Irradiation Performance of HTGR Fuel in WWR-K Research Reactor

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Abstract – A capsule irradiation test with the high temperature gas-cooled reactor (HTGR) fuel is being carried out using WWR-K research reactor in the Institute of Nuclear Physics of the Republic of Kazakhstan (INP) to attain 100 GWD/t-U of burnup under normal operating condition of a practical small-sized HTGR. This is the first HTGR fuel irradiation test for INP in Kazakhstan collaborated with Japan Atomic Energy Agency (JAEA) in frame of International Science and Technology Center (ISTC) project. In the test, TRISO coated fuel particle with low-enriched UO2 (less than 10 % of 235U) is used, which was newly designed by JAEA to extend burnup up to 100 GWD/t-U comparing with that of the HTTR (33 GWD/t-U). Both TRISO and fuel compact as the irradiation test specimen were fabricated in basis of the HTTR fuel technology by Nuclear Fuel Industries, Ltd. in Japan. A helium-gas-swept capsule and a swept-gas sampling device installed in WWR-K were designed and constructed by INP. The irradiation test has been started in October 2012 and will be completed up to the end of February 2015. The irradiation test is in the progress up to 69 GWD/t of burnup, and integrity of new TRISO fuel has been confirmed. In addition, as predicted by the fuel design, fission gas release was observed due to additional failure of as-fabricated SiC-defective fuel.

I. INTRODUCTION

Japan Atomic Energy Agency (JAEA) has been developing a 50 MWt small-sized high temperature gas cooled reactor (HTGR) for multiple heat applications, named HTR50S, with the reactor outlet coolant temperature of 750 °C and 900 °C [1]. It is first-of-a-kind of the commercial plant or a demonstration plant of a small-sized HTGR system to deploy it in developing countries in the 2020s. The design concept of HTR50S is to satisfy the user requirements for multipurpose heat application, to upgrade its performance compared to that of the High Temperature Engineering Test Reactor (HTTR) developed in JAEEA [2] without significant research and development (R&D) utilizing the knowledge obtained by the HTTR design and operation, and to fulfill the high level of safety by utilizing the inherent features of HTGR and a passive decay heat removal system.

Both HTR50S and HTTR employ so-called TRISO CFP (Tri-structural ISOtropic coated fuel particle) as fuel. TRISO CFP has a fuel kernel made with uranium dioxide and coating layers in turn with a low dense pyrolytic carbon (PyC) as “buffer” for carbon monoxide (CO) and fission gas generated by fission, high dense inner PyC, silicon carbide (SiC) and high dense outer PyC having roles to retain fission products and to keep structural integrity against internal gas pressure. In a reference specification of HTR50S, the fuel is targeted at 120
GWd/t of average burnup, which is five times higher than that of the HTTR (22 GWd/t in average). The conventional HTTR CFP fails systematically under the excess design burnup due to internal gas pressure, although its intactness at twice of the design burnup has been demonstrated experimentally. [3, 4] On the other hand, for the fuel fabricated in commercial scale based on the HTTR fuel technologies, no irradiation data achieving in high burnup over 100 GWd/t has been obtained so far. Therefore, a specification of CFP shall be newly designed to reduce internal gas pressure for burnup extension, i.e., by employing smaller fuel kernel and thicker buffer layer.

The objective of this study is to demonstrate irradiation performance of the new TRISO CFP designed for burnup extension under normal operating condition of a practical small-sized HTGR. The studies are to confirm reliability and safety of the new TRISO CFP designed by JAEA and fabricated by Nuclear Fuel Industries, Ltd. (NFI) when high level of the fuel burnup is reached. In this study, a capsule irradiation test with the fuel is being carried out using WWR-K research reactor in the Institute of Nuclear Physics of the Republic of Kazakhstan (INP). This is the first HTGR fuel irradiation test for INP in Kazakhstan collaborated with JAEA in frame of International Science and Technology Center (ISTC) project.

Activities under preparatory phase of the study cover the following:
- design new TRISO CFP for extended burnup by JAEA,
- fabricate fuel specimens on a base of the HTTR fuel technologies and deliver the fuel specimens to Kazakhstan by NFI,
- develop and fabricate the special irradiation test device such as a gas-swept capsule and a gaseous fission products (GFP) monitoring system at WWR-K reactor by INP and
- carry out irradiation test in WWR-K under the condition surrogated normal operating condition of the small-sized HTGR to achieve a maximum level of burnup, and evaluate fuel integrity by GFP monitoring during the entire period of irradiation.

This paper describes irradiation performance of new HTGR fuel designed for extended burnup, based on results of the measured and predicted fission gas release fraction from HTGR fuel during irradiation.

II. EXPERIMENTAL

II.A. Design of New TRISO CFP for Burnup Extension

In accordance with the design philosophy of HTR50S [1] and with experiences on developing CFPs for high burnups through irradiation tests and post-irradiation examination by the Japan Materials Testing Reactor (JMTR) in JAEA [3, 4], etc., the reference specification of the new TRISO CFP was determined as shown in Table 1 according to the following requirements:
- The diameter of the new TRISO CFP was determined to be same as the HTTR, because of minimizing fuel failure fraction in the HTTR fuel compaction condition and keeping accuracies in fuel compact inspections.
- The diameter of UO₂ kernel was determined to be smaller than that of the HTTR in order to increase the volume of buffer layer for avoiding the pressure vessel failure of the CFP. A reference diameter of 500 µm was settled considering the fuel kernel fabrication process and a past experience on 550 µm kernel fabrication. [3]
- The thickness of buffer layer was determined to be larger than that of the HTTR. In order to avoid two modes of failures due to amoeba effect [6] and to the internal gas pressure.
- The thickness of SiC layer was determined to be larger than that of the HTTR considering the fuel kernel fabrication process and a past experience on 35 µm thick fabrication. [3]
- The thickness of the OPyC layer is settled as same as the HTTR CFP to avoid the failure of the CFPs with handling during the fabrication.

Table 1: Dimensions of new TRISO CFP.

<table>
<thead>
<tr>
<th></th>
<th>HTTR (Spec.) [5]</th>
<th>New TRISO (Spec.) [8]</th>
<th>New TRISO (as-fabricated, mean value)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kernel diameter (µm)</td>
<td>600</td>
<td>500</td>
<td>495</td>
</tr>
<tr>
<td>Buffer thick (µm)</td>
<td>60</td>
<td>95</td>
<td>94</td>
</tr>
<tr>
<td>OPyC thick (µm)</td>
<td>30</td>
<td>40</td>
<td>40</td>
</tr>
<tr>
<td>SiC thick (µm)</td>
<td>25</td>
<td>35</td>
<td>33</td>
</tr>
<tr>
<td>OPyC thick (µm)</td>
<td>45</td>
<td>40</td>
<td>41</td>
</tr>
</tbody>
</table>

Figure 1 shows a probability of the pressure vessel failure of the newly designed CFP as a function of burnup, which was calculated in the conditions; 1300 °C of irradiation temperature,
1,100 effective full power days (EFPDs) and \(4 \times 10^{25}\) m\(^{-2}\) of fast neutron fluences (E>0.18 MeV). According to the result, it was suggested that the burnup of the new CFP could achieve more than 100 GWd/t with \(\text{UO}_2\) having 10% of \(^{235}\text{U}\) enrichment. In addition, it was suggested that as-fabricated SiC-defective CFP would fail additionally to through-coatings (TC) failed CFP before achieving the target burnup of 100 GWd/t due to internal gas pressure increasing.

\[
\text{Probability of fuel failure}
\]

**Fig. 1:** A probability of the pressure vessel failure of the newly designed CFP (HBu) as a function of burnup in comparison of the HTTR fuel.

### II.B. Fabrication of Irradiation Fuel Specimen

The fuel kernels made with \(\text{UO}_2\) are fabricated by external gelation process [7]. Metal solution is prepared with the mixture of the uranyl nitrate solution and additive to control the viscosity of the solution. Droplets of the metal solution are generated at the vibrating nozzles and fall into ammonia water to be aged to ammonium diuranate (ADU) particles. The reaction products of ammonium nitrate etc. are washed off, then the particles are dried and calcined to \(\text{UO}_3\) particles at 500 °C in air. The \(\text{UO}_3\) particles are reduced and sintered to \(\text{UO}_2\) particles with about 97% T.D. at 1600 °C under hydrogen atmosphere.

Coating layers are deposited on the kernels in a chemical vapor deposition (CVD) process using a fluidized bed type of coater. The coating process adopted so-called “continuous” coating process, the process without unloading and loading of the particles at the intermediate coating process to reduce as-fabricated coating failures. Mixing gases of acetylene (C\(_2\)H\(_2\)) and argon (Ar) are used for the deposition of porous and low density PyC for the first layer; propylene (C\(_3\)H\(_6\)) and Ar for the deposition of dense PyC for the second and fourth layer; methyl-trichloro-silane (MTS) and hydrogen for the deposition of SiC for the third layer. Dimension of as-fabricated new TRISO CFP is given in Table 1. [8]

The fuel compacts are produced by warm-pressing of the CFPs with graphite powder. In the first step, the CFPs are overcoated by resinated graphite powder with alcohol. The resinated graphite powder is prepared by mixing electro-graphite powder, natural graphite powder, and phenol resin as a binder in the ratio 16:64:20, followed by grinding the mixture to powder. The thickness of overcoating layer is determined by the specification for the volume fraction of the CFPs in the fuel compact. Then the overcoated CFPs are warm-pressed by metal dies to form annular green fuel compacts. The final step of the compaction process is the heat-treatment of the green fuel compacts at 800 °C in flowing N\(_2\) to carbonize the binder and at 1800 °C in vacuum to degas the fuel compacts.

In this study, 3 fuel compact specimens (Fig. 2) were supplied for the irradiation test using WWR-K reactor. Dimension of the fuel compact having cylindrical shape was \(\phi 10\) mm (outer diameter) \(\times \phi 2\) mm (inner diameter) \(\times 12\) mm (height). Volume packing fraction of the CFPs in a fuel compact was settled in 30%. As-fabricated exposed uranium was less than \(1 \times 10^{-7}\) which is better than that of the HTTR fuel (\(2 \times 10^{-6}\)). On the other hand, as-fabricated SiC-defective fraction was 0 to \(1.8 \times 10^{-3}\), that is, at most 1 SiC-defective CFP could include in a fuel compact containing about 660 CFPs.

**Fig. 2:** Fuel compact specimens.

### II.C. Design and Construction of Irradiation Facility at WWR-K

WWR-K is a research reactor for material testing, isotope production and neutron activation analysis commissioned at INP in 1967. [9] WWR-K has 6MW of reactor power, \(1.0 \times 10^{18}\) m\(^2\)s\(^{-1}\) of the maximum thermal neutron flux. A scheme of the WWR-K research reactor is shown in Fig. 3.
Fig. 3: A scheme of the WWR-K research reactor and universal loop facility (ULF) system (solid line colored in blue): 1; reactor core, 2; experimenting box, 3; gas-vacuum systems, 4; electric power/gas supply system, 5; control room, 6; microcontroller.

Fig. 4: A design version of the capsule with three fuel specimens.

In order to design the irradiation capsule, irradiation conditions were settled at $1.0 \times 10^{18}$ m$^{-2}$s$^{-1}$ of thermal neutron flux and $1,050 \pm 100$ °C of irradiation temperature corresponding to HTGR normal operating condition. According to the condition, levels of the heat release under impact of the reactor gamma/neutron emission in the samples and structural materials of the irradiation device of various designs have been calculated. The neutronic calculations was performed using the computer code MCU-REA which implements the Monte Carlo method in the three-dimensional geometry to determine the burnup in uranium samples during reactor operation cycles [10]. The thermal calculations of the irradiation capsule were carried out by using the software ANSYS Fluent v.13 based on finite volume technique. 2-dimensional axisymmetric and 3-dimensional models were developed for the calculations. For symmetry reasons 3-dimensional model used a segment of $\frac{1}{4}$ capsule. The generated power density was calculated from 230 to 290 W/g-U (Watts per 1 gram of uranium) when the irradiation channel having $1.0 \times 10^{18}$ m$^{-2}$s$^{-1}$ of thermal neutron flux and $1,050 \pm 100$ °C of irradiation temperature were chosen.

Based on the result, main requirements to design of the irradiation device have been identified, and the final design of irradiation capsule was fixed as shown in Fig. 4. Molybdenum and zirconium have been chosen as structural materials of the irradiation capsule. Also, it was determined that 3 fuel compact could be loaded into the capsule.

II.D. Procedure of Swept Gas Monitoring

The swept gas monitoring system prepared for HTGR fuel irradiation test has been developed by INP. A block scheme of the system constructed in ULF is shown in Fig. 5 [11]. The highly-purified helium gas (99.999%) goes via pressure controller, to irradiation capsule. The gas pressure controller assures constant pressure of helium at a level of $10^5$ Pa. When taking a sample, helium from irradiation capsule, via throttle device, fills in a preliminary pumped-out reference vessel. The throttle device limits a rate of helium gas passing via capsule. Gas is pumped out by means of the ULF gas-vacuum system; and the gas, pumped out, is collected in special reservoirs. Radioactivity of the gas sampled in a sampling vessel is measured by the germanium detector.

Fig. 5: Block scheme of ULF prepared for irradiation HTGR fuel (EDV - electrically-driven valve; MCV - manually-controlled valve; PS - pressure sensors;
**II.E. Preliminary Analyses for Irradiation Test**

Neutronic and thermal hydraulics analyses have been conducted by INP for determination of irradiation condition to attain 100GWd/t of burnup. In the neutronic analysis by MCU-REA code, the calculation model takes into account actual configuration of the core, the values of the burnup in each fuel assembly in the core, obtained under assumption that the generated power density is 230 W/g-U and the duration of the reactor operation cycle is 21 days. As a result, 6.6% of $^{235}$U and 0.12% of $^{238}$U could burn out up to the end of the first cycle of irradiation. Figure 6 gives time-dependent changes of $^{235}$U and $^{238}$U during the irradiation. It was concluded that the target burnup 100 GWd/t could be attained for 400 days of irradiation, i.e., nearly 70% of $^{235}$U and 2.65% of $^{238}$U could burn out being correspondent to 93.4 GWd/t.

Integrity of the fuel during the irradiation was evaluated with the release rate to birth rate ratio (R/B) of fission gas $^{88}$Kr measured in the helium swept gas, because $^{88}$Kr is not influenced by precursor nuclide. According to the irradiation condition fixed by INP, fission gas release fraction as function of burnup under the condition was evaluated to confirm the integrity of the fuel during the irradiation by JAEA.

The predicted R/B value was evaluated by the fission gas release model developed in the HTTR project. In the model, both failed CFP and as-fabricated uranium contamination are considered as sources of the fission gas. The model treats two modes of the fission gas release, diffusion and recoil, for each source. In addition, the diffusional release takes two ways of diffusions, in-grain and in grain boundary, into consideration. Parameters and equations used in this model are reported in detail in the reference. [12] Finally, R/B of $^{88}$Kr is written as following equation:

$$
\frac{R}{B} = \left( f_{k,r} \times f_{m,d} + f_{k,d} \times f_{m,ad} \right) \times \phi_t + \left( f_{k,r} + f_{m,d} \right) \times \phi_c
$$  
* (eq. 1)*

where (R/B) is fractional release of fission gases from fuel compact, $f_{k,r}$ is recoil release fraction from kernel, $f_{m,d}$ is fractional release from fuel compact matrix, $f_{k,d}$ is fractional release from kernel by diffusion, $f_{m,ad}$ is fractional release by grain boundary diffusion, $\phi_t$ is fraction of through-coatings failed particle, $f_{k,r}$ is recoil release fraction from fuel compact matrix and $\phi_c$ is uranium contamination fraction in fuel compact matrix.

The above prediction was compared with measured data to determine additional failure fraction of the coated fuel particle during irradiation. The measured value of R/B of $^{88}$Kr was calculated by the following equations [12]:

$$
A(t) = A_{sat} \left[1 - \exp(-\lambda t)\right]
$$  
* (eq. 2)*

$$
A_{sat} = \frac{A(t_{amp})}{1 - \exp(-\lambda t_{amp})} \left( t = t_{amp} \right)
$$  
* (eq. 3)*

$$
\frac{dA(t)}{dt} = \lambda A_{sat} \left[\exp(-\lambda t)\right]
$$  
* (eq. 4)*

$$
R = \frac{dA(0)}{dt} = \lambda A_{sat} = \frac{\lambda A(t_{amp})}{1 - \exp(-\lambda t_{amp})}
$$  
* (eq. 5)*

$$
B = \lambda \cdot \frac{W}{3.2 \times 10^{-11}} \cdot \frac{Y}{100}
$$  
* (eq. 6)*

where $A(t)$ is radioactivity of $^{88}$Kr at time (Bq), $\lambda$ is decay constant of $^{88}$Kr ($6.88 \times 10^{-5}$ s$^{-1}$). $A_{sat}$ is saturated radioactivity of $^{88}$Kr (Bq), $t_{amp}$ is time of sampling after when the heat released from fuel are stable (s), $R$ is release rate of $^{88}$Kr (Bq/s), $B$ is birth rate of $^{88}$Kr (Bq/s), $W$ is heat released from fuel (W) and $Y$ is fission yield of $^{88}$Kr (3.58 % [13]).
III. RESULTS AND DISCUSSION

By June 2014, the irradiation test has performed 13 operational cycles and 257 EFPDs of irradiation duration. Up to this period of time, 6.9% of uranium (55% of uranium-235 and 1.6% of uranium-238) in fuel specimens burned out. The calculated level of burnup in the HTGR fuel samples has comprised about 69 GWd/t to an end of Cycle 13 as shown in Fig. 7.

![Fig. 7: Burnup in HTGR fuel for 13 cycles.](image)

During the irradiation, fission gas release fraction was evaluated. Figure 8 shows a comparison of measured and predicted R/B of $^{88}$Kr as function of irradiation time. As results, the measured R/B of $^{88}$Kr up to 26 GWd/t of burnup at around 104 EFPDs agreed well with the predicted which was calculated with as-fabricated uranium contamination (of which the fraction was $1 \times 10^{-7}$) and with no additional fuel failure. Gradual R/B increase with no additional fuel failure was shown up to 60 GWd/t-U at around 160 EFPDs. After that, R/B build-up was observed at 160 - 220 EFPDs. In accordance with the prediction of fission gas release fraction, R/B rising above 60 GWd/t-U corresponded to 3 to 6 through-coatings CFP failures.

As predicted in Fig. 1, probability of as-fabricated SiC-defective CFP to through-coatings failure would increase above 60 GWd/t of burnup. On the other hand, as-fabricated SiC-defective fraction of the fuel specimen supplied for this irradiation test was about $1.8 \times 10^{-5}$ corresponding to about 3 SiC-defective CFPs in the capsule, which agreed with a level of R/B increase in this time. Therefore, although further discussions shall be needed for quantitative accuracies of both R/B prediction and measurement, it was suggested that R/B rising above 60 GWd/t could cause additional failures of as-fabricated SiC-defective CFPs in fuel compact specimens.

![Fig. 8: A comparison of measured and predicted R/B of $^{88}$Kr as function of irradiation time.](image)

IV. CONCLUSIONS

The first HTGR fuel irradiation test targeted 100 GWd/t of burnup is in the progress at WWR-K of INP in Kazakhstan. Newly designed TRISO and fuel compact specimens have been designed and fabricated in basis of the HTTR fuel technologies in Japan. A helium-gas-swept capsule and a gas sampling device in WWR-K have been designed and constructed by INP. By June 2014, the burnup has attained about 69 GWd/t. Although fission gas release by additional failure of as-fabricated SiC-defective fuel might be observed as predicted by the fuel design, integrity of new TRISO fuel has been confirmed at this level of burnup. The irradiation test will continue by the end of February 2015 to confirm the integrity of the fuel under irradiation up to 100GWd/t.

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