UDC 539.4

Study of Irradiation Effects at the Research Reactor

F. Gillemot

MTA KFKI Atomic Energy Research Institute, Budapest, Hungary

УДК 539.4

Изучение радиационного облучения на исследовательском ядерном реакторе

Ф. Жильмо

Институт ядерных исследований, Будапешт, Венгрия

Описаны эксперименты по радиационному облучению материалов в ядерном реакторе мощностью 10 МВт с использованием новой оснастки. В качестве примеров приведены результаты испытаний покрытий сосудов высокого давления реакторов из сталей, содержащих 9% Cr (Euroferr), использование метода Master Curve для реакторных сталей, титановых сплавов и вольфрама.

Ключевые слова: радиационное охрупчивание, оборудование для радиационного облучения, покрытия сосудов высокого давления реакторов, титан, вольфрам, стали на основе Cr–Mo–V, метод Master Curve.

Introduction. Radiation embrittlement is one of the main ageing mechanisms of the materials of the fission and fusion reactors. In many cases, it determines the lifetime of these devices.

The Budapest research reactor (VVR-SM type) – after full reconstruction and power upgrade to 20 MW – was started again in 1993. The reconstructed reactor passed successfully the 1 year test period, and since January 1994 it is in normal operation with 10 MW power. Radiation embrittlement research is available since then. The Budapest research reactor and the BAGIRA 1 and 2 rigs are widely used to study the irradiation effects on the structural materials to evaluate the safety and lifetime of the presently operating and future nuclear energy sources. The Atomic Energy Research Institute participated in the IAEA co-ordinated research programs (CRP-3 to CRP-9) with irradiated material testing. The irradiated material testing is also used in many EU framework programs (FRAME, GRETE, COBRA, ATHENE, PERFECT, COVERS, NULIFE), as well as in the European Fusion Program. National irradiation programs support the lifetime extension of the Paks NPP. We present some examples of the presently performed tests.

The BAGIRA Irradiation Rig System. In close co-operation with the Paul Scherer Institute (Switzerland), two new rigs called BAGIRA (Budapest advanced gas-cooled irradiation rig with aluminum structure) have been constructed [1, 2]. The irradiation volume of the largest rig is $360 \times 20 \times 30$ mm and it can operate in the temperature range of 150–650°C. The heating is provided by combined gamma and electrical heating devices; the temperature measurement is performed by 6 thermocouples, the temperature is controlled by a helium-nitrogen gas mix flow. The fast fluence within the rig is $(2–6)\cdot 10^{13}$ n/cm$^2\cdot$s. Similar but smaller irradiation channel has been constructed for low temperature long-term irradiation testing of fusion materials.

The design includes four basic parts:

1. Dry channel tube (remains continuously in the reactor core).
2. Removable sealed plug.
3. Replaceable target holder.
4. Temperature control and safety system, which includes cooling gas system, analogue and computerized control and data acquisition system.

Dry Channel Tube. The shape of the dry channel tube is identical to the shape of 3 fuel elements in the core. The dry channel tube is made of forged RAlMg2.5 type aluminum. Inside the dry channel, tube channels for the cooling gas and a cavity for the target material with an area of 26×40 mm are prepared.

The outer shape of the dry channel tube is designed to fit the core supports for the fuel elements (Fig. 1), while the gap between the neighboring fuel elements and the channel is big enough for proper cooling water flow.

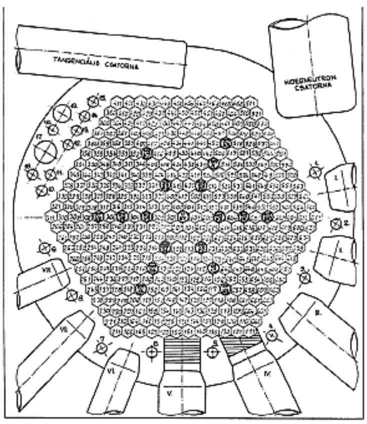

Fig. 1

Fig. 1. The reactor core and locations of rig 1.

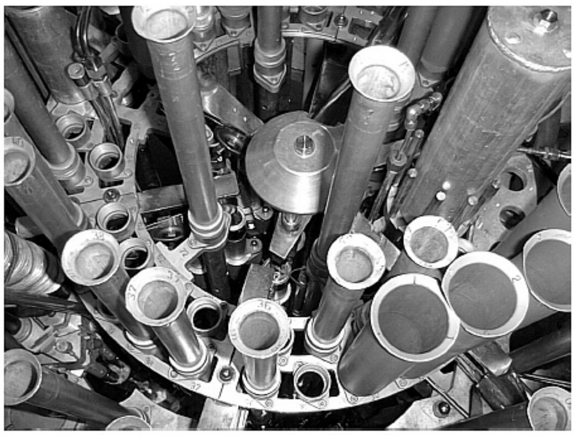

Fig. 2

Fig. 2. The upper part of the Budapest research reactor with the head of BAGIRA 1 rig.

The shape of the dry channel tube remains unchanged up to one meter over the core level to facilitate the replacement of the neighboring fuel elements, it continues in circumferential pipe until it reaches the water level, and ends about 500 mm above it. At the end of the tube there is a removable sealed cover (Fig. 2) including a foil which fails in case of over-pressurization. By removing the cover and disconnecting the thermocouples the sealed plug and the target holder can be removed.

The upper end of the dry channel tube is fixed to the reactor wall.

The Removable Radiation protection Seal. The removable seal is a long tube closed at both ends, and filled with water. It fits into the upper part of the dry channel tube with a small gap. The purpose of the seal is shielding the neutron radiation during operation. The lower part of the seal is a connecting mechanism ensuring easy connection and disconnection of the target holder. After irradiation remote-controlled crane pulls out the seal and the target holder from the dry channel and a hydraulic scissor cuts down the target holder, which is carried to the hot cells by a remote handling car.

The Replaceable Target Holder. The target holder is replaced for every irradiation campaign, while other parts of the equipment are not moved. This type of design reduces the irradiation procedure costs and increases safety.

The general scheme of the target holder of the BAGIRA irradiation equipment is shown in Fig. 3.

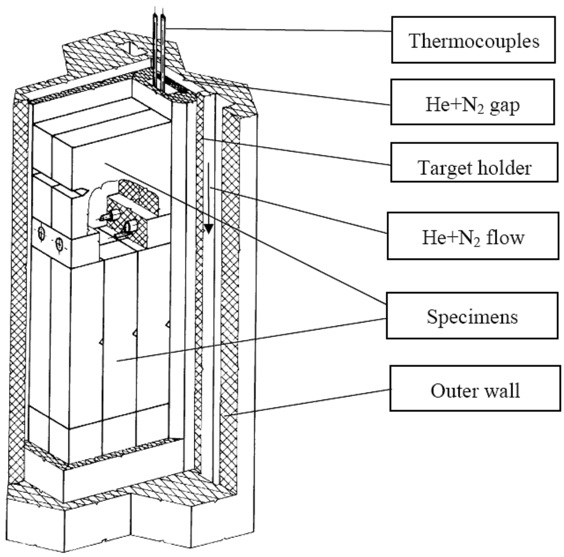

Fig. 3. Cross section of the BAGIRA irradiation rig.

The functions of the target holder are: to fix and protect specimens, remove the induced gamma heat and maintain the same temperature in the specimens, fix and protect thermocouples and the dosimetry foils during transportation, replacement and irradiation tests.

The maximum volume which can be used for specimens inside the target holder is $20 \times 30 \times 360$ mm, which is enough for irradiation of 1700 gr steel material. The target holder is made from aluminum 99.9 for low temperature irradiation and from titanium or stainless steel for high temperature irradiation. Special target holder has been developed for re-irradiation of radioactive specimens.

Temperature Control and Safety Features. Without continuous adjustment, the target temperature follows quickly the reactor power changes. The gamma heating generated in steel at 10 MW reactor power is up to 3 W/gr (it is about 1.5 W/gr at irradiation of titanium and below 1 W/gr at irradiation of aluminum alloys or ceramics). During the design of each irradiation test the material and the mass of the target have to be considered, and the gap between the outer wall and the target holder has to be selected according to the required irradiation temperature. Experience shows that the flow and pressure of the gas mix only slightly affects the quantity of the removed heat, the He–N$_2$ ratio and the gas gap. Therefore the auxiliary electric heating can be used for temperature adjustment. After the start of the reactor gamma heating has a short peak, then during the following two days it slowly diminishes until stable conditions are reached.

The temperature adjustment is controlled by a PC connected to a 16 channel a-d converter collecting the thermocouple and gas pressure data. The software evaluates and stores the data and gives orders to the control system according to the measured values. The software can cope with the normal operation and emergency conditions (target overheating, cooling system leakage, pump fault, dry channel tube leakage, etc.). The operation parameters use transferred through a local net/Internet, and adjustment of the temperature is remote-controlled.

Dosimetry. Dosimetry foils are placed in special foil holders between the target materials. Every foil is coded and its location recorded. After the opening of the target holder, the foils are transported to the neutron activation laboratory, and the gamma spectrum is measured.

Safety Considerations. The gas-mix removes the gamma radiation-generated heat even without any flow, therefore no significant target overheating is possible even at the malfunction of the control system.

The dry channel tube integrity is checked for an inner pressure 20% higher than the rupture pressure of the burst foil and at least 50% higher than the opening pressure of the safety valve.

Leakage of the dry channel tube can cover only a very small volume inside in the core level (the gas volume is limited), so the water inflow would not seriously affect the reactivity.

The control system automatically restarts the controlling computer, in case of system freezing. The design ensures that the water flow between the dry channel tube and the fuel elements is sufficient for proper cooling of the fuel elements, as well as the dry channel.

These features make the BAGIRA rigs inherently safe.

Future of Irradiation Tests in VVR-SM Reactors. Presently the main interest of the nuclear industry is the development of fusion reactors and generation of fission reactors. In order to increase the efficiency and decrease the environmental impact, high operation temperature irradiation tests will be used. Consequently, high temperature irradiation tests combined with in-pile creep and fatigue testing are the future tasks of the irradiation devices.

The future reactors will be exposed to very high fluences (5–100 dpa) during their operation. Presently the medium research reactors (like the VVR-SM) can produce only 1–5 dpa/year. Many specialists share the opinion that the role of these research reactors will decrease in the future. However, two facts support the future use of them:

(i) at very high flux the so-called flux rate effect causes high bias in the material ageing results;

(ii) a considerable part of the ageing process occurs shortly after the irradiation starts.

The radiation embrittlement is the result of at least four different ageing mechanisms: presence of precipitations and segregations, increase in the dislocation density and annealing at irradiation temperature [3]. The first two ageing mechanisms occur at low fluences (generally much less than 1 dpa) and quickly saturate. To study this mechanism, the present middle-power research reactors are ideal devices.

The time-dependent annealing and thermal ageing also cannot be studied in high flux devices. Large-size specimens cannot be used in high flux irradiation devices due to the high heat generation in the middle section of the specimens. These facts imply the combined use of the high flux devices (high flux reactors, spallation neutron sources, two- and three-beams' accelerators) and the available research reactors. In the Atomic Energy Research Institute, the development of future irradiation devices (in-pile creep and fatigue testing at high temperature, corrosion testing in supercritical water during irradiation) has been initiated.

Material Studies. *RPV Cladding Tests*. Most of the reactor pressure vessels (RPV) of the PWRs are inclad with a stainless steel layer, which is generally made by welding and is 3–10 mm thick. At Pressurized Thermal Shock (PTS) analysis the cladding was neglected. If the RPV cladding is ductile and free from defects, no assumption of hypothetical surface cracks is required during the PTS calculations. Without hypothetical surface cracks, the PTS calculations yield 20–30% longer lifetime, which is very important for lifetime extension [4]. Moreover, the irradiation damage of cladding is different from that of ferrite and austenitic steels.

Tensile curves obtained for cladding 15 mm long specimens 3 mm in diameter are shown in Fig. 4. The results indicate that the cladding ductility is optimal at room temperature, while at 300°C the elongation of the unirradiated cladding is less than that of EOL irradiated cladding at room temperature. The irradiation at 300°C only slightly affects the elongation [4].

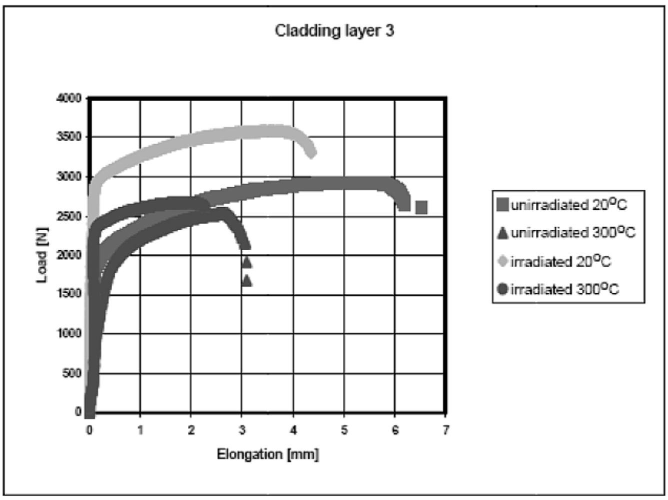

Fig. 4. Tensile results for RPV cladding.

Application of Cr–Mo–V Steel at High Temperatures. The Cr–Mo and Cr–Mo–V steels are used in the PWR power vessels. The irradiation resistance of these materials is widely studied within the operation temperature range of the present PWRs (250–320°C). To improve the efficiency of the future reactors, the operation temperature should be increased. One option is to shift the operation temperature range to 350–560°C. The fissile plants operate within this temperature range and are manufactured from Cr–Mo and Cr–Mo–V steels. These materials are widely used in the industry, their application requires not too much technology development, and their long-term creep and thermal ageing properties are well-known. These are candidate materials for the future fourth generation supercritical water reactors. In the BAGIRA rig, irradiation of Cr–Mo–V steels is presently performed at 450°C.

Ti-Alloys. Ti-alloys have been irradiated and tested for ITER fusion device. In the Tokamak type fusion devices, the first wall and the vessel are connected with elastic connection elements. The high strength and low Young's modulus of the Ti-alloys allow one to use them as elastic elements. This part of the device is exposed to irradiation of about 0.5 dpa. We have irradiated Ti–6l–4V alloy and studied its mechanical properties.

Tungsten. Tungsten is resistant against high temperature. Diverters and other first wall elements of fusion devices and some parts of the high temperature gas-cooled reactors are planned to be produced from tungsten alloys. The radiation embrittlement of tungsten alloys has been studied on samples irradiated in the BAGIRA rig. The load–deflection curve obtained from three-point bending test of irradiated tungsten bars is shown in Fig. 5.

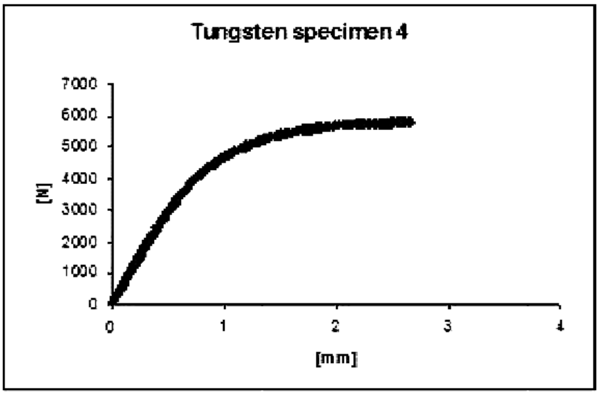

Fig. 5. Load-deflection curve from three-point bending test of irradiated tungsten bars.

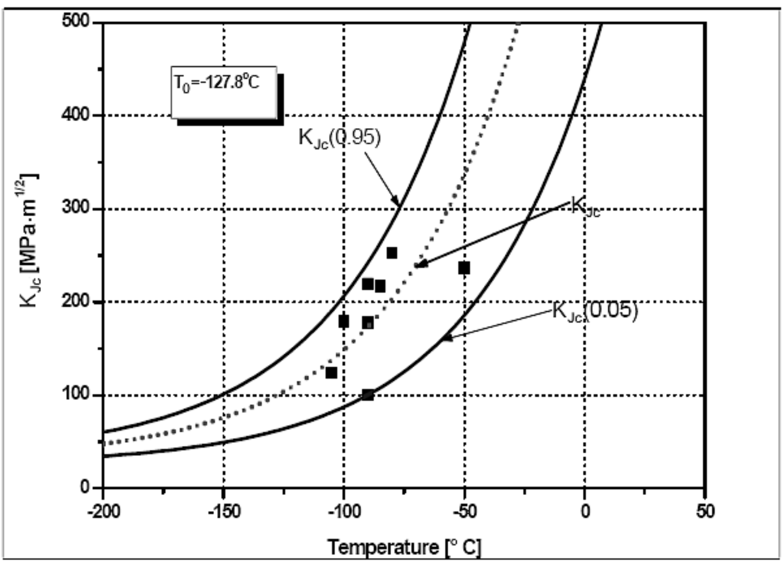

Fig. 6. Master Curve toughness testing results for 9% Cr steel.

9% Cr Steels. The 9% Cr ferrite-martensitic steels have high strength at high temperature, are creep-resistant and can be applied in the 650–900°C temperature range. As these steels consist of low half-decay time elements, after limited decay time they can be treated as low activity waste, and after 100 years' decay can be reused. Since similar steels are regularly used in the power industry, their production and machining technologies are well-known. We have studied the possibility of measuring the fracture toughness on small-sized irradiated Euroferr specimens – which are the limitations of the use of reconstituted specimens and the so-called Master Curve method [5]. The Master Curve obtained for unirradiated 9% Cr steel is shown in Fig. 6.

Conclusions. Our studies imply that research reactors are critical for the testing of the irradiation toughness of the structural materials of the available RPVs and future fission and fusion reactors. Even when high-flux spallation sources will come to operation, the role of available research reactors will be important, for the following reasons:

(i) the leading embrittlement mechanisms (segregations and precipitations) occur at the fluence range which can be easily reached by the available middle- and high-flux research reactors;

(ii) the research reactors can be used for irradiation of specimens large enough for valid fracture toughness testing;

(iii) the middle-flux research reactors can be used to study the flux rate effect.

Р е з ю м е

Описано експерименти по радіаційному опроміненню матеріалів у ядерному реакторі потужністю 10 МВт із використанням нового оснащення. Як приклад наведено результати досліджень покриттів посудин високого тиску реакторів зі сталей з вмістом 9% Cr, використання методу Master Curve для реакторних сталей, титанових сплавів та вольфраму.

1. F. Gillemot, G. Uri, T. Fekete, et al., "The new high temperature irradiation rig of the Budapest research reactor," in: Specialist Meeting "*Irradiation Embrittlement and Mitigation*," Espoo, Finland, 23–26 October, 1995.
2. F. Gillemot and G. Uri, *Development of the BAGIRA Irradiation Rig*, Yearly Report of the Budapest Neutron Centre (2000).
3. L. Debarberis, B. Acosta, A. Zeman, et al., "Effect of irradiation temperature in PWR RPV materials and its inclusion in semimechanistic model," *Scripta Mater.*, **53**, 769–773 (2005).
4. F. Gillemot, M. Horvath, H. W. Viehrig, and L. Debarberis, "Behaviour of irradiated RPV cladding," in: 23rd Symp. on *Effect of Radiation on Materials*, ASTM, San Jose, California, 11–16 June, 2006.

Received 21. 06. 2009