IRRADIATION TESTING OF U_3Si_2-Al-FUELS UP TO VERY HIGH FISSION DENSITIES

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ABSTRACT

During the licensing procedure of the FRM-II the Technische Universität München established a fuel-test program in order to confirm the swelling data given in the literature. Because it was desirable for several reasons to extend the region tested, a second test program was planned and performed in cooperation with the CEA-Saclay. Within the framework of this program two fuel plates (a so-called "Homogeneous Plate" and a so-called "Mixed Plate") were irradiated. Even though in particular the Mixed Plate was irradiated until considerable high fission densities in the fuel-particles were reached, both plates showed a stable swelling behavior.

1 Introduction

The U_3Si_2-Al-fuel that will be used in the FRM-II was developed and intensively tested by the RERTR-Program (**R**educed **E**nrichment for **R**esearch and **T**est **R**eactors) and therefore can be considered as qualified up to uranium densities of 4.5 gU/cm^3 to 4.8 gU/cm^3 in the fuel meat. Since it was the aim of the RERTR-Program that was mainly carried out by the ANL (**A**rgonne **N**ational **L**aboratory) to develop a new fuel that would allow most Research Reactors to convert from high-enriched uranium (HEU) to low-enriched uranium (LEU) most tests were performed with the latter. However, some test irradiations with medium enriched uranium (MEU) and two test-irradiations with HEU-samples were also performed in order to test whether there is a maximum fission density beyond that the swelling behavior is not stable any longer or not. These tests revealed so far that U_3Si_2-Al-fuel exhibits a stable swelling behavior up to very high fission densities with no indication of breakaway-swelling.

2 The compact core concept of the FRM-II

Based on these excellent results it was decided that U_3Si_2-Al-fuel is the optimum fuel for the FRM-II, the new Research Reactor of the Technische Universität München (TUM), which will operate with high-enriched fuel in order to realize a "compact-core" providing the FRM-II with unique chracteristics – such as high thermal neutron flux at low power, a high spectral purity of the energy distribution of the neutrons and a large usable volume outside of the reactor core. The core is cooled with light water (H_2O) and is placed in the center of a moderator tank filled with heavy water (D_2O). It consists of only one cylindrical fuel element that contains the fuel (U_3Si_2-Al) in 113 involutely curved fuel plates. Because of this involute shape the coolant channels between the plates have a constant width of 2.2 mm. The plates themselves have a thickness of 1.36 mm (fuel 0.60 mm, cladding 0.38 mm). The outer diameter of the fuel element is 243 mm and the height of its active zone is 700 mm.

Since the reactor core is very small the leakage of fast neutrons out of the core is very high (more than 50 %) and neutrons which have been thermalized in the D_2O and diffuse back into the core make a big contribution to the nuclear chain reaction. However, they would cause a big peak in the distribution of the power density at the outer edge of the core. This peak is reduced in the FRM-II using the concept of fuel grading: the uranium density in the fuel meat is 3.0 gU/cm^3 in the inner part of the fuel element and 1.5 gU/cm^3 in the outer part.

3 The fuel-qualification program at the TUM

During the licensing procedure of the FRM-II the authorities asked the TUM to confirm the results obtained by the RERTR-Program. This was the birth of a fuel-qualification program performed by the TUM in very close cooperation with the French Commissariat à l'Energie Atomique (CEA); the latter provided the TUM with irradiation time at two of its reactors (SILOE and OSIRIS) and performed the plate thickness measurements. In addition the CEA performed post irradiation examinations (PIE) in its Hot Cells.

3.1 The "SILOE test program"

The "SILOE test program" was established in 1996 and consisted of the irradiation of a test-plate containing fuel with an uranium density of 1.5 gU/cm^3 in the fuel meat. At the end of the irradiation a fission density in the fuel-particles of about 12×10^{21} f/cm^3 was reached. γ-scanning-measurements were performed in the course of the irradiation (i. e. after the 2nd and the 5th irradiation cycle) as well as after the last (10th) irradiation cycle. Following the irradiation PIE were performed on two samples punched out of the plate. These two samples were chosen in such a way that the fission densities in the fuel were about 8×10^{21} f/cm^3 and about 12×10^{21} f/cm^3, respectively.

3.2 The "OSIRIS test program"

The "OSIRIS test program" was established in 1999 in order to gain additional information concerning the irradiation behavior of high-enriched high-density U_3Si_2-Al-fuel. Within the scope of this program it was planed to irradiate two plates until fission densities comparable to or somewhat higher as those at the end of a cycle of the FRM-II [1] were reached. One of the two plates – the so called "Homogeneous Plate" – contained fuel with an uranium density of 3.0 gU/cm^3 and was irradiated until a maximum fission density of about 8×10^{21} f/cm^3 was obtained in the fuel-particles. The second plate – the so called "Mixed Plate" – contained fuel with two different uranium densities and was irradiated until maximum fission densities of 10.4×10^{21} f/cm^3 in the 3.0 gU/cm^3-zone and of 14.3×10^{21} f/cm^3 in the 1.5 gU/cm^3-zone, respectively, were obtained.

4 Results

4.1 Results of the SILOE test program

As reported earlier ([2], [3]) the measurements of the plate-thickness (that were performed after each of ten irradiation cycles) confirmed the stable swelling behavior as expected. The increase of the thickness was small and continuous (see figure 1). In contrast to this a "sudden" strong increase of the swelling rate would have been an indication of "breakaway-swelling" and of a failure of the plate.

In order to be able to compare the data obtained in this test with the data given in the literature the increase of the volume of the fuel-particles was determined. Therefore, the thickness of the oxide-layer on the plate was calculated using the same correlation that was used in designing e. g. the FRM-II [4] and subtracted. After that the volume-increase was calculated based on the increase of the thickness. As one can see from figure 2 all values are in the range of the straight line that was deduced from the data given in the literature. The exceptional point at a fission density of about 3.4×10^{21} f/cm^3 is not realistic, but resulted from a problem with the measurement device.

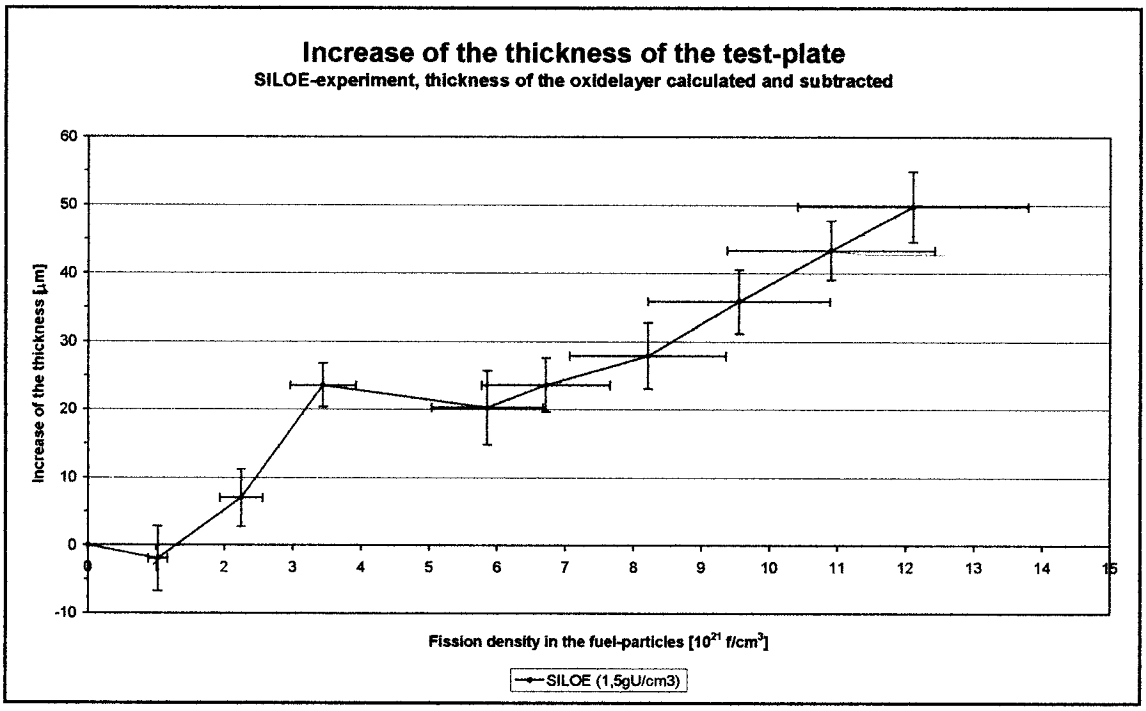

Figure 1: Increase of the thickness of the test-plate with an uranium density of 1.5 gU/cm^3 as a function of the fission density in the fuel-particles. The thickness of the oxide-layer has been subtracted. The error bars for the thickness measurements represent the statistical error obtained from taking the mean value in a small area of the test-plate; the error bars for the fission density are related to the inaccuracy of the γ-scanning-measurements [2]. The exceptional point at about 3.4 x 10^{21} f/cm^3 was caused by problems with the electronic equipment which have been solved in the course of the irradiation test, and therefore is not due to irradiation effects.

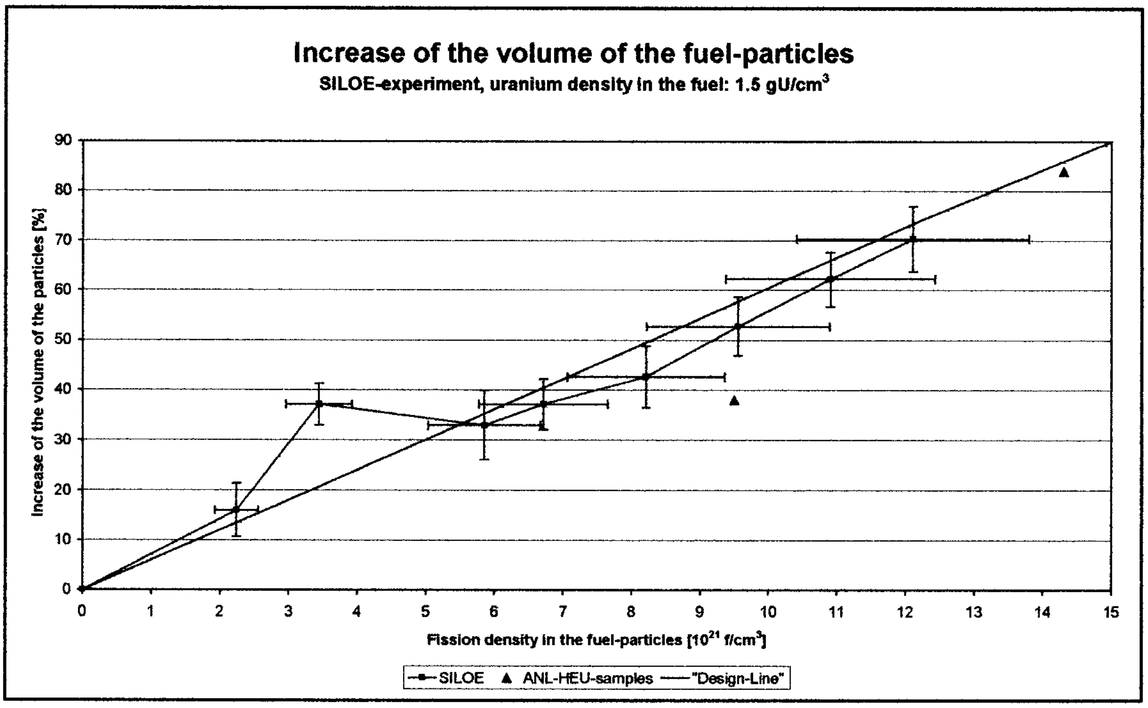

Figure 2: Increase of the volume of the fuel-particles in the test-plate with an uranium density of 1.5 gU/cm^3 as a function of the fission density in the fuel-particles. The values were calculated from the thickness increase of the test-plate. The error bars are related to the ones given in figure 1.

Following the irradiation experiment with this plate (1.5 gU/cm^3) PIE were performed in the Hot Cells LAMA of the CEA-Grenoble in order to obtain information about the fine structure of the U_3Si_2-grains and the thickness of the U-Al-interdiffusionlayer that is formed at the grain-boundaries. The microscopic pictures (one of them is shown in figure 3) demonstrate that the morphology of the fission gas bubbles is as expected. One substantial aim of the PIE was to show that fuel with an uranium density of 3.0 gU/cm^3 in the meat can be used up to fission densities of about 8×10^{21} f/cm^3. The essential condition for this is that there remains enough "free" aluminum[1] at the end of the irradiation. In [5] a conservative value of 15 *vol.%* for the minimum Al-content has been established for stable fuel swelling. In order to calculate the Al-content in the fuel with 3.0 gU/cm^3 at a fission density in the particles of about 8×10^{21} f/cm^3 out of the data obtained from the test-plate with 1.5 gU/cm^3 a relation was formulated [6]. The applicability of this relation was tested by means of a quantitative analysis of the microstructure. The knowledge of the thickness of the interdiffusionlayer is necessary for determining the loss of "free" Al; it was calculated with the help of correlations taken from the literature ([7], [8]) and tested against the results of the microscopic examinations. The analysis showed that at a fission density in the particles of 8×10^{21} f/cm^3 the fuel with a uranium density of 3.0 gU/cm^3 still contains between 50 *vol.%* and 68 *vol.%* of "free" aluminum - this value is far above the required 15 *vol.%*. As a consequence it is proved that this fuel can be used up to fission densities of at least 8×10^{21} f/cm^3.

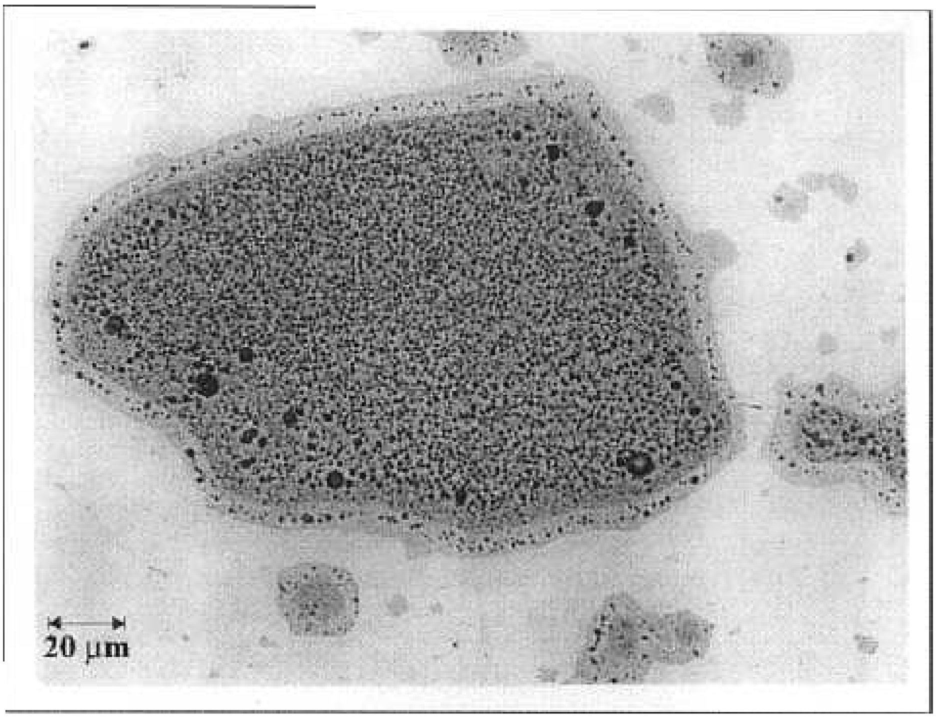

Figure 3: Picture of a U_3Si_2 particle taken in the region of the test-plate with a fission density of 12×10^{21} f/cm^3 in the fuel particles. The morphology of the fission gas bubbles is as expected. Moreover, the thickness of the U-Al-interdiffusionlayer (light-gray area at the grain-boundary) is of the order of 6 μm and in good accordance with the theory [6].

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[1] The term "free" means that the aluminum in the U-Al-interdiffusionlayer is not taken into account.

4.2 Results of the OSIRIS test program

The irradiation of the Homogeneous Plate (3.0 gU/cm^3) was finished on November 6, 2000, and the results of a preliminary assessment are given in figure 4 and figure 5. In figure 4 the evolution of the thickness of the plate is shown whereas figure 5 shows the increase of the plate-thickness as a function of the fission density in the fuel-particles. The values given in this diagram were corrected by estimating and subtracting the oxide-layer. As one can see from both diagrams the increase of the thickness is a roughly linear function of the fission density.

The results of this part of the OSIRIS program clearly confirm the statement made after the PIE (see section 4.1): U_3Si_2-Al-fuel exhibits a stable swelling behavior up to fission densities of at least 8×10^{21} f/cm^3. As we will see later this is even true for much higher fission densities.

Because of a slightly different irradiation schedule and – even more important – because the fission densities aimed at were extended to higher values, the Mixed Plate was irradiated until the end of March 2001. The maximum fission density reached in the fuel-particles was 10.4×10^{21} f/cm^3 in the 3.0 gU/cm^3-zone and 14.3×10^{21} f/cm^3 in the 1.5 gU/cm^3-zone, respectively. Figure 6 shows the evolution of the thickness of the Mixed Plate measured in the "transversal" direction, i. e. across the border between the zones with the different uranium densities. The difference in the swelling of the two zones is clearly visible and it can be stated that – as expected – the density grading has absolutely no negative impact on the swelling behavior of the fuel.

It is of great interest, of course, to compare the results obtained from the OSIRIS-measurements with those obtained from the SILOE-experiment and the data given in the literature. For this reason again the oxide-layer was calculated using the correlation mentioned above [4] and subtracted.

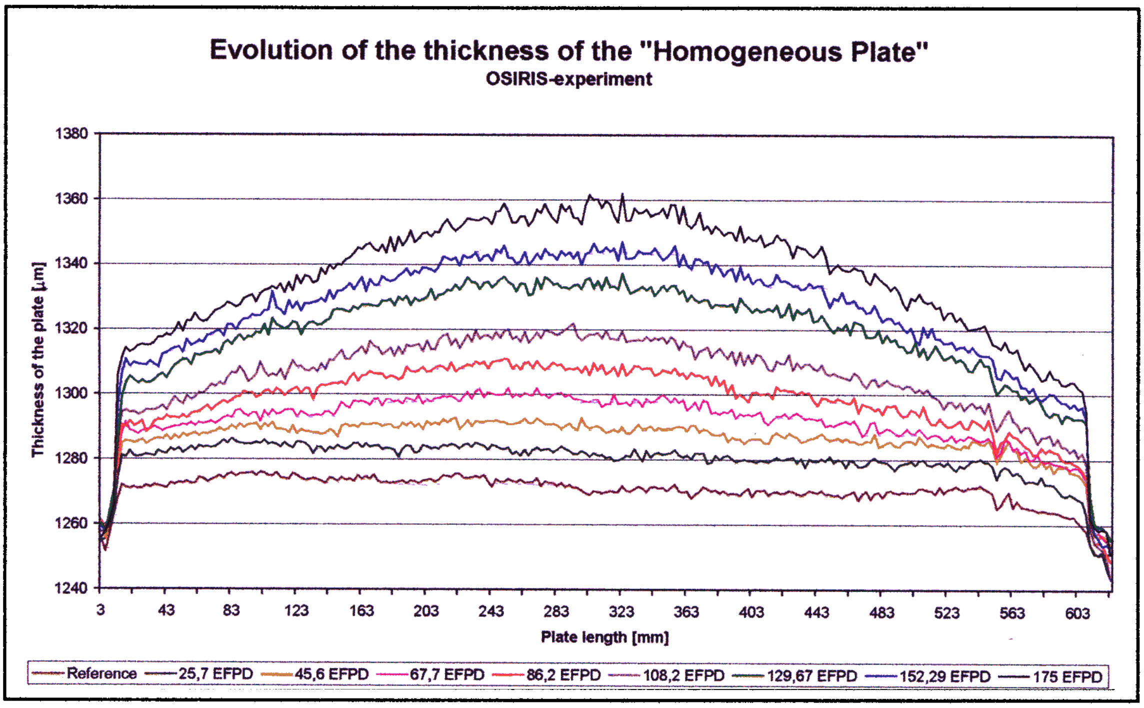

Figure 4: Measurements of the thickness of the "Homogeneous Plate" that contains fuel with an uranium density of 3.0 gU/cm^3 in the fuel meat. It is clearly visible that the curves are nearly equidistant, i. e. that the plate is in the stable swelling stage.

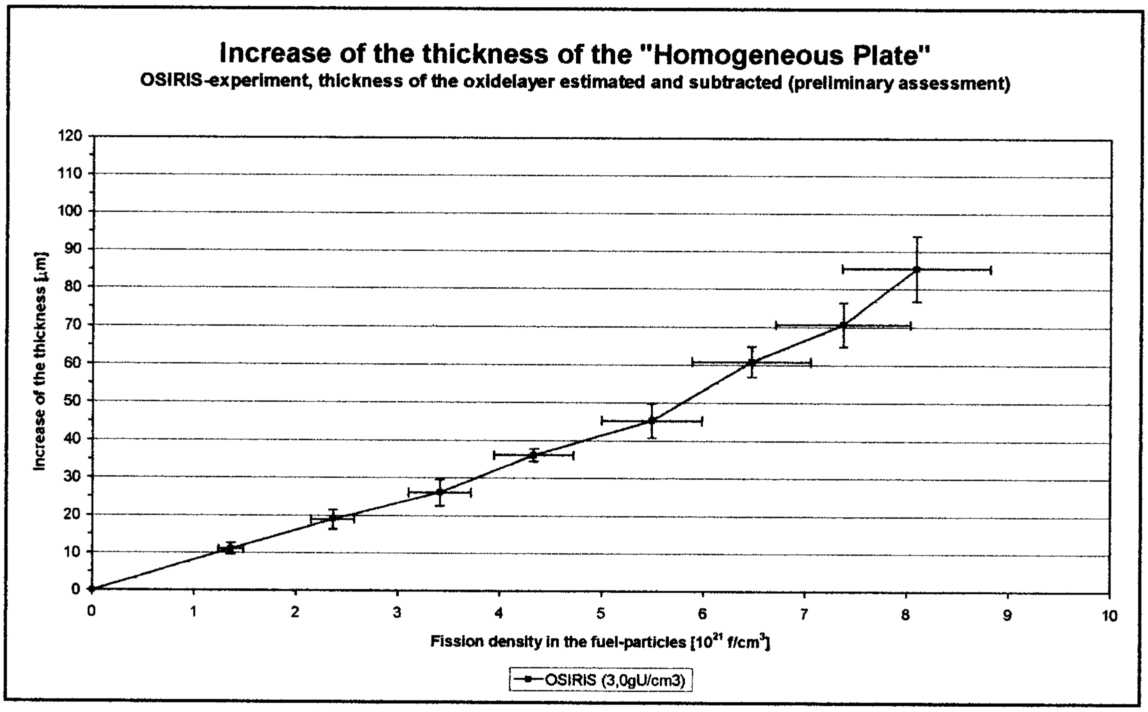

Figure 5: Increase of the thickness of the "Homogeneous Plate" with an uranium density of 3.0 gU/cm^3 as a function of the fission density in the fuel-particles. The thickness of the oxide-layer has been estimated and subtracted. The error bars for the thickness measurements represent the statistical error obtained from taking the mean value in a small area of the test-plate; the error bars for the fission density are related to the inaccuracy of the γ-scanning-measurements.

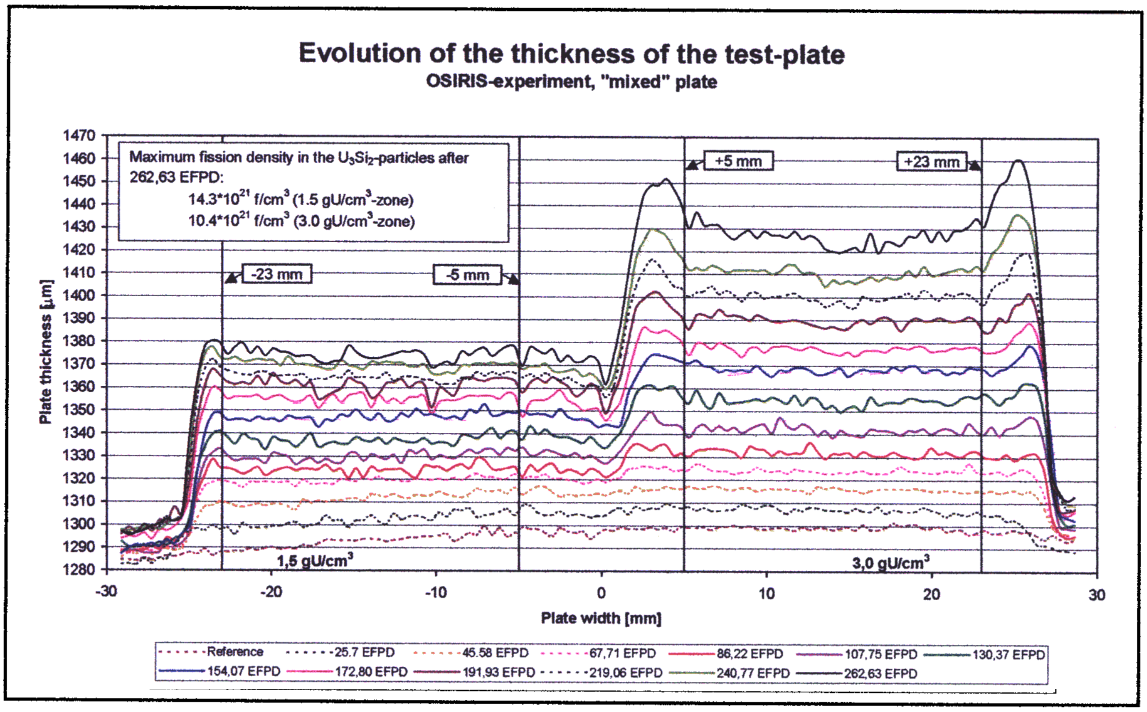

Figure 6: Measurements of the thickness of the "Mixed Plate" in the transversal direction, i. e. perpendicular to the border between the areas containing fuel with an uranium density of 1.5 gU/cm^3 and 3.0 gU/cm^3, respectively. The fission densities reached are considerable high (see insert in the figure). Due to the fact that the different curves are nearly equidistant it can be stated that the plate is in a stable swelling stage and that the grading of the uranium density has absolutely no impact on the swelling behavior of the fuel plate.

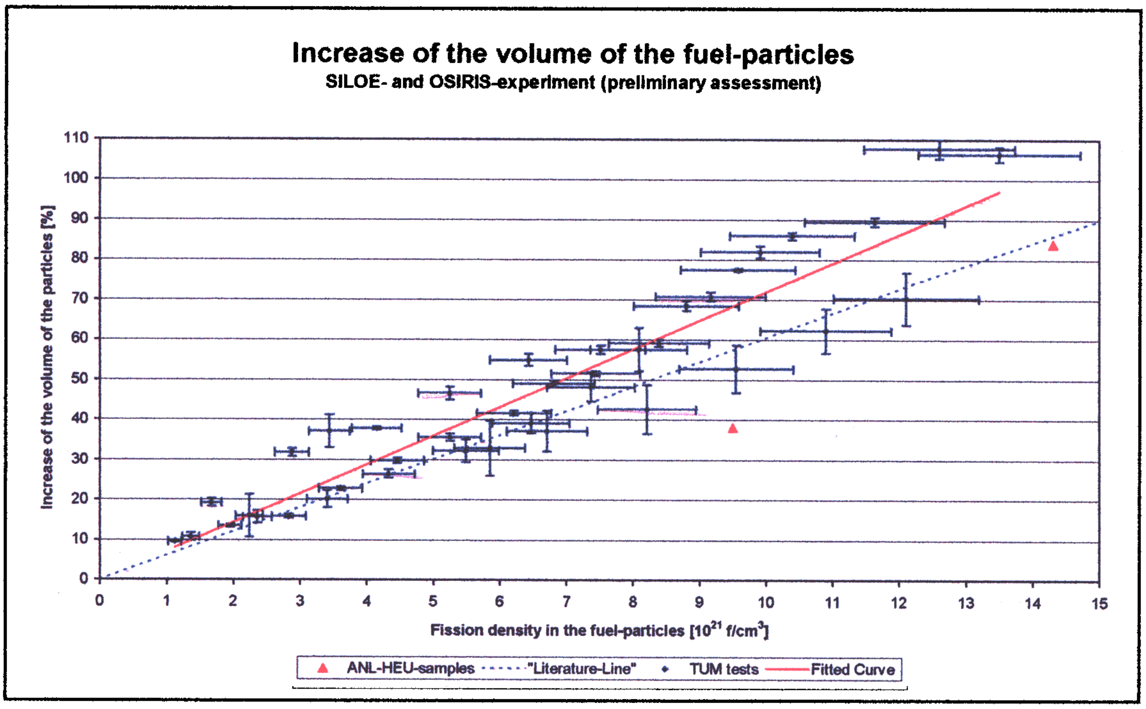

Figure 7: Increase of the volume of the fuel-particles as a function of the fission density. The data points were obtained from the SILOE test program and from the Homogeneous Plate and the Mixed Plate (preliminary data) that were both irradiated within framework of the OSIRIS test program. The dotted straight line was deduced from the data given in the literature (see text). The "ANL-HEU-samples"-points are the results of the two HEU samples irradiated during the RERTR-program. The solid straight line is a linear least-square fit to the data points measured by the TUM.

The thickness-increase of the Homogeneous Plate as well as of the Mixed Plate was converted into the increase of the volume of the fuel-particles (as in figure 2). In figure 7 a compilation of nearly all data obtained so far in the course of the fuel test program of the Technische Universität München is given. All values are in the expected range: the SILOE-data are somewhat below the dotted straight line (as mentioned above this line was determined using the results obtained by the RERTR-Program, i. e. by irradiating LEU- and some MEU-samples) and the OSIRIS-data are somewhat above that line. It must be pointed out that:

1. In particular the results for the Mixed Plate represent preliminary data, since the final γ-scanning-measurements are still underway at present. We have learned that the calculations concerning the fission densities have to be re-calibrated by means of γ-scanning-measurements.
2. The volume-increase of the fuel-particles was determined by converting the thickness measurements rather than by immersion measurements. The latter generally lead to lower values [9].

The solid straight line in figure 7 is a linear least-square fit to all data points. Even though it is slightly above the dotted line it is clear that the fuel exhibits a stable swelling behavior. The volume-increase of the fuel-particles is 7.2 % per 10^{21} f/cm^3; this value matches very well the value of (8 ± 2) % per 10^{21} f/cm^3 as given in [10].

5 Conclusions

The U_3Si_2-Al-fuel was developed and intensively tested by the RERTR-Program. Since it was the aim of that program to qualify this fuel for routine use in combination with LEU only two HEU-samples were irradiated. In order to confirm the results of these measurements and to extend the tested region with respect to uranium density and fission density in the fuel-particles the Technische Universität

München established two test programs. The results of the SILOE test program were used to qualify the U_3Si_2-Al-fuel for the use in the FRM-II; the results of the OSIRIS test program extend the knowledge concerning the irradiation behavior of high-enriched high-density U_3Si_2-Al-fuel. Both test programs revealed that U_3Si_2-Al-fuel exhibits a stable swelling behavior up to very high fission densities. Because there was clearly no indication of breakaway-swelling it may well be that fission densities even higher than tested so far can be reached in U_3Si_2-Al-fuel without any impacts on reactor safety.

6 References

[1] **Feltes, W.; Strömich, A.:** „Brennstoffqualifikation FRM-II"; Report-Nr. A1C-1300334-0; Siemens KWU.

[2] **Nuding, M.:** „FRM-II: Auswertung des Brennstoff-Bestrahlungsexperiments am Reaktor SILOE und Bewertung der Ergebnisse auf mikroskopischer Ebene"; Report-Nr. OPA 218; Technische Universität München, ZBE FRM-II-Bau; Dezember 10, 1998.

[3] **Nuding, M.; Rottmann, M.; Axmann, A.; Böning, K.:** "FRM-II Project Status and Safety of its Compact Fuel Element"; Proceedings of the *RRFM International Meeting 2000*, Colmar; March 19 - 21, 2000.

[4] **Gysler; Ehrich; Sperber:** „Thermohydraulische Kernauslegung"; Bericht Nr. A1C-1301735-1; Siemens KWU.

[5] **Hofmann, G. L.; Rest, J.; Snelgrove, J. L.:** „Aluminum-U_3Si_2 interdiffusion and its implication for the performance of highly loaded fuel operating at higher temperatures and fission rates (a preliminary assessment)"; Argonne National Laboratory; Oktober 1995.

[6] **Nuding, M.:** "Auswertung der Nachbestrahlungsuntersuchungen an der Testplatte FRM-1501 in den Heißen Zellen LAMA im Rahmen der Brennstoffqualifikation für den FRM-II – Eignungsnachweis des Brennstoffs mit einer Urandichte von 3.0 gU/cm^3 im Brennstoffkern"; Report-Nr. OPA 272; Technische Universität München, ZBE FRM-II-Bau; November 19, 1999.

[7] **Hofmann, G. L.; Rest, J.; Snelgrove, J. L.; Koster van Groos, S.:** „Aluminum-U_3Si_2 interdiffusion and its implication for the performance of highly loaded fuel operating at higher temperatures and fission rates"; RERTR International Meeting, Seoul; October 7 - 10, 1996.

[8] **Copeland, G.:** „Status of the fuel development"; Presentation; R&D Staff Meeting of the ANS-Project team; January 11, 1994.

[9] **Copeland, G. L.; Hofman, G. L.; Snelgrove, J. L.:** „Examination of U_3Si_2-Al Fuel Elements from the Oak Ridge Research Reactor"; RERTR International Meeting, Gatlinburg; November 3 – 6, 1986.

[10] **Cahn, R. W.; Haasen, P.; Kramer, E. J. [Hrsg.]:** „Materials Science and Technology - A Comprehensive Treatment, Volume 10 A"; VCH Weinheim; 1994.