

ZIRATTM Zirconium Alloy Technology Programme



The Annual ZIRAT Programme is focused on fuel assembly material issues and open to nuclear utilities and laboratories. In total, 33 organisations were members in 2015, representing more than 93 nuclear units worldwide. This programme was started in 1996.

Deliverables

ANT International will provide the ZIRAT Members with the following:

- Searchable CD-ROM(-s) with the following contents:
 - High-resolution pdf files with the complete ZIRAT Annual Report and the Special Topic Reports in colour.
 - > The files can be copied to a company server, with full read access for everybody with access to the server.
 - > The contents from ZIRAT Annual Report and the Special Topic Reports in pdf-format can be printed. Also, the contents from the pdf-files can be copied and pasted electronically into

other documents, e.g. Word files.

The CD-ROM(-s) of the ZIRAT AR and of the STRs will be provided before the Seminars (see below).

- Hardcopy, four colour reports will be provided if requested. The printed ZIRAT Annual Report and Special Topic Reports will be provided as soon as they are printed with the aim of delivery before the Seminars in February–March.
- Three similar Seminars will be held to present the results of the ZIRAT Programme in USA, in Europe in February-March and in China. The number of full time employees per Member that may attend meetings is limited to eight (8) people per organisation. The language of the ZIRAT Programme will be English.
- The authors will be available for consulting throughout the year. A few telephone or e-mail consultations requiring no additional work are provided at no additional cost to Members.

Listen to Frank Holzgreve, BKW.





Listen to Jean-Paul Dalleur, Tractebel.

Listen to Jorge Benavides Peña, CNAT.





ZIRAT22 Programme

The overall objective of the ZIRAT Programme is to enable the nuclear utilities and laboratories to:

• Gain increased understanding of material behaviour related to successful core operation and evaluations of options for the back end of the fuel cycle.

The objective is met through review and evaluation of the most recent data on zirconium alloys, identification of the most important new information, and discussion of its significance in relation to fuel performance now and in the future. Included in the review are topics on materials research and

development, fabrication, component design and in-reactor performance.

The evaluations are based on the large amount of non-proprietary data presented at technical meetings, published in the literature and provided through discussions with zirconium materials manufacturers.

The open literature information will be collected throughout the year and the data most important to the utilities and laboratories will be selected for the Annual Report. The large collective experience gained by the reviewers in past and current projects is an important factor in making the evaluation, hence ensuring that the presented compiled information is put in perspective, and that the most important information is emphasised. The data will be useful to utilities and laboratories to assist them in evaluating:

- New and potential fuel performance problems and performance limits.
- The effect of new data on current fuel design bases.
- Qualifications desirable for new design features.
- The effect of modified or new fabrication processes on properties.
- Potential use of new Quality Control (QC) methods.
- QC requirements for new materials features.
- Qualification needs for new alloys.
- Lessons learned from fuel performance regarding design bases, fabrication process control, QC, and reactor operation.
- Issues related to reactor (plant) life extension and refurbishment.

This information will help utility staff to implement actions to maintain or improve fuel reliability.

Although the value of recent data endures, the specific technical issues affected by the recent data tend to change with time.

The ZIRAT22 Annual Report will start with a short introduction that will give the background and the current understanding of the topic typically based upon previous ZIRAT reviews. The introductory part will be followed by the review of the relevant data presented since the last ZIRAT review, i.e. ZIRAT21. In addition, each topic will have a final summary sub-section that will provide conclusions and an updated view of the understanding given in the introductory part.

The last section in the ZIRAT22 Annual Report will summarise the current issues related to fuel performance and list the data needed to resolve these issues. The following, currently important, issues are intended to be specifically addressed.

Burnup Achievements and Fuel Performance Issues

- Trends in fuel operating conditions.
- High burnup fuel performance summary.
- Fuel reliability.
- Fuel performance related utility concerns.
- Fuel related regulatory issues of concerns to utilities and laboratories.

Fabrication

Changes in zirconium alloy fabrication and QC methods and their potential effect on performance.

In-Reactor Performance of Zr Alloys

- Irradiation effects on microstructure of Zr alloy components such as: Fuel cladding, liner, guide tube, grid/spacer, fuel channel and pressure tube materials.
- The impact of alloying elements, microstructure, and irradiation conditions (temperature, power history, fast flux, fast dose, PWR, BWR and VVER water chemistry) on:
 - Corrosion and hydrogen pickup mechanisms, redistribution, effects on mechanical and corrosion properties, and dimensional stability.
 - Mechanical properties (e.g. yield and ultimate yield strength, ductility, fracture toughness, fatigue, Delayed Hydride Cracking, Pellet Cladding Interaction)
 - Dimensional stability (irradiation growth, creep, relaxation).
- Recent primary fuel failures (fretting, corrosion, hydriding, Pellet Cladding Interaction (PCI), Pellet Cladding Mechanical Interaction (PCMI)) and secondary degradation, suggested remedies to improve failure resistance, important design and fabrication issues, and impact of plant operation.
- Relationship of fuel rod characteristics to performance in Loss of Coolant Accident (LOCA) and Reactivity Initiated Accident (RIA).
- The direct and indirect impact of water chemistry, CRUD, and chemical additions on the fuel performance.
- Utility and regulator perspectives including burn-up limits based upon the data presented in the ZIRAT21 base Report.

In-Reactor Performance of Accident Tolerant Fuel (ATF)

Prior to the accident at Fukushima, the emphasis of advanced LWR fuel development was on improving nuclear fuel performance in terms of increased burnup for waste mini-

> "We find ZIRAT a great help and, in a way, it's like having an extra person in the team"

> > TED DARBY Senior Co. Specialist, Rolls-Royce PLC





mization, increased power density for power upgrades, and increased fuel reliability. Fukushima highlighted some undesirable performance characteristics of the standard fuel system during severe accidents, including accelerated hydrogen production under certain circumstances. Thus, fuel system behaviour under design-basis accident and severe-accident conditions became the primary focus for advanced fuels, along with striving for improved performance under normal operating conditions to ensure that proposed new fuels will be economically viable. Fuel vendors, nuclear research laboratories, and universities have embarked on an aggressive schedule for the development of enhanced accident-tolerant fuel (ATF) systems. The programs are in the early phases of R&D and are currently supporting the investigation of a number of candidate technologies that may improve the fuel system. This chapter of the annual report provides a brief review of the various ATF systems that are being developed.

Intermediate storage

Fuel related issues in dry storage.



ZIRAT22 Special Topic Reports

In addition to the ZIRAT22 Annual Report, two Special Topic Reports will be prepared on Irradiation Growth of Zirconium Alloys and New BWR and PWR Alloys.

The Special Topic Reports will cover the range from basic information to current knowledge and be written and explained in such a way that engineers and researchers not familiar with the topic can easily follow the STRs, find and grasp the appropriate information. This means that the STRs could be used by the organisations in the training of their internal staff with or without the additional assistance of ANT International staff. The background for selecting topics and proposed contents of the Reports are discussed in detail below.

Irradiation growth of zirconium alloys

Unique aspects of material behaviour in a nuclear power plant include the component's dimensional stability. In fast breeder reactors the Fe and Ni-based alloys creep and swell, that is, they change dimensions in response to a stress and change their volume in response to radiation damage. In light water reactors, zirconium alloy structural components creep, do not swell, but do change their dimensions through the approximately constant volume process called irradiation growth. Radiation effects are not unexpected since during the lifetime of a typical component every atom is displaced from its normal lattice position at least 20 times. With very few exceptions, the mechanical and physical properties needed for reliable fuel assembly performance are affected by irradiation.

Practical effects of dimensional instabilities are well known and it is a rare technical conference in the reactor performance field that does not include discussions on the topic. In addition to lengthening due to irradiation growth, many components are subjected to creep stresses. Because of the difference in pressure inside and outside the fuel rod, clad-ding creeps down on the fuel early in life, and then creeps out again later in life as the fuel

begins to swell. A major issue is to have creep strength sufficient to resist outward movement of the cladding if fission gas pressure becomes high at high burnups. PWR guide tubes can creep downward or laterally due to forces imposed by fuel assembly hold down forces or cross flow hydraulic forces – both leading to assembly bow which can interfere with smooth control rod motion. BWR channels can creep out or budge in response to differential water pressures across the channel wall, again leading toward control blade interference. Fuel rods, water rods or boxes, guide tubes, and tie rods all can lengthen due to irradiation growth, possibly leading to bowing problems. (For calibration, a recrystallised (RX or RXA) Zircaloy water rod or guide tube could lengthen due to irradiation growth more than 2 cm. during service; a cold worked/stress relieved (SRA) component could lengthen more than 6 cm.) Even RX spacer/grids could widen enough due to irradiation growth (if texture or heat treatment was not optimised) to cause uncomfortable interference with the channel.

In addition, corrosion leading to hydrogen absorption in Zircaloy can contribute to component dimensional instability due, at least in part, to the fact that the volume of zirco-nium hydride is about 16% larger than zirconium.

The above discussion leads to the concept that understanding the empirical details and mechanisms of dimensional instability in the aggressive environment of the nuclear core is important for very practical reasons. Reliability of materials and structure performance can depend on such understanding.

The sources of dimensional changes of reactor components (in addition to changes caused by mechanical loading, which is almost always in the elastic range, and conventional thermal expansion and contraction) are: irradiation growth, irradiation creep, thermal creep, stress relaxation (which is a combination of thermal and irradiation creep), and hydrogen and hydride formation.

Irradiation effects are primarily related to the flow of irradiation-produced point defects to sinks such as grain boundaries, deformation-produced dislocations, irradiation-produced dislocation loops, and alloying and impurity element complexes. In zirconium alloys, crys-tallographic and diffusional anisotropy are key elements in producing dimensional changes.

In the past, hydrogen effects have been considered to be additive to and independent of irradiation; however, recent data have brought this assumption into question. It is certain that corrosion-produced hydrogen does cause significant dimensional changes simply due to the 16-17% difference in density between zirconium hydride and zirconium. A length change of on the order of 0.25% can be induced by 1000 ppm hydrogen in an unirradiated material. Recent experiments indicate that the presence of hydrogen and/or hydrides does contribute to the mechanisms of irradiation growth but probably not creep.

Fuel rod diametral changes are caused by stress dependent creep processes, as irradiation growth in the hoop direction is very small.

Fuel rod length changes are caused by several phenomena:

• Stress free axial elongation due to irradiation growth.

• Anisotropic creep (before pellet/cladding contact) due to external reactor system pressure. Because of the tubing texture, axial elongation results from creep down of the cladding diameter; however for heavily cold worked material, it has been reported that some shrinkage may occur. In a non-textured material such as stainless steel, creep down of the cladding would only result in an increase in cladding thickness, with no change in length (more detail below).

• Creep due to pellet-cladding mechanical interaction (PCMI) that results from differences in the thermal strains between fuel pellets and cladding, pellet swelling and cladding creep (closure of the pellet-cladding gap) and that increases with the intensity of contact. PCMI begins early in life due to pellet cracking and relocation in the radial direction, is moderated by the effects of pellet densification and then increases after hard contact between the cladding and fuel. This occurs in mid-life of typical LWR fuel, depending on the cladding creep properties and the dimensional stability of the fuel, and early in life for PHWR (CANDU) fuel.

• Hydriding of the cladding due to corrosion.

Bow of a component such as a BWR channel or PWR control rod assembly can occur if one side of the component changes length more than the other side. Such differential length changes occur due to differential stress and creep, to relaxation of differential residual stresses, or to differential irradiation growth due to differences in flux-induced fluence, texture, material cold work, and hydrogen content.

Irradiation growth occurs simultaneously with irradiation creep if there is an applied stress. The two processes are considered to be independent and additive, even though they compete for the same irradiation-produced defects mechanistically. When assessing dimensional changes of any component all sources of change must be taken into account. This is especially true for fuel rods, which will invariably have length-change components due to irradiation growth and anisotropic creep, and perhaps in addition to pellet-imposed stresses at higher burnups.

In this STR review, aimed specifically at irradiation growth, we will primarily address conditions of direct interest to LWRs and CANDUs, unless the information has mechanistic implications. Specific issues mentioned above will be addressed separately below.

Irradiation creep was covered earlier by ZIRAT 14 Special Topic Report: In-reactor Creep of Zirconium Alloys, authored by Ron Adamson, Friedrich Garzarolli and Charles Patterson, 2009.

The STR addresses all data deemed relevant to understanding irradiation growth, broad review and new aspects of growth mechanisms, and a summary of practical effects of growth on component performance.

Preliminary out lines of the STR sections follow:

Data, Ron Adamson

- 2.1 Irradiation growth of zirconium alloys
- 2.1.1 Introduction
- 2.1.2 Basics of irradiation growth
- 2.1.3 General effects
- 2.1.4 Effects of cold work
- 2.1.5 Texture and crystallography
- 2.1.6 Alloying element effects
- 2.1.7 Temperature dependence of irradiation growth
- 2.1.8 Effects of hydrogen and hydrides
- 2.1.9 Grain size
- 2.1.10 Volume
- 2.1.11 Single crystals of Zr

"An interesting seminar with very useful information and a close overview of the state of art of nuclear fuel. I will recommend the seminar to my colleagues."

IGNACIO COLLAZO Iberdrola

- 2.1.12 Thermal stability implications to mechanisms
- 2.1.13 Stages and introductive mechanisms

Mechanisms and Modelling, Malcolm Griffiths

- 3.1 Review of relevant data that provides insight and support for mechanistic models
- 3.2 Normalisation of dose data (by fluence equivalence or dpa)
- 3.3 Fundamental factors controlling irradiation growth as a function of irradiation dose
- 3.4 Review of past models on irradiation growth
- 3.5 Summary and discussion

Fuel and structural issues, engineering correlations and design approaches, Charles Patterson

- 4.1 Introduction and overview
- 4.2 Fuel and structural issues
- 4.3 Engineering models
- 4.4 Design approaches for accommodating irradiation growth
- 4.5 Summary and discussion
- 5. Conclusions (RB, MG, CP)

New BWR and PWR Alloys

To meet the current situation with more aggressive reactor environments (higher burnups, changing water chemistries and loading patterns), and resolving fuel performance issues such as BWR channel bowing and PWR assembly bowing, a large number of zirconium alloys have been and are being developed. The main driver for the initial material development in Pressurized Water Reactors (PWRs) has been to reduce corrosion rates and Hydrogen Pick-Up Fractions (HPUFs), which have occasionally limited the maximum discharged burnup.

However, to ensure that the new Zirconium Alloys performs satisfactory during normal operation, Anticipated Operational Occurrences (AOOs), postulated accidents and intermediate dry storage, it is crucial to assess the projected performance of components of the new zirconium alloy materials and relate the performance to the material characterises. This assessment is the objective of this Special Topic Report (STR).

This ZIRAT22 STR is an update and expansion of the ZIRAT16 Report since the ZIRAT22 STR also includes BWR material development.

Below is an intended content list

Section 1 describes the background history of Zr alloy material development for PWRs/ VVERs and BWRs.

Section 2 describes the relevant fuel design criteria of the Fuel Assembly (FA) and its components relevant to the Zr alloy during normal operation, AOO, accident conditions

"I enjoyed the meeting and the interactions. The annual report always provides not only a concise summary of work worldwide in the Industry over the past year, but also knowledgeable opinions as to its meaning and significance."

> BRUCE KAMMENZIND Bechtel Marine Propulsion Corporation

and intermediate dry storage.

Section 3, 4 and, 5 covers the performance of current and new improved Zr alloys for PWR/VVER and BWR FA applications during normal operation, AOO, Design Basis Accidents (DBAs) and intermediate dry storage, respectively.

Report authors

The authors are: Mr. Peter Rudling, President of ANT International, Dr. Ron Adamson, formerly at GENE, Vallecitos, Mr. Friedrich Garzarolli, formerly at Framatome ANP, Dr. Charles Patterson, formerly at GNF, Dr. Kit Coleman, earlier at AECL, Dr. Tahir Mahmood formerly at GENE, Vallecitos and Dr. Malcolm Griffiths earlier at CNL (earlier AECL).



Dr. Ron Adamson retired from GE Nuclear Energy in 2000, where he was the manager of Materials Technology. Earlier he graduated from the University of Wisconsin with a B.S. in Mechanical Engineering, an M.S. in Nuclear Engineering and a PhD in Metallurgy. Post-doctoral work on irradiation effects was conducted at AERE, Harwell, England. At the GE Vallecitos Nuclear Center he led research, development and testing programmes for reactor core materials, with special emphasis

on zirconium alloys. During his 31 years with GE, Dr. Adamson was actively involved with utilities and the technical community worldwide. He holds 17 patents, has published over 90 technical papers involving nuclear materials technology, and has received several important awards, including the Outstanding Technical Contribution Award from GE Industrial Power Systems, the Mishima Award from the American Nuclear Society, and the Kroll Medal from the ASTM/Kroll Institute. Zirconium alloy areas in which Dr. Adamson has particular interest and experience include: in-reactor dimensional stability; in-reactor corrosion performance and mechanisms; microstructure evolution due to reactor irradiation; mechanical properties of irradiated material; high burnup performance; failure mechanisms and remedies; and fabrication technology. Since retirement he has been actively associated with ANT International, EPRI and others as a consultant in zirconium technology.



Mr. Friedrich Garzarolli retired from Framatome ANP in March 2002, where he has held various managerial and research positions, dealing with fuel rod performance analysis, planning and evaluation of irradiation tests, materials characterisation and evaluation of irradiation effects in materials. His degree as Diplom Ingenieur in metallurgy was obtained from the University of Leoben, Austria, in 1963. He has been active in the following fields:

- Development of new fuel assembly materials, especially cladding for BWRs and PWRs
- Modelling of corrosion for zirconium alloys and stainless steels
- Effect of water chemistry on cladding corrosion
- PCI failures of cladding
- In-reactor dimensional stability
- High-burnup performance
- Failure mechanisms and remedies
- Microstructure evolution due to reactor irradiation



Dr. Charles Patterson retired from Global Nuclear Fuel in 2008 as a Consulting Engineer for Fuel Engineering. During 44 years with GE Nuclear Energy/GNF, he was actively engaged in the development of fuel manufacturing processes, fuel materials, thermal-mechanical and fuel performance models and in the improvement of fuel reliability. This activity involved irradiation and hot cell Programmes in Asia, Europe and the United States to identify in-core material behaviour,

validate analytic models and improve fuel reliability. Chuck holds patents in the areas of fuel and cladding materials, fuel assembly design and fuel inspection technology. Dr. Patterson has particular interest and experience in the thermal and mechanical behaviour of fuel, cladding and structural materials, the development of analytic models to describe their behaviour and in the improvement of fuel reliability.



Dr. Kit Coleman has, after receiving a PhD in the UK, spent his working career at the Chalk River Laboratories of AECL. Research interests on zirconium alloys included in-reactor creep, development of improved fuel cladding and pressure tube materials. He retired in 1999 as manager of Material and Mechanics Branch but retains an attachment to CNL (formerly AECL) as a Researcher Emeritus. He has published over 100 papers on zirconium technology and has

received the Russ Ogden Award from ASTM and the Kroll Medal from the ASTM/Kroll Institute. He is on the Advisory Editorial Board of the Journal of Nuclear Materials.



*Dr. Sheikh Tahir Mahmood r*etired from Global Nuclear Fuel in 2012 as a Senior Engineer/Technologist for Fuels Engineering at the Vallecitos Nuclear Center. Earlier he received Masters degrees in Physics and Nuclear Technology from abroad and doctorate in Nuclear Engineering from North Carolina State University. His Post-doctoral work on mechanical anisotropy of zirconium alloys and radiation effects on reactor structural materials was done at NCSU and ORNL, respectively.

At GE Nuclear Energy/GNF, he was actively engaged in fuel performance and materials technology. This activity involved failure root-cause investigations through hot cell PIE of the failed in-core components, and development and evaluation of material property data bases for new materials developed for in-core use. Tahir has particular interest and experience in mechanical metallurgy, mechanical behaviour of fuel, cladding and structural materials, and in-reactor behaviour of these materials for improved fuel reliability. He has actively participated in various international nuclear industry research programs.



Dr. Malcolm Griffiths obtained his PhD in Physical Metallurgy from the University of Birmingham in 1981. After a three-year post-doctoral term working on radiation damage in Ti-alloys he joined AECL at the Chalk River Laboratories in 1984. He has worked on various aspects of materials performance in nuclear reactor cores during his 32 years with AECL. As manager of the Radiation Damage and Deformation Branch Malcolm was instrumental in developing strategies

to support CANDU refurbishment that addressed improving pressure tube performance

and the degradation of Inconel X-750 core components. Up until his retirement he was chair and project manager for the Candu Owners Group (COG) R&D program on pressure tube deformation and the COG joint project on Inconel X-750 spacer degradation. From 2003-2013 Malcolm was on the editorial advisory board for the Journal of Nuclear Materials and was an editor from 2013-2016. In 2007 he was recipient of the 2005 Kroll medal from the American Society for Testing and Materials for his pioneering work on microstructure evolution in zirconium alloys during irradiation. He has published nearly 100 papers on material characterisation and reactor core materials performance. He is currently adjunct professor at Queens University, Department of Mechanical and Materials Engineering.



Mr. Peter Rudling is the President of ANT International, managing the ZIRAT/IZNA/LCC Programmes as well as providing seminars and Handbooks on various fuel related topics to the nuclear industry. Peter was a senior consulting scientist at Vattenfall, the largest Swedish power company. Earlier he has also been a Specialist of Fuel Materials at ABB Atom (now Westinghouse) and a Project Manager at EPRI.

Price and Terms of Payment

The fixed nominal price for the ZIRAT Membership appears in the associated Proposal.

Terms and Conditions

The term of ZIRAT22 Programme starts from the date of the purchase order and lasts 12 months onwards.

ANT International shall exercise its best efforts to meet the objectives in this assignment and shall apply to the work professional personnel having the required skills, experience and competence. If the assignment is found to be significantly deficient by the customer within 6 months of its completion, ANT International shall modify the work done within this assignment in such a way that it will become satisfactory to the customer. This modification shall be done without incurring any additional costs to the customer. The total amount of such additional costs due to the modification shall be limited to be less or equal to the amount originally paid to ANT International for this assignment.

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Contact

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Advanced Nuclear Technology International, Spinnerivägen 1, Fack 5035, SE-448 51 Tollered, Sweden. Phone: +46 (0)31-88 16 00. Fax: +46 (0)31-88 16 01. info@antinternational.com www.antinternational.com