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Electrochemical dissolution of U –5 mass. % Zr alloy, steel 316 and zircalloy in nitric acid

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The advantage of electrochemical dissolution of metal or alloy spent nuclear fuel (SNF) consists in the alloy surface oxidation by application of external oxidation potential without adding the chemicals complicating following recovery of fissile materials during SNF reprocessing. The present work deals with the study of electrochemical properties and dissolution of U –5 mass. % Zr alloy simulating U –Zr alloy based SNF and the possible cladding materials, including steel 316 and zircalloy.

The principle dissolution characteristics of U –5 mass. % Zr alloy in 0.5 –6 M HNO₃ were determined using linear voltammetry technique. The corrosion potential (E_{corr}) value of U –5 mass. % Zr alloy electrode was found to increase with growth of HNO₃ concentration. Its value is expected to be determined by the formation of the passive film containing U(IV) and Zr(IV) hydroxides at the electrode surface and by the cathodic reduction of HNO₃. The overvoltage of the latter reaction at U –5 mass. % Zr in 0.5 –6.0 M HNO₃ was found to be high. The increase of HNO₂ yield due to the reduction of NO₃⁻ ions with the increase of HNO₃ concentration in the electrolyte results in the increase of alloy dissolution rate in absence of applied oxidation potential in the solutions containing more than 4 M HNO₃. At the potentials, exceeding 450 mV / Ag/AgCl (Etr), U –5 mass. % Zr passed to the transpassive state. The data of potential controlled electrolysis (PCE) shown that at the potentials close to Etr and fixed HNO₃ concentration the increase of the electrode potential increased the alloy dissolution rate. The systematic study of U –5 mass. % Zr alloy electrochemical dissolution during current controlled electrolysis in HNO₃ electrolytes revealed that the rate of anodic dissolution of this alloy reached 44 mgcm⁻²h⁻¹ and slightly dependent on HNO₃ concentration in electrolyte. It was demonstrated, that maximum yields of soluble U(VI) during electrochemical dissolution were achieved in 2 M HNO₃ and current density less than 100 mAcm⁻². *Formation of a precipitate consisting of crystalline ZrO₂ and X-ray amorphous U compounds at the anode surface accompanied the process of U- 5 wt. % Zr alloy dissolution, causing the decrease of the dissolution current efficiency from 7,2 to 8,6 F/mol with the increase of anodic current density.*

The process of electrochemical dissolution of claddings, consisting of stainless steel and zircalloy were studied in conditions of current controlled electrolysis. The rate of stainless steel electrochemical dissolution in 2-8 mol/l HNO₃ increased from 12±1 to 98±4 mgcm⁻²h⁻¹ with the increase of current density from 23 to 160 mAcm⁻². Current efficiency of a process was found to vary from 90 to 98%. It was shown that the dissolution parameters had not depend on HNO₃ concentration. Rise of the electrolyte temperature to 80 °C was found to be necessary for the electrochemical destruction of Zr-1wt. % Nb claddings. The rate of anodic dissolution of latter alloy reached 44 mgcm⁻²h⁻¹ and was slightly dependent on HNO₃ concentration in electrolyte.

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