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Qualification program of research reactor fuels manufactured at IPEN–CNEN/SP

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Abstract

In the last years, IPEN–CNEN/SP has developed the project, technical specifications and the manufacturing of U_3O_8–Al and U_3Si_2–Al dispersion fuels. Non-destructive analysis techniques have been an important part of the qualification program of these fuels. Today, for utilization in the IEA-R1 research reactor core of IPEN, the U_3O_8–Al fuel is qualified up to a uranium density of 2.3 gU/cm^3 and the U_3Si_2–Al fuel up to a uranium density of 3.0 gU/cm^3. Also, analogous to the qualification process of U_3O_8–Al and U_3Si_2–Al fuels, an extended bibliography revision on the irradiation performance of U–Mo alloy dispersed in aluminum matrix (Al) was carried out to establish a set of parameters that could help in the definition of the technical specifications for manufacturing of this type of fuel at IPEN aiming its posterior utilization in the IEA-R1 reactor. The irradiation performance aspects were associated to the neutronic and thermal–hydraulics aspects in order to propose a new core configuration to the IEA-R1 research reactor using U–Mo dispersion fuels. Core configurations using U–10Mo–Al fuels with uranium densities ranging from 3 to 8 gU/cm^3 were analyzed with the computational programs CITATION and MTRCR-IEA-R1. Core configurations for fuels with uranium densities ranging from 3 to 5 gU/cm^3 showed to be adequate to be used in IEA-R1 reactor and would present a stable in-reactor performance even at high burnup.

Keywords: Research reactors; Dispersion fuels; Dispersion fuel qualification

1. Introduction

The IEA-R1 reactor of IPEN in Brazil is a pool type research reactor cooled and moderated by demineralized water and having beryllium and graphite as reflectors (IAEA-TECDOC-1508, 2006). In 1997, the reactor received the operating licensing for 5 MW. Since 1988, IPEN has been producing and qualifying its own LEU (19.9% of ^{235}U) MTR fuels. MTR fuel elements had been constructed with U_3O_8–Al dispersion fuel plates with densities of 1.9 (from 1988 to 1996) and 2.3 gU/cm^3 (from 1996 to 1999). Since September 1999, IPEN has been manufacturing U_3Si_2–Al dispersion fuel with uranium density of 3.0 gU/cm^3. Fuel performance evaluation and nuclear fuel qualification require a post-irradiation analysis. IPEN has no hot cells to provide destructive analysis of the irradiated nuclear fuel. As a consequence, non-destructive methods have been utilized to evaluate irradiation performance of the fuel elements. A complete fuel element is irradiated in the IEA-R1 core and the fuel element evaluation consists of two items: (i) monitoring the fuel performance during the IEA-R1 operation, concerning the following parameters: reactor power, time of operation, neutron flux at the position of each fuel assembly, burnup, inlet and outlet water temperatures in core, water pH, water conductivity, chloride content in water, and radiochemistry analysis of reactor water; and (ii) periodic underwater visual inspection of fuel assemblies and eventual sipping tests for fuel element suspect of leakage. Irradiated fuel elements have been visually inspected periodically by an underwater

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radiation-resistant camera inside the IEA-R1 reactor pool, to verify its integrity and its general plate surface conditions.

The IEA-R1 fuels follow rigorous technical specifications that were developed after a careful bibliography revision, comprising the world experience in the project, fabrication and fuel performance of dispersion fuels (Simões, 1993). Analogous to the development of U_3O_8–Al and U_3Si_2–Al fuels at IPEN, an extended bibliography revision was developed on the irradiation performance of U–Mo alloy dispersed in an aluminum matrix (Al) (Almeida, 2005). On hand of this revision, it was attempted to establish a set of parameters that could help in the definition of the technical specifications for fabrication of this type of fuel and its posterior utilization in the IEA-R1 research reactor. A set of IEA-R1 core configurations using U–10Mo–Al fuel, with uranium densities ranging from 3.0 to 8.0 gU/cm^3, were analyzed. Due to the higher density of the analyzed U–10Mo–Al fuels compared to the U_3O_8–Al and U_3Si_2–Al qualified fuels at IPEN, it would be possible to reduce the number of fuel elements in the IEA-R1 reactor core, which generated the necessity to review the neutronic and thermal–hydraulics reactor core projects. The core neutronic calculation was developed with the computer program CITATION (Fowler et al., 1971). The thermal–hydraulics analysis was developed with the computer program MTRCR-IEA-R1 (Umbehaun, 2000). The MTRCR-IEA-R1 program permits the calculation of the fuel thermal and hydraulics parameters of the reactor core. The analysis has been made for a reactor operating power of 5 MW.

2. Qualification of the MTR fuel elements manufactured at IPEN–CNEN/SP

In 1988, MTR fuel elements begun to be produced in IPEN–CNEN/SP and since September 1997, the IEA-R1 research reactor employs only fuel elements manufactured at IPEN–CNEN/SP.

The qualification of these fuel elements is made in-use, which means that it is based on their irradiation in the IEA-R1 research reactor followed by the use of non-destructive analysis techniques, mainly visual inspections performed regularly with a radiation-resistant underwater camera as well as sipping tests carried out eventually.

Fuel performance evaluation can be summarized by the fuel element average burnup at the end of its whole irradiation period in the reactor core.

Regarding the qualification in-use of MTR fuel elements manufactured at IPEN–CNEN/SP, by the end of July 2006, the highest average burnup achieved in the IEA-R1 research reactor for each type of LEU (19.9% enrichment) dispersion fuel already employed is presented in Table 1.

Table 1
Highest average burnup achieved in the IEA-R1 research reactor by MTR fuel elements manufactured at IPEN–CNEN/SP (end of July 2006)

Dispersion fuel	Uranium density [gU/cm^3]	Fuel element	Status	Average burnup [%]	
				Calculated	Measured
U_3O_8–Al	1.9	IEA-130	Spent	36.10[a]	(36.8 ± 5.1)[b]
U_3O_8–Al	2.3	IEA-166	Spent	40.50[c]	–
U_3Si_2–Al	3.0	IEA-169	In-use	41.03[c]	–

[a] Yamaguchi (1997).
[b] Terremoto et al. (2000).
[c] Declared by the reactor operators.

3. Bibliography revision of U–Mo dispersion fuels

Since the 80s of the last century, countries that detain the nuclear technology have concentrated efforts in studying U–Mo dispersion fuels. This kind of fuel can have uranium densities up to 8 gU/cm^3 and has been studied as a possible substitute fuel for U_3O_8–Al and U_3Si_2–Al fuels in research reactors with high power and high neutronic flux. The uranium density value of 8 gU/cm^3 represents U–10Mo fuel particle loadings of about 50 vol.% in the meat.

Early irradiation experiments with uranium alloys showed the possibility of acceptable irradiation behavior, if these alloys could be maintained in their cubic γ-U crystal structure (Hofman and Walters, 1994). Many experiments have demonstrated that centrifugally atomized U–Mo powder can retain this gamma uranium phase during fuel element fabrication and irradiation and can be compatible with the aluminum matrix, becoming the prime candidate for dispersion fuels for research reactors. A set of irradiation tests have been conducted around the world for this alloy. Fourteen different fuel compositions, including 12 metallic alloys, have been irradiated as part of five separate experiments for high-density dispersion fuel development in the Advanced Test Reactor (ATR) at the Idaho National Engineering & Environmental Laboratory (Hayes et al., 2003). The irradiation performance data obtained from these tests had led the US-RERTR program to narrow its focus toward the U–Mo binary alloy system as its primary candidate for use in a high-density dispersion fuel. In the experiments RERTR-1 and RERTR-2, the tested fuel plates were fabricated with fuel particles loadings of only 25–30 vol.% in the meat, giving meat-averaged uranium densities of $\sim$4 gU/cm^3. The particular focus of these experiments was to observe the phenomena of fuel–matrix interaction and the fuel particle swelling under irradiation. The fuel plate powers, and consequently temperatures, were maintained low. Fuel plates fabricated with the U–4Mo alloy showed poor behavior. The U–Mo alloys fabricated with at least 6 wt.% performed well up to 70% burnup. The RERTR-3 experiment was designed to test experimental fuel plates under irradiation conditions considered aggressive for research reactor fuels. Forty-seven miniature fuel plates were fabricated and irradiated to a nominal U-235 burnup level of 40%. Based on the results of the RERTR-1 and RERTR-2 experiments, the RERTR-3 experiment focused principally on the U–Mo binary alloy fuels with 6Mo–10Mo wt.%. In this experiment, the test fuels were fabricated with fuel particle loadings of over 50 vol.% in the meat, giving meat-averaged uranium densities of up to 8.5 gU/cm^3. Post-Irradiation Examination (PIE) of these fuel plates showed generally acceptable fuel performance. The fuel swelling was relatively low, with no tendency toward break-way behavior from microscopy. However, at the elevated fuel temperatures of this experiment, significant fuel–matrix interaction was observed. In fact, fuel–matrix interaction was so extensive that no matrix Al remained in the hot central portion of the fuel meat in some fuel plates. Nonetheless, acceptable fuel plate performance was achieved even in cases where all of the matrix Al phase was consumed. The experiments RERTR-4 and RERTR-5 were designed to test larger fuel plates irradiated to nominal U-235 burnup levels of 50% and 80%. These experiments continue to focus on the U–Mo binary alloys with 6Mo–10Mo wt.%. The test results of the RERTR-4 (Hoffman and Meyer, 2002) experiments indicated that the formation of the aluminide interaction phase appeared to be the only aspect of fuel behavior that is significantly affected by temperature. The irradiation behavior of the U–Mo fuel alloy itself was deemed athermal over the temperature range tested. The most important observation of the RERTR-4 experiment was stable, apparently athermal swelling of the U–Mo alloy particles with the presence of small uniformly distributed fission gas bubbles. No evidence of unstable, break-way, swelling, characteristic of other high-density fuels, has been found. The formation of a U–Mo/Al interaction phase will be significant, consuming practically all matrix aluminum at higher temperatures (150 °C). The fission induced swelling rate of this compound is, however, low and very stable. The interaction phase occupies a larger volume than its U–Mo and Al constituents and therefore, contributes to swelling. This contribution, however, is limited by the amount of Al matrix available. The main effect of the interaction product formation is the reduction of thermal conductivity of the meat (Hayes et al., 2002), which should be carefully assessed for a particular fuel design. As the interaction proceeds, a low-conductivity reaction-product phase builds up, with the corresponding depletion of high-conductivity Al matrix phase. This leads to a substantial degradation of fuel meat thermal conductivity with time, and fuel centerline temperatures can increase with burnup even plate power decreases. The U–10-Mo–Al dispersion fuel has been the most studied and has presented excellent performance under irradiation to nominal U-235 burnup level of 80% and with uranium densities ranging from 3 to 9 gU/cm^3. Therefore, based on the results above, this type of U–Mo dispersion was chosen to be analyzed for posterior utilization in the IEA-R1 reactor core.

4. Definition of a new core for the IEA-R1 research reactor using U–Mo dispersion fuel

For the definition of a new IEA-R1 research reactor core, neutronic and thermal–hydraulics calculations were developed for the U–10Mo–Al fuels with densities ranging from 3 to 8 gU/cm^3. The uranium density value of 8 gU/cm^3 was chosen because it represents U–Mo fuel particle loadings of about 50 vol.% in the meat, value normally considered as a limit to maintain the mechanical integrity of the fuel plate under irradiation. The uranium density value of 3 gU/cm^3 was chosen as the minimum value utilized, because it is the maximal meat uranium density qualified for the U_3Si_2–Al fuel fabricated at IPEN–CNEN/SP. Uranium densities smaller than 3 gU/cm^3 are not of interest. Nowadays, the IEA-R1 reactor core has a typical configuration with 24 elements, being 20 standard elements of U_3O_8–Al and U_3Si_2–Al dispersion fuels, four control elements, and one beryllium irradiation element in the central position of the core. In the neutronic calculation, the computer program CITATION was utilized for the three-dimensional core calculation and for burnup calculation. The radial and axial power density curves were utilized as input data for the thermal and hydraulics core analyses. The neutronic calculation results showed that the analyzed 3×3 core configuration (nine elements), with four standard fuel elements, four control elements, and one beryllium irradiation element in the central position of the core and with uranium densities ranging from 6 to 8 gU/cm^3 was very reactive and technically inadequate for the IEA-R1 reactor core at 5 MW. The analyses were concentrated in-reactor core configurations using eight standard fuel elements, four control elements and one beryllium irradiation element in the central position and with uranium densities ranging from 3 to 5 gU/cm^3. The beginning of life neutronic calculation showed that the cores with uranium densities of 4 and 5 gU/cm^3 presented high reactivity excess (1.1367 and 1.1604, respectively). In those cases, for uranium densities of 4 and 5 gU/cm^3, a new core configuration was defined having only 11 elements (six standard fuel elements, four control elements and one beryllium irradiation element in the central position of the core).

In the year 2000 a new thermal–hydraulics model, MTCR-IEA-R1, was finalized at IPEN–CNEN/SP through the commercial program Engineering Equation Solver (EES) (Umbehaun, 2000). The use of this computer model enables the steady-state thermal and hydraulics core analyses of research reactors with MTR fuel elements. The following parameters are calculated along the fuel element channels: fuel meat central temperature (T_c), cladding temperature (T_r), coolant temperature (T_f), the Onset of Nucleate Boiling (ONB) temperature (T_{onb}), the critical heat flux (Departure of Nucleate Boiling – DNB), flow instability and the thermal–hydraulics safety margins MDNBR and FIR. The thermal–hydraulics safety margins MDNBR and FIR are calculated as the ratio between, respectively, the critical heat flux and the heat flux for flow instability and the local heat flux in the fuel plate. Furthermore, the MTCR-IEA-R1 model also utilizes in its calculation the involved uncertainties in the thermal–hydraulics calculation as, for instance, fuel fabrication uncertainties, error in the power density distribution calculation, in the coolant flow distribution in the core, reactor power control deviation, in the coolant flow measures, and in the safety margins for the heat transfer coefficients. The calculated thermal and hydraulics core parameters are compared with the design limits established for MTR fuels: (1) cladding temperature < 95 °C; (2) safety margin for the Onset of Nucleate Boiling higher than 1.3, or the ONB temperature higher than coolant temperature; (3) safety margin for flow instability higher than 2.0; and (4) safety margin for critical heat flux higher than 2.0.

As mentioned in the bibliography revision (item 3), due to the fuel particle–aluminum matrix interaction, the fuel meat thermal conductivity reduces during irradiation. For studying the thermal behavior of the U–10Mo–Al fuel with the program MTRCR-IEA-R1, it was necessary to provide the meat fuel thermal conductivity as input data. Two values of thermal conductivity for the U–10Mo–Al meat were utilized: 70 W/m °C and 13 W/m °C. The value 70 W/m °C was utilized because it represents the meat thermal conductivity value for U–Mo fuel particle loadings of about 50 vol.% in the meat (Hayes et al., 2002). For fuel particle loadings smaller than 50 vol.%, the meat thermal conductivity is higher, bringing smaller fuel temperatures when compared with the temperatures calculated for the meat thermal conductivity value of 70 W/m °C. When the aluminum in the meat is completely consumed, with 100% of fuel particle–matrix interaction products, the meat thermal conductivity reaches values around 13 W/m °C (Hayes et al., 2002). The value 70 W/m °C would be more representative for reactor cores where the fuel temperatures would present values under 100 °C, or for cores in the beginning of life. The meat thermal conductivity value of 13 W/m °C would be more representative for cores with higher temperatures, where the fuel particle–matrix interaction is so extensive that no matrix Al remained in the hot central portion of the fuel meat. In the thermal–hydraulics analysis, the computer program MTRCR-IEA-R1 was utilized with the radial and axial power distribution curves provided for the computer program CITATION. The input data for the thermal–hydraulics simulations were obtained from Almeida (2005). The U–Mo fuel plate geometric dimensions used in the simulations were the same of those of U_3Si_2–Al fabricated at IPEN. The core with 13 elements and uranium density of 3 gU/cm^3 in the fuel plate was simulated first with the MTRCR-IEA-R1 (Simulation 1) for a meat thermal conductivity of 13 W/m °C, without uncertainties treatment involved (nominal condition) and with uncertainties treatment involved (Simulation 2). Afterwards the same core was simulated for a meat thermal conductivity of 70 W/m °C, in the nominal condition (Simulation 3) and with uncertainties treatment involved (Simulation 4). The results are presented in Table 2. The same sequence was utilized for the core simulations with 10 elements and uranium densities of, respectively, 4 and 5 gU/cm^3 (Simulations 5–8 and 9–12, Table 2).

5. Main results and conclusion

The simulation results described in Table 2 show that no design limit is achieved for the analyzed cores. The calculated cladding temperatures are under the value of 95 °C, reaching for the reactor cores with uranium density fuels of 5 gU/cm^3 (Simulations 10 and 12) the maximal value of 93.82 °C. This was expected because these analyzed cores had the highest meat uranium density in the fuel plates, which also achieved the highest calculated peak factor ($F_q = 2.1850$) in the axial power distribution calculation of the fuel element hot channel, when compared to the peak factors ($F_q = 2.076$ and $F_q = 2.118$, respectively) for the core configurations with meat uranium densities of 3 and 4 gU/cm^3 in the fuel plates. The temperatures in the simulations, without uncertainties treatment, are well below those obtained with uncertainties treatment. From Table 2 it can be seen that the coolant temperatures (T_f) for all simulations are below the ONB temperature, indicating one-phase flow in the simulated cores. The margins for critical heat flux (MDNBR) and flow instability (FIR) are well above the value 2.0, admitted as design limit. The maximal fuel meat central temperature was 145 °C. At this temperature the fuel particle–matrix interaction would be completed, reaching the meat thermal conductivity value of 13 W/cm °C. The interaction phase U–10Mo–Al would consume practically all of the matrix Al phase. However, as seen in item 3, for U–10Mo–Al dispersions the fuel behavior would be stable even at higher burnup.

These simulation results were utilized to propose the meat uranium density of 5 gU/cm^3 for the miniplates of U–Mo dispersion fuel to be fabricated at IPEN–CNEN/SP and tested in the IEA-R1 research reactor. With this uranium density in the fuel meat, the number of fuel elements used in the IEA-R1 research reactor would be reduced, bringing economic advantages and also reducing the number of spent fuel elements to be stored in the reactor pool.

Table 2
Simulations with the computer program MTRCR-IEA-R1 for reactor cores with 12 and 10 elements, meat uranium densities of 3, 4 and 5 gU/cm^3 and thermal conductivities of 13 W/m °C and 70 W/m °C

Simulation	T_f (°C)	T_r (°C)	T_c (°C)	T_{onb} (°C)	MDNBR	Flow instability (FIR)
1	47.73	69.05	93.37	120.5	7.73	24.06
2	52.95	86.58	127.3	122.3	4.26	14.37
3	47.73	69.05	74.86	120.5	7.73	24.06
4	52.95	86.58	96.36	122.3	4.26	14.37
5	47.79	69.57	98.81	121.1	7.13	29.54
6	53.05	87.43	136.3	123	3.92	17.64
7	47.79	69.57	76.6	121.1	7.13	29.54
8	53.05	87.43	99.19	123	3.92	17.64
9	47.86	73.68	104.3	121.3	6.41	29.53
10	53.16	93.82	145	123.2	3.63	17.64
11	47.86	73.68	81.04	121.3	6.41	29.53
12	53.16	93.82	106.1	123.2	3.52	17.64

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