U.S. DEPARTMENT OF ENERGY
INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE
DOE/ROK

ABSTRACT

Advanced Corrosion-Resistant Zr Alloys for High Burnup and Generation IV Applications

Primary Investigator (U.S.): Arthur T. Motta, Pennsylvania State University

Primary Investigator (Republic of Korea): Yong Hwan Jeong, Republic of Korea Atomic Energy Research Institute (KAERI)

Collaborators: Westinghouse Electric Company, LLC, University of Michigan, Hanyang University

Project Number: 2003-020-K

Project Start Date: January 2003

Project End Date: December 2005

A collaborative United States/Korea research program to develop Zr alloys for advanced nuclear fuel designs is proposed. In addition to Pennsylvania State University, the U.S. collaborators include Westinghouse Electric Company and the University of Michigan. The Korean collaborators are the Korea Atomic Energy Research Institute (KAERI) and Hanyang University. The objective of the program is to develop advanced, corrosion-resistant Zr alloys for extreme environments, focusing specifically on (i) high burnup applications in current light water reactors (LWRs) and (ii) cladding and reactor internal components in the supercritical water reactor (SCWR), a Generation IV reactor concept. Developing such alloys will permit higher duty operation of current fuel as well as fuel for new reactor designs targeted for near term deployment. In addition, development of corrosion-resistant Zr alloys will provide greater design flexibility and allow for economies of operation in the SCWR.

The proposed program builds on two highly successful NERI programs, which are now ending, and includes several world-class experts in Zr alloy corrosion and irradiation effects from both Korea and the United States. The collaborating organizations include, a major fuel vendor, three major research universities (two in the United States and one in Korea), and an internationally recognized research organization (KAERI). The proposed program will also employ the resources and expertise available at a major user facility at a U.S. national laboratory (the Advanced Photon Source at Argonne).

Waterside corrosion and the associated hydrogen pickup can be a limiting factor in the operation of Zr-based fuel cladding in current light water reactors (LWRs) and will be an important concern in future evolutionary and revolutionary designs called for under the Generation IV Reactor Initiative. In order to meet the more stringent economic demands of nuclear technology, advanced LWRs, and Generation IV reactor concepts, materials must operate under more severe conditions. Fuel cladding and structural materials must be able to perform at higher fluences, higher operating temperatures, longer residence times, and higher burnups than current operating limits. Therefore, it is crucial to improve the corrosion resistance of zirconium alloys for both near-term and long-term applications.

The first step in improving corrosion resistance is developing a clear understanding of the mechanisms of corrosion. During a previous NERI program, a combination of detailed characterization studies and modeling was used to identify some of the crucial parameters that govern corrosion behavior. These detailed studies, which are summarized in the proposal, were performed on complex commercial alloys. The studies provided valuable insight to the corrosion process but

2003-020-K (continued)

identification of individual mechanisms was difficult. Accurate determination of mechanisms must come from model binary and ternary alloys that are specifically designed to isolate the effects of individual parameters on the corrosion process.

The objective of this program is to develop and demonstrate a technical basis for improving the corrosion resistance of zirconium-based alloys in aqueous reactor coolants. In particular, the goal of the proposed research is to develop Zr-based alloys with superior corrosion resistance relative to the current state-of-the-art alloys used in LWRs, namely, Zircaloy-2, Zircaloy-4, ZIRLO, and the Zr-Nb alloys containing 1.0 and 2.5% Nb. These existing commercial alloys were formulated largely through empirical methods of alloy addition, testing, evolutionary optimization of composition and thermo-mechanical processing. Incremental improvements using this classical approach are probably still possible. However, a more fundamental understanding of the effect of alloy chemistry and microstructure on the structure and degradation of the protective barrier oxide is necessary to achieve significant improvements in corrosion resistance. The focus of the proposed approach, therefore, is to characterize the effect of individual chemical and metallurgical variables in selected alloys on oxide properties and to identify those factors that significantly reduce the corrosion rate. This knowledge will serve as the basis for the design of new alloy compositions and processing routes.

Specifically, a series of *model alloys* will be prepared by vacuum arc melting small button ingots that will be reduced to strip by thermo-mechanical processing and autoclave tested. Two series of model alloys will be manufactured and tested. The first series is designed to elucidate the role of solute atoms in the Zr matrix on the corrosion rate (focusing on valence effects and solute concentration). The second series is designed to elucidate the role of precipitates in the corrosion process (focusing on precipitate size, volume fraction, and precipitate type).

These alloys will be tested in different autoclave environments to determine the growth kinetics of the protective oxide and the oxide thickness at transition. These oxides will be characterized using an array of advanced characterization techniques to determine the relationship between oxide microstructure and the two parameters that control corrosion rates (oxygen transport and transition thickness). These characterization techniques include synchrotron radiation microbeam x-ray diffraction and fluorescence (techniques developed at the Advanced Photon Source at Argonne in a current NERI program), cross sectional transmission electron microscopy (TEM), oxide stress measurements, and nano-indentation. The synchrotron techniques provide unique and hitherto unobtainable information about the oxide that, combined with the detailed TEM and mechanical characterization, will provide a much more complete and detailed picture of the oxide microstructure than has been obtained to date. Such data will allow the detailed mechanistic modeling of corrosion.

The analysis of the experimental results will help answer fundamental questions related to the corrosion process. In particular, the program will develop a scientific basis for the well-known empirical correlations between corrosion rates and alloy chemistry/processing and investigate the operating range of Zr alloys at high temperature. The scientific and technical benefit of this program will come from an increased ability to predict corrosion behavior and in the availability of alloys that exhibit superior corrosion performance under severe duty cycles in current and advanced LWRs and in the SCWR.