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Long Time Experience with the Development of HTR Fuel Elements in Germany

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Abstract

The development of spherical fuel elements for HTR-designs in Germany are discussed. Special attention is given to the development, production and characterisation (incl. kernel and coatings) as well as to the irradiation and post irradiation examination of the different coated particle systems. It has been demonstrated in various irradiation tests which were supplemented by heating tests that for a modular HTR power plant (with a thermal output of 200 MJ/s) during the specified normal operation as well as in the case of incidents and even accidents, where the maximum fuel temperature will be below 1620°C, the fission product release is very low.

In this context, it must be mentioned that the present coated particle design has not yet been optimised for the combination of high burn-up and high temperature resistance under accident conditions. The TRISO fuel available is a result from fuel development for large HTRs with gas turbines in a time when the modular concept was not yet been invented although its capabilities inspired the design of modular reactors. Thus, there is still a huge potential for improvement of coated particles especially when plutonium or actinide burning is also taken into account.

1) Introduction

The HTR utilises an all-ceramic core, a graphite core structure, ceramic-coated particle fuels and complete ceramic fuel elements. The use of refractory core materials combined with a single phase inert helium coolant allows high coolant temperatures and results in a number of significant advantages including high thermal efficiency of the HTR and its inherent safety advantages resulting from the low-power density and large thermal capacity of the core, the absence of coolant phase changes, and the prompt negative temperature coefficient. These features ease reactor siting constraints by reducing both cooling water requirements and the consequences of postulated accidents.

The development of the HTR has proceeded in two directions: a) the pebble bed concept in the Federal Republic of Germany and Russia (now also in China and South Africa), and b) the prismatic core in the United States, the United Kingdom, Japan and, recently with the GT-MHR, also Russia. The fuel elements for the pebble bed system consist of 60 mm diameter spheres made up a fuel-free carbon outer zone and an inner-fuelled region with coated particles uniformly dispersed in a graphitic matrix. The prismatic fuel element consists of a machined hexagonal graphite block ~750 mm long and 350 mm across flats. Alternate fuel and coolant holes are drilled in a hexagonal array. Fuel rods, consisting of coated particles bonded in a close-packed array by a carbonaceous matrix, are stacked in the fuel holes.

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Although fuel elements in the two HTR designs differ substantially, the basic fuelcontaining unit, the coated particle is essentially the same, and coated fuel particle fuel development have been done as an international effort quite independent of differences in reactor design. 1

The following three experimental reactors have been developed and successful operated:

• <u>AVR</u> (Arbeitsgemeinschaft Versuchsreaktor): 1967-88, Pebble bed core, (Th,U)C₂, (Th,U)O₂, BISO and (Th,U)O₂ TRISO with high enriched uranium as well as low enriched UO₂ TRISO coated particles; 46 MW_{th}/15 MW_{el}; power density: 2.6 MW_{th}/m³; Helium pressure: 1 MPa; He inlet/outlet temperature: 270/950°C.

• <u>Peach Bottom 1</u> Reactor in US: 1967-74, Core with tube elements, (Th,U)C₂, BISO; 115 MW_{th}/40 MW_{el}; power density: 8.3 MW_{th}/m³; He pressure: 2.5 MPa; He inlet/outlet temperature: 377/750°C.

• <u>Dragon Reactor</u> in the U.K.: 1968-75, Core with tube elements, various (UO₂ driver fuel), TRISO; 20 MW_{th}; power density: 14 MW_{th}/m³; He pressure: 1.0 MPa; He inlet/outlet temperature: 350/750°C.

During the successful operation of the experimental reactors both HTR designs have been approached for the commercial operation with the development of the two HTR prototype plants:

The 330-MW_{el} <u>Fort St. Vrain (FSV)</u> reactor, built by General Atomic Company for the Public Service Company of Colorado in USA and

the 300 MW_{el} <u>Thorium High Temperature Reactor (THTR)</u> at Schmehausen in Germany.

The first of these utilises a prismatic core, while the second utilises a pebble bed core.

• <u>FSV:</u> 1976-89, 842 MW_{th}/330 MW_{el}; power density: 6.3 MW_{th}/m³; Prismatic core; (Th,U)C₂/ThC₂, TRISO; He pressure: 4.5 MPa; He inlet/outlet temperature: 405/784°C.

• <u>THTR</u>: 1986-89, 750 MW_{th}/300 MW_{el}; power density: 6 MW_{th}/m³; Pebble bed core; (Th,U)O₂, BISO; He pressure: 3.9 MPa; He inlet/outlet temperature: 270/750 °C.

These generation of HTRs has been operated on conventional steam cycle.

However, advanced designs with direct-cycle helium turbines and reformers for industrial process heat was under active development in Germany over two decades. Comparable HTR research activities has been done in United States and Russia as well as for a process heat HTR in Japan.

Behind the development of steam cycle HTR-plants like the THTR with 300 MW_{el}, the HTR-500 with 500 MW_{el} both with PCRV (Prestressed Concrete Pressure Vessel) in Germany and the FSV with 330 MW_{el} and PCRV in USA in both countries small high temperature reactors with steel pressure vessel are designed to achieve a so called catastrophe-free design, irrespective of probability for all practical purposes. These so called Modular HTRs with power output between 200 MW_{th} for the Siemens/Interatom and for the HTR-100 (250 MW_{th}) from BBC/HRB with a pebble bed core up to ~265 MW_{th} for the General Atomic prismatic core design. These Modular HTRs are designed such that the maximum fuel temperatures in

accidents remain below 1600°C without active control mechanisms. This modular design are intended to replace water-cooled reactors for electricity generation and to provide environment-friendly process heat for application such as heavy oil recovery, coal gasification and liquefaction. etc.

The German Reactor Safety Commission made in their recommendation in January 1990 e.g. the following statements to the HTR Modular Power Plant Concept developed by Siemens/Interatom. This system is characterised by the fact that several standardised nuclear heat production units of 200 MWth output are combined to form a power plant. The limitation of the reactor power to 200 MWth and of the mean power density to 3 MWth/m3 in connection with the core geometry has particular the following advantages: In the case of a failure of the main heat sink in the HTR Modul, residual heat removal is effected via passive heat conduction, heat radiation and natural convection to the surface coolers provided on the outside of the reactor barrel. Residual heat removal does not require any forced circulation inside the primary system. A maximum fuel temperature of 1620 °C is not exceeded, irrespective of whether residual heat removal remains intact per design intent during an incident or there is an additional failure of the residual heat removal via the surface coolers. Adherence to this maximum fuel element temperature is inherent safety feature of this reactor concept [1].

The HTR development is still on the way in different countries as it will be shown in the following:

In Japan has developed the experimental reactor HTTR with a thermal power of 30 MW which became critical in November 1998 and is on its way to full power. The major specifications of the HTTR are: Prismatic Block Core; Low enriched UO2; TRISO; He pressure: 4 MPa; He inlet/outlet temperature: 395/850 and 950°C; Steel containment; Heat removal IHX and PWC (parallel loaded).

China has built the test reactor HTR-10. The HTR-10 with a thermal power of 10 MW represents the features of modular HTR design, it became critical in the end of the year 2000. The HTR-10 main design parameters are: Modular HTR with a Pebble Bed Core; Low enriched fuel with 17 % U-235 UO2; TRISO; He pressure: 3 MPa; He inlet/outlet temperature: 250/300 and 700/900°C. Reactor core and steam generator are housed in two steel pressure vessel which are side-by-side with a connecting vessel between.

In South Africa, ESKOM as the national utility sees a nuclear future in the HTR pebble bed system. ESKOM successful operates the two unit Koeberg PWR station, but it does not see LWRs as a solution for the present. Rather, it is putting its technical and financial resources behind a HTR project which sees as the best approach to take. The concept design is concentrated on a 100 MWel Pebble Bed Modular Reactor (PBMR) with a direct cycle gas turbine.

2) Coated Fuel Particles

Coated particles are in themselves miniature fuel elements on the order of a millimetre in diameter. A commercial reactor core contains between 10⁹ and 10¹⁰ individual fuel particles. The coatings provide the primary barrier to fission product release. The very small size of coated particles is an advantage in testing, since statistically significant numbers of "fuel elements" can be tested. Individual tests typically contain 10³ to 10⁵ coated particles. As it will be shown through properly designed fuel development and test programs, fuel performance in-service can be predicted with high degree of confidence. Processes that lead to loss of coating integrity can be defined in detail and minimised by design either of the fuel particles themselves or of the reactor core.

The two coated particle types most common used are:

- · the two-layer BISO coating with porous buffer and dense pyrocarbon, and
- the four-layer TRISO coating with its interlayer of SiC between two layers of highdensity isotropic PyC.

Both BISO and TRISO particles are capable of complete retention of gaseous fission products and iodine with properly designed and specified coatings. Intact TRISO particles also provide essentially complete retention of metallic fission products at current peak HTR design temperatures. Because of diffusional release of certain metallic fission products, particularly caesium, strontium, and silver does occur at elevated temperatures from BISO coatings, fuel elements with TRISO coated particles are used (fig. 1) in all modern HTR designs.

One of the attractive features of the HTR is its flexibility in use of different fuel cycles. The thorium cycle with both separable and mixed fuel, the low-enriched uranium cycle, and even cycles based on plutonium fissile particles are feasible. Fuel particles characteristic of those required for each of these cycles have been successfully tested in prototype HTRs as well as in Materials Test Reactors.

The state of the art and the results of the international long-time research and experience in the field of HTR fuels up to the end of the seventieth is given in a special issue of Nuclear Technology on "Coated Fuel Particles" [2].

In the following this paper will be concentrated on the HTR fuel development and experimental data in Germany.

Coated particle development began in Germany in the 1960s and, in 1972, led to a particle design qualified for use in the THTR. This particle consisted of mixed thorium-uranium oxide kernel with a methane derived pyrocarbon coating. The (Th,U)O2 HTI BISO fuel design utilised with 93% enriched uranium (HEU-high enriched uranium). In the period between 1975 and 1980, the German reference particle coating design changed to the LTI TRISO coating which afforded a greater degree of resistance to fast neutron bombardment and a significantly higher degree of fission product retention as mentioned above. The thorium cycle was at that time used concerning optimisation the fuel economics and resource conservation. After a thorough study of low-enriched fuel particle performance, Germany and United States adopted low-enriched uranium fuel for all future HTR projects. In Germany, (low enriched uranium) LEU UO₂ was selected as the reference fuel kernel material in 1980. The reason for this action in Germany was the Non-proliferation aspects and therefore the decision to cancel the reprocessing step. The reference coating design remained the TRISO coating. The sequence for HTR fuel particle development is shown schematically in fig. 2.

2.1) Fabrication of Coated Particles and Spherical Fuel Elements

In Germany the coated particles as well as the spherical fuel element development and production has been in hand of the NUKEM Company. The Research Centre Juelich was responsible for the fundamental research of the material behaviour, especially under neutron irradiation conditions and the development of production procedures for small coated particles batches, producing of irradiation test samples, the irradiation tests at different material test reactors and HTR experimental reactors like Dragon and AVR and as well as the post-irradiation examination. 2.1.1) The <u>uranium-thorium kernel fabrication</u> was mainly based on the sol-gel process. This process has been developed for the production of the $(Th,U)O_2$ kernel and the ThO_2 kernel as well as for the uranium-dioxide kernel fabrication. Using this specified process a total of approximately 400 kg UO₂ kernels was successfully produced for the manufacture of all fuel elements scheduled for AVR reloads [3,4].

2.1.2) For <u>coating of the microspheres</u> the fluidized bed technique is used. The pyrocarbon (PyC) is a unique material that has been central to coated particle development from the earliest days. Results showed that the structure and irradiation behaviour of PyC coatings are highly dependent on deposition conditions, which in turn determine coating properties such as density and crystalline anisotropy. Many activities has been done in this area emphasising the optimisation processes and the development of improved characterisation techniques, also for the post-irradiation examination of the coated fuel particles and fuel elements [2].

The UO₂ or the other kernels are batch-wise coated in fluidized bed furnaces. An inert gas, usually argon, is used for fluidisation. First a porous buffer layer of C_2H_2 is deposited. This layer supplies a free volume for kernel swelling and fission gas production during burn-up and protects the following highly dense layers from recoil atoms. Next a high density inner pyrocarbon layer is deposited from a mixture of C_2H_2 and C_3H_6 . The layer SiC deposited from CH₃SiCl₃, predominantly for retaining the solid fission products, is brittle and therefore protected finally by an outer highly dense pyrocarbon layer [3,4].

2.1.3) <u>The production of spherical fuel elements</u> for HTRs consists behind the (i) fuel kernel casting and the (ii) coating of microspheres of the following steps: (iii) overcoating of particles; (iv) matrix powder preparation; (v) fuel element fabrication, i.e. pre-moulding of fuel zone, high-pressure isostatic pressing of complete element, machining, and 800/1950°C heat treatment; and (vi) quality control [3,4].

Table 1 shows the main particle and fuel data with German reference HEU and LEU particles.

3) Irradiation Testing of Coated Particles, Graphitic Matrix and Spherical Fuel Elements

The overall objective of the HTR fuel element development program was to qualify an element which minimises fission product release under normal and transient conditions for all types of HTR application as well as under accident conditions for small HTRs with a pebble bed core. Apart from fuel elements, the coated particles, the graphite matrix and the reflector graphite have been tested in several MTRs like HFR-Petten, R2-Studsvik, BR2-Mol, Siloé-Grenoble, FRJ2-Jülich as well as in the HTR test reactors AVR and Dragon. Long time tests have been carried out over more than 20 years with about twelve different spherical fuel element types in the AVR as a large-scale test bed [7].

A typical irradiation program for testing was directed to (i) "determination of particle failure rate" under conditions exceeding the demands of the HTR projects with reference to fast fluence and burn-up to investigate performance margins at irradiation temperatures (800-1200°C); (ii) "investigation of burn-up influence", irradiation of fuel in thermal test reactors with low fast neutron fluxes to separate burn-up controlled effects from neutron-induced effects (800-1300°C); (iii) "reference tests", demonstration of reference fuel elements under condition enveloping the demands on of different HTR systems on temperature, fast neutron fluence, burn-up and transient conditions. Fig. 3 shows the fluence/burnup correlation of the reference UO_2 fuel performance.

3.1) The <u>coated particle fuel systems</u> for HTRs has been investigated for nearly three decades and its fundamental performance characteristics and fission product release mechanisms are well understood. The testing range (temperature up to 1300 °C, burn-up up to 16 % FIMA and the fast fluence up to $8 \times 10^{25} \text{ m}^{-2}$) covers the conditions encountered in large steam-generating HTRs, in direct cycle, and in process heat applications. In individual cases, irradiation tests have been conducted beyond the testing range.

Information gathered from these tests form the basis for the understanding of particle fuel performance capabilities. Mechanisms that may result in particle failure and lead ultimately to fission product loss can be classified as: (i) coating damage during fuel manufacture, particularly during sphere pressing; (ii) pressure vessel failure during irradiation or during a temperature transient caused by internal gas pressure of irradiated particles and resulting in complete coating failure; (iii) failure of pyrocarbon layer due to neutron-induced embrittlement: (iv) failure of the SiC layer due to fuel kernel migration and interaction with the coating ("amoeba effect"); (y) failure of the SiC laver by chemical reaction with internally generated fission products; (vi) thermal decomposition of the silicon carbide at extremely high temperatures. Within the reguired operating conditions and projected accident scenarios, all the limits have been overcome either by coated particle design and material choices or by limiting reactor design parameters like the HTR Modul design. The particle manufacturing damage has been reduced to a fraction of less than 6x10⁻⁵ (design value) and a negligible heavy metal contribution in the spherical fuel element due to the natural uranium contamination of the graphite [6].

Fig. 4 shows a typical example of in-pile data measured or calculated in an irradiation experiment: specimen temperature, fast neutron fluence, burn-up and fractional fission gas release rates (released/birthrate) Kr-85m. The fission gas release during irradiation is an indicator of fuel quality and fuel performance [7].

3.2) The <u>graphitic matrix in the fuel element</u> has a number of essential functions. The fuel matrix acts (i) as a moderator for fission neutrons, the carbon density being of significant influence; (ii) provides for heat transfer from the surface of the coated particles to the surface of the fuel elements and must therefore exhibit good thermal conductivity; (iii) must ensure a non-destructive absorption of external forces and must therefore have high mechanical strength; (iv) a good resistance to corrosive attack due to impurities in the coolant gas and high dimensional stability during irradiation with fast neutrons. Isothermal irradiation and experiments for the determination of creep data were carried out in the HFR-Petten at maximum temperatures between 400 and 1450 °C up to the THTR operation time fluence of 3.5x10²⁵m⁻² EDN and in part beyond this fluence [7].

Fig. 5 illustrates the dimensional behaviour of the A3-3 matrix material and of AVR fuel elements as a function of the fast neutron fluence. The dimensional change are shown as areas limited in each case by the external isotherms. A diameter reduction of below 2% was determined for all spheres, but also for the matrix specimens at temperatures between 900 and 1150°C. The area of dimensional reduction of the AVR spheres is shaded. A general result is that the developed A3-3 matrix materials also in the improved A3-27 form always exhibit good dimensional stability even under high irradiation loads [7].

3.3) The <u>irradiation testing of spherical fuel elements</u> in accelerated and long-time tests was carried out as described above. During irradiation of spherical HTR fuel elements, important information about fuel performance could be obtained from the tests in the MTRs as well as from the AVR operation. But all essential properties of the fuel elements have been controlled mainly in the post-irradiation examinations in the Hot Cells e.g. (i) dimensional change of fuel elements; (ii) mechanical (crushing) strength; (iii) corrosion resistance; (iv) fission product behaviour, (v) accident simulation heating tests.

The source terms for fission products in the primary circuit of an HTR are:

- (i) heavy-metal contamination;
- (ii) particle manufacturing defects and in-pile particle failure;
- (iii) release from intact particles.

The important isotopes are Cs, Sr, Ag, I, Xe and Kr. From all the available data it can be pointed out that fission product release from the spherical HTR fuel elements during normal operation is insignificant. In the German HTR-Modul, for instance, the calculated release of Cs-137 accumulated during normal operation conditions after 32 operating years is approximately 2.6×10^{12} Bq (70 Ci). The coolant gas activity of the AVR after 20 years operating was approximately 5.5×10^{11} to 1.1×10^{12} Bq (20-30 Ci) of the noble gases, a few tenths of a MBq of aerosols and non-measurable amounts of iodine.

In the case of a core heatup accident, higher temperatures lead to enhanced diffusion of fission products out of the particle kernel through the TRISO coating and through the graphite [8,9]. One of the most important diffusion coefficients is that of caesium in SiC. On the basis of a number of heating experiments, it has been shown that an increased permeability of the SiC layer for caesium at temperatures of more than 1600°C. Fig. 6 shows results from fission product experiments in the form of diffusion coefficients in UO_2 , in pyrocarbon and silicon carbide coating layers, and A3 matrix as a function of temperature.

Summarising it can be pointed out that for the German reference LEU-TRISO fuel elements the release of solid fission products, e.g. Cs-137 from coated particles into the fuel element matrix and from there into the reactor core equals to the low release levels of gaseous fission products [3,7].

4) Heating Tests for Accident Condition Performance

Accident simulation tests have been performed since the mid-seventies whereby the Research Centre Jülich has concentrated on heating complete spherical fuel elements rather than single particles or small numbers of coated particles. An early experimental program had consisted of heat-up ramp tests with $(U,Th)O_2$ BISO fuel up to 2500°C. This program was followed by work with fuel elements containing $(U,Th)O_2$ TRISO and UO_2 TRISO particles. Special attention has been given to accident performance testing of the UO_2 TRISO particles for small HTRs [10, 11].

The fission gas release data from spheres during heating tests are shown in Fig. 7. The measured isotope is Kr-85 which give the same release as Xe-133 and I-131. As expected, release increases with heating temperature and duration. All 1600°C release results remain below the level of one particle failure $(6x10^{-5} \text{ fraction for 16400} \text{ particles})$. The shape of the release curves can be explained by the following two phenomena: (i) Deterioration of the SiC layer leads to permeability to fission products, but the remaining intact outer pyrocarbon layer delays the release of noble gases and iodine; (ii) On rare occasions can a burst of gas release be observed which is due to pressure induced complete coating failure.

Caesium is the main indicator of SiC deterioration or failure. Its release is further delayed by retention in matrix graphite. All caesium release curves from spheres are combined in fig. 8 as a function of heating times up to 500 hours. These results show an improvement in release by five orders of magnitude when compared to data from BISO particles.

5) Conclusions

The design of modern HTRs is based on high quality fuel. In manufacturing, this fuel quality has been achieved in the early eighties and than continually improved. This improvement consisted of steps to reduce TRISO particle defects during sphere pressing and minimising uranium contamination in the fuel element. For normal-reactor conditions, irradiation testing has been performed in material test reactors and in operation HTRs. Parameters such as heavy metal burn-up, operating temperature, and fast neutron fluence are varied to assess fuel performance. Continuous monitoring of released fission gas during irradiation tests gave a direct indication of the integrity of fuel coatings. In the German program, relevant irradiation tests with more than $2x10^5$ particles were performed without a single coated particle failure during irradiation. Statistically, this result corresponds to a 95% confidence level that the coating failure fraction is less than $2x10^5$.

Fuel testing under off-normal conditions has provided fuel performance information as a function of fuel temperature, up to 2500 °C. In small, modular HTRs, temperatures are limited to below 1600 °C. Here, the fuel does not suffer irreversible changes and continues to retain all safety-relevant fission products. Experiments have been performed with higher temperatures, longer heating times and with fuel from highly accelerated tests to establish the performance margins under accident conditions.

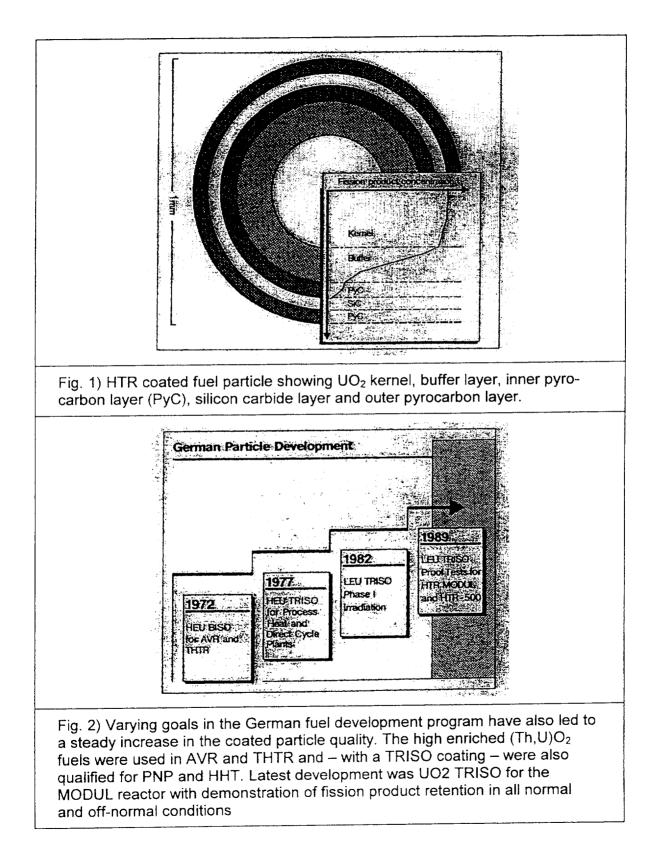
6) References

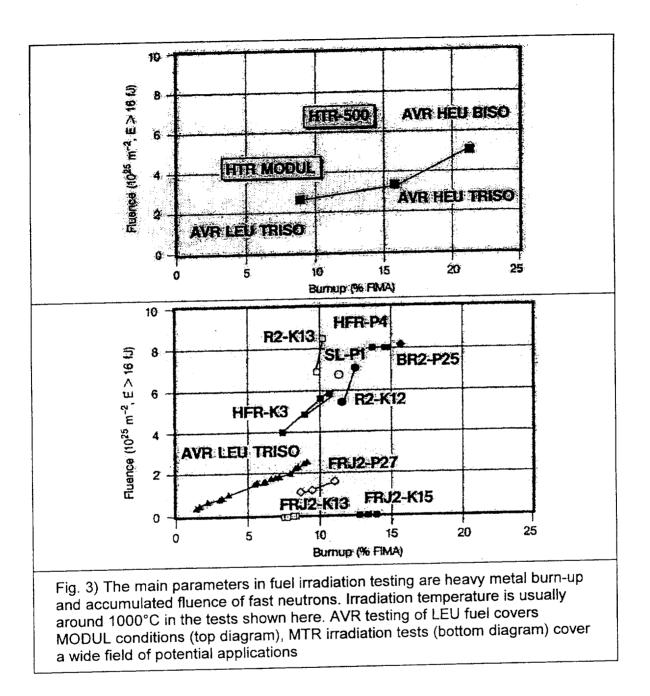
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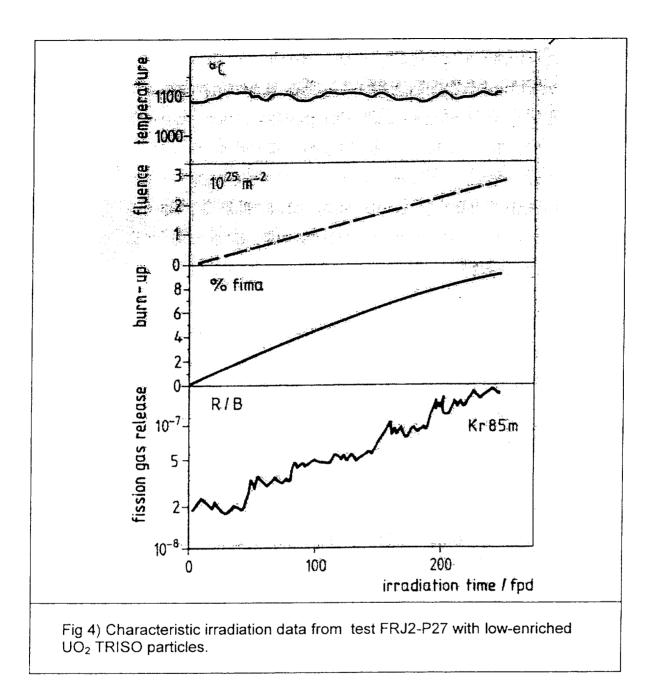
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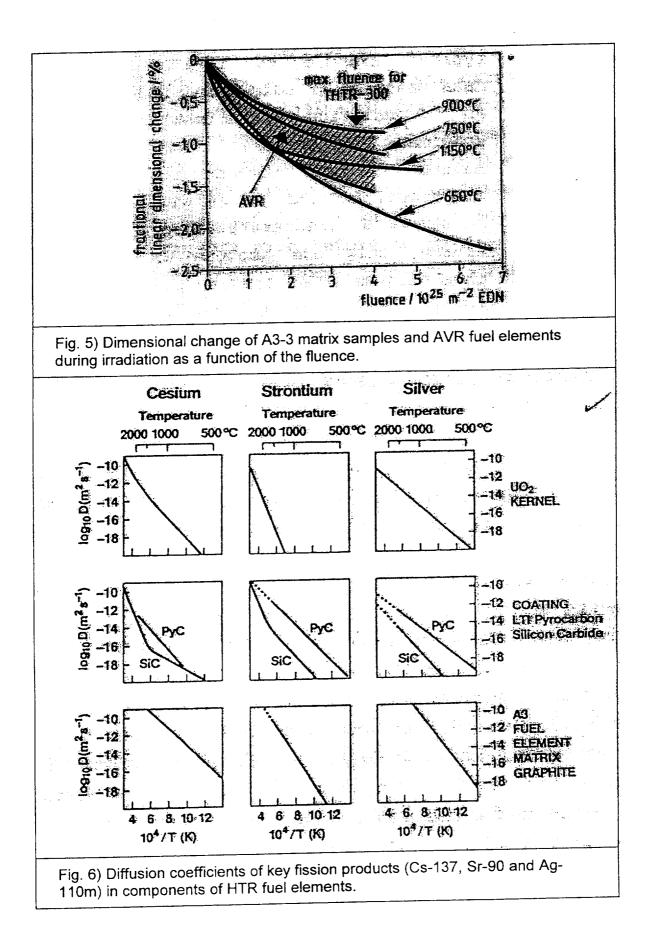
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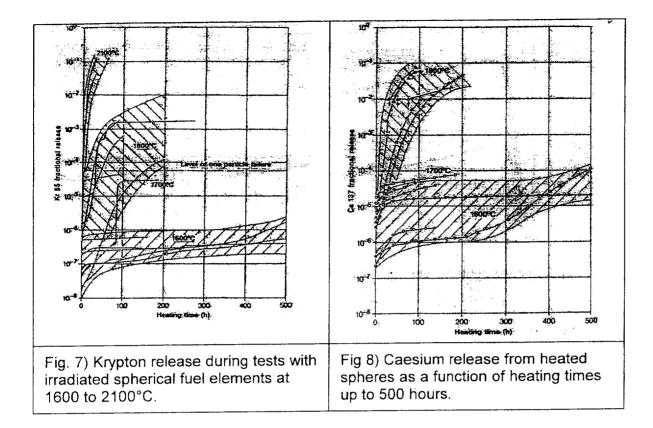
7) Figures



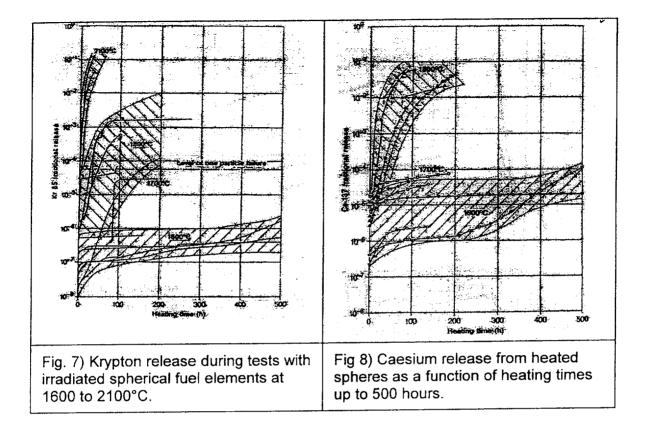




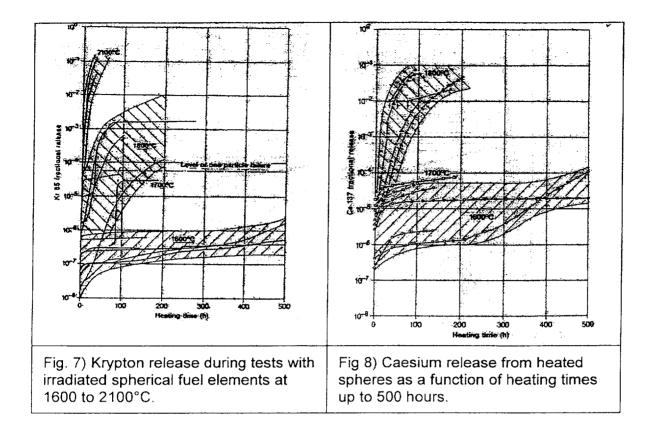




Design Parameter		HEU	LEU
Coated Particles			
Kernel Composition		(Th.U)O ₂	UO ₂
Kernel Diameter	um	500	500
Coaling Layer Thickness	μm	95/40/35/35	95/40/35/35
Coaling Layer Sequence		Buffer/PyC/SiC/PyC	Buffer/PyC/SiC/PyC
^c uel Element			
Heavy Metal Loading	g/Element	11	8-12
J 235 Ennchment		93 %	7-13 %
No. Particles per Element		19,000	13.000-20.000
Volume Loading of Particles		13 °°	10-15 %
Operating Requirements			
Mean Operating Time	d	1100-1500	700
Max, Burnup	MWdA _{HM}	120.000	90.000
Max. Fast Dose (E > 0 1 MeV)	10 ²⁵ m ⁻²	4.5	3.3
vlax, Fuel Temperature	°C	1020	1030
Viax. Power/Element	kW	2.7	4 1



D esi gn Parameter		HEU	LEU
Coated Particles			
Kernel Composition		(Th.U)O ₂	UO2
Kernel Diameter	um	500	500
Coaling Layer Thickness	μm	95/40/35/35	95/40/35/35
Coaling Layer Sequence		Buffer/PyC/SiC/PyC	Buffer/PyC/SiC/PyC
Fuel Element			
Heavy Metal Loading	g/Element	11	8-12
U 235 Enrichment	-	93 %	7-13 %
No. Particles per Element		19,000	13.000-20.000
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Operating Requirements			
Mean Operating Time	d	1100-1500	700
Max, Burnup	MWd/Item	120.000	90.000
Max. Fast Dose (E > 0.1 MeV)	10 ²⁵ m ⁻²	4.5	3.3
Max, Fuel Temperature	•C	1020	1030
Max. Power/Element	kW	2.7	4 1



		HEU	LEU
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Max Burnup	MWd/t _{HM}	120.000	90.000
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Max. Fuel Temperature	°C	1020	1030
Max. Power/Element	kW	2.7	4 1

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