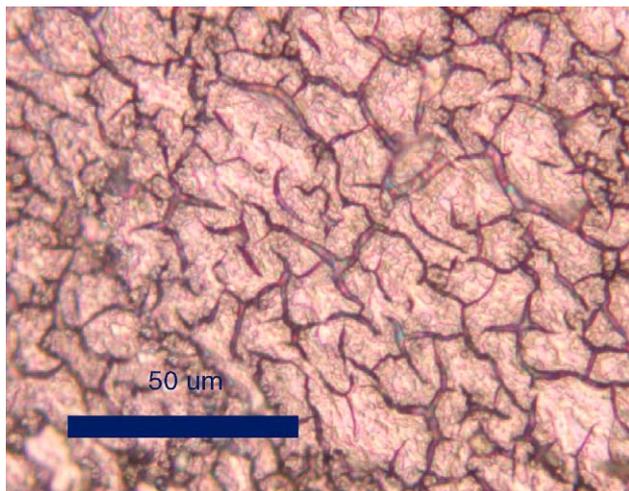


Fig. 6. Digital micrograph of the sample after the OCP measurement shown in Fig. 5. Stopping the reaction before the potential could drop significantly produced a coating that shows less cracking.

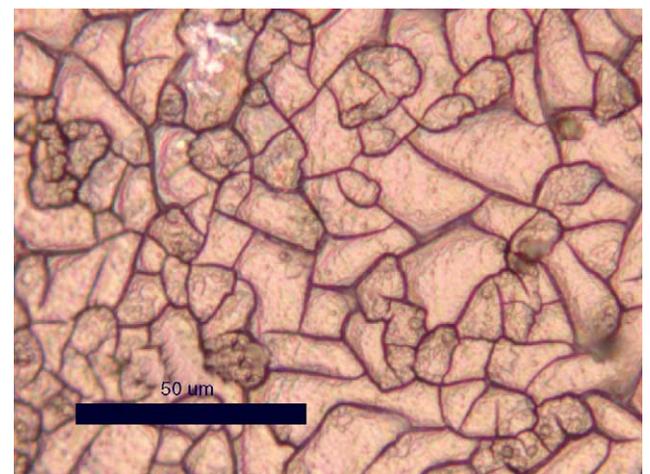


Fig. 8. Digital micrograph of the sample after the OCP measurement shown in Fig. 7. With a nitric acid flush prior to molybdate addition, this sample also shows the characteristic cracking pattern.

removal of the absorbed fluoride ions benefits the coating formation and the coating is more adherent to the alloy. However, fluorides are also hazardous chemicals [20] and it is preferable to find an alternative surface pretreatment technique in order to avoid their usage.

Another method of surface pretreatment that was tested was activation of the sample in 50% HNO₃. Strong nitric acid solutions can be used to remove the native oxide [21]. In these experiments, 50% HNO₃ was used to remove the native oxide and render the surface more hydrophilic. The fluorides provide a similar function; after the 5 min immersion in the fluoride solution, the DU alloy wets with the sodium molybdate solution where as without pretreatment, it does not. If the surface is not wettable with the treatment solution, a continuous and uniform coating cannot be formed. After 5 min of immersion in the 50% nitric acid, the surface became wettable. Shorter times did not render the surface uniformly wettable and there was no additional benefit to longer immersion times in the acid, once the surface had become wettable to the treatment solutions.

After removal from the 50% nitric acid, the sample was immediately immersed into the 0.025 M sodium molybdate

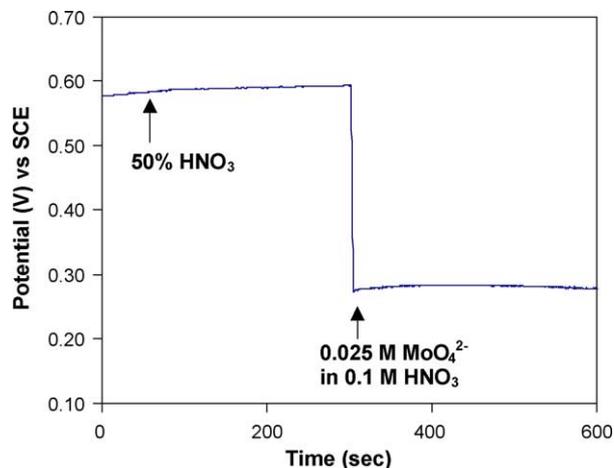


Fig. 9. OCP for immersion in 50% nitric acid. The steep drop was the result of subsequent immersion in 0.025 M sodium molybdate in 0.1 M nitric acid.

in 0.1 M nitric acid solution for 5 min. The OCP for this experiment is shown in Fig. 9. Upon removal from the molybdate solution, it was rinsed with D.I. water to quench the reaction and rinse off any excess solution. This sample exhibited a

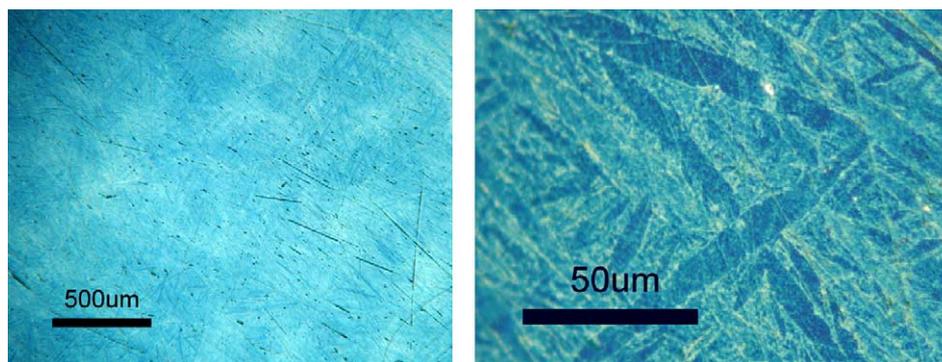


Fig. 10. Digital micrograph of the sample after the OCP measurement shown in Fig. 9. The martensitic structure of the DU alloy can be seen through the transparent blue molybdenum oxide coating.

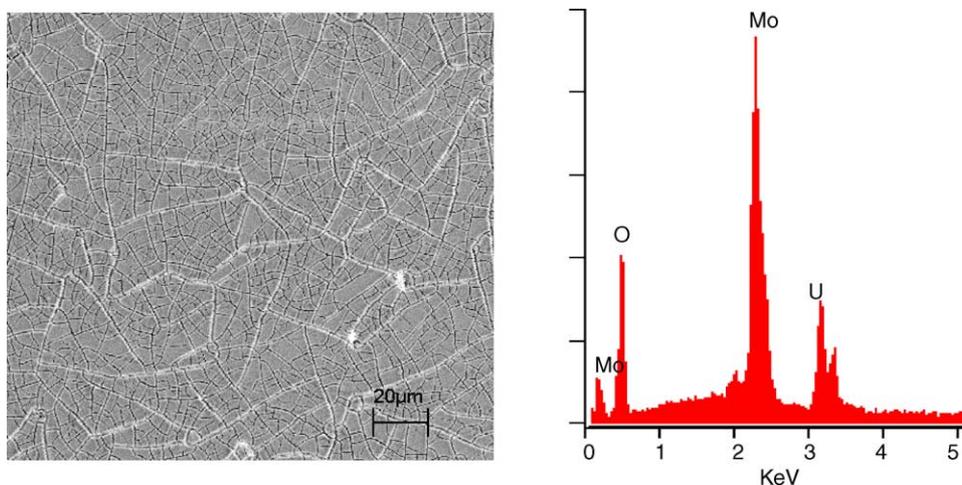


Fig. 11. SEM and EDS of the two-step fluoride-activated coating. SEM shows the cracked plate pattern.

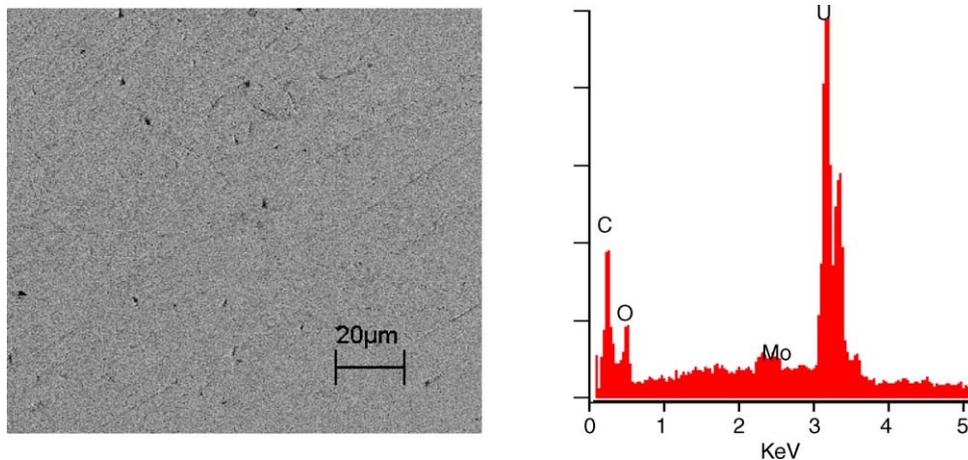


Fig. 12. SEM and EDS of the acid-activated coating. The acid-activated coating appears featureless and does not display the cracked plate pattern.

very thin blue coating as shown in Fig. 10 and does not have the features of the cracked plate pattern that was seen in the fluoride-activated coatings. The martensite structure of the DU alloy [6,22,23] can be seen through this thin coating.

3.2. Coating evaluation

The SEM of the two-step fluoride-activated coating is shown in Fig. 11 and the cracked plate pattern can be seen. The EDS for this coating is included in Fig. 11 and shows a large amount of molybdenum representative of the thick coating that has formed. Fig. 12 shows the SEM and EDS of the acid-activated coating. The SEM appears almost featureless and again demonstrates that this coating does not have the cracked plate pattern. The EDS displays a considerably smaller molybdenum peak and a much larger uranium peak compared to the fluoride-activated coating and is indicative of a comparatively thinner coating. The thickness of the acid-activated coating is illustrated in Fig. 13. The coating was scratched to reveal the sidewall of the coating on top of the depleted uranium alloy substrate. As can be seen from

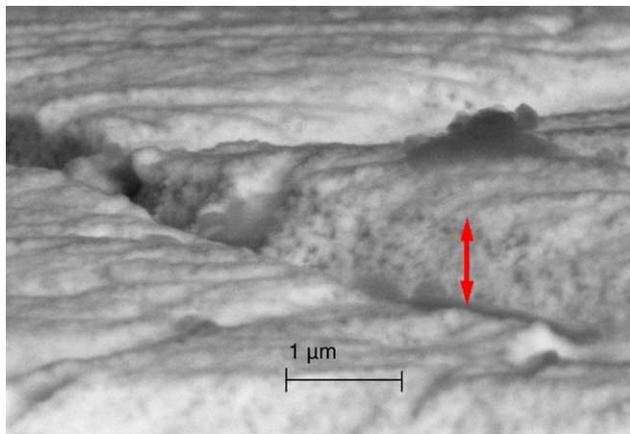


Fig. 13. SEM of a scratch through the acid-activated coating. The thickness of the coating is approximately 600–700 nm.

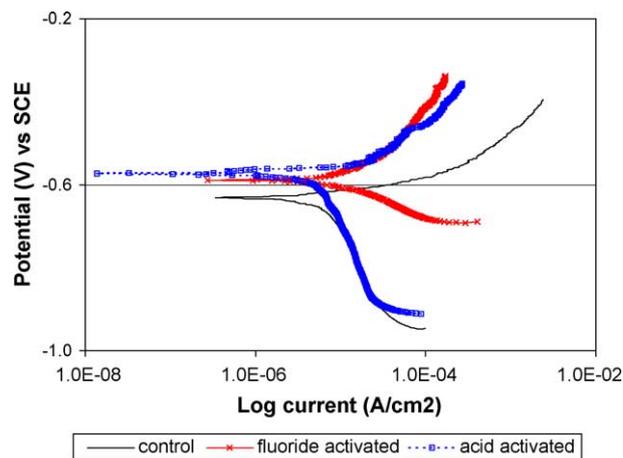


Fig. 14. Polarization of the coated samples as-made in quiescent 0.05 M NaCl shows little difference from the uncoated control sample.

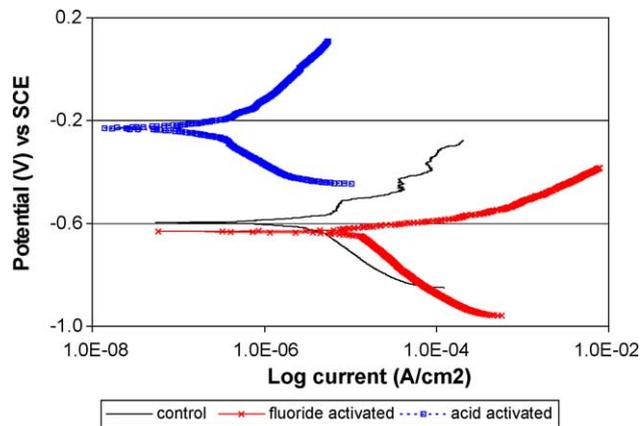


Fig. 15. Polarization of the coated samples in quiescent 0.05 M NaCl. After several weeks of aging, the potential is ennobled and corrosion current drops by an order of magnitude for the acid-activated samples. The fluoride-activated sample shows no change with aging.

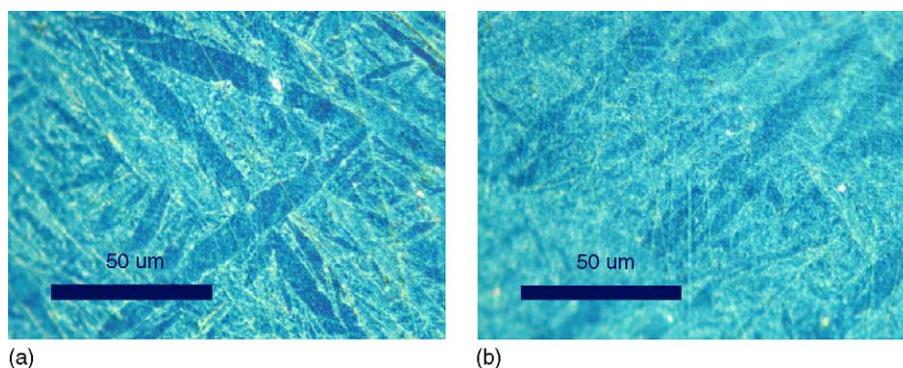


Fig. 16. Digital micrograph of an acid-activated coated sample. (a) Before polarization in 0.05 M NaCl and (b) after polarization in 0.05 M NaCl shows little change.

the scale on the figure, the acid-activated coating is approximately 600–700 nm thick. While the fluoride-activated coating is apparently thicker based on the EDS spectra, it was not explicitly measured because the fluoride-activated coating does not offer any corrosion protection to the DU alloy as will be demonstrated below.

In order to determine the effectiveness of these coatings in corrosion protection, potentiodynamic polarization measurements were made in quiescent 0.05 M sodium chloride solution at room temperature. The coatings were allowed to dry in ambient air for approximately 48 h prior to testing. A freshly polished sample of the DU alloy with no coating was included for a control. Fig. 14 shows the potentiodynamic polarization curves for a sample coated using fluoride activation, a sample coated using 50% nitric acid activation and an uncoated control sample. The as-made coatings show essentially the same electrochemical behavior as the uncoated control sample. However, as can be seen in Fig. 15, after several weeks of aging, the acid-activated sample outperformed the uncoated control sample as well as the fluoride-activated sample. The potential became ennobled

by almost 400 mV and the corrosion current density had dropped by an order of magnitude. The fluoride-activated coating has remained essentially the same as the uncoated control sample. The nature of the changes that occur in the acid-activated coating that enhance its corrosion protection properties during this aging period are currently being investigated.

Fig. 16 shows the surface of the acid-activated coating, Fig. 16a, before and Fig. 16b, after potentiodynamic polarization in quiescent 0.05 M NaCl. There has been no apparent change in the surface of the coating after exposure to the sodium chloride solution. Fig. 17a shows the surface of the fluoride-activated sample after polarization and the coating has been completely removed. Fig. 17b shows the interface between where the coating was in contact with the NaCl solution and where it was not exposed. The unexposed area shows the characteristic cracked plate structure while the exposed area has been completely stripped of the coating. This demonstrates that the fluoride-activated coatings lack the ability to impart any corrosion protection to the DU alloy when exposed to 0.05 M sodium chloride solution.

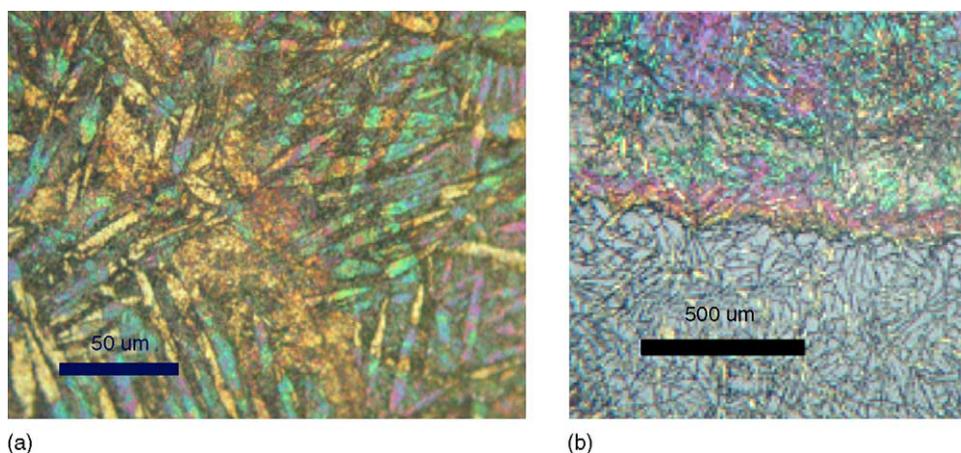


Fig. 17. Digital micrograph of a fluoride-activated coated sample. (a) After polarization of the sample in 0.05 M NaCl, the coating has been removed. (b) The interface of the area exposed to 0.05 M NaCl and the unexposed area.

4. Conclusions

Chemical activation of the DU alloy surface with fluoride solution allows for a molybdenum oxide coating to form on the depleted uranium alloy. However, any residual fluoride remaining from the initial activation step appears to interfere with the coating formation in the second step. The two-step fluoride activation process leads to a thick coating but this does not transpire into better corrosion protection. As-made samples performed no better than uncoated control samples in potentiodynamic polarization measurements done in quiescent 0.05 M NaCl and there was no improvement when the samples were allowed to age. For both as-made and aged samples, exposure to the sodium chloride solution during potentiodynamic polarization completely strips off the coating from the DU alloy.

The two-step nitric acid activation process yields a thinner yet more protective coating than the fluoride-activated process. The coating thickness is approximately 600–700 nm. The as-made acid-activated samples perform the same as the as-made fluoride-activated samples but after aging, the acid-activated samples show a significant improvement in potentiodynamic polarization measurements in 0.05 M NaCl, while there is no change in the performance of the fluoride-activated samples. The potential became ennobled by almost 400 mV and the corrosion current density dropped by an order of magnitude. These acid-activated coatings did not exhibit the damage that is seen in the fluoride-activated samples after potentiodynamic polarization in quiescent 0.05 M sodium chloride.

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