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Zirconium in the Nuclear Industry

16th International Symposium

JAI Guest Editor
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Foreword

This publication, *Zirconium in the Nuclear Industry: 16th International Symposium*, contains papers presented at the symposium with the same name held in Chengdu, Sichuan Province, China, May 9-13, 2010. The sponsor of the symposium was ASTM International Committee B10 on Reactive and Refractory Metals and Alloys.

The Symposium Chairman was Magnus Limbäck, Westinghouse Electric Sweden and Co-Chairman Zhao Wenjin, Nuclear Power Institute of China (NPIC), Chengdu, Sichuan Province, China. Serving as Guest Editors of this publication are Magnus Limbäck and Pierre Barbéris, Areva/Cezus Research Centre, Ugine, France. Arthur Motta, Pennsylvania State University, acted as Associate Editor for the publication of these papers in Journal of ASTM International (JAI).

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Hydriding – Hydrogen Effect

Corrosion – Oxide Layer Characterization

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Failure Mechanisms and Transients

Overview

This STP contains the papers presented at the 16th International Symposium on Zirconium in the Nuclear Industry held in in Chengdu, Sichuan Province, China, May 9-13, 2010. The first symposium was held in Philadelphia in 1968, and symposia have been held ever since in two to three year intervals. The proceedings of each symposium in the series have been documented with an STP.

This symposium series remains, after forty years, one of the top presentation and information source for the research in the area of zirconium alloy performance in a nuclear reactor environment. 42 papers and 32 posters were selected for presentation at the 16th Symposium from 130 abstracts submitted. The forty-two papers published in these proceedings were peer reviewed and edited, and are also published in the ASTM online journal, JAI. In addition, the most significant parts of the discussions that followed the oral presentation of each paper at the symposium are included in these proceedings.

Four experts in zirconium area received their Kroll Awards at the 16th Symposium: J. Banker, D. Franklin, V. Shishov and B. Cheadle. Noteworthy is the fact that the first one deals with zirconium outside the nuclear industry. These papers as well as the 26 previous Kroll award papers are now gathered in "The Kroll Medal Papers 1975-2010" published by ASTM, and covering all aspects of zirconium technology.

137 attendants from 19 countries attended the 16th Symposium. North and South America, Europe, and Asia were represented.

The papers were presented during seven sessions, covering the whole spectrum of zirconium metallurgy, from basic metallurgy to accidental conditions and transport, through fabrication, creep and growth, and corrosion and hydriding. Looking back from the beginning of this symposium series, it appears that these topics remained quite constant over the time.

Besides the historical alloys (Zircaloy-2, Zircaloy-4, Zr2.5Nb and Zr-lNb), several studies were devoted to advanced alloys and alloys under development: ZIRLO™, M5™, X5A, VB, N18, N36, NZ2, E635, ZrNbSnFe with low tin content, Ziron. Some optimisation of Zircaloy-2 was proposed.

Modelling appears more and more intricate with the experiments, from precipitation to VAR melting, from the effect of texture and dislocations on creep to the oxygen distribution during LOCA situations or the outside-in cracking in BWR fuel cladding.

As noted in the past few symposia, advanced techniques are more systematically utilised: high temperatures studies of phase transformations using

synchrotron radiation or neutrons diffraction were presented; EBSD allows measuring local texture, studies on corrosion and hydriding benefit from the precise positioning of thin foils with FIB.

Last, it is worth noting that several studies were presented by enthusiastic young searchers or PhD students, which demonstrates the dynamism of the research in the zirconium metallurgy area.

In the field of basic metallurgy, progresses were reported in the following fields:

- plotting the dynamic recrystallisation domain thanks to processing maps, which were shown to depend on the alloy composition.
- experimental quantification of the precipitation of beta Nb second phase particles (SPP) in Zr-Nb alloys, which was investigated by synchrotron radiation and then modelled.
- texture change and variant selection through the α-β-α phase transformation, wich generated some controversial results on the influence of an externally applied stress.

The processing of zirconium was illustrated first by the VAR melting, with modelling of the alloying element segregation allowing efficient control on the ingot chemistry, then by the investigation of the damage mechanism during pilgering, which is alloy dependent through the number and size of the second phase particles. Transmission electron microscopy was used with the aim of investigating the dislocation microstructure of recrystallised Zircaloy-4 after 400°C creep test to develop a model of the anisotropic viscoplastic behaviour.

Three papers were devoted to Zr-2.5Nb tubes. The first one presented a self-consistent modelling of their anisotropic behaviour, which takes into account not only the texture but also the dislocation microstructure resulting from the last cold drawing pass. A second paper showed that the tubes for the future ADC will procure reduced creep variability, owing to a better understanding of its dependence on the microstructure and to an improved extrusion process. A detailed study of the microstructure evolution during annealing and of its relation with texture and mechanical properties of the tubes was also conductive to improvement in the tube fabrication sequence.

The investigation of hydrogen effects and hydriding constituted an important topic of this symposium with eight papers dealing with this topic. Besides a paper showing the capability of neutron radiography to investigate this area, an investigation of Zr-2.5Nb hydriding during autoclave tests showed that the hydrogen pick-up was linked to the sample position in the tube (back end/front end) and correlated to various microstructural parameters. An investigation of autoclave corroded samples from different alloys

by TEM, RAMAN and SIMS led to the hypothesis according to which the rate controlling process for hydrogen absorption was the diffusion of hydrogen ions in the oxide barrier layer, the best alloys having a higher protective layer due to compressive stresses and Fe dissolution from the second phase particles. Another paper confirms that the hydrogen pick up during corrosion is closely related to the size, area fraction, and compositions of the second phase particles.

The hydride dissolution, re-orientation and stress could be followed in situ by synchrotron radiation diffraction during temperature and stress cycles. The hydride re-orientation was the subject of a second paper coupling mechanical tests on irradiated cladding and finite element modelling, showing the importance of the hoop stress, and evidencing some other parameters. Two papers dealt with delayed hydride cracking (DHC). The first one which was derived from an IAEA coordinated research program investigated the effect of the microstructure on DHC, showing its main role is to control the material strength, while the second one presented in situ observation in an SEM of the crack propagation during a DHC experiment and a FEM of the accepted mechanism.

Two papers on the corrosion mechanism and oxide layer characterization showed on the one hand that the delayed oxidation of the SPPs in the oxide layer lead to the formation of small cracks, and on the other hand the influence of the grain substrate orientation on the oxide layer thickness and epitaxy during steam corrosion tests.

The in-pile behaviour was the subject of numerous studies. In the aim of mitigating the accelerated HPUF tendency at high burn up, it was proposed to decrease the nickel content and increase the iron and chromium contents in Zircaloy-2 within the ASTM specification. Zircaloy-2 was also the subject of a detailed study of the SPPs' evolution, showing that Zr(Fe,Cr)2 SPP amorphize while Zr2(Fe,Ni) SPP remain crystalline during irradiation. The shadow corrosion due to galvanic coupling between a zirconium alloy and a more noble metal was investigated by photo-electrochemistry; the alloy influence was evidenced, and it was postulated that a coating on the fuel assembly spacer may mitigate the shadow corrosion.

Annealing and DSC experiments on irradiated Zircaloy-4 enabled the study of the influence of the neutron damage on the hydrogen solubility, while the morphology of hydrides and SPP was investigated by TEM.

The in pile behaviour of advanced alloys were presented: X5A (for PWR) with two different final heat treatments showed improved properties and that the irradiation creep of ZIRLO™ is linear with the (deviatoric hoop) stress. The same phenomenon is observed in tension and compression. The increase in tin content up to 0.3 % in Zr1NbSnFe does not significantly modify the corrosion resistance nor the hydrogen pick-up compared to Zr1Nb

alloy, while ensuring a higher creep resistance and an improved dimensional stability.

Radiation damage of E635 in VVER (SPP evolution, ⟨c⟩-dislocation loop formation) was investigated and found in agreement with the model samples in the BOR-60 reactor.

The creep and deformation during or after irradiation, already evoked in the previous section, was illustrated by four more papers. No effect of commercial reactor irradiation temperature or hydrogen content was found on the Zircaloy-4 guide tube creep. Zircaloy-4 irradiation creep law was deduced from relaxation experiments on bent beam specimens in OSIRIS. Creep deformation of Zr-1%Nb and Zircaloy-4 during transportation was associated to a significant recovery of irradiation damage, preventing the dislocation channelling. Shadow corrosion-induced bow of Zircaloy-2 channels was associated to differential hydrogen concentration on channels sides adjacent to and away from the control blades.

Four papers dealt with accidental conditions. In LOCA conditions, a diffusion model was developed to compute the oxygen distribution, closely related to the mechanical properties, and compared with experiments. An investigation on the effect of hydrides on mechanical properties during RIA concluded that high strain rates did not seem to impact the stress strain behaviour when the hydrogen content is higher than 400ppm, threshold over which the failure elongation at room temperature decreases drastically. When radially oriented hydrides are present in BWR cladding, an outside-in cracking can occur, which was investigated, in isothermal conditions or with a radial thermal gradient: the outside-in cracking during the power ramp seems strongly dependent on the distribution of dissolved hydrogen as a result of thermal diffusion. Finally, a high burn-up BWR fuel rod, subjected to RIA tests in a research reactor, resulted in cladding failure at room temperature, but not at elevated temperature. A mechanical test was developed to reproduce the ramp test and was shown to be predictive.

The John Schemel Award is awarded following each symposium for the best paper presented at the symposium. The selection is based upon the technical content of the paper, the usefulness of the work reported to the worldwide reactor components community, and the technical difficulty in doing the work. This year, a committee of technical experts in several aspects of the zirconium industry selected the paper entitled “Photoelectrochemical Investigation of Radiation Enhanced Shadow Corrosion Phenomenon” by Y.-J. Kim, R. Rebak, Y-P. Lin, D. Lutz, D. Crawford, A. Kucuk and B. Cheng to receive the John Schemel Award.

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