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Corrosion and Dissolution of Intermetallic Compounds UPd3 and URu3 in 0.5-6.0 M HNO3

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The intermetallic compounds (IMC) of uranium and plutonium with Ru-subgroup metals are expected to be as a principal chemical state of Ru, Rh and Pd in spent (U,Pu)N fuel (SNF) of fast breeder reactors. Their effective dissolution is required for quantitative recycling of fissile materials during the fuel reprocessing.
The corrosion and dissolution of IMC UPd3 and URu3 in 0.5 to 6 M HNO3 electrolytes were studied using linear voltammetry (LV) and current controlled electrolysis (CCE). Corrosion potentials (Ecorr) and corrosion rates (vcorr) of UPd3, determined from Tafel plots, were found to increase from 0.22 to 0.699 V / Ag/AgCl and from 0.07 to 1.2.104 μg.cm-2H-1 correspondingly with the increase of HNO3 concentration from 0.5 to 6 M. The changes in Ecorr and vcorr for URu3 had the same trend and increased from 0.314 to 0.702 V / Ag/AgCl and from 90 to 4.3.103 μg.cm-2H-1. Transpassivation potentials (Etr) of UPd3 and URu3 were found to increase slightly from 0.45 V to 0.55 V / Ag/AgCl and from 1.12 to 1.2 V / Ag/AgCl correspondingly with the increase of HNO3 concentration from 0.5 to 6 M. The data of LV measurements indicate that UPd3 may be dissolved in 6 M HNO3 with the significant rate without application of external oxidation potential. The dissolution rate of UPd3 obtained using quantitative determination of U(VI) and Pd(II) in the electrolyte increased from 2.16 to 45 mg.cm-2H-1 with the growth of HNO3 concentration from 4 to 8 M. At the same time URu3 dissolution with significant rates required the electrochemical techniques, for instance, the CCE.
The rate of URu3 anodic dissolution in CCE conditions was practically independent on the HNO3 concentration in the electrolyte but increased significantly with the increase of anodic current density. At current densities below 12.5 mA.cm–2 (E = 1500 ± 40 mV / Ag/AgCl) the dissolution was slow and the accumulation of soluble U(VI) and Ru(IV) in the electrolyte was found to increase linearly in time. At greater anodic current densities the IMC dissolution was accompanied by the accumulation of a thick black film, apparently RuO2, at the electrode surface and corresponding slowing down the uranium leaching rate. At current densities 100 –180 mA cm-2 the part of Ru was oxidized with formation of the soluble species of Ru(IV) and Ru(VIII), confirmed by the measurements of UV-VIS spectra of the electrolyte. The part of Ru was found to be removed from the solution in a form of volatile RuO4. The material balance of URu3 dissolution in course of CCE in 6M HNO3 at 180 mA cm-2 allowed to estimate the rate of U(VI) leaching as 34.6 mg cm-2 H-1. The calculation of Ru content in the electrolyte based on the UV-VIS spectrophotometry shown that only 62±2 % of the metal were found in the solution, while the rest metal was distributed between gaseous stream and the solid film, covering the electrode surface.
The results of the presented research indicate that the retention of U and Pu in the insoluble residues of SNF dissolution could be determined by the Ru content in the IMC formed during fuel burn-up. To increase the rate of the IMC dissolution the electrochemical technique is proposed.

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