

Qualification of advanced LEU fuels for high-power research reactor conversion designs

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Abstract. The two running EURATOM-funded projects EU-QUALIFY and EU-CONVERSION, coordinated by the HERACLES consortium, are developing and further qualifying advanced LEU fuel systems for conversion purposes of high-power research reactors (HPRRs). Three fuel systems have been identified as potential candidates to replace the use of highly-enriched uranium fuel, namely the uranium silicide dispersion fuel (U_3Si_2/Al), the U-10Mo monolithic fuel and the dispersed U-7Mo/Al fuel system. The U-Mo monolithic and the U_3Si_2/Al dispersion fuel systems are currently being advanced in technology readiness level, towards more representative generic test assemblies, mimicking more closely target reactor specific conditions. A number of irradiation experiments have so far been conducted on the two dispersion fuel systems and the first monolithic irradiation experiment at the Belgian BR2 reactor is targeted to commence during 2025. The post-irradiation examinations of the historical and ongoing tests will be concluded within the projects and the complementary HERACLES work programme. These projects are also important contributors in establishing industrial-level fabrication processes of high-loaded and high-density uranium silicide fuels as well as U-Mo monolithic fuels. The operational times of the current HPRRs are foreseen to be extended by their conversions and, thus, have an essential impact on the future European supply of reactor-produced medical radioisotopes.

1 Introduction

Europe is the world’s largest supplier, and among the world’s largest users, of medical radioisotopes. A secure supply of these isotopes is key to support a safe, high-quality and reliable use of radiological and nuclear technology in healthcare. As most medical radioisotopes are produced by the European HPRRs (high-power research reactors) and MPRRs (medium-power research reactors), these reactors play a major role for the time-critical supply chain of these radioisotopes, but also for fundamental and applied research using neutrons. To ensure the long-term operation of the European research reactors, the reliable supply of safe Low-enriched Uranium fuels (LEU), especially for the upcoming High-enriched Uranium (HEU) to LEU conversion, is crucial. As such, operators of European HPRR and the leading European experts for fuel development and fabrication – CEA (France), Framatome (France), ILL (France), SCK CEN (Belgium), and TUM (Germany) – united into the HERACLES consortium in

2015 with the aim to leverage existing know-how and synergies in this field.

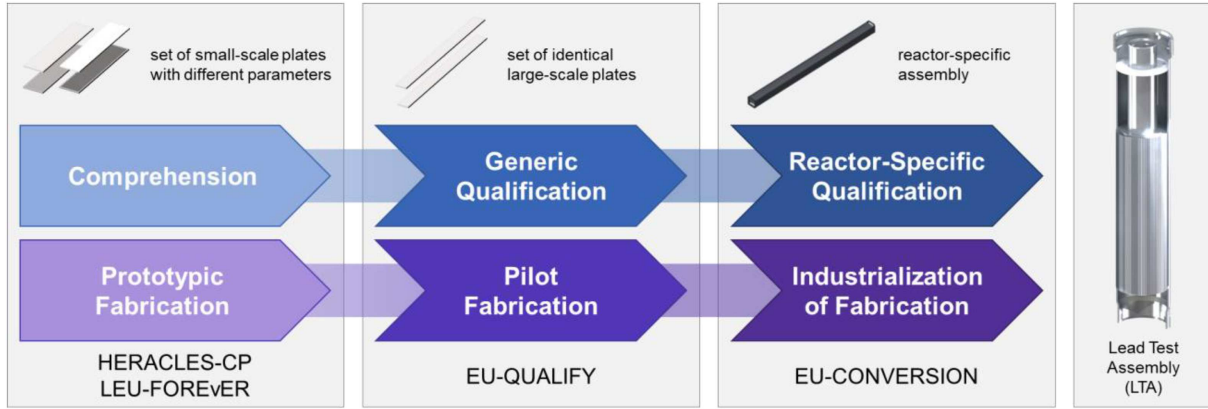
Two EURATOM-funded projects, coordinated by members of the HERACLES consortium, are currently being executed and target the qualification of such LEU research reactor fuels: EU-QUALIFY (2020–2025) and EU-CONVERSION (2024–2028). These projects were preceded by two earlier EURATOM-funded projects laying the groundwork for the high-loaded and high-density fuel systems. Table 1 summarizes these projects and the various participants.

Each of these projects are complementary and overlapping and contribute to the HERACLES fuel development and qualification plan, which is outlined in Figure 1 below. Via successive advancements in fuel development and fabrication, complemented by reactor irradiation tests of increasingly representative fuel plates, the Technology Readiness Level of the fuel systems is advanced. Ultimately, the successful deployment of first-of-a-kind fuel elements to the respective research reactors, also known as Lead Test Assemblies (LTA), concludes the fuel qualification process.

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Table 1. HERACLES projects since 2015, the ones marked in blue are currently ongoing.

Project	Time scale	Participants
EU-CONVERSION [1]	2024–2028	HERACLES + UGA + CVR + SURO + TECHNICATOME
EU-QUALIFY [2]	2020–2025	HERACLES + INP Grenoble
LEU-FOREvER [3,4]	2017–2022	HERACLES + CVR + NCBJ + TECHNICATOME + LGI
HERACLES CP [5,6]	2014–2020	HERACLES

**Fig. 1.** Overview of the HERACLES fuel development and qualification plan.

2 LEU fuel systems for HPRR conversion

In the late 1990's and beginning of the 2000's a 'classical LEU (4.8 gU/cm³) silicide fuel' [7], made it possible to convert the many MPRRs in Europe and worldwide. However, the achieved fuel loading was insufficient for direct application in any of the HPRRs. For these challenging reactors, the main route was to find a solution with another fuel type that had an intrinsically higher U loading, e.g. the U-Mo alloy. Since 2000, this alloy has been tested extensively as a dispersion type of fuel [8] and as a solid metallic fuel foil, called monolithic U-Mo. For monolithic U-Mo, the fuel dispersion in an aluminum matrix is exchanged with a solid fuel foil, leading to very high fuel loadings, unlocking LEU conversion solutions for even some of the most challenging HPRRs.

Up until 2015, the main focuses of the international LEU fuel development and qualification community were the U-Mo dispersion and monolithic types of fuel. The focus of the EU HPRRs was U-Mo dispersion, however, in 2015, it was determined to also look at other options using the previously qualified silicide fuel. These new options increase the amount of fuel in dispersion-type research reactor fuel plates by increasing the volumetric loading of the fuel material in the aluminum matrix, e.g. from 4.8 to 5.3 gU/cm³ for U₃Si₂, or by increasing the typical fuel volume by increasing the meat (core) thickness, or a combination of both.

This has led to the current situation where three novel fuel systems, namely U₃Si₂ fuel with increased density or loading, U-7Mo dispersion fuel (with 7 wt.% Mo), and U-10Mo monolithic fuel (with 10 wt.% Mo), require demonstration of acceptable fuel performance behavior

at high-power conditions through irradiation testing and post-irradiation examination (PIE) programs.

For a HPRR targeting conversion, the currently available fuel systems cover a wide range of possible uranium loadings. The volumetric loading ρ_U in gU/cm³ of a given fuel system can be easily calculated from the fuel material density ρ_{fuel} of U₃Si₂, U-7Mo and U-10Mo in gU/cm³, for dispersion fuels by multiplying with the respective volumetric percentage v of fuel particles in the aluminum matrix, and for monolithic by unity ($v = 100\%$):

$$\rho_U = \rho_{\text{fuel}} \cdot v.$$

For research reactor operators, the so-called surface loading ς_U is more relevant as it allows the determination of expected fissions per fuel plate surface area, an essential part of assessing the reactor performance using a new fuel. Due to the different fuel core thicknesses d_{core} in the fuel plate designs of different research reactors, this value is, contrary to the volumetric loading, not to be seen as generally valid for all reactors.

$$\varsigma_U = \rho_U \cdot d_{\text{core}}.$$

As an example for the Belgian BR2 reactor, following Figure 2 shows the experiments to be conducted in the EU-QUALIFY project and their respective volumetric and surface loadings.

2.1 U₃Si₂/Al dispersion fuel

The experience and fabrication status of the LEU 4.8 gU/cm³ U₃Si₂/Al fuel system is explained in greater

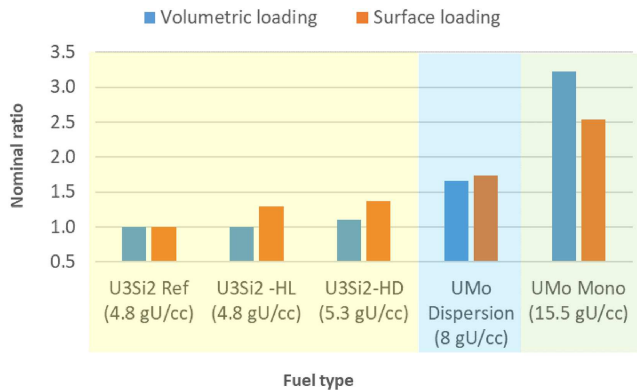


Fig. 2. Nominal differences between a 4.8 gU/cm^3 “reference” plate and the fuel systems represented in the EU-QUALIFY experiments. HL = High Loaded (thicker meat), HD = High Density (higher U loading).

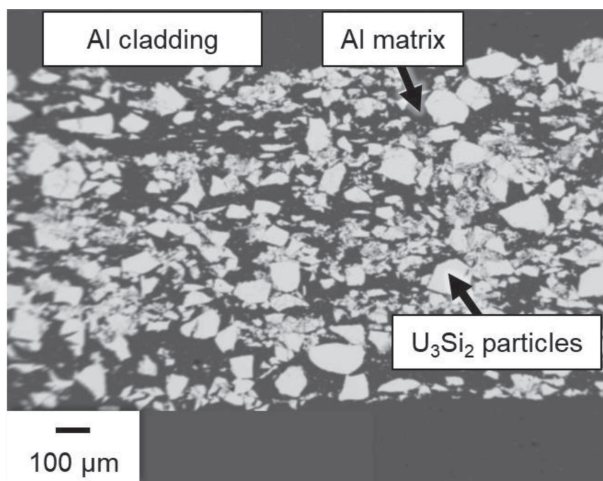


Fig. 3. Electron micrograph of a cross-section of a $\text{U}_3\text{Si}_2/\text{Al}$ dispersion fuel plate with 4.8 gU/cm^3 loading.

detail in [9]. This fuel type is widely used in research reactors throughout the world. It is a dispersion fuel core with U_3Si_2 fuel dispersed in a pure aluminum matrix, roll-bonded to aluminum-alloy cladding plates, as shown in Figure 3 above.

The silicide fuel at 4.8 gU/cm^3 loading is fully qualified for low- and medium-power research reactors. It has been manufactured by Framatome since the 1980’s, with a well-established manufacturing process, thus with high manufacturing and technology readiness level.

However, despite this extended knowledge, some challenges are remaining for the fabrication of a high loaded silicide fuel, either through the increase of volumetric or surface loading, e.g. from 4.8 up to 5.6 gU/cm^3 . Even the well-established 4.8 gU/cm^3 fuel used in the conversion of medium-power research reactors still need qualification for high-power conditions. This necessitates additional development regarding optimized fabrication processes, and actions for reducing cost and fabrication time [9–14]. This also requires that the optimizations are irradiation tested at high power conditions and the fuel system undergo

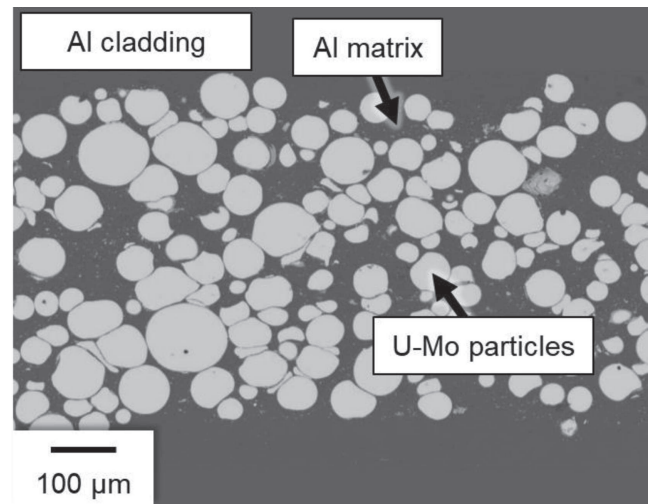


Fig. 4. Electron micrograph of a cross-section of a U-7Mo dispersion fuel plate with 8.0 gU/cm^3 loading.

fuel performance assessments in post-irradiation examinations.

2.2 U-7Mo dispersion fuel

The U-Mo dispersion fuel system, with 7 wt.% Mo, has a high intrinsic uranium density of about 16.0 gU/cm^3 . The actual volumetric fuel loading is approximately half of this (8.0 gU/cm^3) due to the fabrication limit of 50% volumetric percentage of fuel particles in the aluminum matrix. Figure 4 shows a representative cross-section of this fuel system.

The U-Mo dispersion fuel behavior has been studied in various irradiation experiments in the BR2 reactor such as FUTURE, IRIS, E-FUTURE, and SELENIUM [15–18]. The most recent experiment, conducted in both the HERACLES-CP and LEU-FOREvER projects, was SEMPER FIDELIS [19]. Following detailed PIE, this led to a down-select of the detailed fuel system which is to be tested in the E-FUTURE-III experiment, currently planned for irradiation in mid-2025. This detailed fuel system consists of atomized spherical U-7Mo fuel particles, which are heat-treated and coated with $\sim 1 \mu\text{m}$ of ZrN as diffusion barrier, in a pure aluminum matrix. The U-Mo dispersion fuel is currently not a primary choice for conversion for the European HRRRs. The general qualification and PIE reporting of this fuel system will nevertheless be finalized within the HERACLES consortium’s own work programme, facilitating the use of this system for other reactors in the future, if needed.

2.3 U-10Mo monolithic fuel

The U-Mo monolithic fuel system with 10 wt.% Mo, has a high intrinsic uranium density of about 15.5 gU/cm^3

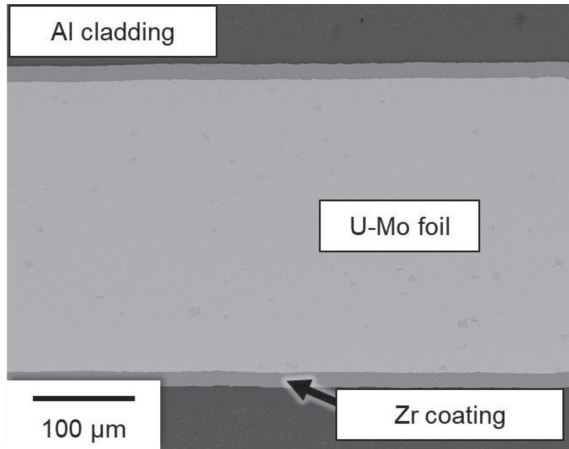


Fig. 5. Electron micrograph of a cross-section of a U-10Mo monolithic fuel plate with 15.5 gU/cm^3 loading.

equal to the volumetric fuel loading due to the complete omission of a matrix material. Instead, a solid metallic foil of U-10Mo with a thin Zr coating, serving as diffusion barrier, is placed directly between the aluminum-alloy cladding plates, as illustrated in Figure 5 above.

As such, it is currently the only fuel system that delivers enough uranium density for the most challenging HPRRs, e.g. FRM II in Germany and the ATR reactor in the United States (U.S.). The U.S. Department of Energy's High-Performance Research Reactor (USHPRR) Conversion Project has investigated LEU fuels since 1978, and due to the high uranium density and good irradiation behavior, U-10Mo was chosen as the most promising LEU fuel for the conversion of all USHPRRs except HFIR [18, 20–27]. The main challenge of this fuel system is related to the largely different fabrication process compared to dispersion-type fuels. The monolithic plate fabrication technique developed at Framatome-CERCA for the European irradiation experiments is the proprietary so-called C2TWP method [28,29], which has been developed during the HERACLES-coordinated EURATOM projects. The FUTURE-MONO-1 experiment to be started in 2025 will be the first European irradiation test of this fuel type.

3 Demonstrating fuel performance

The conversions of HPRRs from HEU to LEU are very complex in nature, and the complexity also varies greatly between the different reactors. In addition, as is the case with nearly all testing conducted at nuclear facilities, the irradiation testing and subsequent PIE are both time consuming and very costly.

The typical progression in a fuel system qualification, building upon the knowledge base from the previous experiments and minimizing the risk of fission product release, starts with irradiations of mini-scale plates, followed by full-scale plate irradiations starting from moderate performance conditions towards more demanding conditions, e.g. from a typical peak heat flux range of $250\text{--}350 \text{ W/cm}^2$ (for low and medium power reac-

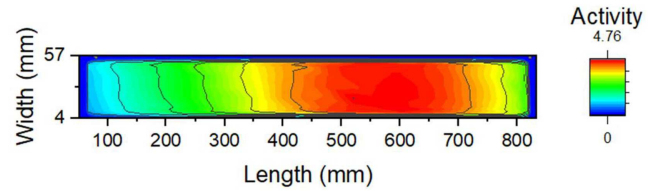


Fig. 6. Example of MCNP estimated Cs-137 activity at End-Of-Cycle (EOC), generated for ‘burnup’ comparison between calculations and post-irradiation measurements.

tors) towards $>500 \text{ W/cm}^2$ and peak U-235 burnup from 40–50% towards $>70\%$. In addition to increasing the irradiation conditions in a step-wise fashion, the size and nature of the physical components and total inventory of fuel is also increased at a reasonable rate to manage risk; e.g. usually flat plate irradiations are then followed by formed plates in baskets or assemblies irradiated at more reactor-specific conditions. In parallel, the fabrication processes are subsequently matured from lab-scale fabrication demonstrations, over pilot fabrication lines towards the full industrialization of the novel techniques as the ultimate goal.

In the end of a fuel qualification process, the preferred fuel system should have sufficient data to demonstrate:

1. fabricability of prototypes (LTAs) and understanding of scale-up issues for relevant throughput
→ fabrication demonstrations that have been irradiation tested and confirmed in PIE
2. acceptable fuel performance at relevant or bounding reactor-specific requirements (including reactor-specific safety requirements)
→ e.g. peak values of fuel/cladding temperature, U-235 burnup, heat flux, volumetric power, etc.
→ Stable, predictable, and safe fuel swelling, plate oxidation, etc.
3. Be affordable, so that the reactor can sustainably obtain and dispose of the fuel.

3.1 Neutronic calculations

A main tool for the design and analysis of an irradiation test is neutronic calculations, ensuring that the planned experiment conditions are obtainable, and reporting the irradiation experiment results (as-is). At SCK CEN, the MCNP software is used. MCNP utilizes the Monte-Carlo method and is used for both pre- and post-irradiation calculations. The typical test condition variables for the tests are:

- peak and average heat Flux (W/cm^2), based on test ‘targets’ (design) or ‘acquired’ (post-irradiation).
- Peak and average U-235 burnup (%), target and acquired.
- Peak and average fission density (e.g. fissions/ cm^3 fuel), target and acquired.

As an example of an MCNP output, Figure 6 shows the distribution of Cs-137 activity (10^9 Bq/cm^2). The Cs-137

Table 2. TRL descriptions for HPRR fuel systems.

TRL	Equivalent description for research reactor fuels
TRL-4	Lab scale fabrication demonstration
TRL-5	Mini-scale fuel plates successfully irradiated
TRL-6	Full-scale fuel plates successfully irradiated in relevant conditions
TRL-7	Successful irradiation of a representative fuel assembly, e.g. a GTA ⁽¹⁾
TRL-8	Successful LTA ⁽²⁾ /first of a kind of specific fuel assembly + approved for use in specific research reactor
TRL-9	Operational use of fuel assemblies in a reactor

⁽¹⁾Generic Test Assembly (GTA); an experimental device used at the BR2 reactor, mimicking the plate and assembly design as well as target irradiation conditions of a ‘client’ HPRR assembly. ⁽²⁾Lead Test Assembly (LTA); a full scale reactor specific fuel assembly.

Table 3. Summary of current TRLs, status, and next development steps for the envisaged fuel systems.

Fuel system	Current estimated TRL (in EU)	Current status	Next steps in EU development
U ₃ Si ₂ fuel with 4.8 gU/cm ³	TRL-9	Readily used in research reactors	Improvements in fabrication yield for increased loading
U ₃ Si ₂ fuel with increased density (or loading)	TRL-7	Representative fuel assemblies tested in relevant irradiation conditions, validation ongoing	Expand window of experience via PIE of MEs ⁽³⁾ and GTA test. Increase applicability for different HPRR conditions.
U-7Mo dispersion fuel	TRL-5	Full-scale plate tested in relevant irradiation conditions, with different improved design, validation pending	Advancement to TRL-6 via E-FUTURE-III test
U-10Mo monolithic fuel	TRL-5	Mini-scale plates successfully irradiated	Advancement to TRL-6 via FUTURE-MONO-1 test and to TRL-7 via GTA-MONO test

⁽³⁾Mixed Element (ME), modified BR2 driver fuel element, outermost HEU plates replaced with LEU.

activity is used as a measurand (or monitor) for burnup comparisons between MCNP calculations and gamma spectrometry measurements.

nup analysis reports, considered to be the most accurate measure of burnup.

3.2 Post-irradiation examinations

Post-irradiation examinations (PIE) are conducted in two phases: non-destructive examinations (NDE) and destructive examinations (DE). The NDE is performed to visually identify any abnormal conditions on the outside of the irradiated fuel plate, quantify the fuel plate thickness, oxide layer thickness and estimate the fuel meat swelling, quantify the burn-up using gamma spectrometry, and verify the comparability to post-irradiation neutronic calculations. The DE aims to evaluate the post-irradiation fuel state, composition, fuel swelling, interaction layers, oxide growth, inter-diffusion, impurities, and other relevant observations at the macro- and microscopic levels and to further confirm the neutronic calculations.

Recent advancements in NDE and DE techniques in the ongoing PIE campaigns have established new tools for burnup estimation, Gamma spectrometry and Electron Probe Micro Analysis [30,31]. Both tools are based on the analysis of a large number of radiochemical bur-

4 Technology readiness levels

It is important to note that the different fuel systems, fuel assembly designs, and production methods are all situated at different technological readiness levels (TRLs). Further, the fuel systems, taking into account their current TRL and their recent testing experiences, have different expectations in their rate of advancement in their readiness levels. In Holmstrom and Wight [32], the TRLs for research reactor fuels are generally defined based on the European Commission TRL definitions [33]. In Table 2, the estimated TRLs are given for the fuel systems currently under development (Tab. 3).

5 Conclusions future work

Within the ongoing EURATOM-funded projects, supported by HERACLES consortium internal actions, a sound technological ground has been made for converting HPRRs in Europe. All of the potential fuel systems

require further demonstrations to increase their technology readiness levels, but great advances have already been made and the scope is nearly fully planned in the current EU projects. It is envisaged that the dispersion silicide fuel system and the U-Mo monolithic fuel system are in a position to conduct assembly-type tests tailored to reactor-specific design requests [34]. These irradiations, named GTA-SILICIDE and GTA-MONO are an integral part of the recently started EU-CONVERSION project.

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Conflicts of interest

The authors declare that they have no competing interests to report.

Data availability statement

The fabrication and irradiation experiment specific data of the ongoing projects are currently confidential within the project consortium.

Author contribution statement

The SCK CEN authors have collated, analyzed and reported the sections on general fuel performance of the U_3Si_2 and dispersed UMo fuel systems as well as the neutronics and post irradiation analysis. TUM has provided the state-of-the-art for the European development of the UMo monolithic fuel system. The paper structure, conclusions and determination of the current technological readiness levels have been done as a combined effort.

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