Neutron Irradiation of Zirconium Alloys in the Budapest Research Reactor

I Szenthe†, F Gillemot†, M Horvath†, Z Hózer†

† Centre for Energy Research, Budapest, Konkoly-Thege Miklós út 29-33.

E-mail: szenthe.ildiko@energia.mta.hu

Abstract. Sustainable and increasingly safe operation is one of the constant goals of various technical and economic developments related to nuclear reactors. Reliability of the fuel elements cladding is basic for the safe and economic operation, to select the best fuel clad large number of simulations must be carried out in different operational conditions and supported by experimental results. Two types of cladding tube material had been irradiated in the a BAGIRA irradiation loop at to obtain aged clad properties. The special design of the irradiation target holder allowed the specimens to be irradiated with different fluences and at a prescribed temperature. The irradiated specimens will be tested and the measured properties will be used to simulate the aged fuel clad behaviour. This paper introduces the design, preparation and implementation of the irradiation of fuel clad materials.

1. Introduction

Ensuring structural integrity is essential for safe, reliable and economical operation of the power reactors. Knowledge of the key failure mechanisms of the fuel elements is fundamental requirement for economic production of electricity, and it requires research data.

The failure of the fuel elements clad under normal and extreme operating conditions causes large economic loss for the plant owner. To avoid the fuel elements failures, the fuel clad material is aged in the laboratory using simulated operational accidental conditions. The ageing mechanisms were also studied by computer simulations, which were compared to the experimental studies. One of the major ageing mechanism is the irradiation embrittlement caused by high fluence of fast neutron irradiation. These simulations must be carried out with validated computer programs supported by experimental results. The effect of the different ageing mechanisms should be studied, including the neutron irradiation damage.

Based on experiments performed and evaluated earlier, the fluence range where the mechanical properties of the clad suffer considerable changes is in the range of 0.5-1.5 dpa, and saturation is expected above 1-1.5 dpa. To reach this damage rate within the foreseeable time a competitive and economical neutron source has to be used.

Nowadays, there are only a few research reactors in Europe that deals with neutron irradiation. In the IAEA Research Reactor Database (RRDB) there are only 14 pieces of research reactor in Europe, except the Russians [1]. From these there are 3 TRIGA type reactors designed for education, private commercial research, non-destructive testing and isotope production, other five research reactor power have about 0-0.1 KW thermal power, they are too small for metallic material irradiation. The Belarus Giacint reactor was designed to simulate different reactor core configurations, VENUS-F reactor in
Belgium is an experimental reactor of the “zero-power critical facility” type, the SUR-100 in Germany is „zero power” reactor, suitable for teaching and research, CROCUSET in Switzerland is an experimental „zero-power” reactor, mainly dedicated to teaching radiation and reactor physics, which has 200 KW thermal power, and $10^9$ neutron cm$^{-2}$ s$^{-1}$ fast neutron scale only. At present time, IR-100 reactor in Ukraine is used to train personnel for the Ukraine’s nuclear power industry. The 25 MW CABRI reactor in France is used to tests accident situations which must be considered in the safety analysis of PWRs. In addition, a pool type 30 MW thermal power MARIA in Poland is dedicated mainly for isotope production. From the remaining six reactors the 7 MW tank type WWR-M in Ukraine is available for material irradiation, but its geopolitical situation today is not good enough. The other five possibilities to perform material irradiation tests: BR-2 tank-in pool type 100 MW reactor in Belgium, HFR tank-in pool type 45 MW reactor in Netherlands, LWR-15 WWR tank type 10 MW reactor in Czech Republic, and the WWR tank type 10 MW reactor in Budapest, Hungary [2].

The Budapest Research Reactor (BRR) is operated by the Centre for Energy Research inside the Budapest Neutron Centre (BNC), which was formed in 1993. [3]. Currently 16 experimental stations are connected to the reactor: the cold neutron three-axis spectrometer (TAS), Low-Level Gamma-Spectroscopy Facility, Time of Flight Small Angle Neutron Scattering instrument, Neutron Reflectometer with Polarized Beam Option, The M(aterial) TEST neutron diffractometer, Neutron activation analysis, Neutron-Induced Prompt Gamma-ray Spectroscopy, Neutron Optics and Radiography for Material Analysis, Prompt Gamma Activation Analysis, neutron diffractometer, Small Angle Neutron Scattering, and an irradiation loop named BAGIRA [4]. Hot material testing and other laboratories are also existing within the campus providing good opportunity to carry out pre- and post-irradiation material tests.

The BRR is a VVR-type Russian designed reactor. It went critical in 1959. The initial thermal power was 2 MW. A full-scale reactor reconstruction and upgrading project began in 1986, following by 27 years of operation since initial criticality. The upgraded 10 MW reactor received the operation license in November 1993. The light-water cooled and moderated tank-type reactor with beryllium reflector has 90 fuel assemblies, the thermal neutron flux density in the flux trap is $2.5 \times 10^{14}$ neutron cm$^{-2}$ s$^{-1}$. The approximately maximal fast flux in the fast channel is $10^{14}$ neutron cm$^{-2}$ s$^{-1}$.

Within the framework of a research and development contract sponsored by the National Research, Development and Innovation Fund of Hungary, two types of cladding tubes´ material was selected (named E110 and E110G) for study. The project entitled „Effect of material structure changes on zirconium alloys used in nuclear power plants on fuel element integrity and environmental load”. As part of the contract, Centre for Energy Research irradiated Zirconium alloys. Before the irradiation different treatment performed to simulate the environmental ageing effects during operation and accidental cases, to evaluate the combined effect of irradiation, heat treatment and corrosion.

Irradiation was performed in the BAGIRA (Budapest Advanced Gas-cooled Irradiation Rig with Aluminium structure) equipment in the BRR. The preparation and fulfill of the irradiation campaign will be presented below.

2. Planning of the irradiation campaign
To perform an irradiation, we made a detailed plan for the following:

- selection of the material to be tested and type of the samples
- determine the number of the samples
- prescribe the time of the irradiation
- prescribe the temperature of the irradiation and elaborate the design for excess gamma heat removal during irradiation
- determination of the irradiation matrix

A lot of other demand also had to be taken into consideration (e.g. fragility, encapsulation, labelling/coding, transportation, storage etc.)
2.1. **Selection of the samples**

Because of the goal of irradiation procedure is to investigate the radiation damage of the material, to perform the post-irradiation measurement to the foreseeable time after the campaign the decay time of the relevant elements of the chemical composition has to be estimated. Based on this estimation the needed shielding for protection during the post-irradiation material testing can be assessed and planned.

The material of the tested two types of zirconium binary alloys are used as the material of the cladding tubes in the VVER type pressurized water reactors. The widely used Russian type zirconium alloy Zr1%Nb is named E110. The Russian manufacturer of the E110 cladding material is switching from an earlier electrolytic process to metal “sponge” technology. In the new E110G cladding 70% of the zirconium metal comes from this “sponge”, the residual 30% is made by the iodide process. The chemical composition of E110G alloy remains the same, 99% zirconium and 1% niobium, and does not change significantly according to the manufacturer but permissible levels of certain trace element concentrations change. Independent domestic measurements are an important complement to the information provided by the manufacturer. There is a need for comparative testing of the E110 and E110G cladding materials, which have been pre-treated with different basic processes that simulate the power plant burnout sub-processes [5-11].

2.2. **Determine the number of the samples**

To specify the optimum number of the samples, a test matrix has been defined. The elaborated irradiation matrix of the pre-treated specimens is shown in table 1.

| Type of pre-treatment | For irradiation or reference | Planned test type | Material E110 | Material E110G | Summa pieces of specimens |
|-----------------------|-------------------------------|------------------|--------------|---------------|--------------------------|
| Native                | for reference                 | tensile test     | 6            | 6             | 12                       |
|                       |                               | compression test | 3            | 3             | 6                        |
|                       | irradiated                    | tensile test     | 6            | 6             | 12                       |
|                       |                               | compression test | 3            | 3             | 6                        |
| Heat-treated          | for reference                 | tensile test     | 36           | 36            | 72                       |
|                       |                               | compression test | 18           | 18            | 36                       |
|                       | irradiated                    | tensile test     | 36           | 36            | 72                       |
|                       |                               | compression test | 18           | 18            | 36                       |
| Hydrogenated          | for reference                 | tensile test     | 72           | 72            | 144                      |
|                       |                               | compression test | 36           | 36            | 72                       |
|                       | irradiated                    | tensile test     | 36           | 36            | 72                       |
|                       |                               | compression test | 36           | 36            | 72                       |
| Hydrogenated with electrolysis | for reference | compression test | 3 | - | 3 |
|                       | irradiated                    | compression test | 3 | - | 3 |
| Axial                 | for reference                 | tensile test     | 1            | 2             | 3                        |
|                       |                               | compression test | 1            | 2             | 3                        |

The following main factors were considered:
- the available volume for samples in the irradiation rig,
- the flux and temperature distribution in the target holder,
• the excess gamma heat removal during irradiation,
• minimum three measurements per test has to be performed to the evaluation,
• the temperature and the time of the different heat treatment simulating the operational and accidental conditions,
• the Hydrogen content in the clad material,
• the production technology (e.g. the number of the electrolysis steps)

The nominal dimensions of the Zirconium alloy tubes E110 and E110G are: outer diameter D = 9.1 mm, wall thickness = 0.685 mm. Three type of specimens are selected for material testing: 8 mm high compression test specimens, 2 mm high tensile test specimens and 50 mm high axial tensile specimens.

2.3. Calculation of the irradiation time

Irradiation time planning was based on data from irradiation performed previously in the BRR. [3-7]. Based on these experiments considerable change of the mechanical properties of the Zirconium alloys change will occur after a fast neutron irradiation with 0.5-1.5 \times 10^{20} \text{ neutron cm}^{-2} \text{ fluence}.

More than 30% strength increase expected above the level of fast fluences 1 \times 10^{20} \text{ neutron cm}^{-2}.

These data are in agreement with other literature data [12].

The fast flux distributions calculated from previous irradiations, were used to design the irradiation target. To measure the real fluence, dosimetry monitors were inserted into different positions of the target. Long decay type of the monitors used, due to the long term of the irradiation.

The fluxes calculated from the data on the dosimetry monitors in the irradiation probe, as a function of the position of the 6 sample holders, are shown on Figure 1.

The BRR operated in 10 days per cycles. To reach the required ageing minimum 5-6 reactor cycles needed. Considering the reactor schedule an optimal irradiation campaign \approx 1700 hours long time selected.

2.4. Irradiation temperature control

The irradiation temperature was selected to simulate the operation conditions. To control of the irradiation temperature within the required range the heating and heat removal had to be balanced.

During neutron irradiation strong nuclear heating occurs. It depends on the position and the properties of the target. Auxiliary electric heating was applied to adjust continuously the temperature distribution. The excess nuclear heat from the target was removed through a gap between the target holders and the wall of the rig, which is cooled by the reactor cooling water. In the gap helium nitrogen gas mix is circulated. Changing the ratio of the nitrogen and helium the heat removal is controlled.

Six thermocouples measured the actual temperature. The data together with other measured parameters are stored in a computer memory. The temperature history during reactor operation can be checked remotely from the office of the research group.

3. Tool for irradiation: the BAGIRA irradiation rig

The BAGIRA rig has been operated since 1998 at BRR. Thirty-two different irradiation campaigns have been performed, testing irradiation ageing of different fission and fusion structural materials, as low alloyed and stainless steels, Al, Ti and W alloys, ceramics etc.
Aluminium sample holder can be used up to 350 °C, for higher temperature irradiation titanium sample holders can be built.

The BAGIRA device can be divided into 3 main parts:
- cooling gas mix preparation system,
- PC based temperature control and data acquisition system (gas recirculation unit, data acquisition and control unit),
- the irradiation channel, sample holder (including specimens and space filler)

The rig is operating inside of the core in place of three fuel elements.

3.1. Sample holder and targets
The rig capacity is 6 pieces of 65*21*25 mm targets. The target must be designed in such a way that it can be easily dismantled under hot-cell conditions.

To simulate the different burnout states, the samples were placed axial positions to receive different fast neutron flux.

After the pre-treatments the E110 and E110G samples were cleaned and filled into the targets. A dosimetry (Cu, Fe, Al-Co1%, Nb) monitor package for measuring flux was included in each sample packet.

4. The irradiation
The following data acquisition were stored by the computer at 15 second periods:
- Date,
- Time (s),
- Temperatures on 6 levels,
- Pressure of cooling gas,
- Absorbed powers of 4 electric heaters,
- Alarm messages

From the recorded data the average temperature and deviations evaluated. The average temperature of the targets summarized in table 2.

| Cycle | Time                  | Hours | T1   | T2   | T3   | T4   | T5   | T6   |
|-------|-----------------------|-------|------|------|------|------|------|------|
| 1.    | 26.09.2017-06.10.2017 | 240   | 297  | 276  | 241  | 211  | 181  | 149  |
| 2.    | 10.10.2017-20.10.2017 | 240   | 300  | 282  | 246  | 206  | 172  | 148  |
| 3.    | 07.11.2017-21.11.2017 | 336   | 298  | 280  | 244  | 201  | 166  | 144  |
| 4.    | 28.11.2017-17.12.2017 | 456   | 300  | 295  | 265  | 215  | 174  | 152  |
| 5.    | 06.02.2018-16.02.2018 | 240   | 291  | 294  | 281  | 240  | 176  | 136  |
| 6.    | 27.02.2018-09.032018  | 240   | 285  | 293  | 286  | 250  | 188  | 148  |
| Summa hours |                  | 1752 |      |      |      |      |      |      |

Figure 2. Preparation and filling of the 65*21*25 mm targets, front- and back view of a filled target
During the irradiation campaign of the E110 and E110G alloys no critical alarm was detected.

After the campaign the rig was lifted from the reactor, the targets were reloaded separately and transported into the hot-cell with a special remote controlled system. In the hot cell the licensed and trained staff removed the samples from the dismantled targets. Appropriate assistance mechanisms, specially designed for this purpose, helped the work in the hot-cell. The irradiated samples are stored in labelled, covered aluminium holders.

The activity of the dosimetry monitors generally measured and evaluated according the ASTM E-261 and ASTM E-693 standards. Flux, fluence and dpa values for the actual core configuration are calculated by the Reactor Analyses Department of the Centre for Energy Research. All the received information from the irradiation campaign, the parameters of the irradiation, the measured and calculated values are documented according to quality standards and regulation of the Centre for Energy Research.

The purpose of this paper was to introduce the neutron irradiation of the fuel clad samples. Presently the samples must be stored in the hot-cell for the decay time until their activities will be reduced enough to test them. The design and production of the testing devices are going on. After testing the results will be published in a separate paper.

Acknowledgement
The study was supported by the National Research, Development and Innovation Fund of Hungary (contract number: NVKP_16-1-2016-0014).

References
[1] IAEA RR Database: https://nucleus.iaea.org/RRDB/Content/Util/MaterialIrradiation.aspx
[2] IAEA RR Database: http://nucleus.iaea.org/RRDB/RR/HeaderInfo.aspx?RId=196
[3] Horváth A, Balaskó M, Patriskov G, Fekete T, Gillemot F, Horváth M and Uri G 2012 The contribution of Budapest Neutron Centre to the fusion materials and diagnostics development program Hungarian Plasma Physics and Fusion Techn. Workshop (HPPW)
[4] Gillemot F, Horváth M, Tatár L, Fekete T and Horváth Á 2008 Structural material investigations in the high temperature irradiation facility of the Budapest Research Reactor Research Reactor Application for Materials Under High Neutron Fluence TM-34779 IAEA
[5] Nikulin S, Rozhnov A, Belov V, Li E and Glazkina V November 2011 Influence of chemical composition of zirconium alloy E110 on embrittlement under LOCA conditions – Part 1: Oxidation kinetics and macrocharacteristics of structure and fracture J. of Nucl. Materials 418 Issues 1–3 p 1-7.
[6] Király M, Hózer Z, Horváth M, Novotny T, Perez-Feró E and Vér N 2019 Impact of thermal and chemical treatment on the mechanical properties of E110 and E110G cladding tubes *Nucl. Eng. and Technology* **51** Issue 2 p 518-25

[7] Budapest Database of E110 Claddings Under Accident Conditions 2007 AEKI-FRL-2007-123-01/01, NEA-1799/01 IFPE/AEKI-EDB-E110 HAS Atomic Energy Res. Inst.

[8] Király M, Antók D, Horváth M and Hózer Z 2018 Evaluation of axial and tangential ultimate tensile strength of zirconium cladding tubes *Nucl. Eng. and Technology* **50** no.3 425-31

[9] Ambient and high temperature mechanical properties of ZrNb1 cladding with different oxygen and hydrogen content 1999 *Enlarged Halden Programme Group Meeting*

[10] Griger Á 2004 Mechanical properties of irradiated and non-irradiated Zr1%Nb and Zircaloy claddings *International Nuclear Information System (INIS)*

[11] Kaplar A, Yegorova L, Lioutov K, Konobeyev A and Jouravkova N 2001 Mechanical Properties of Unirradiated and Irradiated Zr-1% Nb Cladding *NUREG/IA-0199*

[12] Griger Á, Horvath M, Pintémé Csordás A and Uri G 2005 Budapest Behaviour of irradiated cladding material AEKI-FRL-2005-732-01/04 Documentation, *HAS Atomic Energy Res. Inst.*