

Effect of niobium element on the electrochemical corrosion behavior of depleted uranium



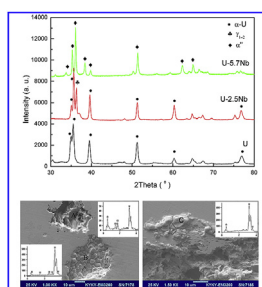
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HIGHLIGHTS

- New method (scanning Kelvin probe) was used to study the corrosion property.
- Three types of corrosion morphologies were found after potentiodynamic polarization.
- The effect of impurity elements on corrosion property was mentioned.
- The corrosion mechanism of DU and U-Nb alloys was discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Depleted uranium (DU) has many military and civilian uses. However, its high chemical reactivity limits its application. The effect of Nb content on corrosion behavior of DU is evaluated by scanning Kelvin probe and electrochemical corrosion measurements. The Volta potential value of DU and U-2.5 wt% Nb is about the same level, the Volta potential value of U-5.7 wt% Nb has a rise of 370mV_{SHE} in comparison with DU. The polarization current of U-5.7 wt% Nb alloy is about an order of magnitude of that of DU. The Nb₂O₅ is the protective layer for the U-Nb alloys. The negative potential of Nb-depleted α phase is the main reason of the poor corrosion resistance of DU and U-2.5 wt% Nb alloy.

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1. Introduction

Depleted uranium (DU) is widely applied in national defense and nuclear energy fields because of its high density and nuclear properties. However, DU is easy to react with the oxygen and water vapor in the environment due to its high chemical reactivity. The

corrosion products of DU also present environmental and health hazards [1–3]. A number of methods have been used to improve the corrosion performance, including alloying, surface coatings and ion implantation. Small quantity of metals (such as Titanium, Niobium, Zirconium and Ruthenium) has been alloyed with uranium to improve its corrosion resistance [4–6]. Protective coatings such as Zinc coating [7], Al-based coating [8–10], Ti-based coating [9,11] and Cr-based coatings [12,13], were prepared to enhance corrosion resistance. Molybdenum, nitrogen and carbon ions [14–18] had been implanted into DU to form a passivation layer.

Alloying is a primitive and effective method to improve the

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corrosion resistance [19,20]. The alloy elements content and the homogeneity are the critical factors to increase the corrosion resistance. Several methods have been used to investigate the corrosion property of DU and its alloys. Research was carried out by observing the change of the atmosphere and weight gain of the sample [19–21]. The electrochemical technique is considered as common and effective method to evaluate the corrosion behavior of the materials [22–25]. Researchers mainly paid attention on the electrochemical curves [26–36]. Less investigation remains the corrosion products morphology of the uranium alloys. Scanning Kelvin probe (SKP) technique has also been shown to be useful and versatile tool for corrosion study since Schmutz and Frankel [37] who first demonstrated the existence of linear relationship between Volta potentials, measured by SKP, and the corrosion potential of various metals. Also, Volta potential maps of a certain area are of great interest which can be used to evaluate whether the surface area is homogeneous or not [38,39].

The aim of the present study is to characterize the corrosion properties of DU and its alloys with different niobium content, U-2.5 wt% Nb (U-2.5Nb) and U-5.7 wt% Nb (U-5.7Nb), by SKP and electrochemical method, in order to investigate the influence of the Nb content and the homogeneity on the corrosion behavior of DU and its alloys.

2. Experimental procedures

2.1. Sample preparation

DU, U-2.5Nb and U-5.7Nb alloys were treated by quenching and annealing after casting. Then the samples were cut into disk of $\phi 16 \text{ mm} \times 2 \text{ mm}$. The samples were firstly abraded with up to 1000-grit SiC water paper and mechanically polished, then cleaned with alcohol and acetone for 5 min in an ultrasonic bath. All the electrochemical experiments were carried out immediately after the sample preparation to reduce the surface oxidation.

2.2. The scanning Kelvin probe tests

The SKP is a vibrating condenser and able to measure the difference of Volta potential (ΔV) between the probe (V_p) and the surface of samples (V_s) which are proportional to the difference in corresponding electron work functions, ϕ_p and ϕ_s [27], correspondingly, as shown in Equation (1), and e is the charge of the electron. The potential V_s of the samples can be obtained, the probe potential V_p is calibrated relative to a reversible electrode and kept constant.

$$\Delta V = V_p - V_s = \frac{\phi_p}{e} - \frac{\phi_s}{e} \quad (1)$$

An ac voltage of 1 V was applied between the tip and sample to induce oscillations of the cantilever. The probe of SKP vibrated at a frequency of 2 kHz and 30 μm above the sample surface. The scan step was 30 μm , and the testing area was 2 mm \times 2 mm. The SKP tests were carried out at room temperature, with a relative humidity of 60%. The Volta potential was measured with respect to Cu/Cu⁺ couple as a reference electrode versus the standard hydrogen electrode (SHE).

2.3. The electrochemical corrosion tests

The electrochemical tests were carried out using a PARSTAT 2263 electrochemical workstation equipped with three electrodes. The samples were set as the work electrode. A graphite electrode and a saturated calomel electrode (SCE) were used as the counter

and reference electrodes, respectively. The effective area of the samples was 1 cm² embedded in epoxy resin. The experiments were performed in 50 $\mu\text{g/g}$ Cl⁻ (KCl as the solute) solution with distilled water. The value of the PH was 7. The samples were dipped in the solution before test until the open circuit potential became stable. Then the linear polarization curves were acquired by the potential sweep in the anodic direction in the range of $\pm 30 \text{ mV}$ (vs. SCE). The scanning rate of the linear polarization was 0.167 mV/s. The scanning rate of the potential was 2 mV/s for potentiodynamic polarization tests.

The morphology of the sample surface after potentiodynamic polarization was observed by scanning electron microscopy (KYKY-EM3200) to evaluate the corrosion morphology of the samples. The energy dispersion EDX was used to analyze the composition of the corrosion product.

3. Results and discussion

3.1. Sample morphology characterization

After quenching and annealing, DU has an orthorhombic α crystal structure. The microstructure of U-Nb alloys depends on the niobium content. Fig. 1a shows the optical morphology of U-2.5Nb. It is found that U-2.5Nb alloy has a pearlite structure which is made up of α (Nb-depleted) and γ_{1-2} (Nb-enriched) double phase composition [6,26]. It can be confirmed by Fig. 2. From Fig. 1b, it can be seen that U-5.7Nb alloy has a single structure with several niobium patches which have been confirmed by EDX, randomly dispersed in grain interior and grain boundaries. The single α' martensite structure is a distorted structure of α -U [6,27,28].

3.2. The Volta potential tests

The Volta potential between the probe and sample surface was measured by SKP. The Volta potential maps of a certain area are of great use to judge whether the surface component is homogeneous. The existence of negative potential difference suggests that the area may be having an anodic behavior. Fig. 3 shows the Volta potential maps of DU and its alloy samples. The range of Volta potential value on the DU, U-2.5Nb and U-5.7Nb were $-700 \pm 100 \text{ mV}_{\text{SHE}}$, $-650 \pm 30 \text{ mV}_{\text{SHE}}$ and $-330 \pm 50 \text{ mV}_{\text{SHE}}$, respectively. There has been a small rise in the Volta potential of the U-2.5Nb in comparison with DU. Volta potential value of U-5.7Nb became ennobled by almost 370 mV relative to DU. Martin et al. [40,41] have shown that the open-circuit potential is linearly dependent on the Volta potential. The anodic shift of the Volta potential is due to a noble area which contributes to slow down the thermodynamics of corrosion. The Volta potential of DU and U-2.5Nb is at same range, which implies that the niobium additions below threshold concentrations have little effect on the corrosion resistance in comparison with depleted uranium [29]. Besides, it can be seen that the Volta potential of DU was less uniform compared with U-2.5Nb and U-5.7Nb. It can be concluded that DU has stronger galvanic coupling than the U-Nb alloys since the former presents significantly higher potential differences.

3.3. The electrochemical corrosion measurements

To quantitatively evaluate the corrosion behavior of DU and U-Nb alloys, linear polarization experiments were carried out. The DU and U-Nb alloys were immersed in the 50 $\mu\text{g/g}$ Cl⁻ solution until the fluctuated potential is lower than 2 mV to obtain the open circuit potentials (E_{oc}). A stable E_{oc} was taken to indicate that the system being studied has reached a “steady state”, i.e., the various chemical reactions had reached a constant rate. Table 1 presents the

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