Physical Ageing of The Research Reactor Core Structural Materials Due To Neutron Irradiation Exposure: A Review

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INFORMASI ARTIKEL	ABSTRACT

PHYSICAL AGEING OF THE RESEARCH REACTOR CORE STRUCTURAL MATERIALS DUE TO NEUTRON IRRADIATION EXPOSURE: A REVIEW. A research reactor (RR) is a nuclear reactor that has function to generate and utilize neutron flux and radiation ionization for research purposes and industrial applications. More than 60% of current operating RRs have been operated for 30 years or more. As the time passes, the functional capabilities of structures, systems and components (SSCs) of those RRs deteriorate by physical ageing, which can be caused by neutron irradiation exposure such as irradiation induced dislocation and microstructural changes. To extend the lifetime and/or to avoid unplanned outages, ageing on the safety related SSCs of RRs need to be properly managed. An ageing management is a strategy to engineer, operate, maintenance, and control SSC degradation within acceptable limits. The purpose of this study is to review physical ageing of the core structural materials of the RRs caused by neutron irradiation exposure. In order to achieve this objective, a wide range of literatures are reviewed. Comprehensive discussions on irradiation behaviors are limited only on reactor vessel and core support structure materials made from zirconium and beryllium as well as their alloys, which are widely used in RRs. It is found that the stability of the mechanical properties of zirconium and beryllium as well as their alloys was mostly affected by the neutron fluences and temperatures.

ABSTRAK
PENUAAN MATERIAL STRUKTUR TERAS REAKTOR RISET SECARA FISIK YANG DISEBABKAN OLEH PAPARAN RADIASI NEUTRON: SEBUAH KAJIAN. Reaktor riset adalah reaktor nuklir yang difungsikan untuk pembangkitan dan pemanfaatan fluks neutron dan ionisasi radiasi untuk tujuan penelitian dan aplikasi industri. Lebih dari 60% reaktor riset yang ada saat ini telah beroperasi selama 30 tahun atau lebih. Seiring dengan berjalannya waktu, kemampuan struktur, sistem dan komponen (SSK) tentunya mengalami penurunan yang dikenal dengan penuaan fisik. Penuaan fisik ini dapat disebabkan oleh karena adanya paparan radiasi neutron seperti irradiasi yang menyebabkan terjadinya dislokasi dan perubahan mikrostruktur material. Untuk memperpanjang umur reaktor dan untuk mengurangi terjadinya pemadaman yang tidak diinginkan maka perlu dilakukan manajemen penuaan SSK, khususnya yang terkait dengan keselamatan operasi. Tujuan dari penelitian ini adalah untuk mengkaji penuaan fisik yang terjadi pada material struktur teras reaktor riset yang disebabkan oleh paparan iradiasi neutron. Untuk mencapai tujuan ini maka dilakukan kajian pada berbagai macam pustaka yang terkait. Pembahasan yang komprehensif tentang sifat-sifat iradiasi dibatasi hanya pada bejana reaktor dan material struktur pendukung teras yang terbuat dari zirconium dan beryllium dan senyawanya yang umum digunakan oleh reaktor riset. Dari kajian yang telah dilakukan, ditemukan bahwa stabilitas sifat-sifat mekanik zirconium dan beryllium serta senyawanya sangat dipengaruhi oleh fluent dan temperatur neutron.
Kata kunci: Reaktor riset, penuaan fisik, iradiasi neutron, zirconium, beryllium.

1. INTRODUCTION

A research reactor (RR) is a nuclear reactor whose main purposes are to generate and utilize neutron flux and radiation ionization for researches such as material researches and industrial applications such as medical applications[1-4]. RRs are usually equipped with various experimental facilities such as in-core irradiation and beam tubes[5]. For example, the Indonesian multi purpose reactor (MPR) type not only provides in-core irradiation facilities such as central irradiation position but also off-core irradiation facilities such as six beam tubes[6].

The Ghana RR-1 (GHARR-1) is equipped with 10 irradiation sites, namely: five sites

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located inside the beryllium annulus reflector and the other five sites located outside the beryllium annulus reflector[7]. In addition, the high flux advanced neutron application reactor (HANARO), which is operated by Korea Atomic Energy Research Institute (KAERI), has been successfully utilized for fuel and material irradiation tests, radioisotope productions, and industrial applications as well as academic and research purposes[8].

Currently, more than 60% of operating research reactors have been over 30 years old[9]. As the time passes, the functional capabilities of structures, systems and components (SSCs) deteriorate by ageing [10, 11]. Ageing is a process in which the reliability of any equipment depreciates, and hence the likelihood of that equipment to fail increases [12].

To extend the lifetime and/or to avoid unplanned outages of RRs, ageing on the RRs' safety related components need to be properly managed[9]. RRs, which have been operated more than 40 years, need to be frequently and thoroughly inspected to ensure the integrity of their safety related components[13]. An ageing management is defined as a strategy to engineer, operate, maintenance, and control SSC ageing degradation within acceptable limits[1]. Activities in ageing management include reparation, refurbishment and replacement of safety related-SSCs, which should be proactively realized throughout the lifetime of RRs. Generally, ageing management is performed for periodic safety review and long-term operation[14].

The International Atomic Energy Agency (IAEA) defines two types of ageing that could happen to equipment, i.e. non-physical ageing and physical ageing. The non-physical ageing refers to the process of out of date due to knowledge and technology evolution or code and standard changes such as outdated documentations. Meanwhile, the physical ageing is caused by the physical, chemical and/or biological mechanisms such as thermal and/or radiation embrittlement[1]. Furthermore, Nitoi et al.[10] confirmed four issues that need to be dealt with regarding the ageing management. Firstly, SSCs, which are sensitive to ageing, need to be identified. Secondly, components, which need ageing mitigation, need to be recognized. Thirdly,

material degradation processes need to be understood. Lastly, the effectiveness of any method dedicated to test, maintenance, surveillance, and inspection of those components needs to be measured.

The purpose of this study is to review physical ageing of the core structural materials of the RRs due to neutron irradiation exposure. The behaviors of neutron irradiations, such as irradiation induced dislocation and microstructural changes are investigated. Comprehensive discussions on irradiation behaviors are limited only on reactor vessel and core support structure materials made from zirconium and beryllium as well as their alloys, which are widely used in RRs. This study will provide important information for ageing management personnel prior to developing and managing a proper and effective strategy for maintaining the integrity and functional capability of the RRs' safety-related SSCs.

2. RESEARCH REACTOR CORE STRUCTURE MATERIALS

RRs are mostly open pool type reactors and the temperature of the coolant is maintained below 100^0C to avoid coolant boiling[15]. The volume fraction of the coolant is relatively high and neutron spectrum is quite soft. Consequently, the coolant temperature coefficient is quite small and the power coefficient of reactivity is close to zero or just in slightly negative values[16]. To maintain the temperature of the fuels relatively low when RRs in high performances, the flow rate of the coolant should be quite fast[17]. Based on the size of the core, RRs can be grouped into two types, i.e. a compact core and a large core. The compact core is mainly used for utilizing beam tubes. Meanwhile, the large core is mainly for isotope production and material test irradiation[18].

The utilizations and competitiveness of RRs are directly determined by the neutron spectrum generated in the irradiation facilities[16]. For example, the German FRM-II RR was designed for beam application optimization, which can produce thermal neutron flux up to 8 x 10^{14} ncm^{-2}s^{-1}[19].

Meanwhile, the Australia OPAL RR was designed to allow cold/thermal/hot beam researches. This RR can produce neutron flux of 3×10^{14} ncm^{-2}s^{-1}[20]. The Ghana RR-1 (GHARR-1) has been designed to be mainly used for neutron activation analysis with a maximum neutron flux of 1×10^{12} ncm^{-2}s^{-1}[21-24]. The average fluxes of typical research reactors is shown in Table 1[2].

Table 1. The Typical Average Neutron Fluxes Generated by RRs Based on Their Thermal Powers[2].

Parameter, Unit	Values				
Thermal Power, MWt	0.250	1	2	10	20
Neutron flux, ncm^{-2}s^{-1}	1×10^{13}	2×10^{13}	3×10^{13}	1×10^{14}	2×10^{14}

TRIGA is a typical multi purpose RR design, which is intended to be operated by scientific institutions and universities[25]. TRIGA RRs have been widely operated by various institutions in many different countries, such as Istanbul Technical University in Turkey[26], Nuclear Technology Development Center (CDTN) in Brazil[27], Bangladesh Atomic Energy Commission (BAEC)[3] and Atomic Energy Research Establishment (AERE)[28] in Bangladesh, Applied Nuclear Energy Laboratory (LENA) of the University of Pavia in Italy[29], Thailand Institute of Nuclear Technology (TINT) in Thailand[30], Vienna University of Technology (VUT) in Austria[31], Institute for Nuclear Research (INR) in Romania[32], Centre of Nuclear Study of Maamora (CENM) in Morocco[4], and National Institute for Nuclear Research (ININ) in Mexico[33]. TRIGA reactors commonly use zirconium and beryllium as well as their alloys for reactor vessel and core support structure materials. Zirconium and its alloys are used to manufacture fuel claddings and guide tubes[9, 34]. TRIGA fuels uniformly mix zirconium hydride (ZrH) and uranium[29, 30, 35-38]. Zirconium offers an advantage of having neutron economies[39]. Meanwhile, beryllium is used for neutron reflector and moderator[40].

Since the primary objective of RRs is to provide neutron sources for various applications, high-energy neutron with high intensity will be exposed to those zirconium and beryllium as well as their alloys. For a long service period, zirconium alloys will experience irradiation induced dislocation and microstructural changes[9, 34, 41]. Meanwhile, the main types of the failures of irradiated beryllium specimens are embrittlement and intergranular[42, 43]. Beryllium degradation processes lead to the weakness of the grain boundaries to maintain its integrity[44].

3. METHODOLOGY

Irradiation damage can be defined as the damage on the crystalline materials due to the interactions between impinging particles and atomic lattice. The impinging particles can be neutrons, ions and/or electrons. Meanwhile, the effects of irradiation could be defined as changes in physical and mechanical properties due to the evolution of defects, aggregation or dissolution caused by that irradiation. Mechanical properties of the reactor vessel and the core support structure due to neutron irradiation exposure, such as irradiation induced dislocation and microstructural changes, are investigated. To understand the neutron irradiation behavior on those reactor vessel and core support structure materials, a wide range of literatures on research reactor ageing management are reviewed. In the review process, the effects of neutron fluences and temperatures on those core structural materials are evaluated.

4. RESULTS AND DISCUSSION

In order to guaranty the integrity of the research reactor core structural materials, a good understanding on the neutron radiation behaviour of the structural material properties is necessary. A number of scholars have studied the effect of neutron fluences and temperatures on the core structural materials. This section reviews and discusses irradiation damage to the RR's core structural materials made from zirconium and beryllium as well as their alloys.

4.1. Zirconium

Characteristics of Zirconium such as Zircaloy-2 as well as Zircaloy-4 and their alloys such as Zr-Sn perform low thermal neutron absorption, reasonable mechanical properties, and good corrosion resistance in high-temperature and high-pressure steam or water[39, 45]. These alloys are commonly used as cladding tubes for encapsulating nuclear fuel as well as preventing nuclear fission products leaking from the fuels.

Lobo and Andrade have studied the microstructural stability of zircaloy-2 and zircaloy-4 against neutron irradiation[34]. They confirmed that the stability of the mechanical properties of zircaloy alloys was mostly affected by neutron fluences. Moreover, Chatterjee, Shah and Dubey [46] found that the most important source for damaging zirconium alloys is neutron irradiation. Zirconium alloy damages can be in the form of dimension changes, tensile strength increases and ductility reductions, transition temperature increases, fracture toughness decreases, crack growth rate increases, and microstructural/chemical composition changes[46]. These changes will, indeed, reduce the lifetime of these components.

Yamada and Kameyama studied the formation and microstructural evolution of a- and c-component dislocations of zirconium alloys[47]. A dislocation loop is a typical point defect cluster in an irradiation-induced cascade[48]. It was revealed that the c-component dislocation density increases in line with the fluence increases. However, the a-component dislocation density did not clearly shown its dependency on the fluences. Moreover, the a-component dislocation density turned into saturated condition at low fluences[47]. Another study, which was done by Yan et al. [45], discovered that the size of these two dislocations increases in line with the increase of the irradiation temperatures and doses.

Choi and Kim studied the behaviours of the hexagonal crystals of Zircaloy at fluences of greater than 3×10^{25} m^{-2}[41]. They found that an anisotropic distribution with a high number density of a-loops created on the prism planes and vacancy c-loops created on the basal planes. Another finding showed that the a-loops dominate the irradiation hardening of the zirconium. Furthermore, Barashev, Golubov and Stoller[49] studied the evolution of the number densities of interstitial-type prismatic loop and vacancy-type basal loops as shown in Fig. 1.
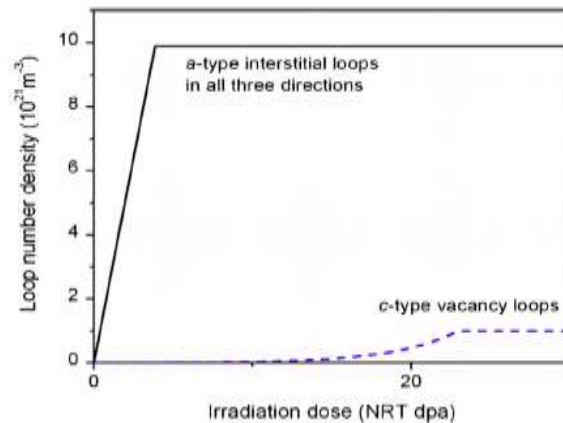

Figure 1. Loop Number Densities Vs Irradiation Dose [49].

Cockeram et al. observed that microstructure changes, strength intensification and uniform elongation reductions of zircaloy are a function of irradiation temperature and fluences[50]. Different from other metals and alloys, zirconium and its alloys exhibit only little void formation when they are exposed to neutron irradiation[34]. Onimus et al. [51] confirmed that mechanical behavior changes happen in recrystallized zirconium alloys. Neutron irradiation increases the yield and the isotropic stresses because of the high loop density hardening.

4.2. Beryllium

According to the study done by Chakin et al.[40], the role of grain boundaries in polycrystalline material of beryllium is essential for maintaining its integrity. Three types of degradations could happen to the beryllium materials, i.e. swelling, embrittlement and microhardness[42-44]. These three degradations are caused by the formation of radiogenic helium and insufficient gas atom diffusion in the microstructure of the berylllium[40, 52-54].

Helium atom deformation, which can significantly distort the crystal lattice of the beryllium microstructure, are mostly caused

by neutron irradiation exposure[40, 52, 55]. Beryllium degradation processes, which lead to the weakness of the grain boundaries, happen in two stages, i.e. a hexagonal closed-packed lattice anisotropy and a powder metallurgy. Helium accumulation kept growing in line with the increase of neutron fluences as shown in Fig. 2[44].
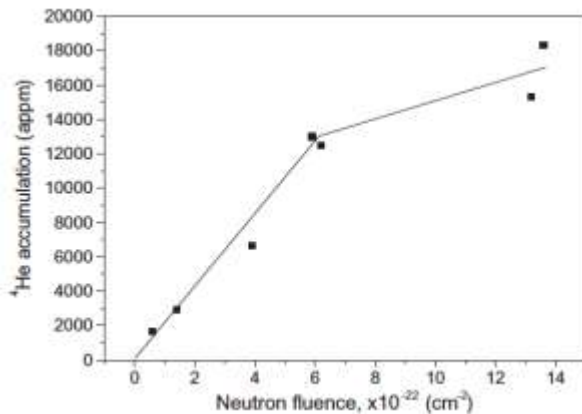

Figure 2. Helium Accumulation Growths vs Fluence Growths in Beryllium[44].

Chakin et al. studied the beryllium swelling affected by various fluences[40]. They found that its swelling grows up in line with the increase of fluence. In their further study[44], beryllium swelling growth rates can be grouped in to three different rates as shown in Fig. 3[44].
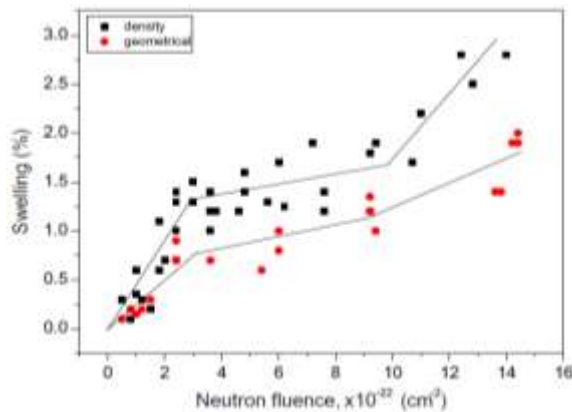

Figure 3. Beryllium Swelling Rates vs Fluence Growth [44].

In the fluence interval between zero and $(3-4)$ x 10^{22} cm^{-2}, the beryllium swelling rate strongly increases with the growth of the neutron fluence. Chakin et al. [44] and Leenaers et al. [53] confirm that, this phenomenon is caused by the accumulation of the radiogenic helium and the absence of the gas atom diffusion. In the fluence interval between 4 x 10^{22} cm^{-2} and $(9-10)$ x 10^{22} cm^{-2},

the growth of the beryllium swelling slows down because helium has been sufficiently transferred to the grain boundary pores. In the fluence interval between 10 x 10^{22} cm^{-2} and $(14-15)$ x 10^{22} cm^{-2}, the swelling rates are back to sharply increase due to "avalanche-like", which accelerates the swelling process.

Similar to the swelling phenomenon, the beryllium strength degradation also differs within three intervals of fluence growths as shown in Fig. 4 [44].
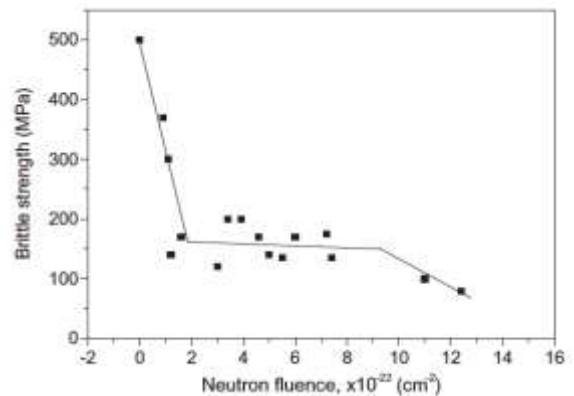

Figure 4. Beryllium Brittle Strength vs Fluence Growths [44].

In the fluence interval between zero and $(2-4)$ x 10^{22} cm^{-2}, beryllium brittle strength strongly drops. In the fluence interval between 4 x 10^{22} cm^{-2} and $(7-10)$ x 10^{22} cm^{-2}, the strength almost stays steady at the value around 150 MPa. However, the beryllium brittle strength back to gradually drop at fluence interval between $(8-10)$ x 10^{22} cm^{-2} to higher fluences [44].

On the other hand, based on microhardness investigation, Chakin et al. [44] confirmed that the beryllium microhardness kept growing in line with the increase of neutron fluences as shown in Fig. 5 [44].
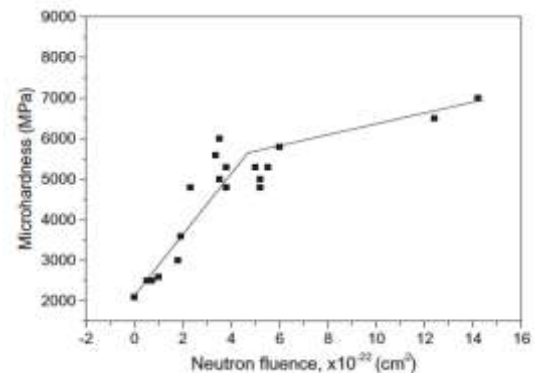

Figure 5. Beryllium Microhardness Growths vs Fluence Growths [44].

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Within the interval between zero and (4–6) x 10^{22} cm^{-2}, beryllium microhardness sharply increase but after 6 x 10^{22} cm^{-2}, the growth is less significant [44].

5. CONCLUSION

Mechanical properties of zirconium and its alloys can be affected by neutron irradiation exposure. It is observed that their microstructure changes, strength intensification and uniform elongation reductions are a function of irradiation fluences and temperatures. The formation and microstructural evolution happen to the a– and c–component dislocations of zirconium alloys. The c–component dislocation density increases in line with the fluence increases. However, the a–component dislocation density did not clearly shown its dependency on the fluences. Furthermore, the size of these two dislocations increases in line with the increase of the irradiation temperatures. Three types of degradations could happen to the beryllium materials, i.e. swelling, embrittlement and microhardness, which are caused by the formation of radiogenic helium in their microstructures. This radiogenic helium keeps accumulated with the increase of neutron fluences. Similar to the helium accumulation phenomenon, swelling and microhardness also keep growing in line with the increase of neutron fluences. On the other hand, beryllium strength degrade with the increase of neutron fluences.

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