INVESTIGATION OF RADIATION DAMAGE IN THE ALUMINUM STRUCTURES OF THE GERMAN FRJ-2 RESEARCH REACTOR

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Abstract

Radiation damage in the structures of the aluminum tank resulting from the reaction of fast and thermal neutrons with aluminum was simulated using the 3D MCNP Monte-Carlo code. Comparison of the local neutron flux with measurement showed a deviation less than 5 %. Radiation damage in term of concentration of silicon was also determined for the front plate of the 4H-2 channel by the calculation of the total rate of Al(n,β)Si reaction. The results are in very good agreement with the measured Si-concentration obtained by chemical analysis.

The results of calculation show that the tensile elongation of aluminum in the most irradiated segment of the 2TAN beam tube is reduced by 62 % of the initial value. The content of Si is considerably lower than the end-of-life limit by 46 % so that from the radiation damage point of view, the power operation of the reactor could be continued for additional 2.82E+6 MWh corresponding to approx. 20 years.

1. Introduction

FRJ-2 is a heavy water moderated tank type research reactor which is operated at 20 MWth for neutron beam experiments and material test. During the reactor operation the reactor aluminum tank and the core components experience radiation damage resulting in a continuous aging and limitation of its life span. Radiation damage and aging of the aluminum components are caused by the interaction and capture of fast and thermal neutrons in aluminum that results in change of microstructure and change of mechanical properties such as tensile strength and elongation.

The mechanism and consequences of radiation damage in different structural materials have beam widely investigated in different studies[1-5]. The results show that the change of microstructure due cavities, dislocations and precipitates primarily depends on the neutron spectrum, thermal and fast fluence. The exiting work is focused on the determination of radiation damage in the individual components by simulation of the interaction process with the MCNP Monte-Carlo code.

Due to the high neutron flux in the inner zone of the core, the 2 inches tangential beam tube (2TAN) represents a sensitive component which is mainly affected by the radiation damage limiting the lifetime of the reactor for power operation. With respect to the extension of the reactor operation in 1995, the licensing authority required a comprehensive inspection and investigation of 2TAN with regard to the actual state of radiation damage. Due to the limited accessibility of the beam tube for material sampling and dose rate measurement, the following three step measure was initiated to meet the requirements of the authority:

- Calculation and measurement of thermal n-flux in a fuel element position adjacent to 2TAN
- Measurement and calculation of radiation damage at the front plate of the horizontal 4H2 channel
- Determination of the dose rate due to neutron absorption at the most sensitive position of 2 TAN.

For the simulation, the 3D MCNP Monte-Carlo code was employed. MCNP is widely used in nuclear engineering to perform complex criticality studies and calculations[2]. It is capable of treating any 3-dimensional configuration of materials in geometric cells of complex form using pointwise continuous-energy cross sections existing for a variety of reactions. The numerical models and features of the code have been extensively validated on the basis of comprehensive benchmark tests and experiments[3]. However, its models do not cover changes in material composition taking place during the operation of the high power research reactor FRJ-2. For this reason, MCNP was coupled with a depletion code and applied for core calculations by recycling the burnup and flux calculations(MCNP). The model developed for the analysis is a detailed nodalization of the core components and surrounding structures of FRJ-2.

2. Mechanisms of Radiation Damage

Radiation damage in the aluminum structures of the core is mainly caused by the interaction and capture process. The effect depends of the composition of aluminum, manufacturing circumstances, irradiation temperature and fluence of thermal and fast neutrons. In the fast energy range (E> 1 MeV) displacement of atoms occurs and nuclear reactions of the type $Al(n,\alpha)Na$ and $Al(n,p)Mg$ takes place. In the thermal neutron field, aluminum atoms are transmuted to silicon according to $Al(n,\beta)Si$. The damage structure consists of dislocations, cavities and precipitates of silicon which grow with increasing fluence. According to [1], approx. 300 displacements are caused in average by fast neutrons (E>1 MeV) resulting in a change of mechanical properties. In case of above nuclear reactions taking place at the neutron energies > 6 MeV and 1,9 MeV swelling of aluminum structure occurs. This effect depends on the material composition. In [3,6], the minimum for the commencement of the radiation damage due to fast fluence is given by 1.E+22 n/cm^2. The effect of the neutron irradiation of aluminum on the tensile properties of aluminum is given in Fig. 1 as a results of investigation in [6].
Due to the high cross section of thermal neutrons and high ratio of thermal to fast fluence, the interaction rate in this range is significantly higher than in epithermal and fast range. The interaction of thermal neutrons with aluminum results in transmutation of aluminum to Si. Under normal condition of a thermal research reactor, the embrittlement of aluminum due to the n,a and n,h production by fast neutrons is lower than the effect of silicon on mechanical properties of aluminum. In general, the solvability of Al for Si is very low so that in the case of the absence of additional alloy like Mg, silicon is precipitated by cristalization at the grain boundaries resulting in an increase of tensile strength and reduction of ductility[6]. The latter condition is considered as a dominating

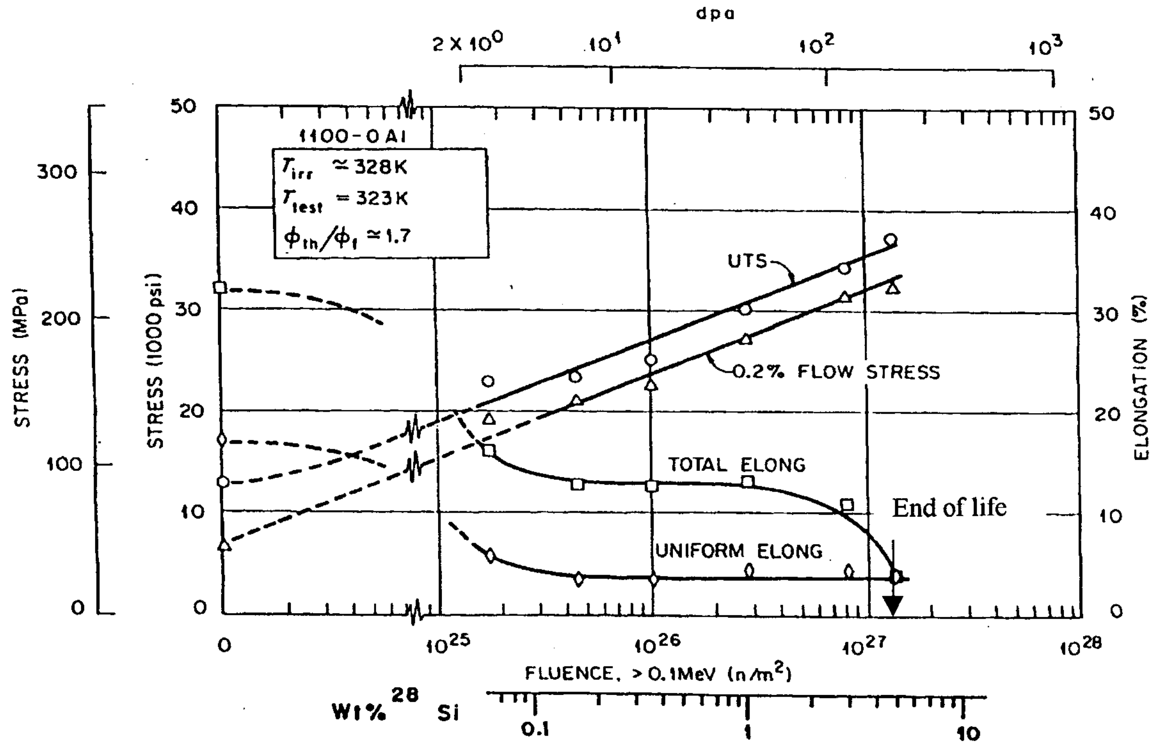


Fig. 1: Fluence dependence of tensile properties of aluminum [6]

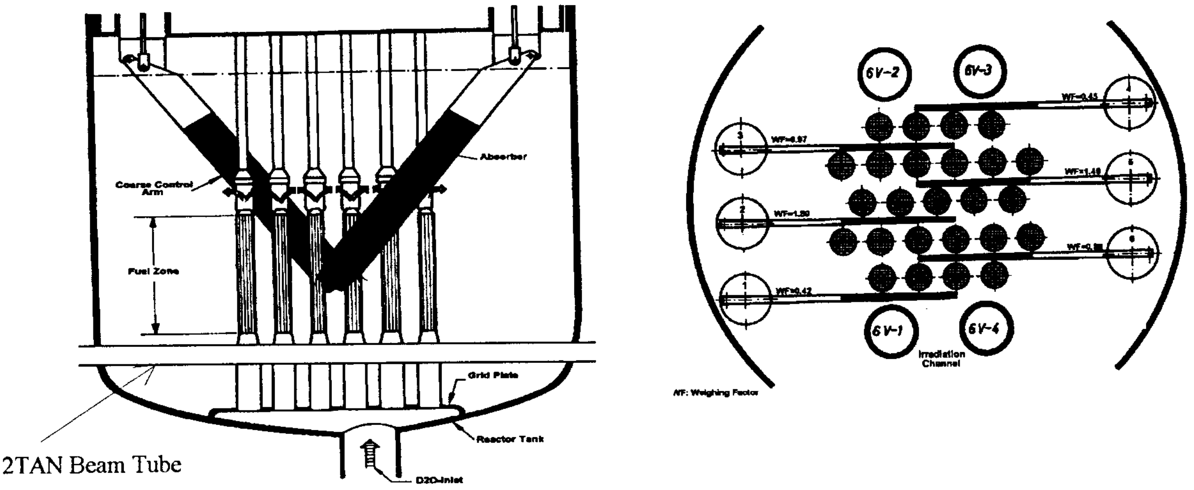
criterion for the quality of mechanical properties of aluminum. According to Fig. 1, the end of life elongation is obtained when the total elongation is reduced to the level of uniform value which is reached at a thermal fluence of 1.18E+27 n/m^2 associated to a Si content of 5 Wt %.

In the following sections the modeling of neutron interaction process in the sensitive core components by the MCNP code is described and corresponding radiation damage is compared with the end of life limit for the future operation of FRJ-2.

3. Description of FRJ-2

The FRJ-2 is a DIDO-class tank-type research reactor cooled and moderated by heavy water. The core consists of 25 so-called tubular MTR fuel elements arranged in five rows of 4, 6, 5, 6 and 4 fuel elements(Fig. 2). It is accommodated within an aluminum tank (RAT) 2 m in diameter and 3.2 m in height. The tank is surrounded by a graphite reflector of 0.6 m thickness enclosed within a double-walled steel tank. The RAT is manufactured from 1080 grade aluminum (99.8% Al), with the longest re-entrant 2 inch tangential beam tube tube (2TAN). Its material specified as 1050 grade (99.5 % Al) with 0.075-0.15 % silicon and 0.01 % (maximum)Cu. The very low copper level was specified to help avoid pitting corrosion. The plate and tubes from which the RAT was fabricated were made from material in the cold worked (half hard) condition. The main longitudinal and circumferential welds in the RAT were fabricated and designed to minimize thermal stresses in the RAT wall. The chemical composition of the filler material used in the welding process was the same as that of the plate.

The active part of the tubular fuel elements is formed by four concentric tubes having a wall thickness of 1.5 mm and a length of 0.61 m. Each tube is formed by three material testing fuel plates containing fuel meat and aluminum cladding. The annular water gap between the tubes has a width of about 3 mm leaving a central hole of 50 mm diameter filled with a thimble for irradiation purposes.



Vertical cross section

Horizontal cross section

Fig. 2: Arrangement of fuel elements and coarse control arms inside the aluminum tank

The reactor is equipped with two independent and diverse shutdown systems, the coarse control arms (CCAs) and the rapid shutdown rods (RSRs). In case of demand, the six CCAs are released from their electromagnets and drop into the shutdown position by gravity, whereas the three RSRs are shot in by pneumatic actuators. The CCAs are lowered and raised manually around a pivot in order to control power levels during normal operation. A large number of horizontal and vertical channels give access to the neutron field in the reactor. The horizontal channels (beam tubes) end either at the tank wall or at the periphery of the core penetrating into the reactor tank.

4. MCNP model of FRJ-2

The MCNP model of FRJ-2 is a complete 3-dimensional full-scale model with a very high level of geometric fidelity. It comprises the reactor core, CCAs, core structures, beam tubes, the graphite reflector and the biological shield. The core region consisting of 25 fuel element was modeled as a cylinder which contains a square lattice with an array of cells representing the individual fuel elements. Each cell in the lattice contains a detailed model of each fuel element comprising the internal thimble, 4 circular fuel tubes and the borated outer shroud tube. Each individual cell is divided into 15 axial, 35 radial and azimuthal material zones.

The D2O reflector in the RAT is represented by a cylinder with an outer diameter of 2.00 m. The lower region of the core down to the bottom of the RAT accommodating the grid plate, unfueled ends and nozzles of the fuel elements, the aluminum structures and corresponding D2O were modeled in detail. The beam tubes of varying diameter and length were modeled in detail and integrated in the corresponding position of the entire model in accordance with the design and construction documents.

Due to the continuous change of the material composition in the fuel meat resulting from the fuel consumption, it was necessary to couple the MCNP code with a depletion code. By this way the variation of the neutronic states of the core could be simulated by multiple linked burnup and MCNP calculations. The details of the burnup recycling has been described in [10] . In the course of coupled calculations, the variation of the nuclide densities is taken into account for all fission products and actinides.

For the reason of high precision, the FE, CCA and structures and beam tubes were segmented in detail and the material zones containing fuel were extensively segmented in axial, radial and azimuthal direction. To consider the variation of the composition in the fuel meat zones the detailed segmentation of the whole core resulted in a model with 11.250 material cells. To achieve sufficient number of neutron tracks and score in all cells and consequently reduce the estimated error of the physical values (keff, local neutron flux and reaction rate) all simulations were run with 2000 cycles each with 1000 particle histories. By the statistical evaluation of 2 millions, a standard deviation of 0.04 was achieved for the value of the local n-flux and reaction rates.

5. Simulation of radiation damage

5.1 Verification of the model

Prior to the simulation of radiation damage in the aluminum structure of the 2TAN beam tube, the MCNP model was required to be verified by calculation of flux profile and reaction rate in selection positions in the core components adjacent to the 2TAN channel. For this purpose the inner thimble of the central fuel element in the C3 position was extended to the bottom to the same height as the 2 TAN channel. In this fuel element, cobalt foils were positioned over a length of 73 cm including 60 cm for the active length of the fuel element. The activation of the foils was performed for a time period of 120 s at full power at a CCA angle of 26.40°. At the time of measurement, the power and average burnup of the central fuel element amounted to 1,25 MW and 24.10 % . The activity of the cobalt foils was determined after the activation by using a Ge-Li detector. For the comparison the reaction rate for the Co59(n,γ)Co60 was calculated by MCNP on the basis of the axial distribution of the neutron flux in the inner channel of the fuel element and corresponding cross sections. To obtain high calculational precision, the number of the history was chosen to 2 millions that resulted in a relative error of approx. 4 %. The results of the comparison are collected in Fig. 3 Accordingly the maximum deviation in the activity of Co-60 between measurement and calculation is limited to 5 % only. With respect to the uncertainty of the measurement (foil mass, measuring time and reactor power) the agreement is very good and MCNP is capable of predicting the flux in a high level of accuracy.

With regard to radiation damage, the reaction rate was calculated for Al(n,β)Si, Al(n,p)Mg and Al(n,α)Na reactions. For the purpose of comparison, the reaction rate of thermal neutrons and total concentration of Si was calculated first for the front plate of the aluminum thimble in the horizontal 4H2 beam tube. The inner face of the thimble is placed at a radial distance of 65 cm from the central axis of the core close to the maximum of the thermal neutron flux in the D2O-reflector. The calculation performed for the core configuration of 5/2000 operating cycle as a function of CCA angle in accordance with the power operation. The simulation resulted –averaged over the CCA angle- in 9,10E+13 n/cm^{2}s for the neutron flux and 0.471E+12 for the reaction rate (Si per s and g aluminum). On the basis of a total energy of 2,6E+6 MWh (Operating time at full power until Nov. 1995), the calculated reaction rate resulted in a total concentration of 0.99 Wt % Si. In parallel to the simulation, the Si content in the front plate of the 4H2 beam tube was determined by chemical analysis of a specimen taken from the aluminum thimble after dismantling. The result of the analysis shows that during the whole years of reactor operation Si has been produced in a concentration of 0.91 Wt %. The deviation of 10 % between the measurement and calculation is attributed to the neutronic effect of the different core configurations and CCA angles during the last 35 years of power history. The comparison for this position shows that also in this case the MCNP model is in a position to precisely predict radiation damage in the different core structures and locations.

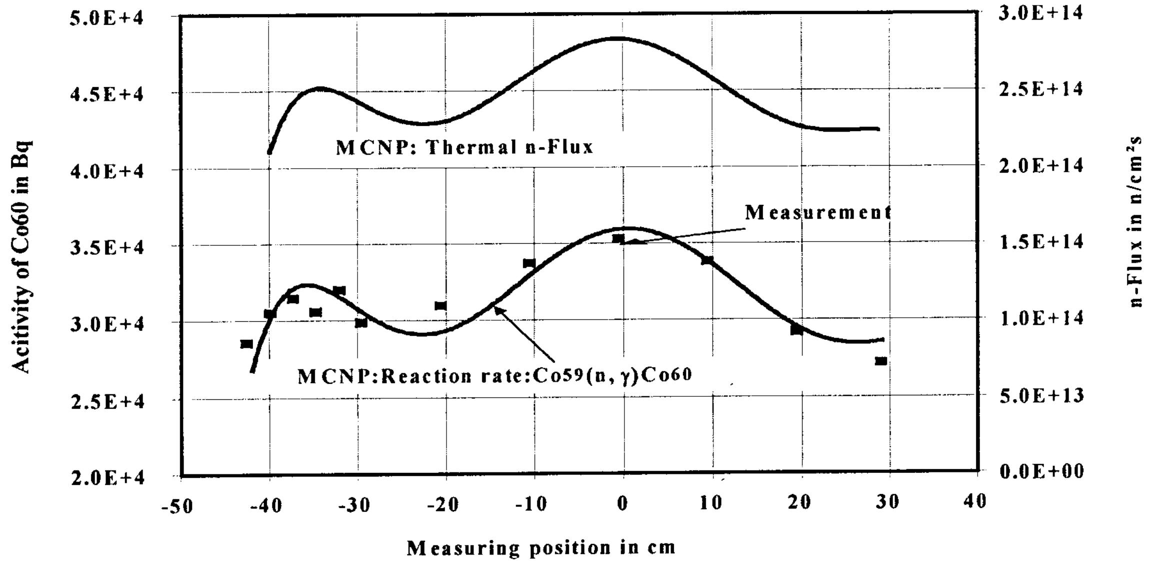


Fig. 3: Distribution of reaction rate (Co-59) in the central channel of the highest rated fuel element (C-3) calculated by MCNP in comparison to the measurement

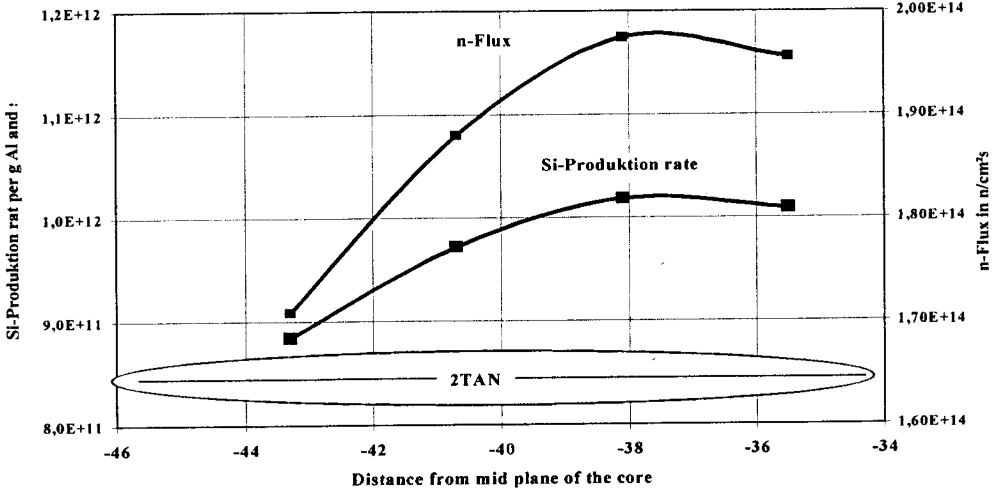
5.2 Neutron Flux and Radiation Damage

The calculation of radiation damage and reaction rate in the aluminum structure of the 2TAN channel was performed in the same manner for the core configuration of the 4/2000 operating cycle for an angle of 26.4°. To find out the critical part of the beam tube with maximum flux, the calculation was concentrated on the central segment of the channel. For the determination of axial distribution of the reaction rate, the cross section of the channel (elliptical shape) was divided in a couple of nodes.

At first the amount of radiation damage due to (n,p) and (n,α) reactions by fast fluence was calculated resulting in 1,45E+13 n/cm^{2}s for the neutron flux with E > 0.1 MeV and 6.42E+12 n/cm2 for E> 1 MeV. Under the consideration of the resulting total fast fluence of 6.78E+21 (E>0.1 MeV) and result of Fig. 2, a displacement density of 9 dpa is obtained for the aluminum structure. This value is in accordance with the earlier investigations which have been resulted in 6-7 dpa for a fast fluence of 4.0E+21 n/cm^2 and 2E+21 n/ cm^2 for neutron energies > 0.1 MeV and 1 MeV[2]. The difference is attributed to the difference in the thermal to fast ratio and test conditions. The amount of the displacement density is significantly low so that no considerable radiation damage is caused by this process of dislocation. With regard to (n,p) and (n,α) reactions, it has been reported that [6] a minimum fast fluence of 1E+22 n/cm^2 is needed to induce a considerable radiation damage in the form of swelling and embrittlement. The existing fast fluence at the 2TAN -6.78E21 n/cm^2- is considerably low and does not results in an change of mechanical properties including tensile strength and ductility in aluminum. The calculation of neutron damage shows that in this case 14 ppm hydrogen atoms and 2 ppm helium are produced in the aluminum structure only.

Due to the thermal nature of the spectrum and high cross section of thermal neutron for (n,β) on the radiation damage is higher than the effect of fast fluence. The results of the simulation of neutron interaction in the thermal range is depicted in Fig. 4. It shows the distribution of the reaction rate for production of Si-28 in the central cross section of the channel. The associated thermal neutron flux is also shown in the figure at the nominal power (20 MW). Accordingly the maximum of radiation damage and the neutron flux occur approx. 4 cm above the mid plane of the channel and amount 1.02E+12 per s and g aluminum for the reaction rate. The neutron spectrum in this location is primarily thermal amounting to 1.96E+14 n/cm²s with a considerable fast component of 1.45E13 (E>0.1 MeV) and 6.42E+12 for E> 1 MeV. Under consideration of the total power history and operating time of 2.82E+6 MWh until beginning of 6/2000 operating cycle (5.9.2000), silicon is produced in a concentration of 2.32 Wt %. The transmuted Si are concentrated in the form of precipitated particles and clusters which cause an increase of tensile strength and reduction of ductility.

Fig.4: MCNP-calculation of reaction rate for Si transmutation in the aluminum structure of the 2TAN channel including the distribution of the thermal n-flux



The loss in ductility originates mainly from the reduction of the plastic deformation by hardening. According to Fig. 1, the critical value for the elongation representing the end of life-value is reached at a neutron fluence of 1.4E+27 n/m² corresponding to a Si content of 5 Wt %. The results of calculation show that tensile elongation is reduced to 62 % of the initial value. The content of Si in the aluminum structure of 2TAN is considerably lower than the end-of-life limit of concentration by 46 % so that from the radiation damage point of view, the power opertion of the reactor could be continued for additional 2.82E+6 MWh corresponding to approx. 20 years.

6. Conclusions

On the basis the MCNP modeling of radiation damage in the aluminum structure of the sensetive beam tube, following conclusions are made:

- Due to the agreement between measurement and simulation, the calculational model is capable of accurately determining radiation damage in any structure and location of the reactor core.
- The change of the mechanical properties of aluminum due to the fast neutron damage is considerably low for any consequences
- As a result of high thermal to fast flux ratio, the precipitate silicon is produced in considerable amount resulting in a reduction of tensile elongation by 62 %.
- Due to the sufficient margin (46 %) to the end of life concentration of Si, the power operation is not limited for the next 10 years from the radiation point of view.

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