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Electrochemical and corrosion behavior of a 304 stainless-steel-based metal alloy wasteform in dilute aqueous environments

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ABSTRACT

The electrochemical and corrosion behavior of a stainless-steel-based alloy made as a prototype metallic nuclear wasteform to immobilize ^{99}Tc, has been studied in a number of reference solutions ranging in pH from 4 to 10. The results showed the 47SS(304)-9Zr–23Mo prototype alloy contained at least five distinct phases with the majority of the Re, used as a Tc surrogate, contained within a Fe_2Mo intermetallic phase. Polarization studies showed this alloy exhibited generally passive behavior in a range of dilute aqueous environments. Impedance measurements indicated passivity breakdown events can occur and lead to localized corrosion, especially in slightly alkaline conditions.

1. Introduction

To meet the need for economic and sustainable nuclear energy production and to satisfy the requirements for controlled, proliferation-resistant nuclear materials management, fuel cycle technologies are under development to deal with the increasing inventory of spent nuclear fuel from commercial nuclear power plants [1–3]. The Department of Energy (DOE) in the USA is evaluating spent nuclear fuel processing options employing a range of aqueous-based and electrochemical processes [4]. Some of these waste streams could be efficiently accommodated in metal alloy waste forms. As part of the Fuel Cycle Research and Development (FCRD) program [5], Fe-based alloys are being developed and evaluated as potential waste forms [4,6–8].

A radionuclide of particular concern under waste disposal conditions is ^{99}Tc, a long-lived beta emitter produced by U fission in the fuel. This radionuclide potentially has a high mobility in oxidizing environments as pertechnetate (TcO_4^-). In the advanced UREX (Uranium Recovery by Extraction) separations process, a portion of the Tc inventory is extracted into the uranium recovery stream during fuel dissolution, in which it occurs as pertechnetate [9–11]. It is proposed that TcO_4^- can be separated from this stream, and reduced to be processed and immobilized as Tc metal, along with other transition metal fission product elements in an Fe-based [12,13] alloy. The development of an Fe-based waste form is being considered since the key high-melting temperature metallic elements (activation products like Tc, Mo, Ru, Pd, and Rh that form the epsilon particles in the fuel [5,14]) present in various waste streams can be dissolved in molten Fe at a processing temperature of 1600 °C and then incorporated in durable intermetallic phases. Initial alloys were made to evaluate the approach by processing a surrogate waste mixture with reagent iron to identify (1) the phases that formed, (2) the distribution of Re (as an initial material for the alloy testing and development of the experimental techniques), and (3) the amount of iron that was required to accommodate the waste metals in durable intermetallics.

Previous studies have attempted to characterize the properties of metallic wasteforms based on stainless steel-zirconium formulations and their ability to act as hosts for Tc, noble metal fission products (e.g., Ru and Pd) and actinides (primarily U but also Pu and Np) [15–17]. Generally, corrosion evaluations were based on short term electrochemical and corrosion exposure tests [18–20]. In this paper the corrosion performance of stainless steel-based alloys has been assessed based on a combination of electrochemical and longer term corrosion experiments in order to assess their capacity to retain Tc and other radionuclides. The waste form under development is composed of several intermetallic phases with a minor amount of an iron solid solution phase. Prototype alloys have been made with Type 304L and Type 316L stainless steels. The results of tests and analyses conducted with the alloy made with Type 304L stainless steel are presented in this paper.

The primary goal of this work is to understand the degradation mechanisms of the component waste form alloy phases and how they influence the radionuclide release behavior under a wide range of exposure conditions. The larger program goal is to develop a model to predict the long term release of radionuclides (most importantly ^{99}Tc) under the range of environmental conditions

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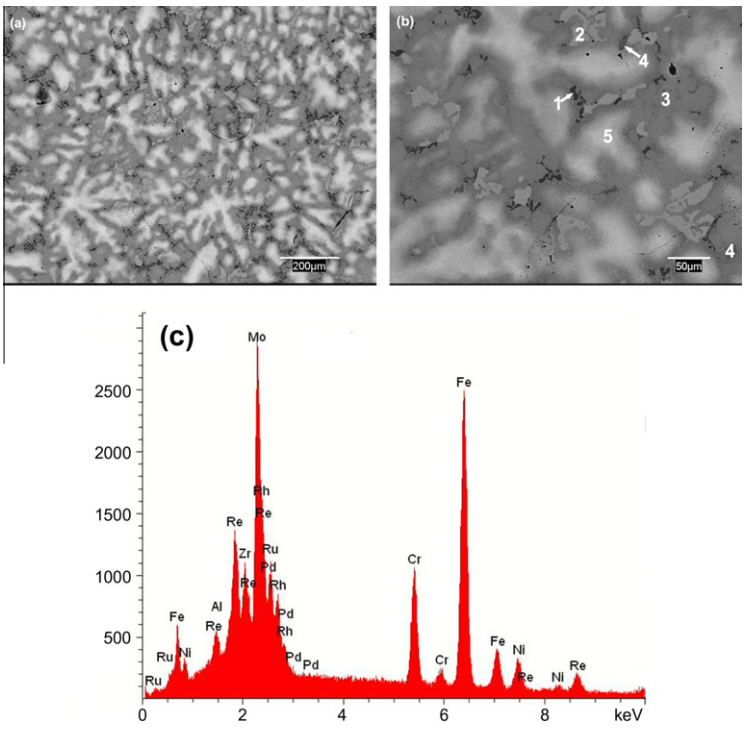

Fig. 1. Microstructure and EDS analysis of Alloy 1: (a) low magnification; (b) high magnification; and (c) EDS area analysis.

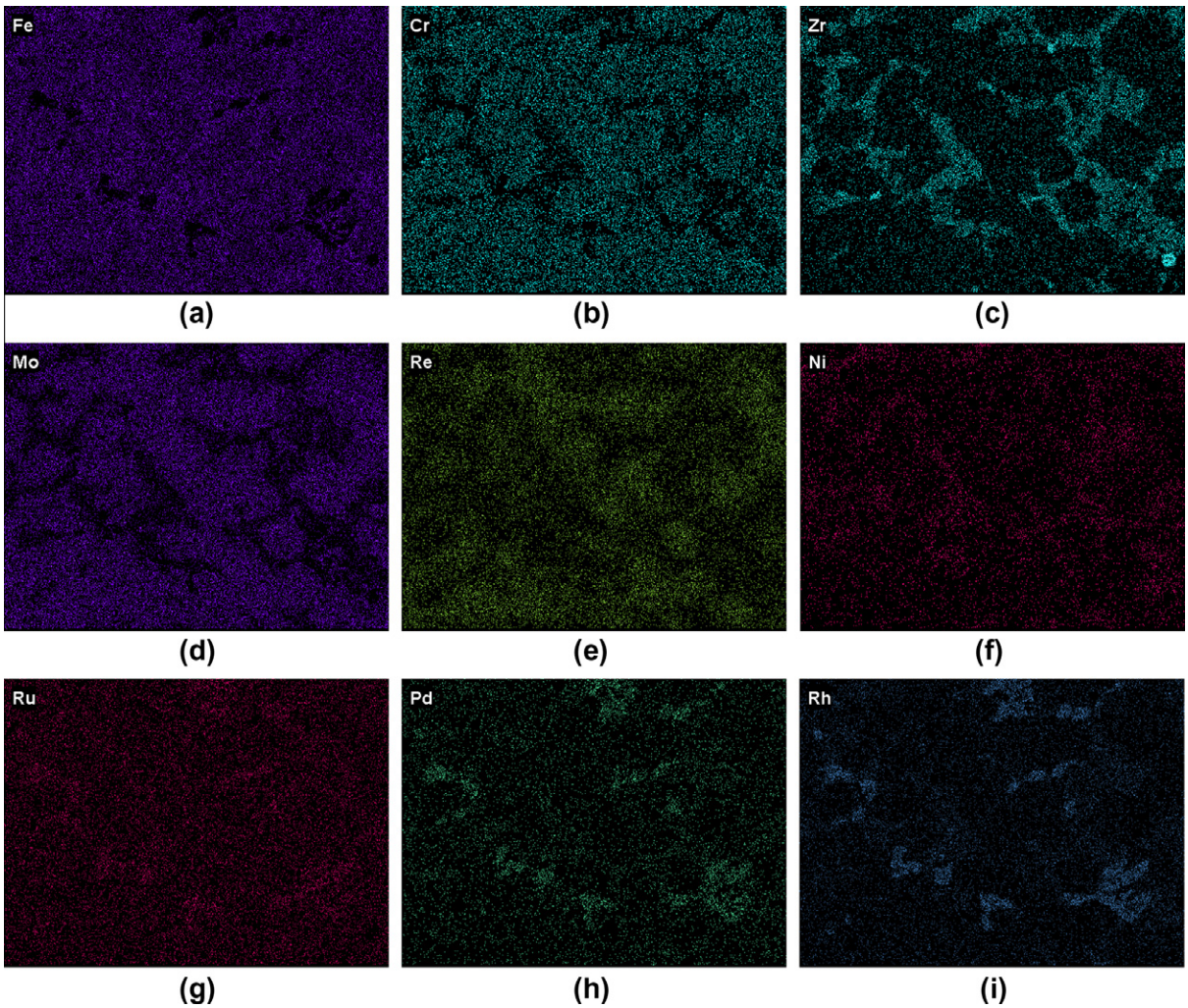

Fig. 2. Element distribution on the surface of Alloy 1 analyzed using EDS mapping (Fig. 1(b) shows the SEM image of this area on the same scale as the EDS maps).