IOP Conf. Series: Journal of Physics: Conf. Series **860** (2017) 012042 doi:10.1088/1742-6596/860/1/012042

Preliminary study of degradation from neutron effects of core-structural materials of Thai Research Reactor TRR-1/M1

P Ampornrat[1], P Boonsuwan, S Sangkaew and T Angwongtrakool

Bureau of Nuclear Safety Regulation, Office of Atoms for Peace
16 Vibhavadi-Rangsit, Chatuchak, Bangkok 10900, Thailand

[1] Corresponding author e-mail: pantipam@gmail.com

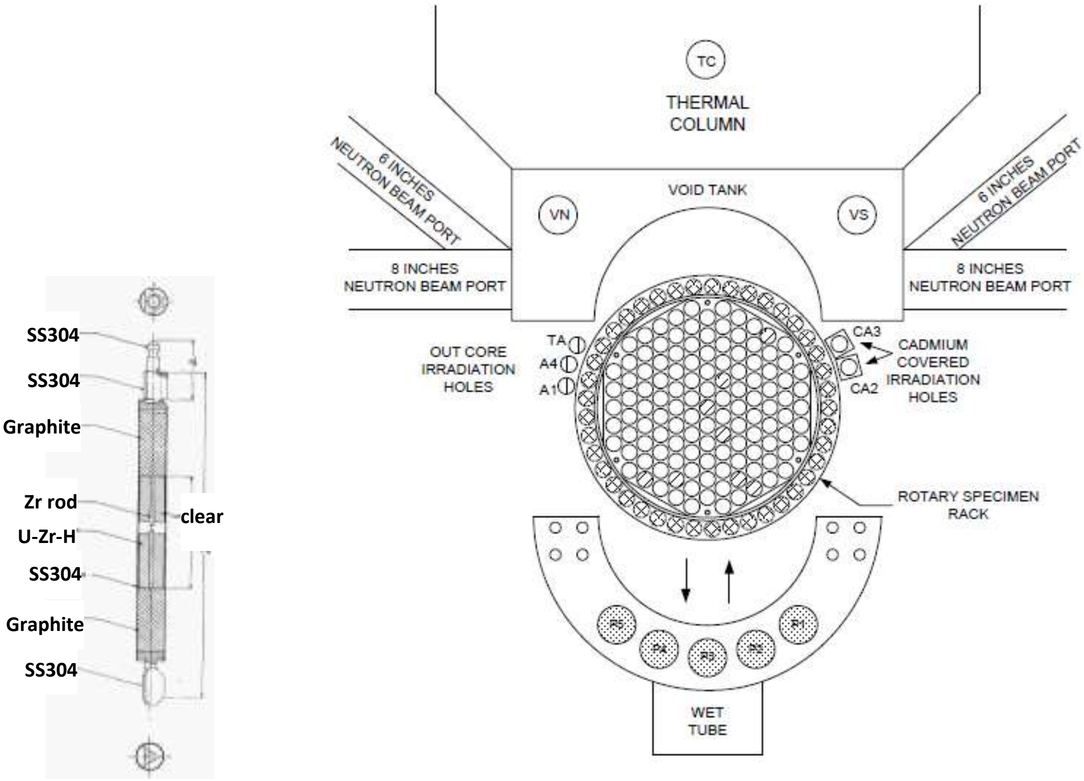
Abstract. Thai research reactor went first critical in 1962. The reactor was converted in 1977 from an MTR-type with high-enriched uranium fuel to a TRIGA-MARK III type using low-enriched uranium fuel, called TRR-1/M1. Since the TRR-1/M1 has been operated for almost 40 years, degradation of reactor structural materials is expected. In this preliminary study, the potential degradation from neutron effects of core-structural materials, e.g., fuel clad (SS304) and core components (Al6061) were studied. Assessment included calculation of neutron energy, flux and fluence in the reactor core to evaluate displacement rate (dpa) and irradiation effects on the material properties. Results showed maximum displacement rates on SS304 was 5.24×10^{-8} per $cm^3\cdot$sec and on Al6061 was 1.14×10^{-8} per $cm^3\cdot$sec. The corresponding maximum displacement levels were ~17 dpa for SS304, and ~4 dpa for Al6061. At these levels of displacement, it is possible for the materials to result in tensile strength increasing and ductility reduction. Further inspection on the core-structural materials needs to be conducted to validate the assessment results from this study.

1. Introduction

Thai research reactor has been in operation since 1962. The reactor was converted from a Material Testing Reactor (MTR) – type utilizing high-enriched uranium fuel to a TRIGA-MARK III type using low-enriched uranium fuel in 1977, called TRR-1/M1. Until now (2016) the TRR-1/M1 has been operated for approximately 39 years. It is possible for its structural systems and components (SSC) to undergo degradation or aging. Especially in-core structural materials (Figure 1) being exposed to cooling water and high radiation simultaneously, these materials would be damaged by radiation as well as corrosion. The International Atomic Energy Agency (IAEA) safety requirements emphasize on Safety of Research Reactor that the aging effects shall be taken into account for all operational states, including periods of maintenance and shutdown [1]. Therefore Office of Atoms for Peace (OAP), as the regulatory authority, has determined degradation of core structural materials of TRR-1/M1 in order to ensure safe operation of the research reactor and determine the SSC that are vulnerable to the aging mechanisms.

Degradation of materials used in nuclear reactors caused by several parameters, namely, temperature, pressure, radiation, mechanical stress and water chemistry. Most of available information arose from nuclear power reactors. Only little information of aging in research reactors is available because the reactors exhibit less degradation evidence than that of the power reactors. The degradation mechanisms usually found in the nuclear reactors are; changes in physical or chemical properties (e.g., swelling, chemical decomposition, phase changed, strength or ductility), irradiation and thermal embrittlement, creep, fatigue, and corrosion (e.g., galvanic corrosion, pitting corrosion, stress corrosion cracking (SCC)).

Aging management of the TRR-1/M1 should be conducted to ensure the safety after long time operation. First aging investigation program of the TRR-1/M1 was conducted with concrete pool structure by the OAP in cooperation with Kasetsart University in 2009. Results showed that there were no leakage or seep from the pool wall. The reactor pool can be continued to use for 25 – 30 years with fluence limit of 10^{17} neutrons per m^2 at the pool wall. The preliminary study reported in this paper focuses on the evaluation of degradation of in-core structural components that have been exposed to neutron radiation. Such in-core structural components consist of core shroud, control rod, fuel elements and instrumented fuel elements. Due to hazardous work conditions caused by high radiation exposure on specimen from the TRR-1/M1, the study on corrosion effects has been planned at a later date. Expected outcomes of the study are: i) to be able to evaluate neutron damage rates and effects on mechanical properties of in-core components after 39 years of service, and ii) to identify the most vulnerable components to such mechanical degradations, which may need immediate attention. The results will be used for further inspection and maintenance to ensure safe operation of the reactor.



(a) (b)

Figure 1. (a) Fuel element [4], (b) Top view of reactor core and out-core irradiation facilities [4].

2. Methodology

Methodology of aging management program (AMP) followed an IAEA safety standard series no. SSG-10 "*Aging management for research reactor*" [2]. Elements of the AMP included; screening of SSCs, identification and understanding of aging degradation, minimization of expected aging degradation, detection, monitoring and trending of aging degradation, and ongoing improvement of the AMP and accurate record keeping.

This work involved first three parts of the AMP listed above. First, the SSCs of the reactor core were classified in terms of materials, importance to safety, ease of replacement, possible aging mechanism (according to their service conditions), and effects of the mechanisms. Identification and understanding of aging degradation were conducted through determination of service conditions and compare with literature data of the structural materials. Major degradation mechanisms of radiation effects were studied in this work.

To evaluate the radiation effects, neutron fluxes of three energy ranges; thermal (0-0.025 eV), epithermal (0.025-0.42 eV) and fast neutrons (1-15 MeV) were evaluated using software tool Monte Carlo N-Particle eXtended Transport Code (MCNPX) developed by Los Alamos National Laboratory. Model was developed with core number 1 arrangement consisting of 97 fuel elements, four fuel follower control rods, a transient control rod, a central thimble irradiation tube, and three neutron detector tubes. Fuel elements were modeled as drawing in Figure 1(a). The control rods were modeled with the sections of boron carbide, fuel follower with zirconium rod through the center of active fuel section and void region. Two types of fuel contained 8.5%wt and 20%wt uranium with hydrogen to zirconium ratio of 1.6. Erbium content of 0.5%wt was added in the fuel with 20%wt uranium. Enrichment of U-235 was 20%wt for both types. More information of modelling were explained in a work of S. Sangkaew [3].

3. Classification of core-internal structural components

The TRR-1/M1 is a pool type TRIGA Mark III research reactor. The pool contains reactor core, irradiation facilities, coolant water, and used fuel storage racks. Geometry of the TRR-1/M1 reactor core and irradiation facilities inside the pool is shown in Figure 1(b). The reactor core is contained in core shroud assembly. Inside the assembly, the core consists of fuel elements, instrumented fuel elements, control rods, neutron detectors, neutron source, grid plates and safety plate.

List of the in-core structure components, material, important to safety, ease of replacement, potential aging mechanism and effects from the aging mechanisms is presented in Table 1. Most of components made of stainless steel SS304 and aluminum alloy Al6061. Clad of fuel elements and control rods is made of SS304 due to its good properties in corrosion resistance, high strength and low susceptible to radiation swelling. Other core structure components are made of Al6061, which has fair corrosion resistance, good fracture properties and ability to withstand irradiation damage. The importance to safety was classified based on their involvement in the reactor protection system. Possible aging mechanisms were listed following guidance in IAEA SSG-10 [2]. It is found that most probable aging mechanisms are corrosion and radiation damages. The neutron radiation damage was reported in this study. The corrosion effects on structural materials will be studied at a later date up on specimen availability.

Table 1. In-core structural components of TRR-1/M1 with properties and possible aging mechanism.

SSC	**Materials**	**Important to safety**	**Ease of replacement**	**Possible aging mechanisms**	**Effects**
Core shroud	Al6061	No	No	Corrosion, radiation effects, temperature effects, mechanical stress	Reduce strength, brittle, crack, physical deformation
Control rod (shim, safety and regulating rods)	Clad – SS304 Absorber – B_4C Fuel–UErZrH (20% U)	Yes	Yes	Corrosion, radiation effects, temperature effects, mechanical stress	Reduce strength, brittle, physical deformation
Control rod drive motor (over pool)	Drive motor assembly	Yes	Yes	Temperature effects, mechanical stress	Unable to control properly

Fuel element and instrumented fuel element	Clad – SS304 Fuel – UZrH (8.5% U) and UErZrH (20% U) Reflector – Graphite	Yes	Yes	Corrosion, radiation effects, temperature effects	Reduce strength, brittle, crack, physical deformation
Neutron detector	Detector assembly	Yes	Yes	Corrosion, radiation effects, supply voltage instability	Reduce strength, physical deformation, unable to count properly
Neutron source	Source – Am-Be Clad – double layers of SS304	Yes	Yes	Corrosion, radiation effects, temperature effects	Reduce strength, brittle, physical deformation
Irradiation tube	Al6061	No	No	Corrosion, radiation effects, temperature effects, mechanical stress	Reduce strength, brittle, crack, physical deformation
Grid plate	Anodized Al	No	No	Corrosion, radiation effects, temperature effects, mechanical stress	Reduce strength, brittle, crack, physical deformation

4. Evaluation of radiation effect

Fission reaction in the reactor core creates various types of radiation, e.g., neutrons, alpha particles, beta, and fission products. Energetic radiation, especially radiation in forms of particles or ions, can cause damage to target materials via displacement or dislocation of the target atoms. This work first focused on effects of neutrons, the dominant radiation in the reactor core.

4.1. Neutron energy and flux

Using the MCNPX software, neutron fluxes of three energy ranges were calculated at central thimble position at power 1 MW, Figure 2. This position yielded the highest neutron fluxes in the reactor core. The fluxes were calculated along axial direction with 10 centimetres increment. The maximum fluxes of three energy ranges were used to determine possible aging mechanisms by applying the conservative approach assessment.

Three ranges of neutron energies determined in this study include; thermal neutrons (0 – 0.025 eV), epithermal neutrons (0.025 – 0.42 eV) and fast neutrons (1 – 15 MeV). Note that the flux at energy 0.42 eV – 1 MeV was omitted in this study to avoid the cross section fluctuation zone.

Results showed that neutron flux of epithermal neutrons is the highest, followed by fluxes of fast and thermal neutrons, respectively. Fluxes of three energy ranges are within magnitude of 10^{13} n/cm^{2}s. Table **2** presents average and maximum fluxes of thermal, epithermal and fast neutrons along axial direction at central thimble position. A plot of neutron flux versus vertical distance from center (Figure 2) shows that fluxes of neutrons at three energy ranges are highest at the center position.

Table 2. Flux and fluence of thermal, epithermal and fast neutrons at central thimble.

Neutron energy	**Average neutron flux (n·cm^{-2}-s^{-1})**	**Maximum neutron flux (n·cm^{-2}-s^{-1})**	**Neutron fluence (39 years) (n·cm^{-2})**
Thermal neutrons (0.0-0.025 eV)	4.72x10^{12} $\pm$0.32%	9.04x10^{12} $\pm$0.84%	2.92x10^{21} $\pm$0.32%
Epithermal neutrons (0.025 - 0.42 eV)	1.58x10^{13} $\pm$0.26%	3.05x10^{13} $\pm$0.66%	9.85x10^{21} $\pm$0.26%
Fast neutrons (1.0-15.0 MeV)	5.39x10^{12} $\pm$0.44%	1.10x10^{13} $\pm$1.10%	3.55x$10^{21}$$\pm$0.44%
Total (0.0-15.0 MeV)	4.01x10^{13} $\pm$0.18%	7.93x10^{13} $\pm$0.43%	2.56x$10^{22}$$\pm$0.18%

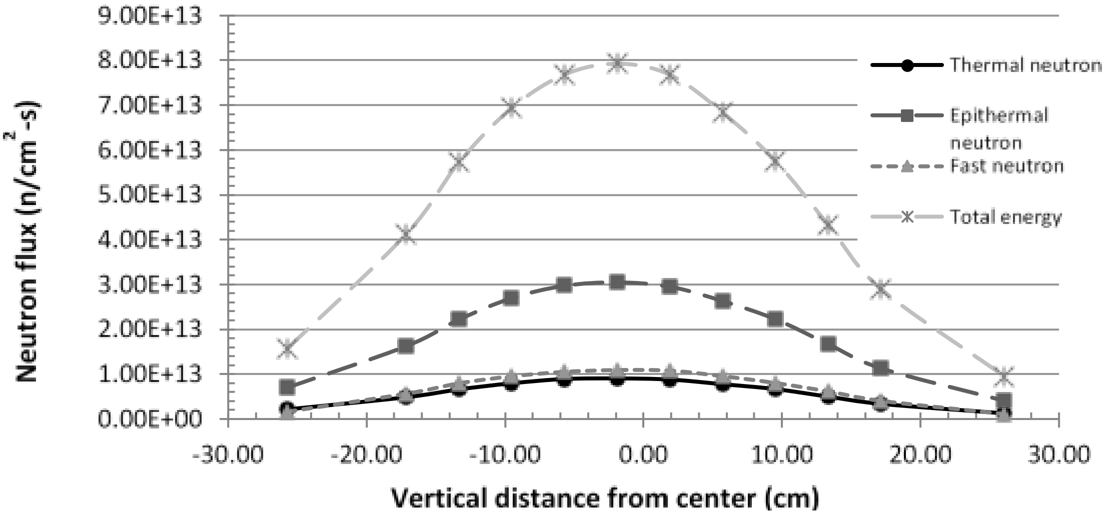


Figure 2. Plot of axial neutron flux at central thimble position at power 1 MW

Neutron fluence was evaluated using assumptions of;

- TRR-1/M1 had been operated for 39 years with 46 hours per week and 50 weeks per year, and
- the operation power is 1 MW.

The fluence was calculated using the maximum neutron fluxes to follow the conservative approach. Results of neutron fluence over 39 years are presented in
Table 2. It was found that the neutron fluence is in the order of 10^{21} $n\cdot cm^{-2}$. The trend of neutron fluence is closely resembling that of neutron flux, where fluence of epithermal neutrons is highest following by fast and thermal neutrons. The fluence was found to reach its maximum at the middle of fuel. Neutron flux and fluence at each energy range shown in Table 2 will be used to calculate displacement rates.

4.2. Displacement rate

Displacement rates of atoms in SS304 and Al6061 were calculated using the radiation damage rate equation;

$$R_d = N \int_{E_1}^{E_2} \phi(E_i)\sigma_D(E_i)dE_i \quad (1)$$

where: R_d is the damage rate
N is the lattice atom density
E_i is the incident neutron energy
$\phi(E_i)$ is the energy dependent particle fluence
$\sigma_D(E_i)$ is the energy dependent cross section

The energy dependent cross section is a probability for displace lattice atoms with the incident neutrons.

$$\sigma_D(E_i) = \int_{T_1}^{T_2} \phi(E_i, T)\, \nu(T) dT \quad (2)$$

where: $\sigma(E_i,T)$ is the energy transfer dependent cross section
T is the energy transfer to the lattice atoms
$\nu(T)$ is the number of displaced atoms

Kinchin-Pease Model determines the number of displaced atoms.

IOP Conf. Series: Journal of Physics: Conf. Series **860** (2017) 012042 doi:10.1088/1742-6596/860/1/012042

$$\nu(T) = \begin{cases} o & for\, T < E_D \\ 1 & for\, E_D < T < 2E_D \\ \frac{T}{2E_D} & for\, 2E_D < T < E_c \\ \frac{E_c}{2E_D} & for\, T \geq E_c \end{cases} \quad (3)$$

where: E_D is the displacement energy
E_c is the cut-off energy

The displacement rates of SS304 and Al6061 of three neutron energy ranges were calculated separately due to discrete values of the flux. The calculation was done under assumption that the dominant neutrons and target atoms interaction was isotopic scattering. Results showed that the displacement of atoms was mainly caused by fast neutrons. This is due to the fact that the incident energy of thermal and epithermal neutrons were not sufficient to displace atoms in SS304 and Al6061. The displacement rates over 39 years were also calculated using the neutron fluence. Calculation results are shown in

Table 3. The results shows that the displacement in 39 years of SS304 reached ~17 displacements per atom (dpa), and that of Al6061 reached ~4 dpa.

Table 3. Calculated displacement rate and displacement in 39 years of SS304 and Al6061.

Neutron energy	Displacement rate (displacement per second)	Displacement in 39 years (dpa)
SS304		
Fast neutrons	$5.24x10^{-8} \pm 18.44\%$	16.91 ± 4
Al6061		
Fast neutrons	$1.14x10^{-8} \pm 7.44\%$	3.67 ± 1.83

In comparison, a nuclear research reactor is typically experiencing significantly less neutron damage rates than those of the power reactors due to much lower fast neutron flux during operation. While fast neutron flux is within the order of 10^{13} n/cm^{2}s in a research reactor, fast neutron flux in a typical power reactor usually is greater than 10^{15} n/cm^{2}s resulting in displacement rate of at least 15 times greater than that of a research reactor. Nonetheless, changes in mechanical properties due to neutron effect can still be observed in a research reactor after many years of service.

4.3. Change in material property due to radiation

The results in previous section showed that fast neutrons caused displacements of atoms of the reactor components. Such displacement can cause changes in the material properties. Most of metal or alloys will undergo radiation strengthening phenomena in which the yield stress will increase and the ductility and the fracture toughness will decrease. These phenomena are caused by various mechanisms in the materials, e.g., displacement cascade and radiation-induced segregation. Due to limitation of specimen and high radiation field in TRR-1/M1, mechanical tests cannot be performed directly on such specimen. In the study, data from literature will be gathered and compared in order to evaluate changes in mechanical properties. Once specimens are available in the future, they will be tested for hardness and other predetermined mechanical properties. Data from other studies has suggested that yield strength (YS) and ultimate tensile strength (UTS) of neutron irradiated SS304 increased tremendously compared to those of neutron irradiated Al6061 [5-8]. Similar trends were observed to have total elongation. The YS and UTS of neutron irradiated SS304 increased rapidly with dpa and reached the maximum of 800 MPa in a sample irradiated to 10 dpa [5, 9]. Neutron irradiation is found to have smaller effects on Al6061, where YS and UTS slightly change after being irradiated up to 5 dpa [8]. Displacements after 39 years of irradiation in SS304 (17 dpa) and Al6061 (4 dpa) were interpolated using the aforementioned data [5-8]. Corresponding YS, UTS and total elongation of both alloys are reported in Table 4. Results indicate that after 39 year of service in the reactor core, YS, UTS and total elongation of SS304 has increased 225%, 58% and 85%, respectively. Meanwhile, YS, UTS and total elongation of Al6061 has found to only increase less than 20%. Therefore, SS304 clad would require careful attention.

Table 4. Comparison of unirradiated and irradiated (literature data) YS, UTS and elongation of SS304 and Al6061.

Property	Unirradiated	After irradiation	Change
SS304		17 dpa	
YS (MPa)	215	~700 [5, 6],	225%
UTS (MPa)	505	~800 [7]	58%
Elongation (%)	70	~10 [5], 8 [6]	85%, 88%
Al6061		4 dpa	
YS (MPa)	276	325 [8]	18%
UTS (MPa)	310	360 [8]	16%
Elongation (%)	12	11 [8]	7%

5. Summary

Analyses showed that the core-structural materials could be effected from aging mechanism after 39 year service in TRR-1/M1. Major possible degradation mechanisms are caused by radiation effects. The neutron irradiation reached fluence of 10^{21} n/cm^2. Determination of displacement rate of SS304 and AL6061 showed that only fast neutrons had influence on the radiation damage. Corresponding displacement per atom of SS304 is approximately 17 dpa, and that of Al6061 is 4 dpa. Comparison with the literature data revealed that the neutron fluence was high enough to effect material properties. Radiation affected YS, UTS and elongation of SS304 tremendously. The effects on Al6061 were found to be smaller than those on SS304. Continuous study should be conducted on corrosion of the structural materials. Moreover, further inspection should be conducted in the reactor based on these results.

References

[1] International Atomic Energy Agency 2005 *Safety of Research Reactor: safety requirements* Safety standard series, no. NS-R-4 (Vienna: International Atomic Energy Agency) p 53
[2] International Atomic Energy Agency 2010 *Aging management for research reactors: specific safety guide* Safety standard series, no. SSG-10 (Vienna: International Atomic Energy Agency) chapter 5 pp 22 – 29
[3] Sangkaew S, Angwongtrakool T and Srimok B 2016 *Proc. Int. Nuclear Science and Technology Conf. 2016* (Bangkok) *Preprint*
[4] Chue-Inta S, Klaysuban N, Tippayakul C and Konduangkaeo A 2010 *Technical meeting on commercial products and services of research reactors* (Vienna) IAEA-TM-38728
[5] Lucas GE 1993 *J. Nucl. Mater.* **206** 287
[6] Chen Y, Alexandreanu B, Soppet WK, Shack WJ, Natesan K, Rao AS 2011 *Proc. 15th Int. Conf. on Environmental Degradation of Materials in Nuclear Power System* (La Grange Park, IL: ANS)
[7] Dadfarnia M, Sofronis P and Robertson IM 2012 *Irradiation Effects on Material Properties for 304L Stainless Steel Base Metal and Welds* Final report to Canadian Nuclear Safety Commission (Urbana, Illinois)
[8] Farrell K and King RT 1979 *Effect of Radiation on Structural Materials* ASTM STP 683 pp 440-449

[9] Maloy SA, James MR, Sommer WF, Ferguson P, Willcutt G, Alexander D, Louthan MR, Hamilton ML, Snead L and Sokolov MA 1996 *The Change in the Mechanical Properties of Alloy 718, 304L and 316L Stainless Steel and Al6061 after Irradiation in a High Energy Proton Beam* UNT Digital Library: http://digital.library.unt.edu/ark:/67531/metadc722804/ (New Mexico)

Acknowledgments
Authors would like to acknowledge assistance from the colleagues from Reactor Center of Thailand Institute of Nuclear Technology for useful discussions and information used in this study.