Study of irradiation effects in materials with high-neutron-flux fission reactors

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Abstract

History of irradiation studies utilizing fission reactors in university-related activities of Japan is briefly reviewed and the future prospect will be described. For a moment, two major materials irradiation fission reactors, JMTR and JOYO are under refurbishment, which resultantly highlights importance of international collaboration.

I. Introduction

Studies of irradiation effects in material with fission reactors are facing to an important turning-point after their long histories with accumulation of vast data-bases. Studies related with present-water cooled power reactors as well as with advanced nuclear power systems such as generation-IV types and nuclear fusion reactors are strongly demanding higher neutron fluence irradiation, where the typical irradiation dose demanded will be far beyond 10 dpa for structural materials. There, needed neutron flux will exceed $10^{18}$ n/m²s and hopefully it will be in the range of $10^{19}$ n/m²s. In the meantime, an accumulated irradiation data-base clearly shows that acquisition of comprehensive irradiation parameters, such as temperatures, oxygen chemical potentials, neutron spectra, and gamma-ray dose rates is indispensable for establishing reliable understandings in behaviors of materials in actual nuclear systems. There, appropriate instrumentations are essential for irradiation rigs used in high-neutron-flux irradiation. Also, recent advance of understandings of irradiation effects highlights importance of in-situ type studies in fission reactors. Examples will be studies for transient behaviors of nuclear fuels and issues related with chemical compatibility of materials with their environments under irradiation, such as stress corrosion cracking studies. Study on dynamic radiation effects in functional materials will be another example. Functional materials are expected to play a more and more important role in advanced nuclear systems, where real time diagnostics of operation conditions and resultant feedback-control of the operation conditions are important.

In a water-cooled type fission reactor, a nuclear heating rate (a gamma-ray dose rate) is usually high, namely about 10W/g ($10^4$Gy/s) for $10^{18}$n/m²s fast neutron flux ($E>0.1$MeV) for iron. There, a higher neutron-flux irradiation means a higher nuclear heating rate. It means that components composing irradiation rigs will be easily heated up to above 500C, if they are not appropriately cooled. Usually, the reliable
instrumentations are very difficult, when the gamma-dose rate exceeds 10W/g for iron. There, mineral insulating cables and thermocouples, if they are thermally isolated and it will easily happen, will be heated up locally above 1000C. Also, the temperature gradient in materials under irradiation is substantial, when materials’ dimension is large. Thus, new instrumentation techniques will be essential for the advanced irradiation studies in a high-neutron flux reactor. Also, some coupling type irradiation utilizing a mixed spectrum reactor and a fast reactor should be taken into consideration. The paper will report present status of instrumentations for reactor dosimetry and in-situ measurements in Japan for fundamental studies of nuclear materials utilizing fission reactors.

II. History of heavy irradiation study utilizing fission reactors in university-related activity in Japan

Materials study utilizing fission reactors in Japan started in 1960s utilizing JRR-2 (Japan Research Reactor 2) of Tokai Research Establishment of Japan Atomic Energy Research Institute(JAERI, corresponding to the present Japan Atomic Energy Agency (JAEA)). There, the materials were irradiated in a small aluminum-made capsule dipped directly into its core coolant water. There was not a temperature control system and irradiation temperature was typically around 50-100C. The fast neutron fluence attained was around $10^{22-23}$ n/m² at the maximum.

Meeting with increasing demands for reactor irradiation, a new fission reactor, Japan Materials Testing Reactor (JMTR) was constructed and went into its criticality in 1969 in Oarai Research Establishment of JAERI. JMTR is a light water cooled and moderated tank type reactor for multipurpose uses. The Oarai Branch of Institute for Materials Research (IMR-Oarai) of Tohoku University was founded in 1971 to work as a coordinator between reactor users in universities and the JMTR. General features of the JMTR are shown in Table 1 and a role of IMR-Oarai in conjunction with JMTR and other research reactors is schematically shown in Fig. 1.[1]

Materials were accommodated in a specially designed irradiation rig and typical irradiation temperature was in the range of 100-600C. The fast neutron fluence attainable in a year will be at the most around $10^{25}$ n/m². Starting with non-instrumented irradiation rigs, several instrumented irradiation rigs were developed as shown in Fig. 3, meeting with demands of university researchers whose interests diverged in a wide research fields.

In the meantime, demand for a much higher fast neutron fluence was elicited in university researchers, whose major concerns were in the fields of development of a nuclear fusion reactor. For the realization of a commercially compatible nuclear fusion reactor, development of structural materials which could survive neutron fluence up to $10^{26}$-10^{27}$ n/m² (more than 100dpa(displacement per atom)). Thus, utilization of high neutron flux fast reactor, JOYO was started in 1978. The JOYO was an experimental
Table 1  Features of JMTR (Japan Materials Testing Reactor)

<table>
<thead>
<tr>
<th>Feature</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reactor Power</td>
<td>50 MWt</td>
</tr>
<tr>
<td>Fast Neutron Flux (Max)</td>
<td>$4 \times 10^{18} \text{n/m}^2\cdot\text{s}$</td>
</tr>
<tr>
<td>Thermal Neutron Flux (Max)</td>
<td>$4 \times 10^{18} \text{n/m}^2\cdot\text{s}$</td>
</tr>
<tr>
<td>Flow of Primary Coolant</td>
<td>6000 m$^3$/h</td>
</tr>
<tr>
<td>Coolant Temperature</td>
<td>49°C / 56°C</td>
</tr>
<tr>
<td>Core Height</td>
<td>750 mm</td>
</tr>
<tr>
<td>Fuel</td>
<td>ETR type, 19.8% $^{235}$U</td>
</tr>
<tr>
<td>Irradiation Capability (Max)</td>
<td>60 capsules</td>
</tr>
<tr>
<td>Fluence/y (Max)</td>
<td>$3 \times 10^{25} \text{n/m}^2\cdot\text{y}$</td>
</tr>
<tr>
<td>dpa of Stainless Steel (Max)</td>
<td>4 dpa</td>
</tr>
<tr>
<td>Diameter of Capsule</td>
<td>30 - 65 mm</td>
</tr>
<tr>
<td>Temp. Control (Max)</td>
<td>2000°C</td>
</tr>
</tbody>
</table>

Figure 1  Role of the IMR-Oarai for utilization of fission reactors for university researches in Japan
sodium-cooled fast breeder reactor (the fast reactor) and its major mission at that time was development and validation of fundamental technologies for the sodium-cooled fast breeder reactor. Thus, the material irradiation for nuclear systems other than a fast reactor was recognized as a side business and the irradiation was carried out mainly with non-instrumented rigs.

By 1980s, importance of appropriately control of irradiation conditions and acquisition of irradiation parameters were well appreciated. For higher neutron fluence irradiation in more defined conditions, university researchers started utilization of FFTF-MOTA (Fast Flux Test Facility – Materials Open Test Assembly) of Pacific Northwest Laboratory of the USA in 1987, under the Monbusho (Ministry of Education of Japan) / USDOE (Department of Energy of the USA) nuclear fusion science collaborative program.[2] In FFTF-MOTA, irradiation temperature was actively controlled. Also, in JOYO, a new instrumented irradiation rig called INTA was developed to realize the active temperature control, and then, more versatile temperature control irradiation rig called MARICO and its successor MARICO-2 were developed. More sophisticated instrumented irradiation rigs were developed for JMTR irradiation as shown in Fig. 2 in 1990s, whose example is shown in fig. 3, for multi temperature and multi neutron fluence control. Instrumented irradiation rigs for in-situ measurement of material-property changes under reactor irradiation were also developed in JMTR in 1980s-1990s.

Figure 2  Development of materials irradiation rigs in the IMR-Oarai
These efforts were extended in the Japan/USA collaboration mentioned above in 1990s in the reactor of HFIR (High Flux Isotope Reactor) of Oak Ridge National Laboratory. Measurements of the electrical conductivity of ceramic insulators and protonic conductors, and of thermal conductivity of nuclear fusion candidate materials such as SiC/SiC composites were carried out in JMTR and HFIR. Optical measurements were also carried out in JMTR using radiation resistant silica-glass optical fibers. Optical measurements were also implemented in BR-2 (Belgium Reactor 2) of SCK/CEN (Belgian Center for Nuclear Energy) under the international collaboration of the ITER-EDA (ITER-Engineering Design Activity). JMTR stopped its operation temporarily in 2006 for refurbishment, aiming for its restart in 2011. Also, JOYO had stopped its operation since 2007, due to a trouble caused by an instrumented irradiation with MARICO-2. The IMR-Oarai exploited a possibility of utilizing fission reactors overseas and extended its collaboration with SCK/CEN, resultantly starting utilization of BR-2 for materials irradiation in 2005. BR-2 is a light water cooled reactor which has many features similar to those of JMTR, with a little higher fast neutron flux. Up to now, three programs, GAYSHA, MICADO-1 and MICADO-2 were accomplished, utilizing irradiation rigs of CALLISTO, ROBIN and BAMI. The project of MICADO-3 is under way.

Figure 3  Multi temperature and neutron fluence control irradiation rig in JMTR
III. Irradiation studies from a view points of university researchers

In an early stage of reactor utilization for study of radiation effects in materials, a fission reactor regarded mainly as a high intensity neutron source. For engineering aspects, validation of wholesomeness of materials and their assembly in a specific nuclear environment was a major target. There, a fission reactor was regarded as an actual practical environment for the application of concerned materials and their assembly and necessity of active control of irradiation conditions were not strongly recognized. In this stage, vast amount and vast kinds of materials were irradiated in materials testing reactors such as JMTR and JOYO. Also, in the fields of materials science, initial concerns were phenomena related with interactions between energetic neutrons and materials and detailed control of irradiation parameters were beyond its scope. (too sophisticated and too expensive)

In the meantime, fundamental aspects of irradiation effects were studied with more irradiation-conditions controllable tools such as ion and electron accelerators and fusion neutron sources (such as RTNS-2 (Rotating target Neutron Source 2) of Lawrence Livermore National Laboratory of the USA, and FNS (Fast Neutronics Source) of the JAEA). Also, advancement of studies by computer simulation techniques was remarkable. To interpret complicated irradiation behaviors of materials in fission reactors from fundamental mechanisms of irradiation effects, irradiation in a fission reactor under more well-defined and well-controlled conditions was becoming more and more needed. A typical example will be an evolution of irradiation studies from the EBR-2 (Experimental Breeder Reactor 2 of Idaho National Laboratory) project to the FFTF-MOTA project in the USA. In Japan, the transition was much more moderate and was not deliberately done, but in retrospect, the trend was clearly identified that the number of instrumented irradiation rigs increased as the time went by in JMTR as well as in JOYO. A general perspective of study of irradiation effects in fission reactors is schematically shown in Fig. 4.

Development of instrumented and irradiation-conditions controllable rigs was not difficult in general in water-cooled type materials testing reactors such as JMTR and HFIR. There, feed-through systems through a pressure boundary was preinstalled and cables needed for instrumentations were easily taken out of the reactor core regions. Some reactors even do not have a pressure boundary such as JRR-3. In the meantime, realization of the instrumentations in a fast reactor is not easy in general. Incarnation of MOTA facility in FFTF was a remarkable demonstration of the advanced reactor-irradiation-technology in the USA. MARICO and MARICO-2 project in JOYO would have demonstrated advanced irradiation-technology in Japan, but resultantly, it reconfirmed difficultness of the instrumentation in fast reactors.

However, in general, reactor irradiation with detailed instrumentations and in well-defined conditions is now a proven technology in many materials testing reactors.
Figure 4 Evolution of irradiation study in conjunction with fission-reactor irradiation studies

The next step will be surely the instrumented and controlled reactor irradiation with higher fast neutron flux and fluence. For the innovation of nuclear power systems, comprehensive understandings of neutron irradiation effects with neutron fluencies higher than that can be realized in a conventional materials-testing-reactors (corresponding to about several dpa). Lifetime extension and improvement of safety/efficiency of a present light water power reactor is demanding irradiation studies with a neutron fluence much higher than $10^{25} \text{n/m}^2$ (preferably higher than 15-20dpa). Development of the fast breeder reactor and nuclear fusion reactor is demanding fast neutron fluence higher than $10^{26} \text{n/m}^2$ (preferable in the range of 200dpa). To achieve the high fast neutron fluence, the neutron flux higher than middle of $10^{18} \text{n/m}^2$’s will be needed. In a conventional water cooled fission reactor, such as JMTR and HFIR, it means that the associated gamma-ray dose rate is higher than $10^4 \text{Gy/s}$, namely a nuclear heating rate of higher than 10W/g for iron.

The electrical conductivity measurements in the JUPITER project in HFIR mentioned above and the study of behavior of magnetic probes for burning plasma fusion devices in JMTR clearly showed the difficulty of reliable instrumentations in the reactor core region where the gamma-ray dose rate exceeds $10^4 \text{Gy/s}$. In the coming decade,
instrumented irradiation in a high flux reactor core region is planned in materials testing reactors such as JMTR. However, it will be a difficult technical challenge and development of technology such as development of reliable mineral insulating cables and their termination techniques as well as long-life electric heaters will be indispensable. Also, probes monitoring irradiation conditions, such as oxygen chemical potential, hydrogen concentration, temperatures, gamma-ray dose rate, neutron flux, etc., are very difficult to develop in such a high gamma-ray dose rate environments, insuring their reliability and long-life.

Also, materials irradiation below 300-350°C in a gamma-ray dose rate higher than 10 W/g is in general very difficult, except for installation of an active special cooling system. The irradiation below 300-350°C is very important for studies related with a light water cooled and moderated reactors.

In the meantime, the sodium-cooled fast reactor has a definite disadvantage for the instrumented irradiation as its boundary structure surrounding its core environment is complicated to satisfy safety regulations. The trouble associated with MARICO-2 irradiation rig in JOYO happened to demonstrate the difficultness. Also, the coolant temperature is higher than that of a light water cooled and moderated type reactors. However, the fast reactor has a definite advantage over a water cooled materials testing reactor, namely, it has relatively a low gamma-ray dose rate, referring to the same fast neutron flux. A typical gamma-ray dose rate will be less than \(10^4\) Gy/s with the fast neutron flux of \(10^{19}\) n/m² s, being about \(10^{10}\) of that in a water cooled fission reactor. Thus, a sodium-cooled fast reactor may be able to play a definite role in instrumented irradiation with a desirable high fast neutron flux, if we could overcome technical difficulties related with complicated boundary surrounding its reactor core.

In total in Japan, JRR-3 will cover the utilization stage of I and II in Fig. 4, and JMTR will cover from stage I to III, Only JOYO will have a potential to cover from stage I through IV, in Japan. It will be very difficult to utilize the next fast reactor, MONJU as a material testing reactor in stage IV, as its main mission is to validate commercially available sodium-cooled fast breeder reactor, and there, complicated instrumentations and related modification of safety boundary structures will not be realistic.

Another important aspect to be addressed finally will be issues related with a use of low enriched uranium fuels and a multi-purpose utilization of a reactor. The use of low enriched uranium is an inevitable result from the non-proliferation problem. And, multi-purpose utilization of a materials testing reactor is also socially inevitable as a countermeasure to deal with a very high cost of a reactor operation. However, the conversion of fuels from highly enriched uranium to low enriched uranium is making a reactor operation more restricted. It sometimes hampered scientific feasibility, though it must be accommodated. To ensure reactivity of a nuclear core for meeting sophisticated demands from advanced materials tests, adaption of more advanced nuclear fuels such as U-Mo will be strongly needed.

Also, in multi-purpose utilization of a reactor, some purposes will strongly interfere with each other. For example, from a viewpoint of radioisotope production and
nuclear doping, long and steady state operation is needed. There, an interruption period of a reactor operation should be minimized. Profiles of neutron flux should not be disturbed in each operation cycle. In the meantime, in advanced materials test, a long interruption period is needed to accommodate sophisticated instrumentation. Irradiation conditions such as neutron flux and fluence would be modified to satisfy specific demands from each experiment. From the view point of materials science, the present “multi-purpose” sounds sometimes “too many purposes” Simplification of reactor purposes are strongly recommended through collaboration among reactors internationally.

IV Summing ups

After its long history, the materials irradiation studies utilizing fission reactors are facing to the turning-point. The major concerns there will be toward instrumented and controlling irradiation in a higher neutron flux. Development of new irradiation technologies, including development new materials needed for the new technologies, will be strongly needed. In the meantime, materials irradiation studies utilizing fission reactors are becoming more and more expensive and time consuming. Collaboration among organizations participating fission-reactor materials irradiation will be inevitable.

References