Material Test Reactors and other Irradiation Facilities

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Contents

Introduction

Irradiation damage in materials for nuclear applications primarily results from the production of energetic particles produced in fission, nuclear reactions, and radioactive decay events. The interaction of these energetic particles (fission products, fast neutrons, protons, alphas, and recoil nuclei) with materials results in the production of atomic-scale defects from ballistic collisions and introduction of new chemical elements from transmutation. In the case of high-energy fission products, the intense ionization along the fission product path can also introduce defects or damage. These irradiation damage processes control the long-term evolution of the materials response to the production and diffusion of defects, fission products and nuclear reaction products.

The response of fuels and materials to radiation is critical to the performance of advanced nuclear systems. Key to understand material performance in a nuclear environment is the analysis of materials irradiated using Material Test Reactors (MTRs) and Ion Beam facilities. Many of the world's non-power-producing nuclear reactors are used for research and training, materials testing, or the production of radioisotopes for medicine and industry. These are basically neutron factories. These reactors are much smaller than power reactors.

Material test reactors, which are the subject of this STR, are a subset of research reactors. In materials test reactors, materials are subject to intense neutron irradiation to study the induced changes. MTRs have provided essential support for nuclear power programs over the decades. Associated with hot laboratories for the post irradiation examinations, they are an important tool in research facilities for the fission and fusion domain. These facilities address the development and the qualification of materials and fuels under irradiation with sizes and environmental conditions relevant to nuclear power plants. They provide a means for improving and to demonstrate safe operations of existing and new build power reactors as well as to support future reactor designs. As MTRs, some being high flux research reactors, are able to reproduce material degradation undergone by materials in power reactors, they provide essential support to the study of ageing of generation II power plants, to the optimization of generation III plants and to test fuels and breeder capacities for generation IV. MTRs are also being used to irradiate new cladding materials and fuels that are being developed to produce enhanced Accident Tolerant Fuel (eATF) systems.

According to the IAEA database there are 83 research reactors performing material irradiations distributed over 29 member states [IAEA, 2018]. Section 1 of this STR provides information about some of the most active and upcoming MTRs around the world. The information on irradiation facilities in these MTRs has been collected from various sources and is summarized in Section 1 of this STR. This information will help in selecting an MTR by the members of a nuclear utility or an engineering laboratory when planning irradiation of their reactor materials to predict the behavior of the existing material at higher burn ups or generation of irradiation data for verification and licensing of new reactor materials such as new cladding and fuel materials for eATF systems.

One value from testing in MTRs comes from the higher neutron fluxes and radiation damage production rates. However, the dose rate and neutron energy spectra in MTRs are different from those in nuclear power plants. Therefore, the irradiated material data obtained from MTRs cannot be directly applied to power plant conditions without first considering the effects of dose rate and neutron energy spectrum. Section 2 of this STR addresses the opportunities and complexities of using materials test reactors with high neutron fluxes to perform accelerated studies of material aging in power reactors operating at lower neutron fluxes and with different neutron energy spectra. Radiation damage and gas production in different reactors have been compared using the code, SPECTER. This code provides a common standard from which to compare neutron damage data generated by different research groups using a variety of reactors [Griffiths et al., 2017].

As mentioned above, traditionally, research to understand radiation-induced changes in materials is conducted via radiation-effects experiments in material test reactors, followed by a comprehensive post-irradiation characterization plan. This is a very time consuming process because of the low damage rates that even the highest flux reactors exhibit. In addition the high cost of research on irradiated materials, in the face of shrinking budgets, puts additional constraints on this approach. When planning an irradiation project, material test reactors should be investigated and down-selected based on a number of considerations, including:

- Technical requirements
- Strategic partners and cooperation
- Cost, transportation and logistics

Beyond the high-energy high-flux material and fuel tests offered by material test reactors, two irradiation source alternatives continue to be considered:

- Power reactors
- Other irradiation systems such as ion irradiation

A promising partial solution to the problem is to use ion irradiation to irradiate materials to very high doses. The advantages of ion irradiation are many. Dose rates (typically 10^{-3} to 10^{-4} dpa/s) are much higher than under neutron irradiation (10^{-7} to 10^{-8} dpa/s), which means that 100s of dpa can be reached in days or weeks instead of years. Because there is little activation, the samples have little or no radioactivity and often can be handled in a laboratory environment. Control of ion irradiation experiment variables is much better than experiments in nuclear reactors.

Challenges to the implementation of ion irradiation as a surrogate for neutron irradiation include rate effects on microstructures and effective temperature, small irradiation volumes, and accounting for transmutations. The unique effects of ion irradiation and the use of ions to study fast neutron irradiation effects is a critical topic, since development and qualification of new structural materials requires neutron doses that are too high to be obtained in existing materials test reactors or spallation neutron sources. The advantages and disadvantages of using ions to simulate nuclear radiation environments, along with available ion irradiation facilities and their capabilities, are reviewed in Section 3 of the STR, with specific examples provided.

1 Material Test Reactors (MTRs) (Tahir Mahmood)

1.1 Introduction

Many of the world's nuclear reactors are used for research and training, materials testing, or the production of radioisotopes for medicine and industry. These are basically neutron factories. These reactors are much smaller than power reactors or those propelling ships, and many are at university campuses or engineering organisations. As of September 2018, there were 261 research reactors operating, in 57 countries (Figure 1-1) [IAEA, 2018]. Some operate with highly-enriched uranium fuel, and international efforts are underway to substitute low-enriched fuel. Material Test Reactors (MTRs), which are subject of sections 1 and 2 this STR, are a subset of research reactors. Therefore, a brief introduction to research reactors is presented here before discussing specifically the MTRs.

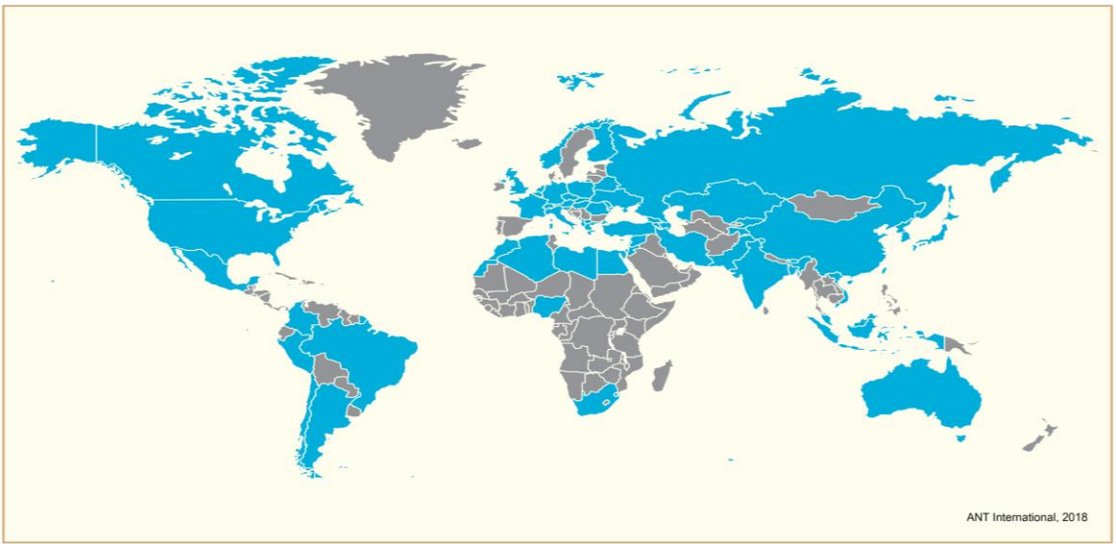

Figure 1-1: Countries (highlighted in blue) that have operational research reactors [IAEA, 2018].

1.1.1 Research reactors

Research reactors comprise a wide range of civil and commercial nuclear reactors which are generally not used for power generation. The primary purpose of research reactors is to provide a neutron source for research and other purposes. Their output (neutron beams) can have different characteristics depending on use. They are small relative to power reactors. The total power of the world's 261 research reactors is little over 3000 MW [WNA, 2018].

Research reactors are simpler than power reactors and often operate at lower temperatures. They need far less fuel, and far less fission products build up as the fuel is used. On the other hand, their fuel requires more highly enriched uranium, typically up to 20% U-235, although some older ones use 93% U-235. They also have a very high power density in the core, which requires special design features. Like power reactors the core needs cooling, though only the higher-powered test reactors need forced cooling. Usually a moderator is required to slow down the neutrons and enhance fission. As neutron production is their main function, most research reactors also need a reflector to reduce neutron loss from the core.

About half the operational research reactors are over 40 years old [IAEA, 2018]. Many research reactors are used with international collaboration. The IAEA has designated two international research hubs based on research reactors, giving them ICERR (International Centres of Excellence based on Research Reactors) status, valid for five years.

- The first is in France, based on CEA's Saclay and Cadrache facilities, and
- the second is in Russia, the Research Institute of Atomic Reactors (RIAR) at Dimitrovgrad, with six research reactors available to IAEA member states.

1.1.2 Types of research reactors

Research reactors come in a wider array of designs than for power reactors. They also have different operating modes, steady or pulsed. Most of the research reactors are either Pool-type with containment or TRIGA (Training, Research, Isotopes, General Atomics) reactors that do not need containment.

1.1.2.1 Pool type reactors

The pool type reactors, also called swimming pool reactors, are a type of nuclear reactors that has a core (consisting of the fuel elements and the control rods) immersed in an open pool of water. Among the fuel elements and control rods are the empty channels for experimental materials. The water both moderates and cools the reactor, and graphite or beryllium is generally used for the reflector, although other materials may also be used. The layer of water directly above the reactor core shields the radiation so completely that operators may work above the reactor safely (Figure 1-2). This design has two major advantages: the reactor is easily accessible and the whole primary cooling system, *i.e.* the pool water, is under normal pressure. This avoids the high temperatures and great pressures of nuclear power plants. Apertures to access the neutron beams are set in the wall of the pool. Tank type research reactors are similar, except that cooling is more active. Some sodium-cooled reactors have sodium pools instead.

Normally the reactor is charged with low enriched uranium (LEU) fuel consisting of less than 20% U-235 alloyed with a matrix such as aluminium or zirconium. Highly enriched uranium (HEU) was the fuel of choice since it had a longer lifetime, but these have been largely phased-out of non-military reactors to avoid proliferation issues. However most often 19.75% enrichment is used, falling just under the 20% level that would make it highly enriched. Fuel elements may be plates or rods with 8.5% to 45% uranium.

Various stations for holding items to be irradiated are located inside the core or directly adjacent to the core. Samples may be lowered into the core from above or delivered pneumatically via horizontal tubes from outside the tank at core level. Evacuated, or helium filled horizontal tubes may also be installed to direct a beam of neutrons to targets situated at a distance from the reactor hall.

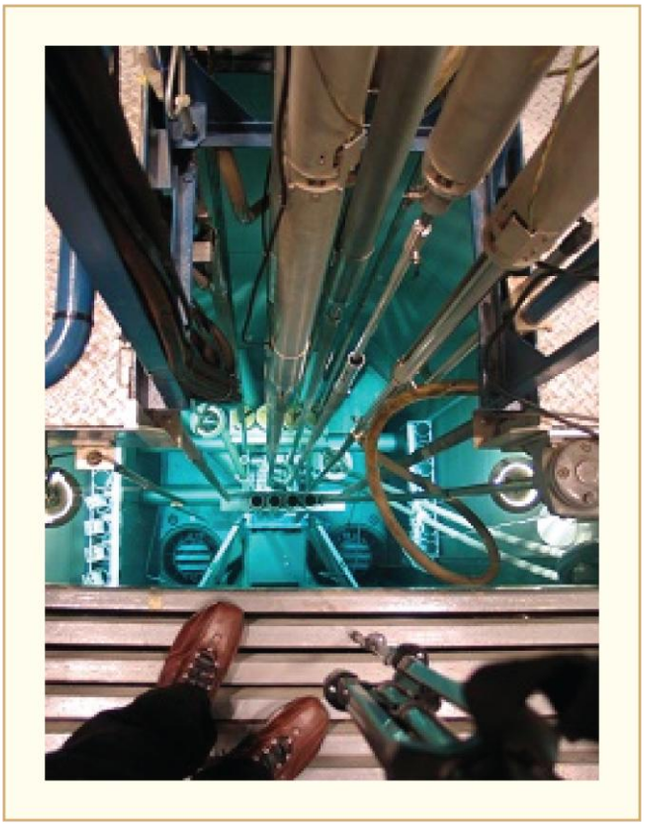

Figure 1-2: PULSTAR reactor at North Carolina State University is a 1 MW pool-type research reactor with 4% enriched, pin-type fuel consisting of UO_2 pellets in Zircaloy cladding.

2 Using Material Test Reactor Irradiations to Assess Materials Performance in Power Reactors (Malcolm Griffiths)

The operating life of nuclear reactors is often dictated by the performance of materials within the reactor core. In the case of power reactors the core components have to operate under extreme conditions of stress, temperature and environment for the life of the reactor (30-40 years), while at the same time being subject to the effects of neutrons, which affect both the dimensions and the physical properties of the materials that make up the core components. Ideally reactor core materials should be radiation-resistant but many reactors were designed and constructed without a full appreciation of how the dimensions and physical properties of these core materials would change over the life of the reactor. Operating experience has shown that material degradation due to radiation damage and transmutation effects can have a significant impact on the material properties that then dictates how a reactor may be operated, e.g. at a lower power or not at all. In order to address the challenges of predicting future behaviour of materials in power reactors, reactor designers have increasingly utilised material test reactors to irradiate different alloys in order to assess the effects of irradiation on the performance of reactor core components over the projected life of the reactor.

Reactor components subjected to a neutron flux will undergo radiation-induced changes that impact their ability to function as designed. Typically, the materials comprising these components undergo changes in dimensions (swelling and creep) and changes in mechanical properties (yield strength, ultimate tensile strength, ductility and fracture toughness) that are an irradiation effect and cannot be simulated in the laboratory. To predict future performance of components in operating reactors, or for the design of new reactors, materials test reactors (MTRs) have been utilized to perform accelerated or controlled evaluations of irradiated materials. The main value from testing in MTRs comes from the higher neutron fluxes and radiation damage production rates. Not only does the MTR provide the researcher with the ability to obtain information on materials degradation in advance of operating reactors, but also one can test material properties using standardized test specimens irradiated under controlled conditions. Occasionally a complete component, or part of a component, may be irradiated in a material test reactor, but such component tests are often not feasible, being limited by the size of the test sites available that can operate at the appropriate temperature, pressure and neutron flux. Also, the most critical reactor components often have complex geometries and, until the advent of nano-scale testing capabilities allowing test specimens to be extracted directly from the irradiated component, information on property changes had to be gleaned from irradiating standardized test specimens only. Testing of ASTM standard specimens (or other qualified design that reduces specimen size) is still the mainstay of reactor materials research but such specimens still need to be made from the same material and subject to the same thermo-mechanical processing as the reactor component of interest.

An MTR provides a platform for irradiating material under controlled conditions of stress and temperature as well as neutron flux (atomic displacement damage rate). The main benefit of using MTRs is to test materials and gain advance knowledge on material performance prior to the point when it would have experienced the same dose in an operating power reactor. One might wish to perform an accelerated test and generate data on material performance equivalent to a neutron exposure expected over 30-40 years of power reactor operation in a short period of time, say 1-2 years. One must be careful that the simulation is not impacted by the accelerated neutron dose rate. Indeed in some cases, creep experiments for example, one needs to irradiate at the same damage rate as in a power reactor and the requirement for a high damage rate accelerated test is not as important as the ability to test under controlled conditions.

A reactor designer who needs information on the dimensional stability or mechanical properties of a given component made from specific alloys will solicit information from materials researchers that can be obtained from materials test reactors. Some of this information may be freely available from previous studies and some may need to be generated from a new irradiation test. In order to choose the best reactor to use for such an irradiation the reactor designer or researcher will need to know which test reactors can best simulate the power reactor conditions being targeted. Also the designer may be working to a schedule that requires accelerated testing in order to meet construction deadlines or, in the case of currently operating plant, to meet life management targets. Many operators of materials test reactors have user guides that provide details of the irradiation capabilities of their specific reactors and a researcher can use these guides to evaluate potential facilities.

For radiation damage studies, for example, researchers may be primarily interested in the fast neutron flux (E > 0.1 MeV) as neutrons in this energy group account for >95% of the atomic displacement damage in most core materials in most reactors [Garner & Greenwood, 2003]. Often neutron fluxes are quoted for higher energies

(E > 1 MeV). Neutrons with energies E > 1 MeV account for fewer atomic displacements (>70% in the core and >85% in the periphery of a PWR, for example). For situations where one wishes to assess material behaviour in a very low fast neutron flux relative to thermal flux (periphery regions in a CANDU reactor, for example), a substantial fraction of the damage could come from thermal neutrons for common engineering alloys containing Fe, Cr and Ni. The problem with using fast neutron fluence as the measure of dose was demonstrated by [Greenwood, 1994]. Greenwood showed that mechanical property data for 316 stainless steel in three different irradiations sites were not consistent when fast neutron fluence was the measure of dose. However, the mechanical property data became aligned when plotted against displacements per atom calculated using the full neutron energy spectrum, Figure 2-1. In this case there were substantial contributions to the atomic displacement damage coming from atom recoils associated with (n,Υ) reactions in two of the three sites. The contribution to the damage from low energy neutrons is less significant for Zr-alloys because these alloys have very low thermal neutron absorption cross-sections [Griffiths et al., 2017] and the dpa per unit of fast neutron fluence does not vary significantly for most reactors except fast reactors [Walters et al., 2018]. As a rule of thumb the contribution to atomic displacement damage from thermal neutrons (E < 0.1 eV) in engineering materials containing Fe, Cr and Ni is about 1% that of fast neutrons (E > 0.1 MeV). Therefore the contribution to radiation damage from thermal neutrons will be significant when the thermal/fast neutron flux > 100; this is particularly important in the periphery regions of heavy-water reactors. Spectral effects can also be very important for Ni-alloys whenever there is a high thermal neutron flux, i.e. everywhere in conventional power reactors. Ni is unusual because the major isotope (^{58}Ni) has a high thermal neutron absorption cross-section and forms ^{59}Ni, which is not naturally occurring and is only created in a nuclear reactor or from cosmic rays. ^{59}Ni itself has very high (n,Υ), (n,p) and (n,α) reaction cross-sections over a very large neutron energy range and can substantially enhance the atomic displacement damage rate [Greenwood, 1983; Greenwood & Garner, 1996]. Not only does the Ni transmutation lead to a significant enhancement in displacement damage rate (depending on neutron spectrum and dose) but it also results in the production of significant amounts of H and He gas (sufficient to affect material properties) when enough ^{59}Ni has been produced, reaching a maximum concentration of about 4 at % Ni, on average after 5 years of operation mid-core in a CANDU reactor core and about 3 at% Ni, on average after about 20 years of operation mid-core in a PWR, or 25 years in a BWR.

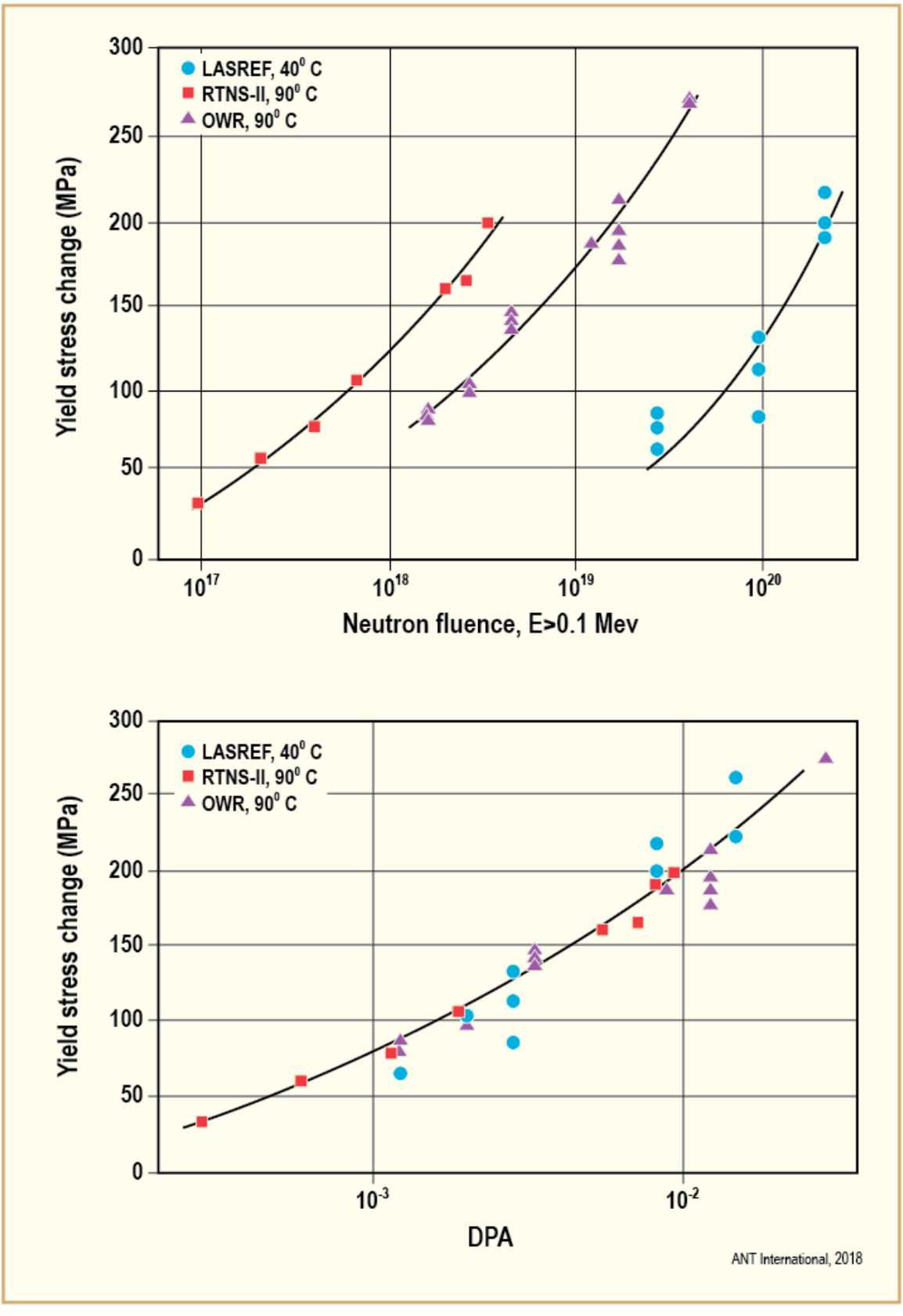

Figure 2-1: a) Increases in yield strength arising in a single heat of identical 316 stainless steel tensile specimens during irradiation at relatively low temperatures (40°C to 90°C) and low neutron exposures, but in very diverse neutron spectra, plotted vs. neutron fluence above 0.1 MeV; b) Same data plotted vs. dpa [Greenwood, 1994].

For accelerated tests it is often not sufficient to know which facilities/sites will give the desired fast neutron flux. It is important in many cases to know what the holistic effect of the neutron spectrum has on the radiation damage produced in a given material. Different neutron spectra will affect atomic displacement damage and gas production in different alloys for a given measure of neutron dose, e.g. fast neutron fluence [Griffiths et al., 2017]. Because the radiation damage is a function of both alloying elements and neutron flux, some facilities may be suitable for irradiating certain alloys but not others, depending on what one is aiming to achieve. Ultimately, any researcher has to perform detailed assessments of the effect of the neutron spectrum on the alloy of interest. This section aims to provide information that will assist a researcher in choosing a suitable irradiation facility but, more importantly, to highlight the potential impact of spectral effects when planning a material irradiation test. Each test reactor will have a number of irradiation sites for a researcher to choose from, with a range of neutron fluxes and spectra. Such information can be gleaned from the site documentation, often in the form of user guides, e.g. for ATR, BR2, HFIR, HFR, that are provided by each site to potential users. Ultimately, if one wishes to be able to simulate the effects of power reactor operation on materials in controlled environments one may need to match the irradiation conditions of the component in question with a test in the appropriate conditions, or in controlled conditions, where one is tailoring an irradiation to study a particular aspect of material behaviour. In either case one needs to know about the neutron spectra and, more importantly, the impact that particular irradiation conditions have on material behaviour. In the first instance one needs to consider what neutron flux intensity and neutron spectra one is dealing with for both the power reactors being studied and the materials test reactors being utilised for assessing materials behaviour.

The science behind the effects of neutron spectrum on atomic displacement damage and gas production will be described in Section 2.1 with some examples from specific reactors. Note that the neutron fluxes for each energy group are often represented by dividing by the lethargy for that group, i.e. flux per unit lethargy decrement. Neutron lethargy, or logarithmic energy decrement, u, is a dimensionless parameter that is the logarithm of the ratio of the energy of source neutrons to the energy of neutrons after a collision:

Equation 2-1: $u = \ln(E_o/E)$ or $u_2-u_1 = \ln(E_1/E_2)$

Dividing the neutron flux by lethargy decrement is preferred for plotting neutron spectra since it gives a more accurate impression of the spectrum because the flux spectra are often calculated in groups having differing widths of energies defining each group. Dividing by lethargy decrement compensates for the weighting inherent in the group width. Sections 2.2 and 2.3 will describe the spectra in current operating power reactors and materials test reactors respectively. Section 2.4 will give examples of the damage and gas production for selected engineering alloys in specific reactor environments. The focus will be on current generation power reactors and existing materials test reactors, including those like the Jules Horowitz Reactor (JHR) currently under construction. Materials test reactors that are no longer operating will not be assessed in detail, except for comparison with existing reactors and also to provide some context with historic data. Throughout, reference will be made to some of the historic data from ex-service MTRs: NRU, OSIRIS and DIDO reactors. For the most part historic dose information is available only in terms of fast neutron dose (E > 1 MeV or E > 0.1 MeV) but it is clear that spectral effects can have an impact on the interpretation of dose data that are given in terms of fast neutron fluence [Greenwood, 1994; Garner & Greenwood, 2003]. Data from these reactors (together with data from ATR) has been used extensively in the past for assessing radiation damage in Zr-alloys (using fast neutron damage as the measure of dose). Although Zr-alloys are not as sensitive to neutron spectra as other engineering alloys containing Fe, Cr and, in particular, Ni, the differences in atomic displacement damage rates due to differences in spectra can still be significant in different reactors and should be considered in any assessments of Zr-alloy irradiation creep and growth that are sensitive to radiation damage rates. In particular, corrections to dosimetry assessments for irradiations in DIDO prior to 1998 need to be applied to historic, published neutron dose data. In Section 2.5 other factors, post-cascade, that affect microstructure evolution, and thus material property changes, will be discussed. Finally, Section 2.6 will provide a discussion and summary of the main considerations in conducting materials testing in nuclear reactors and section 2.7 is a short conclusion.

2.1 Irradiation Damage and Gas Production – Spectral Effects

The primary effect of neutron irradiation on material properties is through the displacement of atoms [Thompson, 1969]. Transmutation is another effect, but the changes in elemental composition resulting from transmutation do not often have a significant effect on material properties. Production of gaseous atoms (He and H) from (n,α) and (n,p) reactions, however, can have serious deleterious consequences for component properties. Some transmutation reactions create secondary isotopes that can have a large effect on the displacement damage rates and also on gaseous atom production (He and H) [Greenwood, 1983; Garner & Greenwood, 1996]. The effect of neutrons on materials is dictated by the neutron spectrum and flux intensity. Typical neutron fluxes and coarse (3 group) neutron energy groupings for many power reactors and test reactors

are shown in Table 2-1 and Table 2-2, respectively. The values shown are maxima and give an indication of the range up to which neutron dose rates are exhibited in different facilities. For the materials test reactors the data are limited to materials test sites; neutron spectra and fluxes in sites used for testing fuel and isotope production are not considered here.

Atomic displacement by neutrons can occur through two main mechanisms:

(i) direct displacement of an atom by an energetic neutron creating a displacement cascade; and

(ii) absorption of a neutron by an atom and emission of a photon (γ-ray), or particle (β, p and α), that can induce further displacements and, more importantly, create a displacement cascade through the recoiling atom.

The mechanism of displacement damage through neutron absorption applies to neutrons of all energies. The creation of a collision cascade by direct displacement of a primary knock-on atom (PKA) only occurs for neutrons with sufficient energy that the PKA itself has a high enough energy to create multiple atomic displacements. For example, a 1 MeV neutron can produce PKAs with energies up to about 70 keV and 40 keV in elastic collisions with Fe and Zr atoms, respectively.

2.1.1 Atomic Displacement by Collision with Neutrons

Neutrons with energies >0.1 MeV are called fast neutrons, as opposed to the slower thermal neutrons, e.g., those with energies <0.5 eV, that are more readily absorbed by material within the reactor core. Some elements (Zr for example) have very low thermal neutron capture cross-sections and by far the main damage mechanism is direct atomic displacement. Assuming an atomic threshold displacement energy of 40 eV, the Zr atom (mass = 91.22 amu) can be displaced by neutrons (amu = 1) with energies >930 eV[4] [Thompson, 1969]. For elements such as Cr, Fe and Ni, a neutron with energy >600 eV is sufficient to cause atomic displacement. For neutrons with energies >100 keV (0.1 MeV), the energy transferred to the primary knock-on atom (PKA) is sufficiently high that it creates many additional displacements as the PKA energy is dissipated within the crystal lattice in the form of a collision cascade (see section 3.4). The calculation of the number of displacements per PKA is based on a consideration of the damage energy, i.e. how much of the energy of the primary knock-on atom is available to create further displacements. The code used to determine displacement damage in this report, SPECTER [Greenwood & Smither, 1985] uses the differential cross-section library [ENDF/B-V nuclear library, http://t2.lanl.gov/nis/data.shtml] to determine the PKA spectrum and the model for damage energy developed by [Norgett et al., 1975] to determine the total number of atomic displacements per PKA.

Typically the neutrons with energies >0.1 MeV create most of the damage and, for Zr in particular, an approximate measure of the relative displacement damage dose can be obtained by measuring and comparing neutron fluxes in these high energy ranges.

2.1.2 Atomic Displacement by Absorption of Neutrons

Neutron absorption, followed by particle emission and subsequent atom recoil, can be an important contributor to atomic displacement damage in materials containing elements such as Cr, Fe and Ni that have high thermal neutron absorption cross-sections.

Neutrons with energies less than that needed to create atomic displacement by direct collision and transfer of momentum, i.e., <1 keV, can produce damage by being absorbed. The subsequent particle or photon emission induces a recoil cascade. It is not just the low energy neutrons that can be absorbed by atoms, thus inducing photon or particle emission, nuclear reactions involving transmutation also occur up to high energies, >1 MeV. The nuclear absorption cross-sections, (n, γ), (n, α) and (n, p) are shown in Figure 2-2 for the major isotopes of

[4] The maximum momentum transferred by a head-on hard-sphere collision is

$$\Lambda = \frac{4M_1M_2}{(M_1+M_2)^2}$$

where M_1 is the mass of the bombarding particle (neutron) and M_2 is the mass of the target atom.

3 Ion Irradiation Techniques and Facilities (Ron Adamson)

3.1 Introduction

During the current century, accelerated efforts have been made to "simulate" or "emulate" the effects of neutron irradiation on material properties by conducting ion "bombardment" (or ion irradiation or charged particle irradiation, used interchangeably in this report and in the open literatures) to gather data. A review of most of such (or related) experiments on zirconium alloys in the time period 1966 to 2014 was provided by [Adamson, 2014]. [Yan et al., 2015] also reviewed ion irradiation effects in zirconium alloys, and reviews for a broader range of materials are provided by [Was, 2007], [Was, 2015], [Griffiths et al., 2017] and, [Zinkle & Snead, 2018]. The book by [Was, 2007] covers many aspects of the effects of various forms of radiation on materials properties.

3.2 General comparison with neutron irradiation

Compared with neutron irradiations, advantages in using ion irradiations include:

- high damage rates, allowing irradiation doses relevant to in-reactor performance to be reached in hours or days
- relatively low overall cost
- radiation activation absent or relatively low
- potential to control experimental variables (temperature, dose or fluence, compositions)
- in-situ (e.g. real time TEM observations) experiments possible
- short experimental "turn-around time" may allow iteration based on "what is learned"
- allows analytical studies with separation of parameters; e.g., multiple irradiations with different parameters.

Compared with neutron irradiations, disadvantages in using ion irradiations include:

- high damage rates
  - is property being studied sensitive to damage rate?
  - what is the effective temperature (temperature shift, [Adamson, 2014], [Was, 2007]) of the irradiation?
- short ion penetration lengths that may introduce surface effect issues (surfaces attract both ions and point defects)
- rastered ion beams that produce macroscopic uniform damage on a specimen may produce different average and instantaneous damage rates
- transmutations do not occur or are different from transmutations caused by neutrons

3.3 Obtaining data

Commercial reactor

The extensive record of material performance as affected by **neutron irradiation** recognizes the large effort required to gather the requisite properties via a commercial power reactor:

- 1 year paperwork for non-standard fuel arrangements
- 2-7 years in-reactor exposure
- several years for in-pool measurements or shipment to hot cells
- extensive effort and precautions needed to handle highly radioactive material
- several years for in-cell or in-laboratory examinations

Test reactor

- 1 year for paperwork
- 1-3 years in-reactor exposure
- 1 to several years to transfer to hot cell
- extensive effort and precautions needed to handle highly radioactive material
- several years for in-cell or in-laboratory examinations

Ion irradiation

- a few months for specimen preparation
- a few months to arrange irradiation in a facility suiting the needs of the experiment
- days or weeks to reach the dose or dpa goal
- possibly a few weeks for any induced radioactivity to diminish (depending on ion energy, dose and material, this time may be zero) and specimen sent to material characterization laboratory
- weeks or months to examine or test specimens

Although a generalization, whereas it could take >10 years to obtain data from a reactor irradiation, it might take just a few years for an ion irradiation.

Cost comparison

- In all cases, ion irradiation significantly less expensive than neutron irradiation
- Ion irradiation facilities exist in national laboratories or universities
  - specific costs for ions differ depending on specific arrangement between user and provider;
  - in some cases cost of ions is zero.

3.4 Basic irradiation damage

The initial radiation damage caused by a neutron, heavy ion or proton is the Brinkman displacement spike (cascade) consisting of a vacancy (V) rich core surrounded by a shell of interstitials (SIAs). Slightly more realistic is the Seegar depleted (vacancy rich) zone. Both are shown in Figure 3-1. In the temperature ranges relevant to LWR materials, the results are clusters of Vs or SIAs, individual Vs and SIAs (collectively called Frenkel pairs) and after some short time ($<10^{-10}$ seconds), vacancy and interstitial dislocation loops. Taken together, these are called "irradiation damage".

In the case of electron irradiations the electron mass is so low that little damage is created (as in the average case in-reactor) unless the electron energy is very high (1 MeV). Even in that case only Frenkel pairs are created.

No single type of radiation can exactly simulate neutron irradiation, as each has its own basic damage-creation characteristics. This is illustrated in Figure 3-2 and Table 3-1, adapted from [Was, 2007] for Ni. Electrons from a high voltage electron microscope (HVEM) create only Frenkel pairs. The average energy transferred to a Zr atom in a "collision" $\overline{T}$, is 60 eV, which is only slightly more than the energy needed to displace the atom ($E_d \approx 40$ eV). This displacement efficiency ($\in$ = the fraction of Frenkel pairs which survive the collapse of the displacement spike or cascade) is near 100%, since no spikes are created. With electrons, very high beam currents are possible, allowing high damage rates and causing high local temperatures. 1 MeV protons do cause small, widely spaced cascades, and 25% of the Frenkel pairs survive. 1 MeV heavy ions create dense, large cascades, more similar to neutrons than the others. Although the goal of many current experiments is to "simulate or emulate" reactor neutron irradiation with ions and electrons, it is really a "different irradiation altogether" [Motta et al., 1996]. Relating to the effects of reactor irradiation is the challenge for the experimenter.

Another difference between reactor neutrons and accelerated ions is the respective energy spreads. For neutrons the energy range is from very low (0.03 eV for thermal neutrons) to several MeV. For ions, the beam is almost

mono-energetic with a very narrow energy spread; e.g., for electrons there is a Gaussian energy distribution with a wide enough spread that could result in dose gradients across the beam.

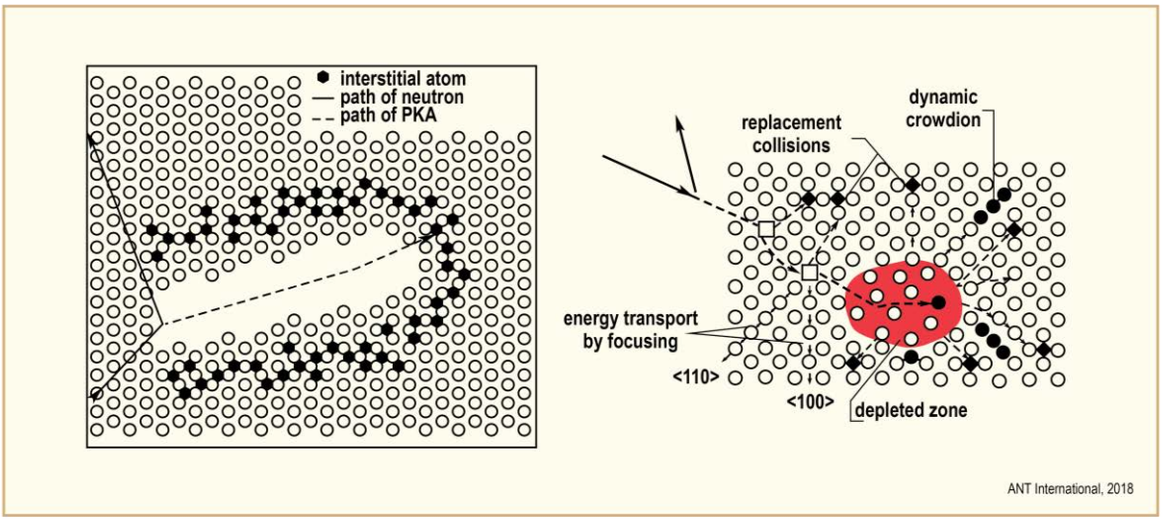

Figure 3-1: Displacement spike (cascade) [Brinkman, 1956] and revised by [Seegar, 1958].

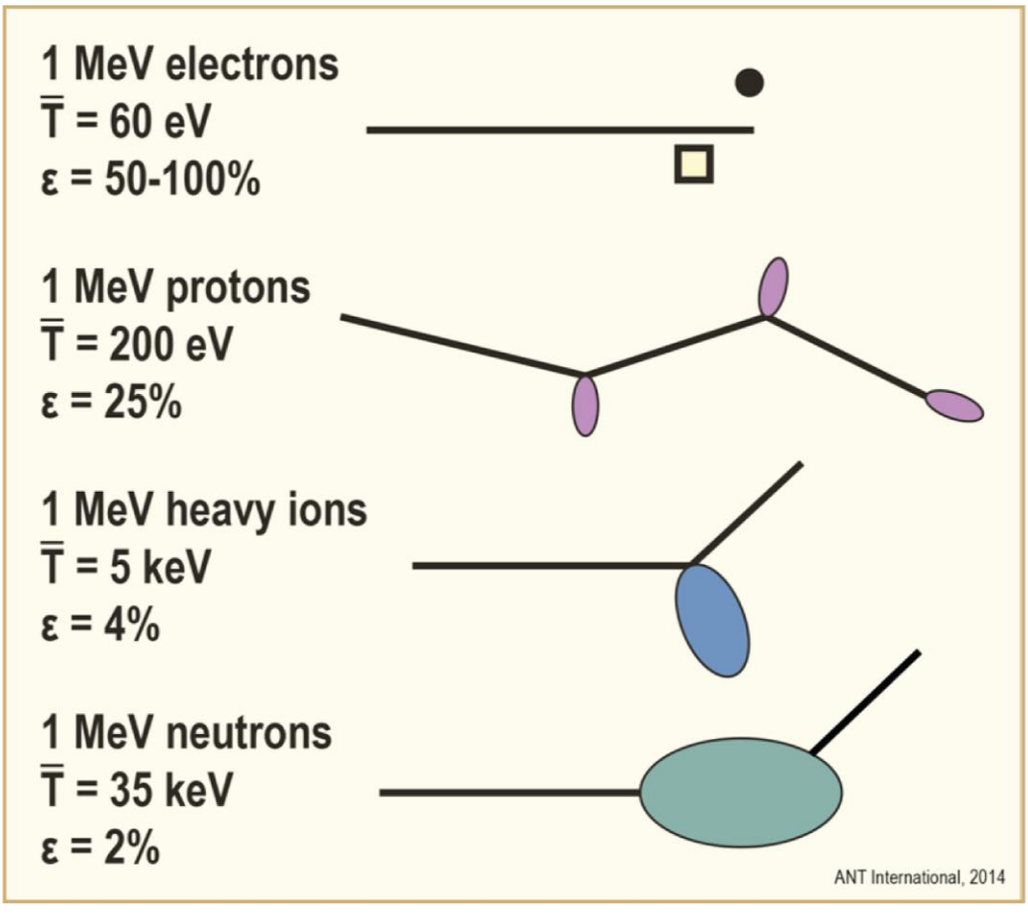

Figure 3-2: Difference in damage morphology, displacement efficiency ($\in$), and average recoil energy ($\bar{T}$) for 1 MeV particles incident on Ni, after [Was & Allen, 1994].

4 Summary

In this STR we have described different facilities that can be used for materials irradiations in support of current reactor operation, for the development of improved components, and also to assess new materials for advanced rectors.

To predict future performance of components in operating reactors, or for the design of new reactors, materials test reactors (MTRs) have been utilized to perform accelerated or controlled evaluations of irradiated materials. MTRs have provided essential support for nuclear power programs over the decades. Associated with hot laboratories for the post irradiation examinations, they are an important tool in research facilities for the fission and fusion domain. One value from testing in MTRs comes from the higher neutron fluxes and radiation damage production rates. Information regarding some of the most active operating MTRs and promising future MTRs has been provided in section 1 of this STR.

For accelerated tests it is often not sufficient to know which facilities/sites will give the desired fast neutron flux. It is important in many cases to know what the holistic effect of the neutron spectrum has on the radiation damage produced in a given material. Different neutron spectra will affect atomic displacement damage and gas production in different alloys for a given measure of neutron dose, e.g. fast neutron fluence [Griffiths et al., 2017]. Because the radiation damage is a function of both alloying elements and neutron flux, some facilities may be suitable for irradiating certain alloys but not others, depending on what one is aiming to achieve. Ultimately, any researcher has to perform detailed assessments of the effect of the neutron spectrum on the alloy of interest. This section aims to provide information that will assist a researcher in choosing a suitable irradiation facility but, more importantly, to highlight the potential impact of spectral effects when planning a material irradiation test.

Material irradiation in an MTR followed by examination in a hot cell facility is a very time consuming process because of the low damage rates that even the highest flux reactors exhibit. In addition the high cost of research on irradiated materials, in the face of shrinking budgets, puts additional constraints on this approach.

A promising partial solution to this problem is to use ion irradiation to irradiate materials to very high doses. The advantages of ion irradiation are many. Dose rates (typically 10^{-3} to 10^{-4} dpa/s) are much higher than under neutron irradiation (10^{-7} to 10^{-8} dpa/s), which means that 100s of dpa can be reached in days or weeks instead of years. Because there is little activation, the samples have little or no radioactivity and often can be handled in a laboratory environment. Control of ion irradiation experiment variables is much better than experiments in nuclear reactors. Challenges to the implementation of ion irradiation as a surrogate for neutron irradiation include rate effects on microstructures and effective temperature, small irradiation volumes, and accounting for transmutations.

On the one hand ion irradiations provide a means of studying some aspects of the irradiation processes at very high damage rates without inducing high levels of activity in the materials, on the other, they are limited in their ability to irradiate large volumes of material and do not induce the same gas atom production (especially He) from transmutation. With the advent of nano-scale specimen machining and testing some of the volume limitations can be addressed but reduced volumes also enable one to work directly with highly activated materials after neutron irradiation. The main advantage then for ion irradiation comes about from the very high doses that can be achieved in small volumes of material in relatively short periods of time. Damage rates are then an issue.

When the vacancy migration energy is high and/or the temperature is low the material heals itself and recombination of point defects is dominant. Under the conditions where the material is affected by recombination increasing the damage rate will enhance the recombination, effectively negating any benefit one might have from displacing the atoms at a higher rate. In ion irradiations where the displacement damage rate can be orders of magnitude higher than in reactor, researchers often apply a temperature shift to compensate for the tendency for more recombination at lower temperatures. Unfortunately such a shift impacts the value of the simulation because one is now not at the temperature of interest. Flux effects are less of an issue for MTR irradiations provided the temperature is high enough. Typically high flux reactors induce damage rates up to about 10^{-6} dpa/s and power reactors operate in the range of 1 -10 x 10^{-8} dpa/s For damage rates in the range of 10^{-6} to 10^{-8} dpa/s there is little concern with recombination for steels and Ni-alloys provided the irradiation temperature is >400 °C. For Zr-alloys with much lower vacancy migration energies, there should be little concern provided the irradiation temperature is >200 °C. It is not sufficient to simply conduct an accelerated test if the increased damage rate is negated by increased recombination. For each irradiation in an MTR there will be an optimum set of conditions for a given material where accelerated testing can be achieved and these depend on the damage rate, the irradiation temperature and the microstructure, as the sink density also affects the

References

Adamson R. B., Bell W. L. and Lee D., *Use of Ion Bombardment to Study Irradiation Damage in Zirconium Alloys*, Zirconium in Nuclear Applications, ASTM STP 551, American Society for Testing and Materials, pp. 215-228, 1974.

Adamson R. B., *Charged Particle Bombardment in Zirconium Alloys – A Review*, ZIRAT19/IZNA14 Special Topic Report, ANT International, Mölnlycke, Sweden, 2014.

Arkhangelsky N. and Izhutov A., *Capabilities and Capacities of RIAR Research Reactors*, IAEA, Vienna, June 10-12, 2013.

ASTM Standard E521 – 16, *Standard Practice for Neutron Radiation Damage Simulation by Charged Particle Irradiation*, 2006.

ASTM Standard E263-13, Standard *Test Method for Measuring Fast-Neutron Reaction Rates by Radio-activation of Iron*, 2013.

ATR NSUF, *Advanced Test Reactor National Scientific User Facility Users' Guide*, 2009.

ATR, https://wikivisually.com/wiki/Advanced_Test_Reactor, 2018.

Bacon D.J. et al., *Computer Simulation of Defect Production by Displacement Cascades in Metals*, Nuclear Instruments and Methods in Physics Research B, Vol. 102, pp. 37-46, 1995.

Bess J. D. and DeHart M. D., *Baseline Assessment of TREAT for Modeling and Analysis Needs*, INL/EXT-15-35372, Idaho National Laboratory, September 2015.

Bess, J.D. et al., *TREAT Multi-SERTTA Neutronics Design and Analysis Support*, Proc. PHYSOR 2016, ANS, Sun Valley, Idaho, May 1-5, 2016.

Bieniawski A., *An Overview of the NNSA Global Threat Reduction Programs Goals for HEU Minimization*, RERTR 2009 International Meeting on Reduced Enrichment for Research and Test Reactors", Beijing, China, November 1-5, 2009.

Bignan G., Colin M. and Pierre J., *A New High Performance Material Testing Reactor Working as an International Facility: First Developments to address R&D on Material*. 2nd International Workshop MINOS, 4-6, , Cadarache, France, November 2015.

Bignan G. et al., *The Jules Horowitz Reactor Research Project: A New High Performance Material Testing Reactor Working as an International User Facility – First Developments to Address R&D on Material*, 22nd Int. Workshop Irradiation of Nuclear Materials: Flux and Dose Effects, EPJ Web of Conferences 115, 01003, DOI: 10.1051/epjconf/201611501003, 2016.

Blaumann H. et al., RA-10: *A New Argentinian Multipurpose Research Reactor*, International Atomic Energy Agency (IAEA): IAEA, https://inis.iaea.org/search/search.aspx?orig_q=RN:44019508, 2012.

Brinkman J.A., Amer J. Phys 24:251, 1956.

BWR Vessel Internals Project, *Weldability of Irradiated LWR Structural Components*, TR-108707, BWRVIP-45, 1997. www.epri.com.

Cann C. D., So C. B., Styles R. C., Coleman C. E., *Precipitation in Zr-2.5Nb enhanced by Proton Irradiation*, J. Nucl. Mat. 205, pp. 267-272, North-Holland, 1993.

Cetinera N. O. et al., *An Advanced Materials Irradiation Facility for Materials and Fuels Irradiations at the High Flux Isotope Reactor*, TopFuel meeting, Boise, ID, September 2016.

Chapman O. J. V., McElroy R. J. and Sheldon B. E., *Irradiation Creep and Growth during Proton and Neutron Bombardment of Zircaloy-2 Plate*, Zirconium in the Nuclear Industry: Sixth International Symposium, ASTM STP 824, D. G. Franklin and R. B. Adamson, Eds. American Society for Testing and Materials, pp. 343-375, West Conshohocken, PA, 1984.

Griffiths M., Mecke J. F. and Winegar J. E., Evolution of Microstructure in Zirconium Alloys During Irradiation, Zirconium in the Nuclear Industry: 11th Int'l Symposium, ASTM STP 1295, E. R. Bradley and G. P. Sabol, Eds., American Society for Testing and Materials, pp. 580-602, 1996.

Griffiths M. et al., *Degradation of Ni-alloy Components in a CANDU Reactor Core*, 16th Int. Symposium on Environmental Degradation in Materials, Asheville, NC, 2013.

Griffiths M., *The Effect of Irradiation on Ni-containing Components in CANDU Reactor Cores: A Review*, AECL Nuclear Review, Vol. 2, No. 1, pp. 1-16, 2013; erratum AECL Nuclear Review, Vol. 3, No. 2, p. 89, 2014.

Griffiths M., Walters L., Greenwood L. R., Garner F., *Accelerated Materials Evaluation for Nuclear Applications*, Journal of Nuclear Materials, 488, pp. 46-62, 2017.

Grossbeck M.L., Ehrlich K. and Wassilew C., *An Assessment of Tensile, Irradiation Creep, Creep Rupture and Fatigue Behavior in Austenitic Stainless Steels with Emphasis on Spectral Effects*, J. Nucl. Mater., Vol. 174, p. 264, 1990.

Heald P.T. and Speight M.V., *Point Defect Behaviour in Irradiated Materials*, Acta. Metall., Vol. 23, p. 1389, 1975.

Hengstler-Eger R. M. et al., *Heavy Ion Irradiation Induced Dislocation Loops in AREVA's M5 Alloy*, J. Nucl. Mat. 423, pp. 170-182, 2012.

HFIR*, Neutron Sciences at Oak Ridge National Laboratory*, https://neutrons.ornl.gov/hfir, 2018.

High Flux Reactor (HFR) - *Characteristics of the Installation and the Irradiation Facilities*, HFR Petten, 2005 https://iet.jrc.ec.europa.eu/sites/default/files/documents/brochures/hfr_mini_blue_book.pdf

IAEA RRDB (Research Reactor Data Base), https://nucleus.iaea.org/RRDB/RR/ReactorSearch.aspx?filter=0 , 2018.

Idrees Y., Yao Z., Kirk M. A., Daymond M. R., *In Situ Study of Defect Accumulation in Zirconium Under Heavy Ion Irradiation*, J. Nucl. Mat. 433, pp. 95-107, 2013b.

Ilas G., Bryan C. and Gehin J., *Needs and Requirements for Future Research Reactors (ORNL perspectives)*, Oak Ridge National Laboratory, Oak Ridge, TN, Feb. 2016.

INL (Idaho National Laboratory), *NSUF Ion Beam Investment Options Workshop*, INL/EXT-16-37857, INL Meeting Center, Energy Innovation Laboratory, March 22–24, 2016.

Izhutov A. L., Starkov, V. A., Pimenov, V. V., Fedoseev, V. E., Maynskov, S. V., conversion of the mir.m1 reactor to LEU-fuel, RERTR 2012 - 34th International meeting on reduced enrichment for research and test reactors, Warsaw, Poland, October 14-17, 2012

Izhutov A. I. et al., *Prolongation of the BOR-60 Reactor Operation*, Nuclear Engineering and Technology, Volume 47, Issue 3, April 2015.

Izhutov A. L. et al., *SM Reactor Core Modernization Program*, Research reactor fuel materials meeting, A0093, 2017a.

Izhutov A. L., Starkov V. A., Pimenov V. V., Marikhin N. Y., *SM-3 Reactor Characteristics After Core Modernization*, Research reactor fuel materials meeting, A0094, 2017b.

Jones R.B., Edens, D.J. and Wooton, M.R., *Influence of Thermal Neutrons on the Hardening and Embrittlement of Plate Steels*, Effects of Radiation on Materials: 19th International Symposium, ASTM STP 1366, M. L. Hamilton, A. S. Kumar, S. T. Rosinski, and M. L. Grossbeck, Eds., American Society, for Testing and Materials, pp. 366-382, West Conshohocken, PA, 2000.

Kai J. J., Huang W. I. and Chou H. Y., *The Microstructural Evolution of Zircaloy-4 Subjected to Proton Irradiation*, J. Nucl. Mat. 170, pp. 193-2009, 1990.

Woo O. T., Hutcheon R. M. and Coleman C. E., *Precipitation in Zr-2.5Nb during 10 MeV Electron Irradiation*, Mat. Res. Soc. Symp. Proc. Vol. 373, 1995.

Wooding S.J. et al., *A Molecular Dynamics Study of High-Energy Displacement Cascades in α-Zirconium*, Journal of Nuclear Materials, Vol. 254, pp.191–204, 1998.

Woolstenhulme N. E. and Wiest J. D., *ATF Transient Testing Pre-Conceptual Design and Engineering Considerations Summary*, INL Report INL/EXT-13-29898, August 2013.

Woolstenhulme N. E. et al., *Capabilities Development for Transient Testing of Advanced Nuclear Fuels at TREAT*, Top Fuel 2016, Boise, ID, September 11-15, 2016.

Yamada S. and Kameyama T., *Observation of C-component Dislocation Structures Formed in Pure Zr and Zr-base Alloy by Self-ion Accelerator Irradiation*, J. Nucl. Mat. 422, 167-172, 2012.

Yan C. et al., *Effects of Ion Irradiation on Microstructure and Properties of Zirconium Alloys – A Review*, Nucl. Eng. Tech. 47, pp. 323-331, 2015.

Yang T. Y., Yu G. P. and Chen L. J., *Precipitate Stability in Beta-Quenched Zircaloy-4 Irradiated by Nitrogen-Ions at 470°C*, J. Nucl. Mat. 150, pp. 67-77, 1987.

Ziegler J. F., Biersack J. P. and Littmark U., *The Stopping and Range of Ions in Matter, Pergamon*, New York, 1996.

Ziegler J., SRIM, <http:www.srim.org>, 2008.

Zinkle S.J. and Snead L.L., *Opportunities and Limitations for Ion Beams in Radiation Effects Studies: Bridging Critical Gaps Between Charged Particle and Neutron Irradiations*, Scripta Materialia 143, 154–160, 2018

Zou H., Hood G. M., Roy J. A., Packwood R. H., *Irradiation-Driven Solute Redistribution in Zr Alloys*, J. Nucl. Mat. 245, pp. 248-252, 1997.

Zu X. T. et al., *Effect of Proton and Ne Irradiation on the Microstructure of Zircaloy-4*, Philosophical Magazine, Vol. 85, Nos. 4-7, pp. 649-659, 01 February-01 March, 2005.

List of Abbreviations

µXRD	Micro X-Ray Diffraction
ADS	Accelerator Driven System
appm	atom parts per million
ATR	Advanced Test Reactor
BU	Burn Up
BWFC	Boiling Water Fuel Capsule
BWR	Boiling Water Reactor
CANDU	Canada Deuterium Uranium
CARR	China Advanced Research Reactor
CEA	Commissariat à l'énergie atomique et aux énergies alternatives (France)
CVN	Charpy V Notch (Impact Chary test specimen)
eATF	Enhanced Accident Tolerant Fuel
EB welding	Electron Beam welding
EBSD	Electron BackScatter Diffraction
EDS, WDS	Energy Dispersive Spectroscopy, Wavelength Dispersive Spectroscopy
EMIR	Étude des Matériaux sous IRradiation
EPMA	Electron Probe Micro-Analysis
HANARO	High Flux Advanced Neutron Application Reactor
HEU	Highly Enriched Uranium
HFETR	High Flux Engineering Test Reactor
HFIR	High Flux Isotope Reactor
HFR	High Flux Reactor
HFR	High Flux Reactor (Petten, NL)
HR-STEM	High-Resolution Scanning Transmission Electron Microscope
HTHF	High Temperature High Flux (irradiation device)
HVEM	High Voltage Electron Microscope
HWBR	Heavy Water Boiling Reactor
IAEA	International Atomic Energy Agency
IB	Ion Bombardment
ICERR	International Centres of Excellence based on Research Reactors
IET	Institute of Energy Technology (Norway)
INL	Idaho National Laboratory
I-SCC	Iodine induced Stress Corrosion Cracking
ISCC	Irradiation Enhanced Stress Corrosion Cracking
JMTR	Japan Material Test Reactor
JRC	Joint Research Centre
LAMDA	Low Activation Materials Development and Analysis Laboratory
LEU	Low Enriched Uranium
LHGR	Linear Heat Generation Rate
LOCA	Loss Of Coolant Accident
LVDT	Linear Variable Differential Transformer
LWR	Light Water Reactor
LWR	Light Water Reactors
MeV	Million Electron Volt
MITR	Massachusetts Institute of Technology Reactor
MOX	Mixed Oxide
MPa	Mega Pascals
MTR	Material Test Reactor
MTR	Material Testing Reactors
MURR	Missouri University Research Reactor
MW	Mega Watt
NDE	Non-Destructive Examination
NPP	Nuclear Power Plant
NRG	Nuclear Research and consultancy Group
NSUF	National Scientific User Facility
ORNL	Oak Ridge National Laboratory
PIE	Post Irradiation Examination
PIE	Post Irradiation Examination Facility
PWR	Pressurised Water Reactor

Unit conversion

TEMPERATURE		
°C + 273.15 = K	°C × 1.8 + 32 = °F	
T(K)	T(°C)	T(°F)
273	**0**	32
289	16	61
298	25	77
373	**100**	212
473	**200**	392
573	**300**	572
633	360	680
673	**400**	752
773	**500**	932
783	510	950
793	520	968
823	550	1022
833	560	1040
873	**600**	1112
878	605	1121
893	620	1148
923	650	1202
973	**700**	1292
1023	750	1382
1053	780	1436
1073	**800**	1472
1136	863	1585
1143	870	1598
1173	**900**	1652
1273	**1000**	1832
1343	1070	1958
1478	1204	**2200**

Radioactivity	
1 Sv	= 100 Rem
1 Ci	= 3.7×10^{10} Bq = 37 GBq
1 Bq	= 1 s^{-1}

MASS	
kg	lbs
0.454	**1**
1	2.20

DISTANCE	
x (µm)	x (mils)
0.6	0.02
1	0.04
5	0.20
10	0.39
20	0.79
25	0.98
25.4	**1.00**
100	3.94

PRESSURE		
bar	MPa	psi
1	0.1	14
10	**1**	142
70	7	995
70.4	7.04	**1000**
100	10	1421
130	13	1847
155	15.5	2203
704	70.4	**10000**
1000	100	14211

STRESS INTENSITY FACTOR	
MPa$\sqrt{m}$	ksi$\sqrt{inch}$
0.91	**1**
1	1.10