5. Detailed Evaluation

After first estimate diffusion analyses showed that an effective diffusion model is still a viable solution to predict silver transport and release from TRISO particle spherical fuel, detailed analyses of all high and medium applicable tests (Table 1) were performed.

Detailed evaluation starts with fuel spheres from the top of the table, performing analyses of the most important tests first.

5.1 HFR-K3

5.1.1 Reactor

The High Flux Reactor (HFR) at Petten, the Netherlands, is a closed-tank in-pool type material testing reactor, being in operation since 1962 with a thermal power of 45 MW [28]. It is a light water moderated, beryllium reflector water-cooled reactor with fast and thermal fluxes in the order of $4.5 \times 10^{18} \text{ m}^2\text{s}^{-1}$ and $2.4 \times 10^{18} \text{ m}^2\text{s}^{-1}$ respectively. The reactor was extensively used in the British and German HTR programmes. Special full-scale rigs were developed to test HTR fuel elements under realistic conditions, with appropriate correlation between burn-up, neutron fluence, and temperature.

Four spherical fuel elements were tested simultaneously in a so-called BEST-rig. Each rig contained four individual and independently-monitored capsules arranged in line. A binary mixture of helium and neon achieved temperature control, with helium providing the best cooling.

Fluence, temperature and fission product release were measured for each capsule in real time. Full sphere irradiation tests of interest performed were HFR-K3, -K5 and -K6. Several other irradiation tests were done on fuel and compacts but only these three experiments are of interest for modern fuel evaluation. Unfortunately, the HFR-K5 and -K6 tests were performed at the end of the German HTR programme when interest had waned. Postirradiation evaluation of tests spheres and fuel rig materials was not done in time and no measurements of silver release from these tests were recorded. The earlier irradiation test HFR-K3 underwent comprehensive post-irradiation testing and heating examinations.

5.1.2 Irradiation test

Four elements from Arbeitsgemeinschaft Versuchsreaktor (AVR) reload 19 with LEU-TRISO fuel were inserted into the core inside a three-capsule BEST rig. The middle capsule contained two spheres with both outer capsules containing one sphere each. Each capsule consisted of a steel container with graphite cups which housed the test spheres. Fuel spheres were numbered 1, 2, 3 and 4 starting from the top with planned irradiation temperatures of 1 020 °C, 700 °C, 700 °C and 1 020 °C surface and 1 200 °C, 920 °C, 920 °C and 1 200 °C centre, respectively [29]. Instrumentation consisted of thermocouples, flux monitors and SPN-monitors, as well as sweep and temperature regulation tubes for each single capsule.

The placement of flux detectors and thermocouples relative to the four fuel spheres in the irradiation rig are shown in Figure 13. Due to flux gradients present in the HFR irradiation positions used, the rig was turned through 180° during the test in order to minimize the effect of gradients inside the fuel elements. HFR-K3 started on 15 April 1982 and ended successfully on 5 September 1983. Burn-up values of 10.6% FIMA were attained, with fast fluences of 6.3 x 10^{25} m⁻². Heating tests up to 1 800 °C were conducted during the postirradiation examination. The objectives for this test were as follows [25]:

- a. Accelerated reference test on LEU-TRISO fuel spheres for an HTR steam cycle process heat applications.
- b. Providing irradiation data and Post-irradiation Examination (PIE) results for a data set of a licensable fuel element for HTR with LEU cycle.
- c. Examination of mechanical performance of particles of 1981 standard quality.
- d. Determination of release of relevant fission product nuclides from the fuel elements.
- e. Testing of the BEST-rig with sweep and regulation gas circuits and out of pile experimental installations at the HFR.

Post-irradiation examination was completed, with ceramographic examinations on particles from test element HFR-K3/1 after a high-temperature annealing test, and fission product inventories measured on all four test elements. All capsule components and graphite cups were leached and fission products in the solutions determined with gamma spectrometry. Further gamma spectrometric measurements were made of drilling samples in the fuel-free zone of the fuel elements to determine $137Cs$ and $110mAg$ profiles in the fuel-free zone. This was a two times accelerated test that ran for 359 Effective Full Power Days (EFPD).

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Figure 13: HFR-K3: Flux Detector and Thermocouple Placement

Due to the neutron flux gradient present in the HFR, the rig was turned 180° several times during irradiation to minimize the effect of uneven azimuthal burn-up. Burn-ups of 7.5, 10.0, 10.6, and 9.0% FIMA and fast fluences of 4.1, 5.8, 5.9 and 4.3 x 10^{25} m⁻² for the four tests spheres respectively were achieved [30]. The $85mKr$ fractional release remained very low $(< 10⁻⁶)$ so it can be assumed that no particles failed during irradiation nor were there any particles defective from manufacture.

AVR reload 19 is comparable with PBMR design fuel, and the irradiation conditions of HFR-K3 are acceptable and comparable to expected PBMR operational conditions. The data received from HFR-K3 progress reports and post-irradiation examination reports is sufficient to evaluate silver transport. Important test element data of HFR-K3 is listed in Table 3 with expected PBMR fuel specification and irradiation data.

Parameter	Unit	1	$\overline{2}$	3	4	PBMR	
Specification							
Uranium content	g	10.22	10.22	10.22	10.22	9.00	
235 U enrichment	$\frac{0}{0}$	9.82	9.82	9.82	9.82	9.6	
CP content		16 350	16 350	16 350	16 350	\sim 14 500	
Failed CP content	per FE	θ	θ	$\boldsymbol{0}$	θ	$\sim 0.2^{(b)}$	
Irradiation data							
Burn-up (FIMA)	$\frac{0}{0}$	7.53	10.02	10.57	8.97	9.8	
Neutron fluence $(> 0.1 \text{ MeV})$	10^{25} m ⁻²	4.1	6.1	6.3	4.3	2.7	
Average centre temperature ^(a)	$\rm ^{\circ}C$	1 2 4 7	1 1 2 1	1 1 1 5	1 2 7 8	1 040	
Max power output	kW/FE	2.72	3.41	3.61	3.42	2.1	

Table 3: HFR-K3 Test Element Specification and Irradiation Data

a. Calculated from the measured surface temperature and FE power output.

b. 1.44 x 10^{-5} failure fraction¹ x 14 500 particles per FE.

 \overline{a}

¹ The failure fraction from manufacture used for PBMR design analysis of 1.44 x 10⁻⁵ was derived statistically from German fuel manufacturing experience.

5.1.3 Evaluation

A first evaluation of HFR-K3 fission product release was performed by Christ in 1985, shortly after completion of the first post-irradiation examination [26]. After this evaluation more information about the irradiation test was released and further examinations of the fuel and rig materials changed original fluxes, estimated burn-ups, and temperatures achieved. Further evaluations were performed on this test (e.g. [31] and, most notably, unpublished work by Venter [32]).

5.1.3.1 Input data

A thorough evaluation of thermocouple performance was performed by Venter [32]. As is the case with most irradiation tests [33], a number of thermocouples failed during irradiation with a resulting loss of important temperature data. Thermocouples placed on the surfaces of fuel spheres indicated temperatures several degrees lower than those embedded in fuel-free regions of fuel spheres. This is especially clear for capsules 1, 2, and 4. It is to be expected that there would be a difference between temperatures measured on fuel sphere surfaces and temperatures measured 3 mm from the surface but, as graphite is a good conductor of heat, one would expect a difference of a few degrees at most and not 100 to 200 degrees Celsius.

Thus all temperatures measured by surface thermocouples are suspect and should be discarded. Unfortunately capsule 3 had six surface thermocouples and only one embedded thermocouple. Temperature values for the embedded thermocouple form the upper bound for capsule 3 temperatures and it is safe to conclude that all surface temperatures for this capsule are too low and should be discarded. There are also some temperature values that are obviously wrong and these were also discarded. Cycle-averaged temperatures calculated using only data from embedded thermocouples are shown in Table 4. Also included in the table for comparison is cycle-averaged temperature data obtained from the RUBICON data handling computer code used at HFR. The evaluation of HFR-K3 test spheres was performed with both sets of temperature data.

Table 4: HFR-K3: Cycle Averaged Temperature Data

Cycle	Capsule 1		Capsule 2		Capsule 3		Capsule 4	
Number	Thermal	Fast	Thermal	Fast	Thermal	Fast	Thermal	Fast
82.04	0.862	1.24	1.10	1.93	1.11	1.95	0.900	1.29
82.05	0.834	1.23	1.06	1.87	1.07	1.89	0.870	1.28
82.06	0.908	1.30	1.14	1.97	1.15	1.99	0.940	1.36
82.07	0.909	1.38	1.15	1.94	1.16	1.95	0.940	1.45
82.08	0.887	1.43	1.13	2.33	1.14	2.35	0.920	1.45
82.09	0.874	1.34	1.12	1.95	1.13	2.07	0.920	1.40
82.10	0.849	1.31	1.08	2.03	1.09	2.05	0.890	1.38
82.11	0.882	1.38	1.13	2.06	1.14	2.08	0.920	1.39
83.01	0.890	1.35	1.14	2.12	1.15	2.13	0.930	1.42
83.02	0.859	1.32	1.11	1.99	1.12	2.01	0.910	1.37
83.03	0.847	1.31	1.08	2.01	1.09	2.03	0.890	1.36
83.04	0.862	1.38	1.17	2.10	1.18	2.12	0.960	1.43
83.05	0.900	1.23	1.16	1.08	1.17	1.90	0.950	1.31
83.06	0.802	1.32	1.03	2.03	1.04	2.08	0.850	1.37
83.07.1	0.810	1.32	1.03	1.88	1.04	1.90	0.840	1.32
83.07.2	0.838	1.36	1.07	1.99	1.08	2.01	0.880	1.32

Table 5: HFR-K3: Thermal and Fast Neutron Fluxes (x $10^{18} \text{ m}^{-2} \text{s}^{-1}$)

Neutron fluxes were taken from the metrology report [30], the first HFR-K3 evaluation [26], and updated with corrections made by Venter [32]. Thermal and fast neutron fluxes are tabled in Table 5. The cross sections used in the burn-up calculation were based on cross sections used in previous HFR evaluations [34] and updated to yield correct fission power and burn-up values.

Cycle	Capsule 1		Capsule 2		Capsule 3		Capsule 4	
Number	Venter	Christ	Venter	Christ	Venter	Christ	Venter	Christ
82.04	619	810	964	900	964	900	644	855
82.05	614	770	934	860	954	860	639	815
82.06	649	830	984	930	1 0 1 9	930	679	880
82.07	689	880	1173	980	1 0 8 3	980	724	930
82.08	714	830	974	920	1 0 9 8	920	724	875
82.09	669	810	1 0 2 4	900	1 0 4 9	900	699	855
82.10	654	740	1 0 1 4	830	1 0 1 4	830	689	785
82.11	689	800	1 0 29	890	1 0 5 8	890	694	845
83.01	674	820	1 0 5 9	910	1058	910	709	865
83.02	659	800	1 0 0 4	880	1 0 3 9	880	684	840
83.03	654	690	999	760	1 0 1 9	760	679	725
83.04	689	810	1 0 5 4	900	1 0 6 8	900	714	855
83.05	614	820	939	910	949	910	654	865
83.06	659	700	1 0 1 4	780	1 0 3 9	780	684	740
83.07.1	659	690	1 0 3 4	770	1 0 3 4	770	659	730
83.07.2	679	710	994	790	1 0 3 9	790	679	750

Table 6: HFR-K3: Gamma Heating (W)

Gamma heating has been calculated by both Christ [26] and Venter [32]. The method used by Christ is not available in the literature but Venter's method is well described in the reference. Gamma power values were deduced from data from the HFR-K6 irradiation test from two different locations in the HFR core. A linear relationship between gamma power and fast neutron $(E > 0.1 \text{ MeV})$ flux was observed and a linear least squares fit was performed. It was found that the gamma power could be expressed by a fitted line:

$$
P = 2.3861 \phi_{E>0.1MeV} \tag{17}
$$

Both Christ and Venter gamma heating results are tabled in Table 6.

5.1.3.2 Evaluation results

Fission powers produced during irradiation and total burn-up achieved were calculated first to ensure that calculated temperatures are as close as possible to actual values. The fission powers were calculated using the best available thermal neutron fluxes (Table 5) and cross sections from later HFR irradiation tests [34] that have been corrected to yield the correct burn-up. These newly calculated fission powers where then used with estimated gamma heating (Table 6) to determine fuel temperatures. He/Ne coolant gas temperatures were adjusted to ensure that calculated and measured surface temperatures (Table 4) agree. Centre fuel temperatures were then calculated from surface temperatures and total power produced (fission and gamma) in each test sphere. Fission powers (in watt), and surface and centre temperatures (all in $^{\circ}$ C) for all four test spheres have been determined using both Christ and Venter data and are listed in Table 7 to Table 10.

Table 7: HFR-K3/1: Fission Power (W), Surface and Centre Temperatures (^{o}C)

Table 8: HFR-K3/2: Fission Power (W), Surface and Centre Temperatures (^{o}C)

Table 9: HFR-K3/3: Fission Power (W), Surface and Centre Temperatures (^{o}C)

The fractional releases of silver from the test spheres during irradiation are listed in Table 11. Fractional releases are simply the total measured released $110m$ Ag activity divided by the total ^{110m}Ag inventory in the fuel sphere. For test sphere 3, no measured value is reported for the graphite cup. It is not possible that there is no silver in the graphite cup. Some silver must be measurable just from contamination sources alone. The absence of a value means that the measurement failed and not that no silver was measurable.

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Comparing fractional releases from sphere 2 and other test spheres, it can be conservatively estimated that the fractional release on the graphite cup should be between 1.6 and 2.2 x 10^{-4} . For the sake of conservatism in nuclear analyses, the higher value is included in the total fractional release.

Fuel Sphere	Steel Capsule	Graphite Cup	Total Fractional Release
	1.0×10^{-3}	1.2×10^{-3}	2.2×10^{-3}
	2.3×10^{-4}	2.2×10^{-4}	4.5×10^{-4}
	1.6×10^{-4}	(2.2×10^{-4})	3.8×10^{-4}
	2.1×10^{-3}	1.6×10^{-2}	1.8×10^{-2}

Table 11: Fractional ^{110m}Ag Release from Fuel Elements of HFR-K3 [25]

Measured values were obtained by dissolving the graphite cups that surrounded fuel spheres during irradiation and by leaching the surfaces of the stainless steel containers and measuring the 110m Ag concentration in the liquids. Thus there are some additional sources of 110m Ag in fuel spheres and graphite cups that should be considered; firstly the natural uranium and thorium contamination of the graphite cups that housed the test spheres.

The total mass of the graphite cups that house each sphere $(\sim 190 \text{ g})$ is approximately the same as the mass of graphite in a test sphere $(\sim 200 \text{ g})$. It can be conservatively assumed that uranium and thorium contamination in the graphite cups is at least as much as the contamination in the fuel sphere. This graphite contamination was reduced to an effective uranium contamination only and added to the fuel-free zone contamination.

In addition to natural uranium and thorium contamination in the cups, the graphite of which the cups and fuel spheres are manufactured also contains silver as an impurity. Christ [26] gives a value of 0.8 ± 0.5 ng/g for the concentration of silver in the graphite used for fuel spheres and graphite cups. Thus a fuel sphere of mass 209 g of which 200 g is graphite, will contain 1.6×10^{-7} grams of silver. The number of silver atoms in the fuel sphere will be 8.8 x 10^{14} atoms. Only 48% of these will be 109 Ag so that the number of 109 Ag atoms is 4.2 x 10^{14} atoms. For a 60 mm diameter sphere, the volume concentration of 109 Ag in a fuel sphere will be between 3.7 and 6.0 x 10^{12} 109 Ag atoms per cm³. In the Gontard report [25] the natural silver contamination in A3-27 is estimated at 2.7 ng/g, which translates to about 1.3 x 10^{13} 10⁹ Ag atoms per cm³.

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The graphite cups and steel capsules also contain silver as a contaminant. At Studsvik during the R2-K12 irradiation tests, the reserve graphite cups used in that irradiation rig were measured prior to irradiation. Silver contamination was found to be as high as 180 ng/g [25], and for low-temperature irradiation tests, these contaminations dominate the measured silver outside the test sphere. Therefore, total silver contaminations between 8 x 10^{12} and 2.6 x 10^{13} 109 Ag atoms per cm³ for each test capsule must be considered.

Using all input data from both Christ and Venter discussed in paragraph 5.1.3.1, and contamination sources discussed above, diffusion coefficients were derived for all four test spheres in Table 12. The temperatures and diffusion coefficients listed are the average centre fuel temperatures for each test sphere and diffusion coefficients at those specific temperatures. For the hotter test spheres there is very little difference between diffusion coefficients derived from the two data sets. For the two cooler test spheres, contamination of irradiation rig materials dominate measured release fractions and derived diffusion coefficients are much more dependent on the data set used. A range of contamination values was considered in order to derive realistic diffusion coefficients. For the purpose of final evaluation of all derived diffusion coefficients, the Venter data coefficients may be seen as the lower limit and the Christ data coefficients as the upper limit of the range of coefficients for this test.

Fuel		Venter Data	Christ Data		
Sphere	Temperature	Coefficient $(m2s-1)$	Temperature	Coefficient $(m2s-1)$	
	1231 °C	7.75×10^{-18}	1247 °C	7.63 x 10^{-18}	
2	1121 °C	6.58×10^{-18}	1 099 °C	1.37×10^{-17}	
3	1 115 °C	5.96 x 10^{-18}	1 099 °C	1.50×10^{-17}	
4	1 278 °C	2.54×10^{-17}	1 285 °C	2.55×10^{-17}	

Table 12: Derived Diffusion Coefficients: HFR-K3

HFR-K3 is the most important irradiation test to evaluate not only silver but also caesium release from fuel from the German irradiation programme. It is the best-documented test that also underwent full post-irradiation examination and irradiation conditions achieved during the test is the most applicable to future HTRs. Diffusion coefficients derived here can therefore be considered the most valuable in deriving final transport data.

5.2 FRJ2-K13

5.2.1 Reactor

The FRJ2-DIDO reactor in Jülich, Germany, is a heavy-water-moderated and -cooled material test reactor with a nominal power of 32 MW_{th} . The facilities for irradiating samples in core are limited to a diameter of 52 mm. Full sphere tests are therefore not possible inside the core and fuel sphere tests are performed in reflector positions. The neutron spectra are therefore very well moderated and high fast fluences are precluded from fuel sphere test requirements. On the other hand, very high thermal neutron fluxes could be achieved as the irradiation rig could be moved into the radial maximum of the thermal neutron flux profile.

Similar to the HFR BEST-rig, four spherical fuel elements are tested simultaneously in a two individually swept capsule irradiation rig shown in Figure 14. Temperatures in the rig are measured by nine thermocouples, and temperature is controlled by a binary mixture of helium and neon. During reactor operation neutron fluxes are measured at various positions in the core in order to evaluate neutron fluxes at the irradiation positions. In addition the integrated fluence is measured with neutron activation wires inserted into ceramic and steel tubes. Full sphere irradiation tests of interest performed were FRJ2-K11, -K13 and -K15. Several other irradiation tests were done on compacts and fuel elements but only these three fuel experiments are considered for silver transport evaluation.

5.2.2 Irradiation test

Four elements with AVR reload 19 fuel spheres with LEU-TRISO fuel were inserted into the reflector outside the core in a two-capsule irradiation rig. Each capsule contained two spheres and consisted of a steel container with graphite cups which houses the test spheres. Fuel spheres were numbered as K13/1, K13/2, K13/3 and K13/4 with planned irradiation temperatures of 985 °C, 990 °C, 990 °C and 980 °C surface and 1 131 °C, 1 149 °C, 1 148 °C, 1 127 °C centre, respectively [29]. FRJ2-K13 started on 24 June 1982 and ended successfully on 12 February 1984. Burn-up values of 7.5, 8.0, 7.9, and 7.6% FIMA were attained, with fast fluences of $\sim 0.2 \times 10^{25}$ m⁻².

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The objectives for this test were as follows [25]:

- a. Irradiation test of AVR reload 19 fuel.
- b. Providing irradiation data under highly controlled irradiation conditions.
- c. Examination of coated particle performance without the influence of fast fluence.
- d. Investigation of transport coefficients of metallic fission products.
- e. Supplying irradiated fuel for accident simulation tests in the KÜFA.

Post-irradiation examination was completed, with full gamma spectrometric analyses and deconsolidation of the rig. All capsule components and graphite cups were leached and fission products in the solutions determined with gamma spectrometry. Further gamma spectrometric measurements were made by drilling samples in the fuel-free zone of the fuel elements to determine the ^{137}Cs and ^{110m}Ag profile in the fuel-free zones of test spheres 1, 2 and 3.

Test sphere FRJ2-K13/2 was heated after irradiation at 1 600 °C followed by deconsolidation of the element to get loose particles. Ceramographic investigations of these particles showed punctuated deposits of metallic fission products in the kernel and coagulated pores. Test sphere FRJ2-K13/4 underwent accident testing up to 1 800 °C. The test spheres were of the same manufacturing batch and all fuel sphere data used in evaluating silver transport are the same as for the HFR-K3 evaluation. The ^{85m}Kr fractional release remained very low $(< 10^{-6}$) so it can be assumed that no particles failed during irradiation nor were there any particles defective from manufacture.

5.2.3 Evaluation

Much less information about the irradiation conditions is available compared to HFR-K3, but measured irradiation temperatures are available from published graphs and good estimates of the neutron fluxes can be made from reported values [35]. Cross sections were selected to achieve correct burn-ups and plutonium contributions supplied in the literature [25]. Fission power values determined from a neutronic calculation and estimated gamma-powers were then used to determine fuel temperatures. Coolant gas temperatures were adjusted to ensure that calculated and measured surface temperatures agreed. Centre fuel temperatures were calculated from surface temperatures and total power produced (fission and gamma) in each test sphere. The fission powers (in watt) and the surface and centre fuel temperatures (in $^{\circ}C$) are presented in Table 13 and Table 14.

Table 13: FRJ2-K13/1 and /2: Fission Power, Surface and Centre Temperatures

Table 14: FRJ2-K13/3 and /4: Fission Power, Surface and Centre Temperatures

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Fractional releases of silver from test spheres during irradiation are listed in Table 15. The fraction of silver on the steel capsules for test spheres 1 to 3 has a constant value of 1.3 x 10^{-2} although fractions on the graphite cups vary between 3.7 x 10^{-3} to 7.5 x 10^{-3} , which suggests that the silver fraction from the steel capsules is dominated by some other source than the fuel sphere. This is out of line with measurements made after the HFR-K3, R2-K12 and R2- K13 irradiation tests where capsule steel fractions were always less than graphite cup fractions. The fraction of silver on the steel cups for test sphere 4 is 3.1×10^{-2} which is much higher than for the other spheres although the factions on the graphite cups remain comparable. The sphere 4 measurement must be questioned and most probably is a transcription error where the 1 and the 3 have been swapped. If this is the case, the silver fraction for all capsules is exactly the same and may be from silver contamination of the steel. Considering a reasonable silver contamination of 10 ppm of the capsule steel could explain the measured silver fraction on the steel capsule.

Fuel Sphere	Steel Capsule	Graphite Cup	Total Fractional Release
	1.3×10^{-2}	5.7 x 10^{-3}	1.9×10^{-2}
	1.3×10^{-2}	7.5×10^{-3}	2.0×10^{-2}
	1.3×10^{-2}	3.7×10^{-3}	1.7×10^{-2}
	3.1 (1.3) x 10^{-2}	8.0×10^{-3}	3.9 (2.1) \times 10 ⁻²

Table 15: Fractional ^{110m}Ag Release from Fuel Elements of FRJ2-K13 [25]

Natural uranium and thorium contamination of the graphite cups that housed the test spheres were treated in the same way as for the HFR-K3 evaluation. Similarly, it was assumed that comparable silver contamination of the graphite cups existed. Much uncertainty remains about silver contamination of the steel capsules. For best estimate analyses it was assumed that the silver fraction measured from the steel capsules was predominantly from natural silver contamination of rig materials and a fraction of 1×10^{-2} was deducted from the measured fraction of this source. This leaves a fraction of 3×10^{-3} from test sphere release which is significantly more than what was measured in HFR-K3 and R2-K12, and very similar to what was measured during R2-K13. By ignoring any silver contamination in the steel capsules and assuming that all measured silver originated from the test spheres, an upper limit diffusion coefficient can be derived.

The resulting diffusion coefficients are shown in Table 16. The lower diffusion coefficient set will be used in deriving the final best estimate coefficients, and the upper limit results to derive final coefficients that may be used for safety and design analyses.

Fuel		Best Estimate	Upper Limit		
Sphere	Temperature	Coefficient $(m2s-1)$	Temperature	Coefficient $(m2s-1)$	
	1 131 °C	1.67×10^{-17}	1 131 °C	2.56×10^{-17}	
$\overline{2}$	1 149 °C	1.71×10^{-17}	1 149 °C	2.57×10^{-17}	
3	1 148 °C	1.43×10^{-17}	1 148 °C	2.37×10^{-17}	
$\overline{4}$	1 127 °C	1.79×10^{-17}	1 127 °C	2.64×10^{-17}	

Table 16: Derived Diffusion Coefficients: FRJ2-K13

5.3 FRJ2-K15

5.3.1 Irradiation test

Three elements from AVR reload 21-1 with LEU-TRISO fuel were inserted into the reflector outside the core in a three-capsule irradiation rig as is shown in Figure 15. Each capsule contained one sphere and was individually swept and temperature-controlled. Fuel spheres were numbered as K15/1, K15/2 and K15/3 with planned irradiation temperatures of 808 $^{\circ}$ C, 980 °C and 803 °C surface and 970 °C, 1 150 °C and 990 °C centre, respectively [36]. FRJ2-K13 started on 4 September 1986 and ended successfully on 21 October 1990. Burn-up values of 14.1, 15.3, and 14.8% FIMA were attained, with fast fluences of $\sim 0.2 \times 10^{25}$ m⁻² [37]. The objectives for this test were as follows [25]:

- a. Irradiation test of AVR reload 21-2 fuel (type GLE-4).
- b. Experimental demonstration of high burn-up potential of LEU TRISO particle.
- c. Determination of coated particle performance under high burn-up.
- d. R/B measurements during transients tests at different burn-ups.
- e. Examination of fission product transport.

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Post-irradiation examination was completed, with gamma spectrometric analyses and deconsolidation of the rig. Only the graphite cups were analysed for fission products with gamma spectrometry. The ^{85m}Kr fractional release remained very low ($\leq 10^{-6}$) so it can be assumed that no particles failed during irradiation nor were there any particles defective from manufacture. All three test elements underwent KORA corrosion and KÜFA heat-up testing after irradiation.

5.3.2 Evaluation

Fuel characteristics and final irradiation data of the test spheres are provided in Table 17 [38]. The silver fractional release for test sphere 3 was not successfully measured and is not discussed further.

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Figure 15: FRJ2-K15: Irradiation Rig

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Fast and thermal neutron fluxes are available from irradiation progress reports. Cross sections were selected to achieve correct burn-ups and plutonium contributions. Fission power values determined from a neutronic calculation and estimated gamma-power values were used to determine fuel temperatures. Coolant gas temperatures were adjusted to ensure that calculated and measured surface temperatures agreed. Centre fuel temperatures were calculated from surface temperatures and total power produced (fission and gamma) in each test sphere. Calculated fission powers, surface temperatures (from irradiation progress reports) and calculated centre temperatures are listed in Table 18. Also included are initial Hochtemperatur Anlage (HTA) calculated fission power values extracted from original irradiation progress reports. Not all the progress reports could be found and in some cases HTA calculations were not performed.

Table 18: FRJ2-K15/1 and /2: Fission Power, Surface, and Centre Temperature

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Fractional releases of silver from the test spheres during irradiation are listed in Table 19. AVR reload 21 fuel represents the best-quality German fuel manufactured and considerably lower heavy metal contaminations in the matrix materials were achieved than for previous batches. Natural uranium and thorium contamination of graphite cups that housed the test spheres were treated in the same way as for HFR-K3 and FRJ2-K13 evaluations. Similarly, it was assumed that comparable silver contamination of the graphite cups existed.

No measurement data for fractional silver release on steel capsules are available in the literature but only fractional release activities of the graphite cups on spheres 1 and 2. Considering the activity ratios of FRJ2-K13, it was conservatively assumed to use a capsule activity equal to the cup activity for best estimate analyses, and two to three times the cup activity for design limit values. Correspondingly, the natural silver contaminations in the steel capsules were also adjusted.

Fuel Sphere	Steel Capsule	Graphite Cup	Total Fractional Release
	7.5×10^{-4}	7.5×10^{-4}	1.5×10^{-3}
	3.2×10^{-3}	3.2×10^{-3}	6.4×10^{-3}

Table 19: Fractional ^{110m}Ag Release from Fuel Elements of FRJ2-K15 [38]

Resulting diffusion coefficients are shown in Table 20. The lower diffusion coefficient set will be used in deriving the final best estimate coefficients, and the upper limit results to derive final coefficients that may be used for safety and design analyses. The diffusion coefficient derived for the lower temperature sphere 1 is highly dependent on the level of natural silver contamination assumed in the evaluation. The natural silver contamination has therefore been selected inside the acceptable range to give a diffusion coefficient as conservative as possible but still in line with other diffusion coefficients derived in the detailed evaluation. The diffusion coefficient for sphere 1 is rather too high than too low and is also the only coefficient derived during this evaluation that exceeds the current IAEArecommended diffusion coefficient (refer to Figure 17).

5.4 R2-K12

5.4.1 Reactor

The R2 reactor in Studsvik, Sweden, is a light-water-moderated and -cooled reactor similar to the HFR in Petten. It generates 50 MW thermal power and utilizes 90% enriched fuel. Suitable irradiation conditions for sphere irradiation are available in both the core and reflector regions, with high thermal and fast neutron fluxes in the core.

5.4.2 Irradiation test

In this experiment, four fuel elements were irradiated in a four-capsule rig in the R2 reactor core. Two elements contained mixed oxide $(Th, U)O₂ TRISO$ particles and the other two elements contained a two-particle system. The two-particle fuel was an investigation into the viability of combining fissile UC_2 and fertile ThO₂ TRISO particles. The two-particle system was abandoned and only the one-particle test spheres are considered here. The two test spheres of interest were inserted into the top two capsules of the irradiation rig shown in Figure 16 [25].

Basic fuel and irradiation parameters are listed in Table 21. The test spheres contained 10 960 TRISO particles imbedded in A3-27 matrix material. The total heavy metal load of 6.08 g per sphere consisted of 1.12 g uranium enriched to 89.6% and 4.96 g thorium. Effective uranium contaminations used in evaluations were 2.2×10^{-5} and 1.0×10^{-6} in fuel and fuel-free zones respectively [39]. This was a two times accelerated test that ran for 308 effective full power days. The ^{85m}Kr fractional release remained very low ($\leq 10^{-6}$) so it can be assumed that no particles failed during irradiation nor were there any particles defective from manufacture.

Each capsule consisted of a steel container with graphite cups which housed the test spheres. Fuel spheres were numbered as R2-K12/1 and R2-K12/2 with planned irradiation temperatures of 950 °C and 1 120 °C surface and 1 100 °C and 1 280 °C centre, respectively [37]. Instrumentation consisted of five thermocouples per sphere and wire flux monitors as well as sweep and temperature regulation tubes for each single capsule. R2-K12 started on 28 November 1978 and ended successfully on 12 February 1980. Burn-up values up to 12.4% FIMA were attained, with fast fluences of 6.9×10^{25} m⁻².

Table 21: R2-K12 Test Element Specification and Irradiation Data

The objectives for this test were as follows [25]:

- a. Accelerated reference test on HEU-TRISO fuel spheres with different particle types.
- b. Evaluating irradiation conditions corresponding to a 3 000 MWth process heat plant.
- c. Examination of mechanical performance of particles of 1977 standard quality.
- d. Evaluating the differences between one- and two-particle fuel systems.

The capsules underwent gamma-scanning and deconsolidation at Studsvik before being sent to the KFA at Jülich for final analyses. Fission product inventories were measured in test spheres and capsule components in order to determine fractional releases

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Figure 16: R2-K12: Irradiation Rig for Spherical Fuel Elements

5.4.3 Evaluation

A first evaluation of R2-K12 fission product release was performed by Acharya in 1983, shortly after completion of the first post-irradiation examinations [39]. In the Acharya report mention is made of other analyses performed by Muncke, but these reports could not be found. These studies did not try to derive transport parameters for silver in the different fuel materials, but attempted to explain the observed results using transport parameters and models accepted at that time. Results from both studies are compared without explanation of all observed results. A more serious attempt to explain observed release fractions are discussed later by Röllig and Muncke (HRB) as reported by Gontard [25], and their recommendations are used in this evaluation.

5.4.3.1 Input data

Measured surface temperatures [40] and thermal neutron fluxes [39] are available directly from the literature and fast neutron fluxes can be derived from fast neutron fluence values. Surface temperatures and neutron fluxes used in the evaluation is presented in Table 22.

5.4.3.2 Evaluation results

Fission powers produced during irradiation and total burn-up achieved were calculated to ensure that calculated temperatures are as close as possible to actual values. Fission power values were calculated using best available thermal neutron fluxes (Table 22) and cross sections that have been corrected to yield the correct burn-up. These newly calculated fission power values were then used with literature-supplied gamma heating to determine fuel temperatures. He/Ne coolant gas temperatures were adjusted to ensure that calculated and measured surface temperatures agree. Centre fuel temperatures were then calculated from surface temperatures and total power produced (fission and gamma) in each test sphere. Fission power values as calculated by HBK and this evaluation are presented with the gamma heating for each cycle in Table 23.

Centre fuel temperatures were calculated for both GA and HBK studies and are presented with this evaluation results in Table 24. There is not a very big variation in calculated centre temperatures between the three calculations, even though all three sets use different thermal conductivity relations for A3-27 matrix material.

Table 22: R2-K12: Surface Temperature (°C) and Neutron Flux $(10^{18} \text{ m}^2 \text{s}^{-1})$

Table 23: R2-K12: Fission and Gamma Power (W)

Table 24: R2-K12: Calculated Centre Temperatures

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Fractional releases of silver from test spheres during irradiation are listed in Table 25. Röllig and Muncke estimated natural silver contamination in A3-27 to be in the order of 2.7 ng/g which translates to about 1.3×10^{13} 10⁹Ag atoms per cm³. At Studsvik during the R2-K12 irradiation tests, reserve graphite cups used in that irradiation rig were measured prior to irradiation. Silver contamination was found to be as high as 180 ng/g or around 8.3 x 10^{14} 109 Ag atoms per cm³ [25]. It is not known if the reserve graphite cups are really representative of the irradiated graphite cups. For example, the irradiated graphite cups may have undergone high-temperature annealing prior to insertion in the irradiation rig that may have reduced the natural silver contamination. By reducing this contamination by a factor of 10 and taking into account the relative weight of the graphite cups, a maximum 109 Ag contamination in graphite materials of 7.8×10^{13} atoms/cm³ was used. The case where natural silver contamination might have been reduced to levels estimated for other irradiation rig graphite cups (1.3 x 10^{13} 10⁹Ag atoms/cm³) have been investigated as well.

Fuel Sphere	Steel Capsule	Graphite Cup	Total Fractional Release
	3.7×10^{-3}	2.9×10^{-2}	3.3 x 10^{-2}
	9.1×10^{-3}	4.9×10^{-3}	1.4×10^{-2}

Table 25: Fractional 110m Ag Release from the Fuel Elements of R2-K12 [39]

After considering recommendations from previous evaluations and all calculated results, diffusion coefficients were derived for both test spheres in Table 26. The temperatures and coefficients listed are the average centre fuel temperatures for each test sphere and coefficients at those specific temperatures. For evaluation where the spread of data is limited due to a lack of detailed information, an upper limit of a factor of two higher than the best estimate value was suggested by Röllig [21].

Table 26: Derived Diffusion Coefficients: R2-K12

Fuel		Best Estimate	Upper Limit		
Sphere	Temperature	Coefficient $(m2s-1)$	Temperature	Coefficient $(m2s-1)$	
	$1\,123\,^{\circ}\mathrm{C}$	7.39×10^{-18}	$1\,123\,^{\circ}\mathrm{C}$	1.48×10^{-17}	
\mathcal{D}	1289 °C	1.68×10^{-17}	1289 °C	3.36×10^{-17}	

5.5 R2-K13

5.5.1 Irradiation test

This experiment was a combined test with high-enriched $(Th, U)O₂$ spherical fuel and lowenriched UCO/ThO₂ block fuel segments from a block fuel assembly. Initially it was planned to test LEU fuel but due to production delays HEU fuel was tested, and this test was therefore the last test performed on HEU spherical fuel. The two fuel spheres were inserted into capsules 1 and 4 of a four-capsule fuel rig.

Basic fuel and irradiation parameters are listed in Table 27. The test spheres contained 19 780 EUO 1674 TRISO particles imbedded in A3-27 matrix material. The total heavy metal loading of 11.3 g per sphere consisted of 1.14 g uranium enriched to 89.0% and 10.1 g thorium. Effective uranium contamination and natural silver content used in evaluations were the same as for the R2-K12 evaluation. This test ran for 517 effective full power days after which burn-up values up to 10.2% FIMA and fast fluences of 8.5 x 10^{25} m⁻² was attained. The ^{85m}Kr fractional release remained very low $(< 10^{-6}$) so that it can be assumed that no particles failed during irradiation nor were there any particles defective from manufacture.

Each capsule consisted of a steel container with graphite cups which housed the test spheres. Fuel spheres were numbered as R2-K13/1 and R2-K13/4 with planned irradiation temperatures of 960 °C and 750 °C surface and 1 170 °C and 980 °C centre, respectively [37]. R2-K13 started on 22 April 1980 and ended successfully on 19 September 1982. Burnup values up to 12.4% FIMA was attained, with fast fluences of 6.9 x 10^{25} m⁻².

The objectives for this test were as follows [25]:

- a. Demonstration test of HEU-TRISO fuel spheres irradiation performance.
- b. Evaluating fission product transport behaviour.
- c. Examination of particles performance under long-term irradiation.
- d. Evaluating A3-27 matrix material irradiation behaviour.
- e. Supplying spherical fuel elements for PIE annealing tests.

The capsules underwent gamma-scanning and deconsolidation at Studsvik before being sent to KFA at Jülich for final analyses. With results from gamma spectrometry on graphite cups, leach tests on steel capsules together with inventory measurements and calculations, relative fission product release from the fuel was investigated. Both fuel spheres were deconsolidated and particles were analysed. It was found that coated particle retention of fission products were exceptional and only $110m\text{Ag}$ had been released in detectable quantities.

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Table 27: R2-K13 Test Element Specification and Irradiation Data

5.5.2 Evaluation

A first evaluation of R2-K13 fission product release was performed by Muncke, but these reports could not be found. A short discussion of this work is provided in the Gontard report [25].

5.5.2.1 Input data

Measured surface temperatures and neutron fluence values are available directly from the literature [41]. Calculated fission powers are also available with measured R/B values for ^{85m}Kr. The surface temperatures and neutron fluxes used in this evaluation are listed in Table 28.

Table 28: R2-K13: Surface Temperature (°C) and Neutron Flux $(10^{18} \text{ m}^{\text{-2}} \text{s}^{\text{-1}})$

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5.5.2.2 Evaluation results

Fission power values produced during irradiation and total burn-up achieved were calculated to ensure that calculated temperatures were as close as possible to actual values. Fission power values were calculated using best available thermal neutron fluxes (Table 28) and cross sections that have been corrected to yield the correct burn-up. These newly calculated fission power values where then used with literature-supplied gamma heating to determine fuel temperatures. He/Ne coolant gas temperatures were adjusted to ensure that calculated and measured surface temperatures agree. Centre fuel temperatures were calculated from surface temperatures and total power produced (fission and gamma) in each test sphere. Fission power values as calculated by HBK and this evaluation are presented with gamma heating for each cycle in Table 29. Centre fuel temperatures were calculated and are also included in Table 29.

Table 29: R2-K13: Fission and Gamma Power (W) and Centre Temperatures (°C)

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Fractional releases of silver from test spheres during irradiation are listed in Table 30. Diffusion coefficients were derived for both test spheres in Table 31 using the same reasoning as for the R2-K12 evaluation. Natural silver contaminations between 1.3 and 7.8 x 10^{13} 109 Ag atoms/cm³ were investigated. For best estimate analyses, the higher value was used and for upper limit evaluations the lower number was used. Temperatures and coefficients listed are average centre fuel temperatures for each test sphere and coefficients at those specific temperatures. For evaluation where the spread of data is limited due to a lack of detailed information, an upper limit of a factor of two higher than the best estimate value was suggested by Röllig [21].

Table 30: Fractional ^{110m}Ag Release from Fuel Elements of R2-K13 [25]

Table 31: Derived Diffusion Coefficients: R2-K13

5.6 FRJ2-K11

Irradiation data, temperature graphs and fuel parameters were summarized in a number of HBK quarterly reports [42], [43] and [44]. The irradiation report [45] contains early handwritten tables of first estimations of thermal neutron fluxes, burn-up and power production. Irradiation test conditions were extracted from all these sources.

5.6.1 Irradiation test

Two spheres from AVR reload 13 and two spheres from AVR reload 15 with HEU-TRISO fuel were inserted into the reflector outside the core in a two-capsule irradiation rig. FRJ2- K11 started on 27 April 1979 and ended successfully on 30 October 1980. Burn-up values of 10.0 and 9.7% FIMA were attained, with fast fluences of $\sim 0.2 \times 10^{25}$ m⁻² [43]. ^{85m}Kr fractional release remained very low $(< 10^{-6}$) so that it can be assumed that no particles failed during irradiation nor were there any particles defective from manufacture. Post-irradiation examinations were performed on all test spheres but fractional fission product releases were successfully measured for spheres 3 and 4 only.

5.6.2 Evaluation

A first evaluation of fission product release from FRJ2-K11 was performed by Muncke, but the report could not be retrieved. Fuel characteristics [44] and final irradiation data [43] for test spheres are provided in Table 32.

Parameter	Unit	3	$\boldsymbol{4}$			
Specification						
Sphere weight	g	200	200			
Uranium content	g	1.12	1.12			
235 U enrichment	$\frac{0}{0}$	89	89			
Thorium content	g	4.9	4.9			
CP content		10 700	10 700			
Failed CP content	per FE	θ	θ			
Irradiation Data						
Irradiation time	EFPD	260	260			
Burn-up (FIMA)	$\frac{0}{0}$	10.0	9.72			
Neutron fluence	10^{25} m ⁻²	0.2	0.2			
$(> 0.1 \text{ MeV})$						
Average centre temperature	$\rm ^{o}C$	1 1 8 3	1 1 7 6			
Max power output	kW/FE	2.55	2.44			

Table 32: FRJ2-K11 Test Element Specification and Irradiation Data

Fast neutron fluxes in the reflector of the DIDO reactor are very low and values similar to FRJ2-K13 evaluation were used. Cross sections were selected to achieve correct burn-ups. Fission power values determined from a neutronic calculation and estimated gamma-power values were used to determine fuel temperatures. In this test it was attempted to keep surface fuel temperatures constant in the region of 1 000 °C. Therefore coolant gas temperatures were adjusted to ensure that calculated and experiment-estimated surface temperatures agreed. Calculated fission power values, surface temperatures and calculated centre temperatures are listed in Table 33.

Post-irradiation-examinations were performed at Harwell in the UK [46]. Fractional releases of silver from test spheres during irradiation were 4×10^{-2} for both spheres according to the project report [43] although a fraction of 5.4×10^{-2} for test sphere 4 has also been reported in other literature [23]. Natural uranium and thorium contamination in the matrix material of the test spheres were in the order of 1×10^{-4} [47]. Heavy metal contamination in the graphite cups that housed the test spheres were treated in the same way as for the FRJ2-K13 evaluation. Similarly, it was assumed that comparable silver contamination of the graphite cups existed. Resulting diffusion coefficients are shown in Table 34. The lower diffusion coefficient set was calculated using best estimate input data and will be used in deriving final best estimate coefficients. Upper limit results are simply a factor of two higher than expected results and may be used for safety and design analyses.

Table 34: Derived Diffusion Coefficients: FRJ2-K11

5.7 Discussion of results

All six applicable irradiation tests have been evaluated with the best available information and assumptions based on the best available engineering judgement. The level of information available for each irradiation test varies greatly from almost complete neutronic and thermohydraulic histories available for HFR-K3 to only brief summaries for FRJ2-K11. Furthermore it is unknown whether the quality of the SiC layers changed significantly from the early tests to the latest. Comparing derived diffusion coefficients for each test sphere of each test in Table 35, there are significant differences between diffusion coefficients derived for the earliest (FRJ2-K11) test and the latest (FRJ2-K15) tests. Similarly, there appears to be an influence from the irradiation facility used. The FRJ2 tests appear to have higher diffusion coefficients compared to the HFR and R2 tests.

What is important for all irradiation tests, irrespective of facility or fuel tested is the effect of natural uranium, thorium and silver contamination occurring in the fuel and irradiation rig materials. Especially for lower irradiation temperatures $(1100 \degree C)$, naturally occurring silver in graphite cups dominate measured fractional release. Natural silver contamination was only measured on R2 graphite cups and estimated for A2-27 matrix material used in fuel sphere manufacture. Silver contamination in irradiation capsule steels is unknown; however, measurements made during FRJ2-K13 appear significant. Another complicating effect that contributes to uncertainty in results is the efficiency of wet chemistry techniques used to leach and remove released silver from irradiation rig materials and the accuracy of gamma spectrometry used to measure ^{110m}Ag activities in leach solutions.

Considering the above, it must be asked whether all irradiation tests should be weighed equally in determining a final diffusion coefficient for silver in SiC. Confidence in diffusion coefficients derived from the HFR-K3 irradiation test is higher than in any of the other evaluations and is significantly higher than for FRJ2-K11. If weighing of results is to be considered, the first question to be answered is, what weight to apply to which evaluation?

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This is highly subjective and since the 'better' evaluations generally produced lower diffusion coefficients, it can be viewed as an attempt to lower diffusion coefficients in order to simplify reactor analyses. It was therefore decided to rather consider all irradiation tests evaluated as equal and err on the conservative side. The only exception is where both evaluations performed for HFR-K3 using Christ and Venter data respectively will be used. In effect HFR-K3 irradiation test evaluation for spheres 1 and 4 will therefore be weighted by a factor of two. For the two colder spheres, 1 and 2, the Venter and Christ data represent the lower and upper limits and are used to evaluate best estimate and design limits.

All derived diffusion coefficients were plotted against average centre fuel temperatures in Figure 17. The following best estimate and design limit diffusion coefficients were derived by fitting all results to a straight line:

Best estimate: $D = 1.14 \times 10^{-13} e^{-109/RT}$ m²s⁻¹

Design limit:

 $D = 2.28 \times 10^{-13} e^{-109/RT} \text{ m}^2 \text{s}^{-1}$

The recommended IAEA diffusion coefficient is also plotted against temperature in Figure 17. All derived diffusion coefficients for all test spheres evaluated are below the recommended IAEA diffusion coefficient line except for the lowest temperature sphere (FRJ2-K15/1). Almost all design limit diffusion coefficients are also below the IAEA line with FRJ2-K11 and FRJ2-K15/1 being the exceptions. It appears that the currently recommended diffusion coefficient is overly conservative. Considering that the current silver diffusion coefficient in SiC was derived from particle heat-up tests after being irradiated in compacts, it might also be possible that the silver retention ability of a TRISO particle somehow improves during the sphere-making process. Also under consideration is the much higher diffusion coefficients derived for FRJ2-K11 and by Amian [48]. Amian performed his investigations on coated particles manufactured before 1978. This evaluation focused on fuel manufactured after 1978 (with the exceptions being FRJ2-K11 and R2-K12). The bestperforming fuels have been the ones irradiated in the later fuel tests: FRJ2-K15 and -K13 as well as HFR-K3. These are fuel spheres from AVR reloads 19 and 21, which were manufactured well after 1980. It could well be that Chemical Vapour Deposition (CVD) coater performance has increased, resulting in higher quality SiC that have superior silverretention abilities.

Table 35: Summary of Derived Diffusion Coefficients

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Both FRJ2-K11 diffusion coefficients' best estimate and upper limit values are approaching the Amian diffusion coefficient line (the official IAEA line in Figure 17). It could possibly be that the Amian line presents the SiC diffusion coefficient for TRISO particles manufactured before 1978 and the new diffusion coefficient derived in this detailed evaluation present the SiC diffusion coefficient for TRISO particles manufactured after 1980. Therefore it might be that there is no discrepancy between the Amian evaluation and this current study. It can then also be argued that diffusion coefficients derived for FRJ2-K11 should be removed as they belong to an 'older' fuel set that have inferior silver-retention abilities compared to the latest state-of-the art German reference fuel. However, justifying such an argument based on only a perceived lower silver-retention ability is difficult, and for the sake of conservatism, it is suggested that the FRJ2-K11 results remain in Table 35 and Figure 17.

Figure 17: Diffusion Coefficients from the Detailed Evaluation