Experience of long-term U–Al fuel performance in research reactors with low energy generation

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Abstract. Life cycle extension practices for U–Al fuel in Al cladding used at the OR and the IRT MEPhI research reactors are discussed. The IRT MEPhI reactor uses tube type IRT-3M FA with dispersed UO₂–Al fuel meat and SAV–1 aluminum alloy as cladding. The OR reactor uses pin type fuel elements with U–Al alloy fuel meat in cladding of Al alloy AMSN. The necessity of life cycle extension for the fuel elements is caused by a large calendar time of fuel assembly operation (10–30 years) and regulatory bodies concerns about this issue. The results of fuel cladding corrosion effects estimation using available empirical correlations for the corrosion rate and the rate of oxide film growth are presented. The results of visual inspection of the fuel elements are shown. Water chemistry monitoring practises in the reactor pool and in spent fuel storage are analysed. The successful experience of the IRT MEPhI (in 2003–2007) on repeated operation of the fuel assemblies previously discharged from the core (after the storage over 5–21 years) is presented.

1. Introduction

Fuel assembly (FA) operation time for low and medium- power research reactors with low coefficient of usage can be up to 10–30 years. In FA technical specifications fuel cladding leakage is a failure criterion. There are no limitations on calendar time of FA operation; however, in some cases, the regulatory bodies require to analyze the safety of FA long-term usage. This paper presents the results of such analysis for the OR and the IRT MEPhI research reactors.

The IRT MEPhI reactor uses tube type IRT-3M FA with dispersed UO₂–Al fuel meat and SAV–1 aluminum alloy as cladding. Before 2009, during the reactor operation, the time of FA usage was 10–15 years (until the maximum burnup reached 70%). After 11-year shutdown of the reactor because of administrative reasons connected with the relationships with the regulator, the time of FAs usage reached 11–25 years, including 11-year storage. In the framework of the reactor start planning, possibility of the fuel usage was analyzed. Visual inspection using underwater camera and leak testing with heating of all FAs were carried out. The successful experience of the IRT MEPhI on repeated operation of FAs previously discharged from the core (after the storage over 5–21 years) was
presented. The literature on the performance of Al alloys during reactor operation and wet storage was analyzed.

The OR reactor uses pin type fuel elements with U–Al alloy fuel meat in cladding of Al alloy AMSN. The FAs were used in the core during 29 years. The regulator requires to carry out analysis of condition of the reactor elements reached 30-year age to extend the life cycle of the element. To estimate life-limiting parameters of the fuel elements and to extend the life cycle of the OR reactor fuel the following activities were performed: operation logs and FA technical specifications analysis, fuel swelling calculation, calculation of cladding thickness reduction because of corrosion, and visual inspection of several fuel elements and FA guide tubes using underwater camera.

2. Reactor description

The IRT MEPPh reactor uses tube type IRT-3M FA with dispersed UO₂–Al meat and SAV-1 aluminum alloy as cladding (0.5 mm – thick). The FA with the longest service time was used over 14 years in the core and then was stored over 11 years in water basin of spent fuel storage. The calculated burnup of the FA is 55%. This FA is planned to be used only to reproduce the core loading before the shutdown period, then it will be replaced with fresh FA. There are four FAs with burnup of 45-51%, the burnup of the rest FAs is in the range 5–42%. The analytic estimation of the corrosion effects was conservatively performed for the FA of the longest service time with thermohydraulic parameters of the “hot” point. The parameters of the “hot” point are as follows: the cladding surface temperature − 74°C, heat flux − 0.24 MW/m², and water velocity − 1.8 m/s (for the power of 2.5 MW).

The OR reactor uses pin type fuel elements with U–Al alloy fuel meat in 1.5 mm - thick cladding of Al alloy AMSN. The FAs were used in the core during 29 years, whereas the integral energy generation of the core was 398 MW·h, and the average fuel burnup reached 0.6%. The analytic estimation of the corrosion effects was conservatively performed for the FA with thermohydraulic parameters of the “hot” point. The parameters of the “hot” point are as follows: the cladding surface temperature − 72°C, heat flux − 0.051 MW/m², and water velocity − 2.8 m/s (for the power of 0.3MW).

The main parameters of FAs of the IRT MEPPh and the OR reactors are presented in table 1.

| Parameter                      | OR      | IRT-3M  |
|--------------------------------|---------|---------|
| Fuel element type              | Pin-type| Tube-type|
| Fuel composition               | U-Al    | UO₂-Al  |
| ²³⁵U enrichment, %              | 36      | 90      |
| Fuel meat diameter/thickness, mm| 7       | 0.4     |
| Cladding thickness, mm         | 1.5     | 0.5     |
| Cladding material              | AMSN    | SAV-1   |
| Active part length, mm         | 500     | 580     |

3. Models for corrosion effects estimation

Aluminum cladding of fuel elements undergoes corrosion during their service. Uniform corrosion is the major form of attack on the cladding during in-reactor operation. Oxide film grows on the surface of the cladding. Formation of oxide film affects fuel performance by increasing fuel temperature because thermal conductivity of the oxide is lower than that of aluminum. The oxide film undergoes dissolution and erosion in flowing water; after it reaches a critical thickness, spallation may occur. Spallation tends to introduce local corrosion attack. The corrosion rate and the rate of oxide film growth depend on cladding-water interface temperature, heat flux at cladding surface, coolant flow rate, and water quality. The oxide films may be 20–50 µm thick in high performance research reactors (~2–5MW/m² or larger) [1].

The kinetic equation of aluminum alloy oxidation is formulated as follows [2]:
where $x, x_0, t, k,$ and $p$ are the oxide thickness, the oxide thickness at time zero, time, reaction constant and rate-law power, respectively. The model was validated against the experimental data for high flux research reactors [3].

According to [4], the uniform corrosion depth of aluminium alloys at temperatures $<100^\circ$C is parabolic with time:

$$K = K_pt^{1/2}$$

where $K, t,$ and $K_p$ are the corrosion depth, time, and corrosion constant, respectively.

The corrosion constant of SAV-1 alloy in water is 0.075, 0.092, and 0.11 $\mu$m·h$^{1/2}$ for the temperatures 40°C, 60°C, and 90°C, respectively (water was saturated with CO$_2$ and contained 0.005 mol/l H$_2$O$_2$, 0.05 mg/kg Cl$-$ and 0.01 mg/kg Cu, statics).

Pitting corrosion of the cladding of spent fuel elements is the major issue during long-term storage in water basins. Pitting of Al is encouraged by the presence of ions of Cu, halides, and bicarbonates, but diligent control of water quality can mitigate these concerns [1, 5, 6]. Also it was pointed out in [7] that the depth of pits does not grow significantly for long time water immersion because the iron-rich intermetallic inclusions that cause the formation of pits are corroded with time and this inhibits the pit growth. Furthermore, pitting corrosion can be revealed by visual inspection.

4. Analysis for the IRT MEPhI reactor

4.1. Corrosion effects analytic estimation

It is known that in low power research reactors problems from corrosion films formed on the aluminum cladding are uncommon if water quality is adequately controlled [1]. However, taking into account long-term fuel service and storage, the oxide film thickness and the corrosion depth were estimated for the IRT MEPhI FAs using available models [2, 4]. It should be noted that the correlations used for the corrosion depth and the oxide film thickness estimation were obtained based on the tests over time periods much shorter than 25 years and without long shutdown periods. Therefore, the estimations are qualitative rather than quantitative.

Using equation (1), the oxide film thickness for the FA with the longest time of operation (14 years of in-reactor operation and 11 years of underwater storage at spent fuel pool) was estimated. Relations for $k$ and $p$ were taken from the model described in [2]. Maximum cladding temperature ($T_w$), heat flux ($q$) and coolant flow rate ($v$) for the reference IRT MEPhI core at the nominal power of 2.5 MW were used in the calculations. The parameters of the FA operation were $T_w = 74^\circ$C, $q = 0.24$ MW/m$^2$, $v = 1.8$ m/s, and pH 6.0. The time is supposed to be 2 years (effective time of the reactor operation at the nominal power of 14 years of the FA in-reactor service). The oxide film thickness was found to be 10 $\mu$m except for the film that has been dissolved. It should be noted that in the pH range of 5.0–6.0 the oxide growth rate is very sensitive to pH. For pH of 6.2 the oxide film thickness was found to be 20 $\mu$m. The oxide film may increase during the storage, but the model does not give the significant oxide film growth with zero heat flux and flow rate for the film thickness $>10$ $\mu$m.

The corrosion depth was estimated using equation (2). Using interpolated value for the temperature of $74^\circ$C (0.0997 $\mu$m·h$^{1/2}$) for the FA with the longest time of operation the depth of uniform corrosion was found to be 13 $\mu$m (over the effective time of in-reactor operation). Then, using the corrosion constant of 0.062 $\mu$m·h$^{1/2}$ (20°C) for the time period of 23 years (12 years of reactor shutdowns and 11 years of storage in the spent fuel pool), over 25 years of in-reactor service and storage the total depth of uniform corrosion was found to be 31 $\mu$m. The estimated minimum cladding thickness (0.47 mm) is within the lower limits specified in the design documentation for the fuel elements IRT-3M in the initial state (0.35 mm).
Since we accept that the thickness of the oxide film on the cladding may increase to some extent over 11 years in a storage pool, the impact of this increase on the possibility of further FA in-reactor operation should be considered. The corrosion product accumulated on the surface of the cladding may lead to a fuel failure if it reaches a critical thickness [8]. According to [8], corrosion product thickness necessary to cause spallation strongly depends on the heat flux. Using the correlation presented in [8], it can be concluded that for the heat flux <0.5MW/m² the corrosion product thickness required to cause spallation is never reached. This confirms the fact that for low power research reactors oxide film spallation usually does not occur.

As a result, it can be expected that uniform corrosion of the cladding of IRT-3M fuel elements for the IRT MEPhI will not lead to a fuel failure in case of further FAs in-reactor operation.

The absence of a significant pitting corrosion was confirmed by the results of the fuel visual inspection and the analysis of water chemistry monitoring in the reactor pool and spent fuel storage presented further.

4.2. Fuel visual inspection
Visual inspection and leak testing with heating of all 16 FAs were carried out. Visual inspection of the external fuel tubes using underwater camera did not reveal significant pitting corrosion. The surfaces of the fuel elements are smooth (figure 1). No significant deposits, stains and mechanical damages was revealed. FAs have different shades of the surface (duller for the FAs of higher burnup). It should be noted that the shades depend on the photo quality (for example, the same FA is shown in figure 1(a) and figure 1(b). The colour is uniform over the height of FA. The surfaces of the internal fuel tubes cannot be examined.

4.3. Water chemistry control
Diligent monitoring and control of water chemistry is the most important factor affecting aluminum fuel cladding preservation, especially during long-term storage in water basin.

During reactor operation, continuous monitoring of water electrical conductivity is carried out. The conductivity is normally kept at the level < 1.5 μS/cm. Regular monitoring of chemical parameters of the water is performed. Water pH is maintained between 6 and 6.2.

In storage basin the same water chemistry parameters as in the reactor pool are maintained. The only difference is that monitoring of all parameters is carried out periodically (every week). Water purification procedure reduces water stagnation in basin.

IAEA recommends the following basin management procedures: "(1) use good quality water in spent fuel storage pools; (2) avoid the use of plain carbon steel as a material of construction of storage pools; (3) avoid the use of incompatible metals and alloys in the spent fuel storage pool; (4) prevent
dust and debris from falling on pool surfaces; (5) avoid any accumulation of sludge in the pool; (6) conduct a corrosion monitoring programme." [5].

IRT MEPhI storage basin meets the most of these recommendations. This allows for preserving fuel cladding during long-term storage in water basin.

4.4. Experience on repeated operation of FAs previously discharged from the core
In 1997–2002 because of the lack of fresh fuel assemblies a few spent FAs with burnup of 44–50% were loaded in the IRT MEPhI reactor core. The parameters of the repeated operation of FAs are presented in table 2.

| FA  | 1st unloading Date | Burnup, % | 2nd loading Date | Burnup, % | 2nd unloading Date | Burnup, % |
|-----|--------------------|-----------|------------------|-----------|--------------------|-----------|
| #133| 07.07.95           | 44.1      | 24.12.97         | 02.03.01  | 52.6               |
| #130| 01.03.96           | 46.1      | 04.05.00         | 30.05.03  | 54.9               |
| #128| 13.07.94           | 46.2      | 15.03.02         | 07.03.03  | 49.0               |
| #129| 13.07.94           | 49.7      | 15.03.02         | 07.03.03  | 52.5               |
| #132| 28.12.94           | 49.2      | 15.03.02         | 07.03.03  | 51.8               |
| #182| 30.04.99           | 50.1      | 15.03.02         | 07.03.03  | 52.1               |

It can be seen from table 2, that FAs #128 and #129 were operated during ~1 year after 7.7 years of wet storage. The increase of coolant activity was not observed during this year.

The set of experiments on the measurement of the power density in spent FAs IRT-3M and IRT-2M was performed at the IRT MEPhI in 2003–2007. Investigated FAs were subsequently placed into the core instead of one of beryllium blocks. The FAs were irradiated during one weekly operation cycle at the reactor power of 2.5 MW. Then the activity of the FAs was measured. Power density axial distribution was determined by γ-spectrometric method using radiation intensity of $^{140}$La ($E_{γ}$=1596.2 keV). Copper indicator was placed in the center of the investigated FAs for the measurement of thermal neutron flux. These experiments were focused on burnup measurement and were described in [9]. In present work we use these experiments to demonstrate that irradiation in the core of high burnup spent FAs did not lead to their cladding leakage.

Table 3 presents the operation history of the measured FAs. It can be seen from table 3, that even FAs stored over 9-21 years did not reveal cladding leakage during the irradiation in the core.

| FA  | FA type, enrichment | Date of the loading | Date of the unloading | Burnup. % | Date of the experiment | Time from the 1st loading | Time of storage |
|-----|--------------------|---------------------|-----------------------|-----------|-----------------------|--------------------------|----------------|
| #47 | IRT-2M, 90%        | 02.01.86            | 14.03.92              | 62        | 25.06.07              | 21.5                     | 15.3           |
| #02 | IRT-2M, 36%        | 03.12.84            | 07.03.86              | 32        | 20.02.07              | 22.2                     | 21.0           |
| #126| IRT-3M, 90%        | 11.05.87            | 31.12.93              | 52.5      | 08.04.03              | 15.9                     | 9.3            |
| #127| IRT-3M, 90%        | 11.05.87            | 03.03.94              | 42.7      | 20.05.03              | 16.0                     | 9.2            |
| #183| IRT-3M, 90%        | 11.03.91            | 28.04.00              | 51.1      | 29.10.03              | 12.6                     | 3.5            |

As a result, it was shown that there is a successful experience of high burnup FAs IRT-3M and IRT-2M operation after long-term wet storage.
5. Analysis for the OR reactor

5.1. Fuel swelling and corrosion effects estimation

Fuel swelling for the fuel element with maximum burnup was estimated using the model described in [10]. The fission product swelling is a linear function of burnup with coefficient based on the measured fuel swelling data. Using the most conservative correlation from [10], the fission product swelling for maximum $^{235}$U burnup of 0.086g (0.95%) was found to be 0.074%. The limiting swelling was estimated as 2% that corresponds to ductility limit of 1%. The time over that this limit could be reached is > 160 years (taking into account usual operation mode of the reactor).

To prolong the established life cycle of FAs, the conservative estimation of corrosion rate presented in [11] was used. According to [11], the rate of uniform corrosion of aluminum cladding of pin-type fuel elements is about 0.01 mm/year. Cladding thickness reduction was found to be 0.29 mm; and, accordingly, the cladding thickness was found to be 1.21 mm. The limiting cladding thickness was selected at 0.4 mm (slightly larger than the lower limit specified in the design documentation for the fuel elements IRT-3M in the initial state). The time over that this limit could be reached is ~110 years. Taking into account previous 29 years of operation and using reliability criterions, the life cycle of the OR fuel elements was extended to 16.4 years.

Additional corrosion effects estimation was performed using the models described in Section 3. Using equation (1), the oxide film thickness for the FA with maximum surface heat flux was estimated. The parameters of the FA operation were $T_{op} = 72^\circ$C, $q = 0.051$ MW/m$^2$, $v = 2.8$ m/s, and pH 6.0. The time is supposed to be 1325 hours (effective time of the reactor operation at the nominal power (0.3 MW) of 29 years of the FA in-reactor service). The oxide film thickness was found to be 7.7 μm except for the film that has been dissolved.

Using the corrosion constant of 0.098 μm·h$^{1/2}$ (72°C) in equation (2), the depth of uniform corrosion was found to be 4 μm (over the effective time of in-reactor operation). The fact that this is less than the calculated oxide film thickness can be attributed to a large error of the models in case of thin oxide layers and low corrosion depth. Using the corrosion constant of 0.062 μm·h$^{1/2}$ (20°C) for the time period of 29 years the depth of uniform corrosion was found to be 35 μm. Accordingly, the cladding thickness was found to be 1.48 mm. It can be seen that the cladding thickness prediction using the relation from [4] is far less conservative than that made using the corrosion rate of 0.01 mm/year, and it can be used for another life cycle extension in future.

5.2. Fuel visual inspection

Visual inspection of the fuel pins using underwater camera was carried out. 12 fuel elements from the hottest FAs (10 ps) and from random FA (2 ps) were selected for the inspection. Visual inspection of the fuel pins did not reveal significant corrosion defects, deposits, and mechanical damages. The colour of the surfaces is opaque and uniform.

5.3. Water chemistry control

The analysis of the reactor operation logs confirmed that water chemistry parameters over 29 years of operation were in the range established by national standard OST 10134-91.

6. Conclusions

As a result, the following conclusions can be made for both reactors based on pre-operation analysis: the results of corrosion effects estimation using the models based on empirical correlations showed that uniform corrosion will not lead to fuel cladding leakage; the fuel elements visual inspection and analysis of water chemistry control procedures enable to conclude that there is no significant pitting corrosion of the fuel cladding, and large fission product release to the coolant is not expected. The main procedure of cladding leakage revealing is coolant activity monitoring during the reactor operation at nominal power level. It should be noted that low-level leakage of fission products is not a...
severe accident and there is no constraints for the long-term FAs operation till their leakage will be revealed (failure criterion in FA technical specifications).

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