A New Nuclear Materials Laboratory at Queen's University

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Abstract

The Reactor Materials Testing Laboratory (RMTL) at Queen's University and the results of commissioning tests are described. RMTL uses energetic protons (up to 8MeV) to simulate fast neutron damage in materials for reactor components. The laboratory is also capable of He implantation (up to 12 MeV) to simulate the effects of transmutation He in reactor components. The $17.5M laboratory comprises a new building, a 4MV tandem accelerator, two electron microscopes, mechanical testing and specimen preparation equipment, and a radiation detection laboratory. RMTL focusses on studying dynamic effects of irradiation (irradiation creep, irradiation growth, irradiation induced swelling, fatigue under irradiation) *in-situ*.

1. Introduction

Materials exhibit fundamentally more complicated behaviour inside a reactor than when outside. This is due to the presence of the high neutron flux which alters the way that deformation, corrosion and failure occur. While mechanisms operating in materials can be studied outside of the reactor, the impact of radiation on mechanisms must also be investigated.

To understand the impact of the reactor environment on material properties, most researchers carry out experiments in a reactor. This is technically very difficult and expensive, since both the test pieces and the equipment used to carry out the experiments will be activated and damaged by the reactor conditions. In addition, fine control of experimental parameters necessary to establish basic mechanisms is very difficult, and often impossible. This drastically limits the possibility of gaining an understanding of the mechanistic process occurring under reactor conditions, requires more conservative design approaches and limits the ease with which new materials or systems can be used to improve performance.

In addition many Materials Test Reactors (MTRs) and facilities have been closed down in the past 20 years (including materials test loops in the NRU reactor at Chalk River). Hence, even the capability for empirical/engineering evaluation of materials for advanced reactor designs is limited.

Many of the problems associated with experiments in MTRs and much of the expense can be eliminated by using high energy ions, produced by an accelerator to simulate the effects, refered to as "displacement damage", produced by fast neutrons in a reactor. Displacement damage is the displacement of atoms from their normal positions in the material by collisions with fast particles (neutrons in the case of a reactor). It is measured in units of displacements per atom (dpa), the average number of times each atom in the material has been displaced from its normal position.

Heavy ions provide the best simulation of the displacement damage produced by neutrons, however, their penetration depths are very limited (a few μm) for energies that will not activate the samples in the same way as a neutron flux. Studies with heavy ions are therefore mainly limited to post-irradiation examination (PIE) using electron microscopy and other techniques. Mechanical properties and dynamic effects of irradiation cannot be studied in any conventional way.

In the late 1970's the United Kingdom Atomic Energy Authority at the Atomic Energy Research Establishment at Harwell, pioneered the use of energetic protons (up to ~4MeV) to produce displacement damage in thin "bulk" specimens of metals of interest for reactor components [1,2], including Zircaloy-2 [3].

Protons with energies of few MeV will penetrate several 10s of μm in most structural metals, and, for example protons of 3.5MeV will pass completely through a stainless steel or zirconium target 50μm thick, leaving no hydrogen to contaminate the sample, and with little activation. They will produce a relatively uniform displacement damage profile through the thickness of such a sample. The displacement damage produced is somewhat different from that produced by fast neutrons, but work at Harwell, and later at Atomic Energy of Canada Limited's Whiteshell Laboratories [4,5] showed that dynamic effects (irradiation creep, irradiation growth, irradiation induced swelling) as well microstructural changes are similar to those seen under fast neutron irradiation.

2. The Reactor Materials Testing Laboratory

The Reactor Materials Testing Laboratory (RMTL) is a M$17.5 project approved for funding in July 2009 after two years of preparatory work. Funding obtained from the Canadian Foundation for Innovation (CFI), the Ontario Ministry of Research and Innovation (MRI), Queen's University, and the equipment manufacturers (in kind) was approved in July 2009.

The project includes a new building, Figure 1, located at 136 Grant Timmins Drive in Kingston, Ontario. The equipment includes a 4MV High Voltage Engineering Europa B.V. Tandetron tandem accelerator, Figure 2, an FEI Tecnai Osiris scanning transmission microscope (STEM), Figure 3, an FEI Nova NANOSEM scanning electron microscope, Figure 4, a MicroMaterials Nanotest-Vantage nano indenter, Figure 5, and ancillary equipment for specimen preparation, radiation detection, etc. An important component of the laboratory's activities is research on personal neutron detectors by the University of Ontario Institute of Technology [6] using the neutron fields that are a by-product of the materials irradiation experiments.

The heart of the project is the 4MV tandem accelerator. It was specified to accelerate protons up to 8MeV with a beam current of 20 μA and He^{++} ions to 12 MeV, with a beam current of up to μA. With 4MeV protons, a 20 μA proton beam will produce a displacement damage rate in Zr alloys at least 30x that produced by the peak fast neutron flux in a CANDU power reactor. 12 MeV He^{++} ions can be implanted up to 50 μm into metals like Ni to simulate the effect of He formed by n-α reactions in nuclear reactors.

Figure 1. The Reactor Materials Testing Laboratory (RMTL) located at 136 Grant Timmins Drive, Kingston, Ontario.

Figure 2. The High Voltage Engineering Europa 4MV Tandetron tandem accelerator at RMTL. The ion injector is partially visible at the left, one of the beam-tubes at the right.

Figure 3. FEI Tecnai Osiris 200kV scanning transmission electron microscope at RMTL.

Figure 4. FEI Nova NANOSEM scanning electron microscope at RMTL.

RMTL has two independently shielded target rooms allowing experimental set-up or disassembly to be carried out in one room while an experiment is being carried out in the other. The accelerator is capable of, and will be licenced for 24 hour unattended operation, seven days a week. The target rooms will have interchangeable target stations for performing “static” irradiations (irradiations at constant conditions) of specimens for mechanical testing and/or PIE. In addition there will be two target stations for in-situ dynamic mechanical properties testing, where steady state rates (e.g., of irradiation creep, irradiation growth and irradiation-induce swelling) can be established, and the effect of changing the experimental conditions (e.g., stress, temperature, displacement damage rate) investigated.

3. Planned Research at RMTL

The capability to perform *in-situ* dynamic testing will allow the identification of sub-microscopic mechanisms of macroscopic behaviour, and understanding the macroscopic effects of changing conditions (e.g., fatigue under irradiation).

The possibilities of such testing have been demonstrated in experiments at Whiteshell Laboratories in which the functional dependence of irradiation creep and growth on displacement damage rate of zirconium at 345K were successfully studied [4]. Figure 5 shows a creep curve under proton irradiation, plotted in the typical fashion for irradiation experiments i.e., strain on the vertical scale vs displacement damage (dpa) on the horizontal scale (proportional to integrated beam current in this case, proportional to fast neutron fluence or effective full power hours in the reactor case). The creep curve looks typical in this case, with an initial primary region, during which the strain rate decreases, followed by a steady state. However, when plotted as strain vs time (as is typical for a conventional creep test), Figure 6, the effect of changing the beam current during the test to determine the sensitivity of the strain rate to displacement damage rate can be clearly seen. The observed linear relationship between damage rate and deformation rate (which results in the smooth creep curve of Figure 6) tells us that the mechanism of irradiation-induced deformation at 347K (close to the operating temperature of CANDU calandria tubes) is similar to that at 570K (the operating temperature of CANDU pressure tubes).

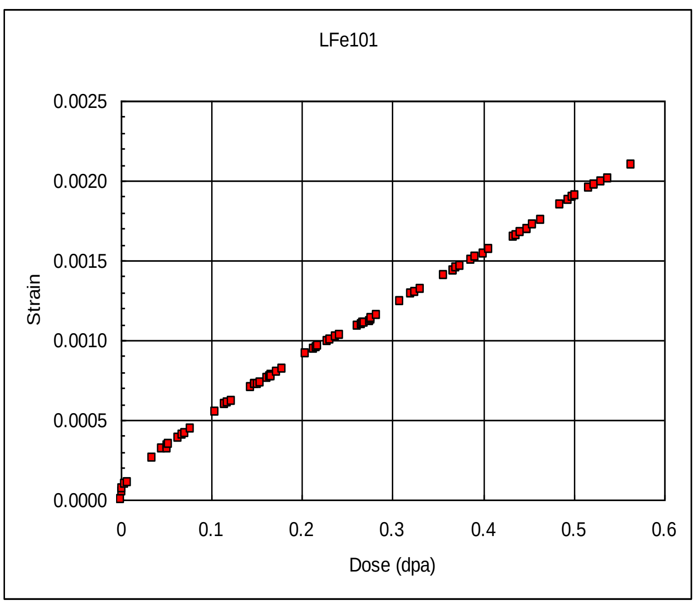


Figure 5. Creep curve (strain vs irradiation dose) for Zr containing 60 ppm Fe, irradiated with 4.4 MeV protons at 347 K with a stress of 50 MPa and various beam currents, from Reference 4.

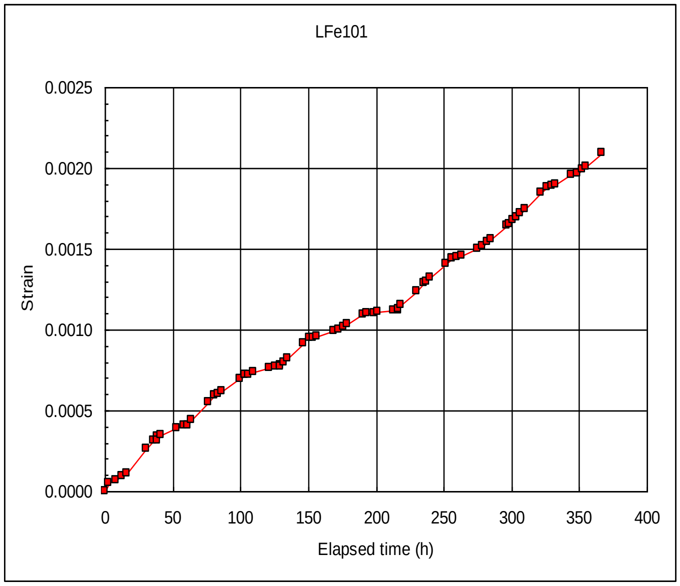


Figure 6. The same creep curve as Figure 5, but strain vs time, from Reference 4. The slope increases and decreases with increased and decreased beam current (proportional to displacement damage rate).

Research at RMTL using such techniques will provide scientific understanding and technology at a critical juncture in the development of nuclear power, and in particular for Canadian nuclear technology. Other facilities are not available in Canada or internationally to perform such research on the necessary scale and schedule for Canada to take advantage of the technology over the next twenty-five years.

This facility will also have an important human resources impact. The opportunity for graduate students and post doctoral fellows to work on this facility will provide a very high level of understanding of the impact of radiation damage on materials for both engineers and scientists working in the nuclear industry. In combination, the provision of high current 8MeV protons together with dedicated experimental and post-mortem examination equipment make this facility unique in the world. We therefore expect major international collaborations to build from this infrastructure. A number of international collaborators have already been identified.

Current CANDU reactors were designed on the basis of empirical or semi-empirical information from limited experimental results from materials test reactors, and much of the information supporting their on-going operation is based on data obtained from in-service inspection and characterization of the components. This results in conservative operating envelopes which impact the capacity factors of the reactors. A 1% gain in capacity factor is typically worth ~$3M/year. Research at RMTL will provide the means to improve capacity factors, by reducing the uncertainties in predictions of safe operating margins, relating to fracture characteristics of the materials (fatigue under irradiation, for example).

It will be several years before the more recently built reactors (i.e., three units in Korea, two in China and one in Romania) will be refurbished. Research at RMTL will provide technology to improve the design of the components and systems during refurbishment to reduce required maintenance, improve margins and increase capacity factors in the refurbished reactors.

Candu Energy Inc. is currently offering an upgraded reactor design to the market. Some of the reactor core materials are different from those in the existing design, e.g., a tight-fitting Zr-alloy spacer. Research at RMTL will provide support for licensing and operation of the improved design.

Canada is a member of an international agreement to develop Generation 4 reactors. Several very different reactor designs are being considered. The main thrust is to design reactors with higher operating temperature, and hence improved thermodynamic efficiency. These higher temperatures will require the use of new materials that are presently not employed in nuclear reactors and materials technology will dictate the success of any given design. Through Natural Resources Canada, Canada is promoting the development of the Super-Critical Water cooled Reactor (SCWR) based on evolution of the CANDU design. Research at RMTL will address materials issues for the SCWR.

RMTL also provides an ideal surrogate to an actual reactor site for research on irradiation field measurement and characterization, and for developing improved protocols for personnel and environmental protection. The beams generated by the accelerator itself, the mixed neutron and gamma fields generated from test specimens during irradiation and the residual radioactivity of the specimens all provide an excellent environment for development of instruments to measure, characterize and quantify radiation, and will contribute to improved worker and environmental protection in current and future reactor designs.

4. Current Status

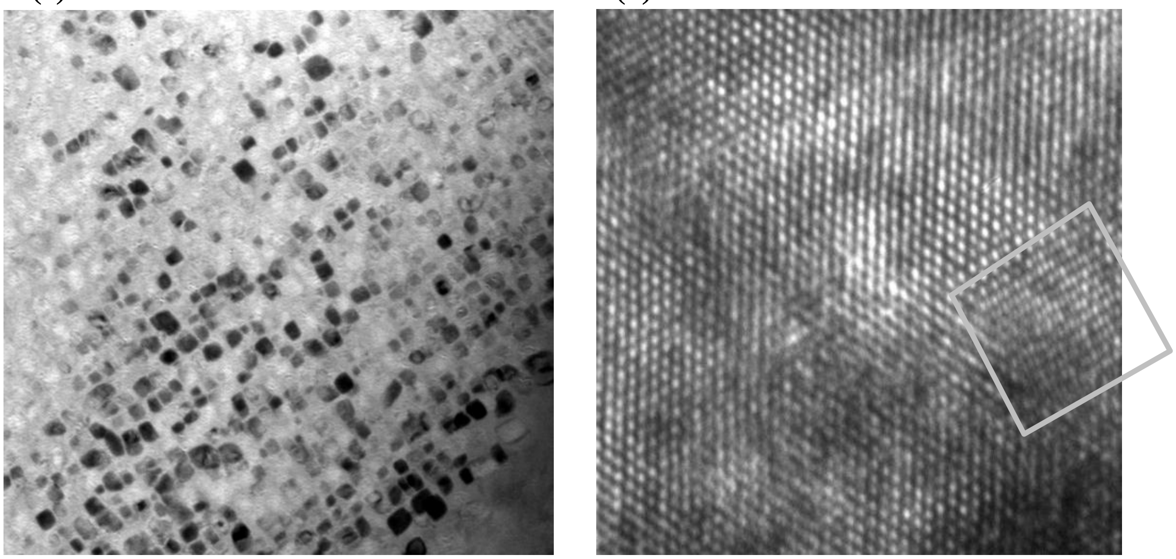
A Canadian Nuclear Safety Commission licence for construction of a Class 2 facility was granted in October 2011. Construction was completed and much of the equipment installed by September 2014. The commissioning licence was granted October 2014. Building systems were completed February 2015, the accelerator was commissioned by the manufacturer during March-April 2015. The laboratory is being commissioned in preparation for its operating licence during April-May 2015.

Neutron and γ fields measured from the Faraday cups in the accelerator hall and target rooms and outside their respective shielding walls during the manufacturer's commissioning of the accelerator were consistent with design calculations [7,8]

The electron microscopes have been in use since mid-2014. An example of the high resolution capability of the STEM is shown in Figure 7. It shows the transformation of the body centred cubic β-

Zr phase in Figure 7(a) and lattice images of both phases in Figure 7(b) in the high strength pressure tube alloy EXCEL [9]. The lattice image of the ω-phase has a resolution of about 0.1nm.
An example of the capability of the SEM is shown in Figure 8. The individual sub-platelets ~15nm wide are resolved in what is probably a hydride in Zr-2.5Nb pressure tube material.

(a) (b)



-------------------- **200nm** --------- **2nm**

Figure 7. TEM of phase transformation, β→ω, in high strength pressure tube alloy EXCEL: (a) cuboidal hexagonal ω particles in a matrix of body centred cubic β–Zr, and, (b) lattice image showing the atomic arrangement of both phases with a resolution of ~0.1nm (ω-phase in the lower right quadrant marked by grey square). From the Ph.D. thesis project of Hongbing Yu.

5. Conclusions

A new laboratory, the Reactor Materials Testing Laboratory (RMTL) has been built at Queen's University. The laboratory has a 4MV tandem accelerator, state of the art electron microscopes and other equipment for the study of irradiation effects in reactor core materials using high energy protons to produce radiation damage and He ion implantation to simulate the effects of n-α reactions in nuclear reactors. The laboratory will emphasize in-situ studies of dynamics effects, e.g., fatigue under irradiation. RMTL is unique in the world in being dedicated to such studies.

6. Acknowledgements

Funding for RMTL was obtained from the Canadian Foundation for Innovation (CFI), the Ontario Ministry of Research and Innovation (MRI), Queen's University, with additional in-kind support from equipment manufacturers HVEE BV, FEI, MicroMaterials Ltd. and Struers Ltd.

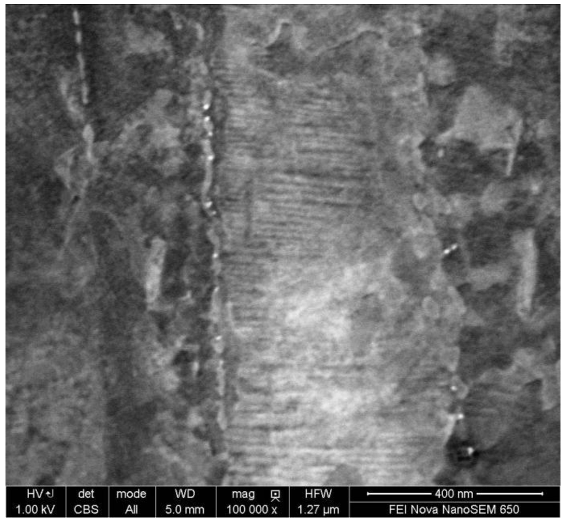


Figure 8. SEM image of what is probably a hydride in Zr-2.5Nb pressure tube material. The individual horizontal platelets of the hydrides ~15 nm wide are resolved.

7. References

[1] J.A. Hudson, R.S. Nelson and R.J. McElroy, *The irradiation creep of nickel and AISI 321 stainless steel during 4MeV proton bombardment,* Journal of Nuclear Materials, 65 (1977) pp 279-294

[2] R.J. McElroy, *Irradiation growth during fission fragment and proton bombardment of uranium,* Journal of Nuclear Materials, 90 (1980) pp 279-306.

[3] O.J.V. Chapman, R.J. McElroy, B.E. Sheldon, *Irradiation creep and growth during proton and neutron bombardment of zircaloy-2 plate,* Proc. 6th International Symposium on Zirconium in the Nuclear Industry, ASTM STM 824, 1984, pp 343-374.

[4] C.K. Chow, R.A. Holt, C.H Woo and C.B. So, *Deformation of zirconium irradiated by 4.4 MeV protons at 347,* Journal of Nuclear Materials, 328 (2004) pp 1-10.

[5] C.D. Cann , C.B. So a, R.C. Styles a and C.E. Coleman, *Precipitation in Zr-2.5Nb enhanced by proton irradiation*, Journal of Nuclear Materials, 205 (1993) pp 267-272.

[6] A.J. Waker and G. Taylor, *Sensitivity and uncertainty in the measurement of H*(10) in neutron fields using a REM500 and a multi-element TEPC,* Radiation Protection Dosimetry, 161(1-4), 2014, pp 205-209

[7] W.T. Diamond, *Shielding design basis for a proton accelerator facility at Queen's University*, Queen's University Report RMTL-CNST-G.9, Appendix B, October 2011.

[8] W.T. Diamond, *Gamma-ray yields during operation of accelerator from the targets planned for use in the facility*, Queen's University Report RMTL-CNST-G.9, Appendix A, June 2011.

[9] B.A. Cheadle R.A. Holt, V. Fidleris, A.R. Causey and V.F. Urbanic, *High Strength, Creep Resistant EXCEL Alloy Pressure Tubes,* B.A. Cheadle, Proc 5th International Symposium on Zirconium in the Nuclear Industry, ASTM STM 754, (1981)193-207.