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#### HANDBOOKS & REPORTS

# THE EFFECT OF HYDROGEN AND HYDRIDES ON THE INTEGRITY OF ZIRCONIUM ALLOY COMPONENTS: Hydride Reorientation



#### Preface:

Hydride orientation has an important effect on fracture toughness of hydride-containing zirconium alloys because hydrides form as approximately linear arrays of platelet-shaped microscopic precipitates with habits on or near the basal planes of the  $\alpha$ -Zr matrix in which they form. In the highly textured fuel cladding and pressure tube components used in nuclear applications these stringers have predominantly two tube orientations: circumferential and/or radial. Fracture of hydride precipitate clusters oriented in the radial direction are considerably more damaging, resulting in a significant reduction in the overall fracture toughness of these tubes compared to fracture of hydride precipitates elongated in the circumferential direction. Early on, as a consequence of the foregoing, manufacturing processes were sought and found that resulted in the precipitation of mostly circumferentially oriented hydride stringers. However, it was also found that

application of a sufficiently large external tensile hoop (circumferential) stress applied during cooling on these tubes could result in the reorientation of hydride stringers from the circumferential to the radial direction. Such external tensile stresses could result from a variety of sources such as during abnormal reactor service conditions or when fuel cladding tubes are removed from wet storage locations, dried and transported for placement in longer-term dry storage facilities.

This Stand Alone Report (SAR) addresses a key aspect of the issues raised in the foregoing by providing a comprehensive, self-contained and up-to-date review and analyses of the results of studies carried out on the conditions governing hydride orientation in zirconium alloy pressure and fuel cladding tubes used in nuclear reactors. The report combines a detailed theoretical and experimental overview of this subject with the author's own analyses of these results. These analyses make use of theoretical advances documented in the author's 2012 book dealing with the effects of hydrogen and hydrides on the integrity of zirconium alloy components. In the author's 2012 book, emphasis is placed on delayed hydride cracking, which is a localised failure mechanism. The present book is a follow-up to the foregoing study but dealing exclusively with the conditions affecting the orientation of hydrides in the bulk of the material. Although the ultimate objective of this study is the development of predictive models for the effect of hydride precipitates on the overall fracture toughness of zirconium alloy components, the topic of this SAR has been restricted to the study of factors controlling hydride reorientation. This restriction was necessitated by the complexity and number of studies found in the literature on this subject. Nevertheless, this report also provides information on the stresses and strains in reoriented and unreoriented hydride clusters of relevance to the susceptibility to fracture of these precipitates. Overall, emphasis is on fuel cladding tubes as these pick up considerably more hydrogen to the end of their design life than do pressure tubes and therefore the former have been more extensively studied.

The report starts with a brief summary of the results of the earliest experimental studies on hydride orientation and their effect on fracture toughness. This is followed by a detailed updated derivation of the Ells/Puls theory of stress-driven hydride reorientation which is then used to predict the results of experimental studies on hydride stress reorientation, starting from the earliest to the most recent ones found in the literature, including a large number of recent results obtained from in situ synchrotron X-ray studies. These latter types of studies have enabled real time observations of the evolution and characteristics of hydride precipitation during cooling and heating with and without external stress. Making use of Eshelby's theory of the stress state within and at the boundary of misfitting inclusions, an analysis of the strain variations in hydride and matrix during heating and cooling is then given by this writer. It is shown by means of this analysis that it is possible to rationalise semi-quantitatively most of the extant in situ synchrotron X-ray results. A detailed derivation is given of the mathematical formulation of the phase field methodology as applied to the study of the evolution of hydride precipitate morphology and stress state in zirconium alloys under various thermomechanical cycles. The emphasis of these calculations is to show how this methodology can simulate graphically the distribution, shape and orientations of hydride precipitates similar to those observed in optical metallography. These phase field methodology simulations are supplemented by the results of ab initio atomistic models, allowing for the determination of the possible crystal structures and hydrogen compositions of hydride precipitates that may form during the nucleation and early growth stages of hydride precipitation. An abridged version covering all of the foregoing topics is given in the summary and conclusion section in which sufficient detail is given to provide the reader with a self-contained, yet still relatively concise, overview of the results of the foregoing, rather complex theoretical and experimental studies.

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## The Author



**Dr. Manfred Puls** has a PhD degree in Physics from McMaster University, Hamilton, Ontario, Canada. After spending a year as a post-doctoral fellow at McMaster, he joined AECL, retiring from AECL in 2006 after 35 years of service. The first 20 years of service in AECL were spent as a researcher, whilst the remaining 15 were spent in various management positions, the last 9 of which were at AECL's Engineering Company located in Missisauga, Ontario. At that site he managed a resource group of engineers consisting of many of the company's experts for the design, build support,

development, life management and service of fuel channel technology in CANDU reactors.

Throughout his graduate studies and his stay at AECL, Puls was involved in research, design and development activities covering a wide range of technical topics and fields, only a few of which are noted here, as follows:

- phase transitions and phase equilibria (hydride formation, critical phase transitions),
- diffusion controlled processes (delayed hydride cracking, creep crack growth);
- fracture (critical crack propagation, delayed hydride cracking, hydride blister fracture);
- properties of point defects and dislocations and their interactions in crystalline solids based on atomistic models of dislocations and point defect/dislocation interactions in ionic and metallic crystals;
- development of an improved calandria tube design and specifications for the manufacture of improved pressure tubes.

After retirement from AECL, Puls worked as a consultant with Kinectrics Inc. (the privatised successor company to the Ontario Hydro Research Division) on topics associated with ensuring that DHC can be prevented in pressure tubes of operating CANDU reactors. In 2012 Puls published a book through Springer-Verlag, U.K. The book has the same main title as the SAR being marketed in this brochure. The focus of the 2012 book was on DHC whilst the SAR being promoted in this brochure deals with the science and conditions for hydride reorientation, given that the orientations of hydride precipitates play an important role in impacting the fracture toughness of zirconium alloys.

Further details of Puls' research activities and publications can be found under his ResearchGate profile <u>https://www.researchgate.net/profile/Manfred\_puls2</u>.

Puls has been a member of various international and national organizations such as:

- Canadian representative to the International Working Group on Life Management of Nuclear Power Plants (IWG-LMNPP); an IWG operating within the Nuclear Power Division of the International Atomic Energy Agency, Vienna, Austria.
- Chairman of the CANDU Owners Group (COG) DHC & Fracture Working Party and member of the Fuel Channel Program Technical Committee.

- Member of, and contributor to, the Subcommittee on Technical Requirements for In-Service Evaluation of Zirconium Alloy Pressure Tubes in CANDU Reactors, a committee responsible for the technical content of a new CSA Standard, CSA N285.8, Technical Requirements for In-Service Evaluation of Zirconium Alloy Pressure Tubes, the first version of which was published in 2005.
- Member of the Editorial Advisory Board of the Journal of Nuclear Materials.

Read Dr. Puls' full biography

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