The development of high density, low enriched fuel for the conversion of research reactors: a historical overview.

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Abstract. In the framework of the joint worldwide efforts to reduce the risk of proliferation by minimizing the use of highly enriched uranium in the civil nuclear fuel cycle, an extensive development and qualification program has been undertaken to develop and qualify research reactor fuel systems with reduced enrichment (< 20 %), that should allow to convert research reactors all over the world without significant loss of performance and significant increase of cost. This paper gives an historical overview on the efforts that have been deployed to this effect up to the present time.

Key Words: Research Reactor, Conversion, Reduced Enrichment

1. The origins of the Conversion Program

Research Reactor fuels typically use a dispersion of intermetallic fuel material in a matrix, obtained by blending enriched uranium-aluminide UAl_X powder with Al powder. Research reactors operate at low temperatures (coolant below 100 °C) but the operating conditions are demanding in other ways: they operate at high power densities and the burnup at discharge is is also very high; so these fuel must be able to withstand structural damage from fission and accommodate an important amount of fission products.

Highly-enriched uranium (HEU) allows more compact cores, with higher neutron flux densities and also extended cycle length. Therefore up to the late 1970's most research reactors used this 90-93 $\%^{235}$U enriched fuel.

From the early 1970's on security concerns have grown, especially since many research reactors are located at universities or other civilian locations where security provisions are not as stringent than in military weapons establishments.
The question of enrichment was a major focus of the UN-sponsored International Nuclear Fuel Cycle Evaluation (INFCE) in the period 1977-79. It concluded that to prevent weapons proliferation from the HEU fuels then commonly used in research reactors, enrichment should be reduced to no more than 20 $\%^{235}$U.

This was followed by a similar initiative by the USA in 1978 when the 'Reduced Enrichment for Research and Test Reactors' (RERTR) program was established to address a concern about the proliferation of HEU in civil commerce. Its goal was to enable research and test reactors to convert to, or back to, low-enriched uranium (LEU; < 20 $\%^{235}$U) by developing higher-density fuels that could accommodate the increase in ^{238}U and that could be used without significant performance loss or cost increase.

The Soviet Union made similar efforts from 1978 on and produced fuel of 2.5 gU_{tot}/cm^3 with enrichment reduced from 90 to 36 %. It largely stopped exports of 90 % enriched fuel in the 1980's to Russian-built research reactors in other countries.

2. Initial LEU Fuel Development: high-density dispersion fuels

The dispersion fuels commonly used in the early 80's were UAl_x and U_3O_8. However the highest uranium densities attainable with these fuels due to fabrication constraints (2.3 and 3.2 gU_{tot}/cm3 respectively) are insufficient to convert most research reactors. Because of the previous ORNL experience, UO2 was not considered.
Therefore the U.S. RERTR program started the LEU development program with the densest uranium silicide phase (U_3Si) dispersed in aluminium, with a density goal of 7.0 gU_{tot}/cm^3. Concurrently a few samples based on the next-lower density silicide phase, U_3Si_2, with a density of 4.8 gU_{tot}/cm^3 were also tested.
The backup fuel system, in case the uranium silicides would not work, was uranium-molybdenum alloy (bulk γ-phase UMo) clad in Zircaloy; however, development of UMo fuel was not pursued at this stage of the LEU fuel development.

Progressively a strong international cooperation developed to carry out the development of the silicide dispersion fuels.
The first irradiation tests showed that the main silicide fuels being tested for use in plate-type fuel elements, U_3Si-Al and U_3SiAl-Al, were affected by the growth of fission gas bubbles in the fuel particles at burn-ups of 85 $\%^5U$ in LEU.
On the other hand the U_3Si_2-Al fuel samples remained extremely stable at a burnup of at least 90 $\%^5U$; the fuel particles showed a uniform distribution of submicron-sized bubbles with no evidence of bubble linkage.

Therefore the U.S. RERTR program redirected the focus on the U_3Si_2-Al fuel system. The development of this fuel continued rather straightforward, and already in 1987, a qualification report was submitted to the U.S. Nuclear Regulatory Commission (NRC). In July 1988, the NRC gave a generic approval for use of U_3Si_2-Al dispersion fuel at a density of 4.8 gU_{tot}/cm^3 in its licensed non-power (research, training and test) reactors under operating conditions within the envelope of the qualification irradiation test conditions (NUREG-1313).
This fuel was then promoted worldwide and many research reactors, with powers up to 50 MW, have been converted since 1988 using dispersed U_3Si_2-Al fuel and this process still continues.

Meanwhile, Atomic Energy of Canada Ltd. (AECL) had also begun a program to develop U_3SiAl-Al and U_3Si-Al dispersion fuels for their research reactors, which use pin-type fuel elements. They succeeded to qualify these fuels because the restraint imposed by the cylindrical cladding prevents a significant growth of the fission gas bubbles so that the bubbles did not interconnect and breakaway swelling did not occur.
Later, AECL also qualified U3Si2-Al fuel in MAPLE-type pins.

A problematic aspect of the silicide fuels concerns the reprocessing, due to the formation of silica gel during dissolution of spent fuel. However around this time the U.S. decided not to use reprocessing as a back-end solution and in 1996 the UKAEA announced that it would stop all reprocessing activities. Consequently the reactors that had by then converted to silicide fuel no longer had a back-end solution for their fuel cycle. This situation has changed in 2014 when AREVA-NC announced that they will be in position to accept silicide fuel for reprocessing in the near future.

3. Development of high density LEU fuels for high performance reactors

Research reactors with high-power-density cores (usually called "high performance research reactors", HPRR) require higher density fuels than what can be manufactured with U_3Si_2. So in the 1990's the world's fuel developers community turned their attention on the development of very high density fuels.

High-uranium-content alloys, such as UMo and UNbZr with uranium contents of the order of 90 wt% had to be considered as dispersants for very-high-density dispersion fuels. Since tests of fast reactor fuels had shown favourable behaviour under irradiation of γ-phase fuel, at least at low burnup, the U.S. RERTR program decided to concentrate on γ-phase UMo as a promising candidate for a very high density fuel and started their efforts around 1996.

Despite the existing historical knowledge on UMo, the qualification of that fuel could not be performed straight away because of the challenging environment of HPRR's regarding density, heat load and burn-up. To address these issues a comprehensive fundamental research program was established to guide the development of UMo into a qualified RR fuel (cfr. the RERTR irradiation test series in ATR).

The initial development of UMo dispersions was focused on finding the lower limit of the Mo addition. The lower the Mo content of the alloy, the higher the U content and the better the neutronic balance: lower parasitic neutron absorption and higher reactivity.

On the other hand the chemical behaviour (reaction with the matrix aluminium) and the stability under irradiation of the alloy decreases with decreasing Mo content.

The initial irradiation tests of UxMo dispersion fuels (x ranging from 4 to 10), showed that the fuel appeared to perform satisfactorily (stabilize the γ-phase) if $x \geq 6$. The effect of γ-phase decomposition into α-phase during fabrication was also tested, and it was found that irradiation caused the α-phase material to convert back to γ-phase material.

It was however experienced that there was considerably more interaction between the fuel and the matrix aluminium than there had been for the silicide fuel. But at the rather low fuel temperature of the first tests, the amount of reaction product was not excessive, leaving an adequate amount of matrix material for good heat transfer at the end of irradiation.

Encouraged on the positive of the first tests with dispersed UMo fuel, the U.S. RERTR program irradiated a few mini-plates containing small foils of metallic UMo clad by roll bonding in Al 6061 cladding. These plates also performed very well, with little interaction between the UMo and the cladding and retention of the fission gases within the UMo foil.

Based on these encouraging results, the RERTR program began serious investigation into the manufacturing techniques for so-called 'monolithic' UMo fuel plates, which allows to reach even higher densities (16 gU_{tot}/cm^3) and convert all research reactors in the world.

The Russian program was also testing low-enriched U9Mo dispersions with a loading of 5.4 gU_{tot}/cm^3 in fuel tubes with diameters large enough that they would act essentially like flat plates with respect to restraining fuel meat swelling.

4. Further development of UMo based fuels for high performance reactors

The European HPRR's have participated in the development of the UMo fuel from the very beginning, with a large number of irradiation tests in the OSIRIS and BR2 reactors. Details on these irradiations are noted in Table 1 and will be discussed in the following.
Besides the physical fuel characteristics like matrix composition, the UMo grain shape and irradiation conditions, fabrication specifics like different heat treatment procedures and other preparation aspects are also important for the behaviour of the fuel under irradiation.

Name	**Year of irr.**	**Powder**	**Matrix materials**	**TC [°C]**	**Heatflux [W/ cm²]**	**Burn-up [% 235U]**	**Res.**
IRIS 1	1999	8g/cc gr. 20%	Pure Al	75	140	67%	OK
IRIS 2	2001	8g/cc at. 20%	Pure Al	93	240	40%	Stop
FUTURE-UMo	2002	8g/cc at. 20%	Pure Al	130	350	33%	Stop
IRIS 3	2005	8g/cc at. 20%	Al+2% Si	83	200	60%	OK
IRIS 4	2008	8g/cc at. 20%	Al+2% Si	100	270	55%	OK
IRIS-TUM	2008	8g/cc gr. 50%	Al+2% Si	98	260	88%	OK
E-FUTURE	2010	8g/cc at. 20%	Al+4 & 6% Si	130	470	71%	OK *
E-FUTURE II	2011	8g/cr at. 20%	Al+7&12% Al-Si alloys	130	470	30% 54%	Stop Fail
SELENIUM	2012	8 g/cc at. 20% coat. Si / ZrN	Pure Al	130	470	70%	OK

*The irradiation was performed until the foreseen end, but the plates were found to have pillowed.

Table 1: Overview of dispersed UMo irradiations in Europe. Burn-up is LEU equivalent.

The French UMo program had started with the irradiation of U7Mo full-sized plates in a moderate power density test (IRIS 1; 140 W/cm²) in the OSIRIS reactor and showed good results. The French program therefore continued its irradiation program with a second irradiation in the IRIS series in OSIRIS and an irradiation in the BR2, called FUTURE-UMo.
Power-density conditions in IRIS-2 and FUTURE-UMo were higher (respectively 230 and 350 W/cm²) than in IRIS-1.

These irradiation experiments revealed a highly undesirable behaviour of UMo/Al fuel systems under high fission rates (FR), which had not been observed in the historical tests for fast reactors with the higher temperatures, lower heat loads and the lower burn-ups. One or more plates showed unacceptable behaviour in each of these experiments: in FUTURE-UMo, an abnormal thickness increase in the plates at the hot spot was detected after the second irradiation cycle (33 % peak burn-up) and the same thing had happened in IRIS-2 by the end of the fourth irradiation cycle (40 % peak burn-up).

Destructive PIE’s showed that large, crescent shaped fission gas bubbles had formed at the interface of the aluminium matrix and an 'interaction' layer formed around the fuel kernels, eventually causing breakaway swelling of the fuel meat. The unreacted UMo particles, however, had behaved in a very stable manner. Fission gas was contained within the UMo lattice structure in a nanobubble superlattice or in small, non-interconnecting bubbles along grain boundaries.
This excessive local swelling of the UMo-Al dispersion plates, described as 'pillowing', was observed in the high power zones of the plates. Extensive PIE work showed that this was the consequence of excessive interaction between the UMo kernels and their surrounding matrix, leading to the formation of an amorphous interaction phase. It appeared that the interaction

layer (IL) is amorphous during irradiation, thereby significantly increasing the fission gas mobility in the interaction material and decreasing the viscosity of the interaction product. Because an important amount of interaction product is formed under high-power-density (high-temperature) conditions and because this IL growth accumulates on the interface with the Al matrix all fission products originally ejected from the UMo kernel into the matrix, this interface is mechanically weakened and the stresses developed due to swelling eventually lead to breakaway swelling. It was concluded that in order to minimize this phenomenon, the growth of the interaction layer has to be controlled and minimized.

Similar failure of the UMo-Al dispersion fuel system were also observed in the Russian tubes irradiated to ~ 60 %^{5}U burnup, even though the fuel loading was considerably lower. Re-examination of several RERTR miniplates irradiated under high-power-density conditions revealed small areas where the same phenomenon appeared to be happening.
The U.S. and French programs therefore decided to postpone full-sized fuel element irradiations and to concentrate on fully understanding and solving the problem.
In addition, the U.S. program decided to accelerate its development of UMo monolithic fuel.

Since the interaction is a diffusion process, the inter-diffusion characteristics can be influenced by alloying the two materials of the diffusion couple (UMo and Al) with third elements. On basis of historical experience, the addition of Si to the dispersion fuel matrix was put forward as a way to mitigate the IL formation.
As a result, miniplates containing amounts of Si ranging from 0.3 wt% to ~5 wt% were irradiated in the RERTR-6 experiment in the ATR reactor, and full-sized plates using an Al-2w%Si matrix were irradiated in the IRIS-3 experiment in OSIRIS.

After the promising results of the IRIS 3 irradiation with 2 w%Si, it was deduced by modelling extrapolations that 4-6 w%Si added to the matrix should suffice to limit the swelling of the plates for heat fluxes up to 470 W/cm². These are the heat fluxes levels representative for the actual conditions in RHF (ILL), BR2 (SCK•CEN) and RJH (CEA) and are therefore the ones required to be tested in the qualification of the fuel.

In the same period (around 2008), the IRIS-TUM plates were irradiated, testing also the addition of Si (2.1 w%Si) to the matrix, but with ground powders as in IRIS-1 instead of atomised powders. The IRIS-TUM campaign turned out to be rather successful, with no break-away swelling observed until the very high final burn-up of up to 88 %^{5}U LEU equivalent. However the observed swelling still was considerable and the heat flux was not representative for HPRR conditions. Furthermore, production of ground UMo fuel on an industrial scale is not available. Nevertheless these experiments showed that the addition of Si to the matrix can significantly slow down the swelling of the fuel plates.

5. Intensified collaboration

In order to optimize the efforts, an EU consortium called LEONIDAS was formed with a strong collaborative link with the US programme.
Increased Si content in the matrix was tested in a high power irradiation, called E-FUTURE, which was performed in the BR-2 reactor, using U-7w%Mo atomised kernels dispersed in Al-4w%Si and Al-6w%Si matrices.
E-FUTURE showed the desired effect for burn-ups up to 60 %^{235}U. However pillowing was once again observed for burn-ups larger than 65 %^{5}U, with smaller pillow size for the higher

Si content of 6 %. This was certainly an important improvement over the burnup threshold of 33 %^{5}U reached with FUTURE-UMo, the more as the power at beginning-of-life (BOL) in E-FUTURE (470 W/cm^2) was well above the power at BOL in FUTURE-UMo (350 W/cm^2).

It was then considered that further development of the UMo dispersion fuel would require more elaborate solutions, unless a further increase of the Si content in the matrix and improved dispersion of the Si particles could resolve the problem.
The latter solution was tried in the E-FUTURE-II experiment, while the former pathway gave rise to a number of modifications that were introduced in the fuel system. The main focus of these ideas has been to modify the UMo-matrix interface, where the IL is formed, and to assure that sufficient matrix material would be left at high burnup.

The E-FUTURE II campaign was designed to test the effect of a further increase the Si content of the matrix. The Si was introduced as finer dispersion in order to be more efficient for the reduction of the IL formation. The selected concentrations of 7 wt%Si as lower boundary and 12 wt%Si as Al-Si eutectic were beyond what had been tested so far as plate-type fuel. However, 12 w%Si had already been tested successfully in Russia in pin-type geometry and had revealed significant suppression of the IL.
But the E-FUTURE II campaign had to be interrupted earlier than foreseen: it was observed that the plates were buckled rather than swollen; this may indicate a different failure mode. It can be speculated that with higher Si concentrations, the hardness and brittleness of the Al-Si matrix had increased and that this effect contributed to the failure of this campaign.

Two other irradiations have been performed to test other parameters than the silicon content in the matrix.
CEA launched the IRIS-4 irradiation using pre-oxidised UMo kernels (obtained by thermally annealing the atomised powder in an oxygen containing atmosphere). The in-pile behaviour of the IRIS-4 plates turned out to differ relatively little from the IRIS-3 plates. On a microstructural level, the IL formation did not appear to be disturbed or delayed by the presence of the oxide layer.
SCK•CEN put forward sputter coated UMo kernels in the SELENIUM project in the BR2 reactor. The atomized UMo powder used for this campaign was directly coated with diffusion barriers, Si or ZrN. thereby preventing the diffusion between UMo and the Al matrix and the build-up of the IL. This was also an attempt to avoid Si use altogether.
SELENIUM completed the foreseen irradiation cycles, accumulating a 70 %^{5}U burn-up without showing anomalous swelling. This experiment showed that the use of a diffusion barrier coating such as ZrN provides superior protection against interaction compared to the addition of Si to the matrix. However, the formation of an IL is still observed at the high fission rate (FR) locations. This is believed to be the consequence of a break-up of the coating. Whether this happened early in the irradiation under high FR conditions or later as the UMo swelling causes the coating to break up, is currently being analysed.

SELENIUM showed that it is possible to separate the mechanisms for failure due to the UMo-matrix IL formation on the one hand and the UMo fuel kernel swelling itself on the other hand: the IL formation is a power, i.e. fission rate dependent effect that generates little swelling, while the UMo kernel swelling is a phenomenon dependent on burn-up / fission density, with no direct effect on IL formation. The two effects need to be mitigated.
Other irradiation experiments testing different means of introducing the silicon or different coatings of the UMo particles have also been performed in the US, Russia and Korea.

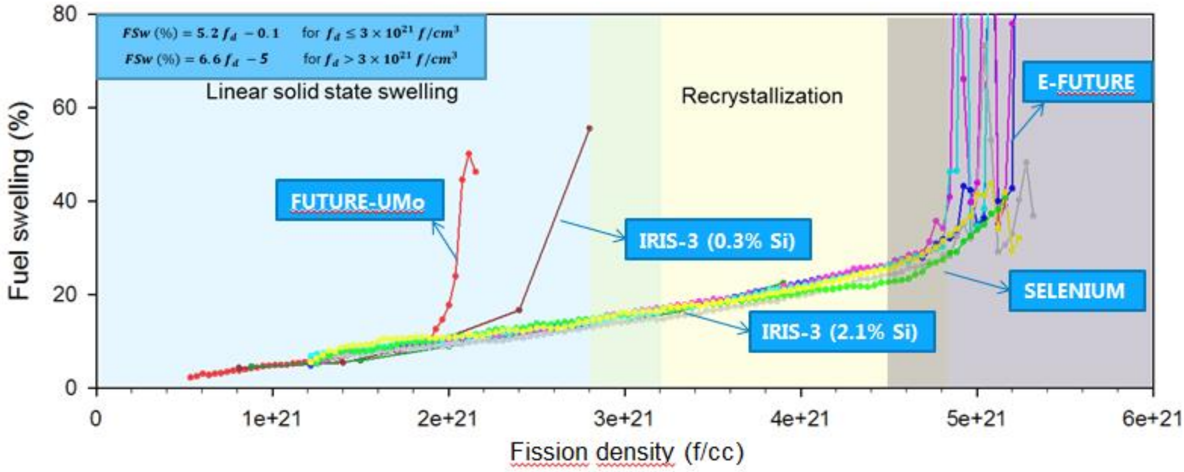

Figure 1: Observed fuel swelling in the E-Future I and SELENIUM irradiation tests. The exponential increase in thickness at high burn-ups is currently the key issue in the development of the fuel.

Two further points deserve mention.
First, as might be expected, a similar problem occurs at the fuel/cladding interface in UMo monolithic fuel. This problem has been addressed primarily by the introduction of a thin (~25 mm) diffusion/fission fragment recoil barrier between the fuel and the cladding. Zirconium is the preferred material for this purpose.
Second, there is an inherent fission density limit for UMo fuel particles or foils; fortunately, this limit appears to be at fission densities above the maximum achievable with LEU. However, if UMo is used with higher-enriched uranium, one can reach the point where the UMo cannot contain the accumulated fission gas, and breakaway swelling could become a problem.

Qualification of UMo dispersion fuel is presently being pursued by an expanded European collaboration, called HERACLES. There's a collaborative relationship with the US program.
The HERACLES roadmap starts with a comprehension phase. The idea is that a better understanding of the fuel behaviour is required to be able to develop adequate engineering solutions.

As part of the comprehension phase for dispersed UMo fuel, a new irradiation test for full-size plates, called "SEMPER FI", is foreseen. The irradiation test matrix for this test is presently being defined and irradiation is planned at BR2 in 2016. The aim is to study IL formation with ZrN coated particles and coated particles at high burn-up, and to inter-compare different coating technologies.

Engineering solutions will then be developed for the fuel evolution under high FR to high FD: It is considered that the fuel system can survive the high FR challenge if IL formation at high FD is limited and by reducing the UMo swelling rate; for this two distinct engineering efforts are required:
- limiting IL formation at high FR: SELENIUM has shown that the protective potential of a ZrN diffusion barrier coating is superior to the effect of Si in the matrix (or as a coating, which are considered essentially equivalent). Our understanding of the mechanism leading to IL formation in the presence of a coating is essential since the solution required depends on this.

- limiting UMo swelling at high FD: UMo swelling is mainly influenced by the recrystallization effect (RC). RC is known to be influenced by the Mo content of the fuel and its grain size. The engineering approach proposed here aims to homogenise the fuel to avoid Mo concentration gradients, with the largest possible grain size to delay and reduce the rate of recrystallization. This comes down to submitting the fuel kernels to a heat treatment prior to plate production.

A sister irradiation to SEMPER FI is also being prepared by the US: a mini-plate irradiation test in ATR, called EMPIRE, aiming at the study coated particles at high burn-up, verification if coating is still required if heat treatment prior to plate production is applied and the test of EU welding procedure (C2TWP) in the production of 'coated' monolithic miniplates.

Concurrently a manufacturing development program leading to industrialisation is carried out: for dispersed fuel it deals with powder production by atomization, heat treatment (annealing) prior to plate manufacturing and coating of the fuel particles; for the monolithic version, the production of the foils and the welding procedure of the recoil barrier on the monolithic foil.

The comprehension phase will be concluded as soon as all the relevant data from SEMPER FI and EMPIRE are available and analysed.

The U.S. program is now essentially concentrated on the qualification of UMo monolithic fuel, aimed at the conversion of the five civil U.S. High Performance research reactors. The main effort is deployed on the industrialization of the production process of the monolithic fuel and the construction of a dedicated fabrication facility.

Other organizations have also undertaken activities in the LEU fuel development field:
KAERI has since many years done concentrated efforts in the development of the production of fuel powders by an atomization process and in the upscaling of this process for the domestic fuel supply of HANARO. Since 2000 KAERI has focussed on qualifying rod-type UMo fuel with 5-6 $gUtot/cm^3$ and irradiation tests are going on. Coating techniques are also being developed. The fuel tests are however limited to maximum linear power values characteristic of medium power reactors.
In Argentina, CNEA has deployed an intensive R&D activity to fabricate both dispersed and monolithic UMo (Zircalloy-4 cladding) to develop solutions for the encountered technical problems.

References:

Within the length constraints for this paper, it's not possible to give a comprehensive bibliography, even limited to the essential papers. A comprehensive overview of the European programs may be found in the review paper by Van den Berghe and Lemoine (Nuclear Engineering and Technology 46 (2014), 125). For more information, one may refer to the proceedings of the annual RERTR and RRFM conferences.