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Zirconium in the Nuclear Industry

17th International Symposium

STP 1543

Editors
Robert J. Comstock
Pierre Barbéris

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Foreword

This Compilation of *Selected Technical Papers*, STP1543, *Zirconium in the Nuclear Industry: 17th International Symposium*, contains peer-reviewed papers that were presented at a symposium held February 3–7, 2013 in Hyderabad, India. The symposium was sponsored by ASTM International Committee B10 on Reactive and Refractory Metals and Alloys and Subcommittee B10.02 on Zirconium and Hafnium.

The Symposium Co-Chairmen were Pierre Barbéris, Areva/Cezus Research Centre, Ugine, France and Srikumar Banerjee, Atomic Energy Commission, Anushakti Bhavan, Mumbai, India.

The STP Editors are Robert J. Comstock, Westinghouse Electric Company, Pittsburgh, PA, USA and Pierre Barbéris.

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Corrosion and Hydrogen Pickup

In Reactor Performance

Irradiation and Hydrogen Effects

High Temperature Transient Behavior

Degradation and Failure Mechanisms

Overview

This STP contains the papers presented at the *17th International Symposium on Zirconium in the Nuclear Industry* held in Hyderabad, Andhra Pradesh, India from February 3–7, 2013. The first symposium was held in Philadelphia in 1968 with subsequent symposia held every two to three years. The proceedings of each symposium in the series have been documented with an STP.

During this symposium, the William J. Kroll Zirconium Medal was presented to John Kearns (2011 winner) and Srikumar Banerjee (2012 winner) for their unique and lasting contributions to the technology of zirconium alloys. Both provided historical perspectives of their research during the symposium and contributed papers that are included in the STP.

The symposium was truly international; with approximately 130 participants from 17 countries attending and representation from North and South America, Europe, and Asia. The 17th Symposium included 42 platform presentations along with a session with 31 posters. This STP contains 38 peer reviewed papers along with the papers from the two Kroll winners. In addition, the discussion of each platform presentation provided an opportunity for further insight and understanding of the paper. As in past symposia, the questions along with written responses by the authors are included at the end of each paper.

The symposium is an opportunity to capture a snapshot of the current research areas that are relevant to the nuclear industry. This symposium was no exception. While the papers are grouped into seven categories (*Basic Metallurgy and Alloying Effects, Fabrication, Corrosion and Hydrogen Pickup, In Reactor Performance, Irradiation and Hydrogen Effects, High Temperature Transient Behavior,* and *Degradation and Failure Mechanisms*), there is often overlap between the topics as you will see when you browse through the STP or delve into papers more deeply.

An important component of the symposium is the in-reactor performance of zirconium alloys with several papers presenting recent results from materials irradiated to high burnups and fluence.

- Alloy E635 was irradiated to 72 MWd/kgU in VVER-1000 reactors with performance data presented from both fuel cladding and structural components. The behavior of the material was correlated to both temperature and neutron fluence.
- The evolution of the microstructure of M5™ fuel rod cladding irradiated in a PWR up to 7 cycles with fast fluence up to 17.1×10^{25} n/m^2 was described. In a separate paper, results from M5™ with 1000 ppm Fe (designated M5-Fe)

included both oxide thickness measurements at burnups of about 65 MWd/tU and free growth at fluences to 20×10^{25} n/m^2 (E > 1Mev).

- ZIRLO® structural components were characterized for both corrosion and dimensional changes following irradiation in PWRs with a maximum fluence of 13.6×10^{25} n/m^2 (E > 1Mev).
- Optimized ZIRLO™ fuel cladding was irradiated beyond the license limit of 62 GWd/MTU to 70 GWd/MTU. Characterization of the cladding included corrosion, dimensional changes, and mechanical properties.
- A detailed study was presented on the influence of temperature and microstructure on the irradiation creep behavior of Zr-2.5Nb pressure tubes.

A dominant theme in this STP is the role of hydrogen on the performance of zirconium alloy components. Issues discussed in this volume where performance was dominated by hydrogen included the following:

- Failure of BWR fuel rods was attributed to the localization of hydrides following accelerated corrosion and subsequent cracking of the hydride lenses. Despite an extensive investigation, the cause of the accelerated corrosion was not definitively identified.
- The growth of beta-quenched Zircaloy-2 BWR channels was driven late in life by accelerated hydrogen pickup that coincided with the dissolution of second phase particles.
- As reorientation of hydrides plays an important role during dry storage, in-situ measurements were performed to gain new insights into the reorientation of hydrides in Zircaloy-4.
- Delayed hydride cracking (DHC) growth rate of in-service Zr-2.5Nb CANDU pressure tubes was controlled by thermal and irradiation effects on the microstructure (e.g, decomposition and reconstitution of the beta phase controlling hydrogen diffusion to the crack tip).

In addition to papers that highlight the impact of hydrogen, several papers focused on understanding the mechanisms of hydrogen ingress into the metal or understanding the interaction of hydrogen with point defects and dislocation loops in the matrix. The latter has potential implications related to hydrogen assisted irradiation growth.

The US Nuclear Regulatory Commission proposal of a rule to amend the current requirements governing emergency core cooling system for light nuclear power reactors has prompted renewed activity in cladding ductility following a high temperature transient. Papers addressed different aspects of the high temperature oxidation behavior of cladding, including secondary hydriding following rod burst, the detrimental role of nitrogen on oxidation kinetics, and the impact of a pre-oxide or steam pressure on oxidation and subsequent cladding ductility. One paper demonstrated

the improved oxidation performance of E110G relative to E110. Unfortunately, the reason for the improvement remains an area for continued research.

In response to the Fukushima Daiichi nuclear accident in March, 2011, significant attention has been given to improving the accident tolerance of zirconium alloy fuel cladding. Researchers presented one approach through the application of coating technology to improve the high temperature oxidation resistance of the cladding. While significant development work remains, this is an area that will likely receive continued attention to identify viable options.

The zirconium community continues to push the limits of state-of-the-art analytical techniques to characterize the microstructure of both non-irradiated and irradiated zirconium alloys. Techniques such as synchrotron radiation (e.g., μ x-ray absorption near edge spectroscopy, diffraction, and stress measurement), electron back-scattered diffraction, atom probe tomography, secondary ion mass spectroscopy, and electron energy loss spectroscopy have become routine analytical tools in multiple laboratories around the world. Cold neutron prompt gamma activation analysis was successfully used in one study to non-destructively measure hydrogen content in corrosion coupons. Complementing the analytical characterization techniques are modelling efforts designed to facilitate mechanistic understanding of performance phenomena.

Following the symposium, a committee of technical experts covering a breadth of experience in the zirconium nuclear industry selected the best paper based upon technical excellence, relevance to the nuclear industry, and 'groundbreaking' research. The winner of the John H. Schemel Best Paper Award was the paper entitled "Effect of Hydrogen on Dimensional Changes of Zirconium and the Influence of Alloying Elements: First-Principles and Classical Simulations of Point Defects, Dislocation Loops, and Hydrides" by M. Christensen, W. Wolf, C. Freeman, E. Wimmer, R. B. Adamson, L. Hallstadius, P. Cantonwine, and E. V. Mader. Congratulations to the winners.

Robert J. Comstock
Pierre Barbéris

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