## **PROGRESS REPORT**

**CONTRACT** N° :

#### <u>CONTRACT N°: FIKI-CT-2000-00099 (HTR-F)</u> CONTRACT N°: FIKI-CT-2001-00150 (HTR-F1)

HTR-F

HTR-F1

**PROJECT N° :** 

ACRONYM :

# •

TITLE :

## **High-Temperature Reactor Fuel Technology**

**PROJECT CO-ORDINATOR :** 

COMMISSARIAT A L'ÉNERGIE ATOMIQUE

FRANCE

**DURATION:47** 

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**PARTNERS** :

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# **<u>1. Executive summary</u>**

First of all we have to recover this experience acquired in the past on HTR fuels. The particular design based on fuel coated particles that ensure leak tightness to the fission products needs to master a high quality of fabrication fuel. So it is necessary to restart the activities on fabrication in order to improve later on the coated fuel technology. Concerning the fuel performance under irradiation it is needed to assess the limit of operation of such fuel particles in particular in the case of higher burn-ups and higher temperature to be investigated. This better understanding of fuel can be met by fuel modelling but also by performance of an irradiation and PIE programme.

The global objective of the HTR-F and F1 is the coated fuel particle qualification at high burn-up and high temperature with a high reliability. For doing so it is needed to restore the fuel fabrication capability in Europe. The main challenge is to reach a very low level of defective particles and the main essential step is the coating of the particle by CVD in a fluidised bed in order to obtain a high standard fuel fabrication quality. A second objective is to assess the fuel qualification keeping a temperature limit fixed at least at 1600°C for fuel leak tightness even for very high burn-ups. A third objective is to start studies on innovative fuels to be used for applications that are different from the former HTR designs, it is interesting to develop new fuel particles with in particular alternative coatings such as zirconium carbide.

The content of the HTR-F project is devoted to fuel technology development and HTR-F1 project is complementing the activities of HTR-F project.

A first task is to gather in a common data base the available data from the various types of fuels tested in the past in European reactors. The architecture is completed and the Fuel DB is now operational and the filling of the database is in progress, almost finished.

A first irradiation HFR-EU1 is under realisation in the HFR reactor on pebbles from the last German high quality fuel production and Chinese pebbles of the HTR-10 fabrication with the objective to reach a burn-up of 200 000 MWd/t. An additional simple experiment HFR-EU1bis with only German pebbles will be performed with the objective to reach a burn up of about 150 000 MWd/t at a temperature which is compatible with the VHTR requirement. This irradiation will be completed in the HTR-F1 project. PIE will be performed on these irradiated pebbles to check the irradiation behaviour of irradiated fuel particles or elements. Another specific task is in this project is to have operational the KUFA furnace which was transferred from FZJ to the JRC/ITU centre. The first tests will be performed on three irradiated German pebbles and the results will be interpreted in the HTR-F1 project.

In order to understand and to predict the irradiation behaviour of HTR fuels it is necessary to model the different phenomena occurring in the fuel in reactor. A selection of properties and models has been made. A common code taking into account these relevant existing models is under development at the European level and first calculations on HFR-P4 have been done. Pre-calculations of the HTR-EU1 test will be done and a first qualification of the code will start on selected cases in the HTR-F1 project.

Concerning the fuel manufacturing in a first step at laboratory scale the best suited fabrication processes has been studied and tested for kernel fabrication in Germany and France. For the coatings of particles first studies has been conducted in facilities at CEA with the benefit of the past know-how. Innovative coatings has been be also considered for preliminary studies. In the HTR-F1project first Pu based kernels will be fabricated at ITU/Karlsruhe.

# 2. Objectives and strategic aspects

The investigation of fundamental characteristics of HTGR fuels has taken place since 30 years. All around the world, the fuel was based on BISO coated fuel particles only having two pyrocarbon layers. Currently TRISO particles having an additional SiC layer for improved fission product retention are being used. Such fuel particles are used in block, pin-in-block and pebble type fuel elements. Their kernels consist of uranium and/or thorium oxide, carbide or oxicarbides. These kernels have 235U or Pu as fissile material. The kernels are embedded in a buffer layer and the subsequent layers ensure leak tightness to the fission products.

The design and safety of future HTR's are based on high quality fuel. There is an absolute need to be able to demonstrate the advantage of the first robust barrier offered by the fuel particle under high burn-up and accident conditions.

In order to develop HTGR technology in Europe it is essential to maintain in the strategic advantage of mastering a high quality of fuel fabrication. The manufacturing of coated particles includes two steps : manufacture of the kernel and coating process. Different technologies exist for the fabrication of the fuel kernels and the more crucial step of the fabrication is the coating of the particles by Chemical Vaporisation Deposition process (CVD) in a fluidised bed.

For reaching such a high standard in the fabrication, a large experience had to be accumulated on the process and indeed on the control of the fabrication. This fuel quality has been reached (in Germany, US, Japan and Great Britain for example) in the past but due to the long delay after stopping the programs, a part of the expertise is decaying fast. So, the objective in Europe is to restart as soon as possible the activities on fabrication and particularly on particle coating on an experimental level and to recover the maximum of the past technical know-how for further improvement of the coated particle technology. This objective was started in the HTR-F project where fabrication processes were selected and first  $UO_2$  kernels and coated particles will be produced. In the HTR-F1 project, the effort is put on characterisation of the coated particles but also studies are extended to the fabrication of Pu based kernels.

Concerning the fuel performance under irradiation, the past experience showed that if the quality of the fabrication is good, the behaviour of the particle under irradiation is also good with extremely low releases of fission products both under normal operation (which results in a very clean primary circuit) but also under simulated accidental conditions. Generally speaking irradiation tests on HTR fuels have demonstrated acceptable performance in fuel operated to temperature above 1300 °C and to burn-ups higher than 15 % FIMA (not at the same time). Moreover we know that a temperature of about 1600 °C does not cause particle failure on a fuel element for longer exposure periods during core heat up experiments.

The qualification of the HTR fuel needs to perform an irradiation programme with two main objectives :

- Demonstration of a defect level ( $\sim 10^{-5}$ ) of coated particles under normal irradiation conditions up to high burn-up,
- Out-of-pile accident simulation tests, in order to ascertain that the fraction of failed particles remains low, even up to high temperatures.

The qualification of the HTR fuel under irradiation is started in the HTR-F project : the definition and the realisation of two experiments on pebbles in the HFR reactor are on going. The transfer of the KUFA facility was made in the HTR-F project and the licensing of this facility will be obtained in order to be operational and to be able to perform tests in the HTR-F1 projects (on irradiated pebbles).

In order to improve the performance of the fuel element, it is necessary to understand the fuel behaviour under irradiation by modelling the different phenomena, which occur during irradiation. In particular it is important to predict the time-to-failure; several mechanisms can lead to a failure of the particle: kernel migration, interaction of fission products with the SiC layer, pressure vessel failure. The modelling of fission products release is also of relevance. The development of European code is the objective of HTR-F project. In the HTR-F1 the objective is to qualify this code and to apply it for irradiation test pre-calculations.

The work in the HTR-F and F1 project will in part be undertaken in interaction with other European HTR projects within the 'HTR-Cluster' as well as in international collaboration especially with regard to the test reactors HTR-10 and HTTR or other international projects. Besides that, archiving of know-how obtained on earlier research projects, will be continued.

The Fifth EU Framework Programme aims at exploitation of the potential of nuclear energy by making nuclear technology safer and more economical. The aspect of sustainability is strongly emphasised in its full scope addressing environmental compatibility, social acceptance, waste management, disposal and non-proliferation aspects, protection of plant personnel, energy supply diversity, competitiveness etc. Against this background, the exploration of innovative concepts should also contribute to the general programme objectives including conservation and advancement of relevant know-how and its transfer to a new generation of nuclear engineers. New applications of nuclear power are indicated in complementary market segments beyond dedicated electricity production like co-generation of heat and power and incineration of long-lived radiotoxic elements.

The HTR-F and F1 projects are widely compatible with these programme objectives as it goes back to the fundamentals of the HTR philosophy to keep fission products and actinides safely encapsulated in the coated-particle fuel under all conceivable operational and accidental conditions of the reactor and of the final repository for spent fuel. The transparency of the safety concept with self-stabilisation of nuclear and core heat-up transients even in hypothetic accident sequences may help to reconcile nuclear power with public opinion. The limitation of the maximum releases is assured by the design of the reactor and can even be demonstrated, as was the case in the AVR and DRAGON projects, or will be possible in future again with the new test reactors in Japan and China now under commissioning.

The HTR-F and F1 projects also comply with the programme strategy of the 5th EU Framework Programme with a visible European added value in combining complementary expertise from different European organisations. Together with the other HTR-related projects, it strengthens the position of European industry and R&D organisations on a global basis and helps to advance the level of competence in HTR key technologies for future European application or participation in international projects.

# **<u>3. Scientific and technical performance</u>**

## **<u>3.1 WP1 Collection and Analysis of data on Fuel Behaviour under</u></u> <u>irradiation</u>**

Contribution: FZJ (2, WP leader), CEA(1), JRC-IE (3), BNFL (5),

The task of the WP1 is to create and load two databases, which are built and located at JRC/IE, Petten :

- □ A documentary database (DDB) which will contain two types of fuel documents :
  - General or synthesis documents,
  - Specific documents concerning tests and experiments.

These documents have been selected by CEA, FZJ, JRC/IE and BNFL among all their own available documents.

□ A technical fuel database (FDB) which contains into specific fields all the available data concerning past and future experiments. A few suitable experiments have been selected by CEA, FZJ, JRC/IE and BNFL among their own past fuel development programmes.

The Fuel – DB database was developed using the platform ODMD (On-line Data Management and Dissemination) (JRC/Petten). The database relies upon a four – entity data model to describe the test results : Source Address, Fuel Element, Test Facility and Test Result. The Fuel-DB database was established on a server at JRC Petten.

Progress

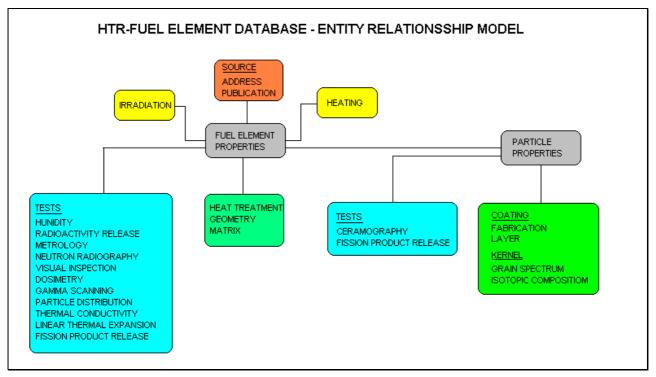
Documentary database	CEA	FZJ	JRC/IE	BNFL
Development and operating manual			Done	
Selection of documents	Done	Done		Done
Digitalisation	Done	Done		Done
Loading	Done	Done		To be done

Technical database	CEA	FZJ	JRC/IE	BNFL
Structure agreement		Don	e	
Development and operating manual			Done	
Selection of experiments	Done	Done		To be done
Loading	Done	Done		To be done

The last step is to include the BNFL data in the database and to issue the final deliverable which will present the database, will include the user's manual and will synthetize the work done in this work package.

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Documentary database



Simplified entity relationship model of the technical fuel database

# **3.2 WP2 Irradiation and PIE**

<u>Contribution to irradiations in HFR and PIE</u>: NRG (7, WP leader) and JRC-IE (3) and FZ-Juelich (2), Contribution to safety tests (KUEA): IBC ITU (4) and FZ Juelich (2)

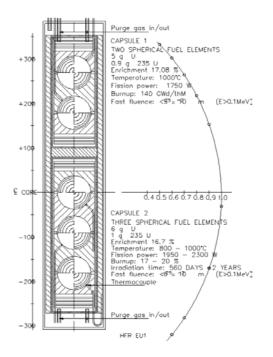
Contribution to safety tests (KUFA): JRC-ITU (4) and FZ-Juelich (2),

# Irradiations

### HFR-EU1

In this experiment 5 HTR fuel pebbles will be irradiated in two separate capsules up to a burn-up of 20% FIMA. During the experiment the fission-gas release from the experiment will be determined. FZJ had delivered 23 fuel pebbles from former AVR production to JRC Petten. Three of these pebbles are to be used for HFR-EU1, while the other two are from current HTR-10 fuel production and made by INET/China. These Chinese fuel elements will be irradiated as a part of the EU/China co-operation within HTR-TN.

In order to be able to optimise the performance of the experiment in the HFR it is of importance that the fuel elements tested are as spherical and smooth as possible, since this influences the heat transfer between the fuel elements and the graphite half shells in which the elements are contained in this specific experiment. For designing this experiment it is also of crucial importance to ascertain that the fuel free zone is sufficiently thick (5 mm) so that 4 mm deep holes can be drilled in this layer without damaging the coated particles located inside the fuel element. The holes are used to accommodate thermocouples, which measure the fuel element outside temperature during the experiment. In order to select from the 23 fuel elements available those elements that are most suitable for the current test all pebbles were X-rayed in two different orientations. This selection is only done on grounds which are of specific importance to this experiment (roundness and thickness of the 5 mm layer). The selected pebbles also underwent a precise 3-d mapping by laser metrology.



The fabrication of parts for the rig head and sample holder was completed and significant progress was made in the assembly of the irradiation rig.

Significant delays with respect to the initial planning have occurred due to technical, organizational and manpower reasons. A new planning was presented and approved. The initial objectives of this irradiation were confirmed, i.e. maintaining the nominal central fuel temperatures and irradiating to a burn-up of 20% FIMA in the fuel elements at the highest flux position.

#### Sweep-Loop facility

The Sweep-Loop facility will be used to perform on-line measurements of fission gas release in the two separate capsules in HFR-EU1. The assembly of the gas mixing panels as well as the glove box and its internals for gas analysis were completed. The specifications for instrumentation and computer aided data analysis and rig control were defined and the work is close to completion. The technical specifications for the electric and electronic cabling were produced.

### HFR-EU1bis

This experiment was initially proposed by JRC Petten to catch up with the delays experienced in HFR-EU1. It relies on an existing and simplified rig design that had been used in the 1980's and 1990's for testing HTR fuel pebbles. The gas analysis is also simplified (batch-wise instead of on-line) and a reactor position with higher flux was chosen to further accelerate the accumulation of burn-up.

In summer 2003, Euratom has joined the Generation IV International Forum and JRC Petten has proposed to adjust the irradiation objectives such that HFR-EU1bis would become immediately relevant for the VHTR fuel project. This means that the central fuel temperatures should be raised by 150 °C to 1250°C, while the target burn-up should reach approx. 15% FIMA. This increase in fuel temperature was achieved with a minor design modification.

The fabrication of parts for the rig was completed and assembly has started.

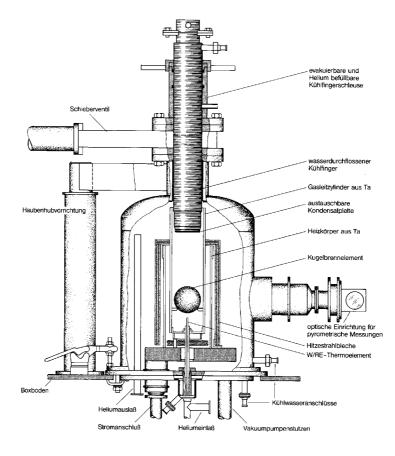
A first draft of the Design and Safety Report was issued and will be submitted to the HFR Reactor Safety Committee.

The next step foreseen in 2004 will be to put in pile the two experiments EU1 and EU1bis. *Nevertheless, the PIE will not be allowed to be completed within the HTR-F1 contract due to the delay above mentioned. An extension of the HTR-F1 contract will be necessary to perform the PIE of EU1bis.* 

# **KÜFA device**

The aim of the heat up experiment of irradiated pebbles and coated particles is to determine the behaviour of these pebbles and particles under accident conditions. The release of fission products can be monitored as a function of time and temperature. The information thus obtained is required for the determination of the consequences of accident conditions in a HTR. The heat up experiment is done with the so-called KÜFA device, which was used up to the begin of the 1990's in Fz-Jülich (Germany).

As a matter of interest, within the HTR-F project, the KÜFA basic device was transferred from FZ-Jülich to ITU-Karlsruhe, ancillary equipment were put out to tender and bought (control electronics for the furnace, detectors for Kr-85 release, detectors for the solid fission products, mechanical lifting device for the furnace and standards for the  $\alpha$ -measurement) and the reinstallation of the KÜFA-device for the cold testing previous to the installation in the hot cell was finished.



KUFA device

Since November 2002, the following activities have been performed for the KÜFA equipment :

- □ The cold test of the KÜFA has been outside the hot cell up to 1800°C successfully completed
- □ The old alpha-containment has been removed from the hot cell and the hot cell was decontaminated.

- □ The new alpha-containment (where the KÜFA is already installed) was brought to the hot cell and the connection to the water supply and to the cold traps and all the electrical connections started
- □ All the calibrations needed (NaI-detectors with a known amount of <sup>85</sup>KR, Ge-detector for the gamma measuring of the plates, etc.) have been started and are in progress
- Development of a conical collimator for the gamma measurements before and after the KÜFA-tests and for the quantitative determination of the burnup terminated. Installation is in progress and first measurements are foreseen by the end of March.

The above-mentioned activities have been done by staff of JRC-ITU with the assistance of staff of FZ-Jülich, who have detailed knowledge on the KÜFA-device from their past experience.

The installation has a six months delay due to problems with the decontamination of the old alpha-box, which was placed in the hot cell position. Nevertheless, the extension of the HTR-F project duration allows the commissioning of the KUFA furnace within the HTR-F contract. The test programme to be done within the HTR-F1 project is under discussion and is foreseen to be performed just after the first measurements.

# <u>3.3 WP3 – Fuel Modelling</u>

Contribution to all Tasks: CEA (1, WP leader), FZ-Juelich (2), BNFL (5), FRA-ANP (6)

## **Properties and models**

The task has ended in 2003.

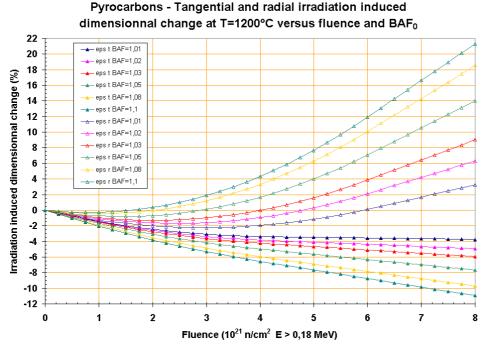
As a matter of interest, the collection of data and models was done in 2002. The objective was to collect the best properties and models for the HTR fuel particle. In this framework and after the sharing of the tasks among the contracting parties, CEA had in charge the collection of properties and models for the  $UO_2$  and  $(U,Pu)O_2$  fuel kernel, FZJ and BNFL had in charge the data and models on coatings (PyC and SiC) and FRA-ANP had in charge the description of fission products diffusion.

The next step, performed in 2003 was to select the best properties and models for the HTR fuel particle in order to integrate them in the code itself.

The physical properties retained for  $UO_2$  fuel arise from both CEA and international recommendations and those for the mixed oxide result from a common work realized at the European level for the EFR project in 1990.

The main models of behaviour under irradiation we have considered for the kernel are in pile densification, fuel swelling, fission gas release and CO's production. For the three ones, the proposed models come from the CEA experience on PWR and FBR oxide fuels. Concerning CO's production, we have chosen in the open literature empirical models built on experimental data and which were already used in the past.

Concerning the layers, properties was given by BNFL and FZJ. CEA added a set of data coming from CEGA corporation. The main considered models and properties for the layers are the thermal conductivity  $\lambda$ , the coefficient of thermal expansion  $\alpha$ , the Young's modulus E, the Poisson's ratio v, the irradiation creep law and the irradiation induced dimensional change rate. The diffusion coefficients data, used in different countries, are given, extracted from literature,



especially from IAEA-TECDOC-978, Fuel performance and fission product behaviour in gas cooled reactors for Fuel kernel, Buffer layer, Pyrocarbon layer and Graphite.

## **Code development**

The task has ended in 2003.

The code intends, in order to understand and to predict the irradiation behaviour of HTR fuels, to model the different phenomena occurring in the fuel in reactor.

A FEM modelling of the fuel particle, codenamed ATLAS, was developed, including the properties and models selected in the project. In the framework of the HTR-F and F1 projects, it is a deterministic approach which allows the calculations of temperatures, strains and stresses in the coated fuel particle. A particle and eventually an additional layer around the particle to simulate the particle's environment are modeled.

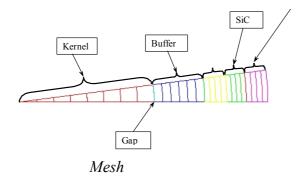
The finite element method allows :

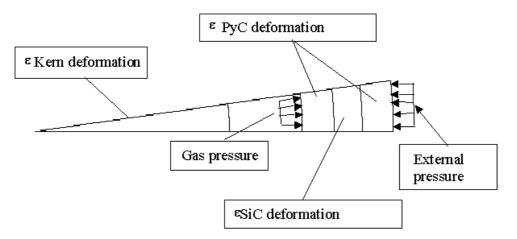
- □ A thermal calculation giving the temperature field in the meshing nodes.
- □ A mechanical calculation allowing access to the displacements fields, stresses and strains in the meshing nodes. The particle model is axisymmetrical, two-dimensional and represents a part of the particle.

The thermal model treats the conduction in the particle. Between the buffer and the kernel and between the buffer and the IPyC, meshes simulate a gaseous joint. The boundary conditions of this model are two-fold : a condition of nil flux on each side of the model and an imposed temperature on the particle external surface nodes. The conductivity of the layers and of the kernel is re-calculated at each time step and depends on the temperature, the integrated fluence and the fabrication and gaseous porosities. The porosity is re-calculated at each time step.

The mechanical model treats the visco-elasticity in the particle. The non-linearities are of two types : first, the material non-linearity through irradiation creep laws and second, the geometrical non-linearity through a contact condition between kernel and buffer and between buffer and IPyC. The layers are connected to each others. The main characteristics of the model are as follows :

- **□** The thermal load is the temperature field resulting from the thermal calculation.
- □ The pressure load is calculated at each time step from the free volume (itself calculated from the deformed geometry), the temperature and the quantity of gas (Xe, Kr, CO) present. It is applied to the IPyC layer internal skin and to the kernel external skin.
- □ The loads imposed by the swelling of the kernel and the irradiation induced deformation of the layers are considered as loads of imposed deformation type. These are taken into account by making an analogy between swelling and thermal expansion.





Loadings and boundary conditions of the mechanical model

The next step will be proposed in the FP6. It will include mainly the statistical approach of the particle behaviour.

# **Code application**

The objective is to start the qualification of the code on the basis of selected experiments.

In 2003, the first deterministic thermal and mechanical calculations on a free particle using the ATLAS.V1.0 European code was performed. These calculations were based on the HFR-P4 experimental irradiation of little pebbles in the HFR reactor.

Concerning the PyC and SiC layers, and in addition to a simplified reference set, two property sets were used : one from BNFL and one from the German research organization FZJ.

The thermal and mechanical behavior of the particle under irradiation is a complex phenomenon with many parameters. The different property sets led to rather different results, from both a thermal and mechanical point of view.

This first approach helped identify the following important parameters :

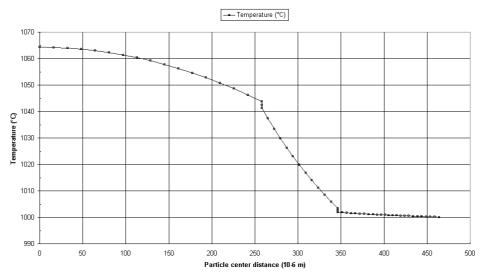
- □ The deformation kinetics under fast flux of the pyrocarbon layers and particularly at the beginning of irradiation,
- □ The irradiation induced creep coefficient of the PyC layers, but also that of the SiC layer,
- □ The failure modes of the layers.
- □ The behavior of the buffer. Here it would be useful to know its structural evolution as well as its thermal conductivity under fast flux.

This improved knowledge on these properties of PyC and SiC layers is an important step for the design basis and understanding of the in pile behavior of future fabrications. It entails the fine reinterpretation of past experiments and the achievement of analytical irradiation experiments on materials elaborated under the same conditions as those for future fabrications.

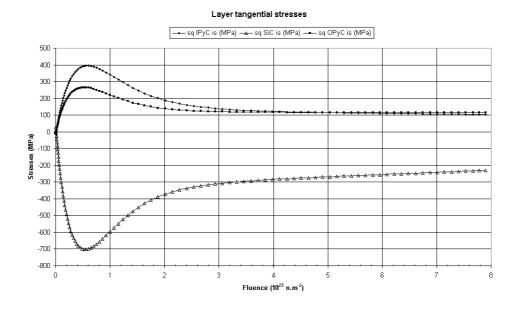
The next step will be the pre-calculation of EU1 which is underway and the first qualification of the ATLAS code has been defined. The selection of cases have been made : HFR-K3 and HFR-K15 have been selected and FZJ is in charge of the collection of all the data (fabrication, irradiation, properties, models...). BNFL is in charge of the definition of the expected results. Concerning the calculations, the following grid has been established :

Exp./Code	FZJ	BNFL	FANP	CEA
HFR-K3	CONVOL	STRESS3		ATLAS
HFR-K15	CONVOL	STRESS3	ATLAS	

#### Last time step temperature cross section



HFR-P4 thermal calculation results with BNFL data



HFR-P4 mechanical calculation results with FZJ data

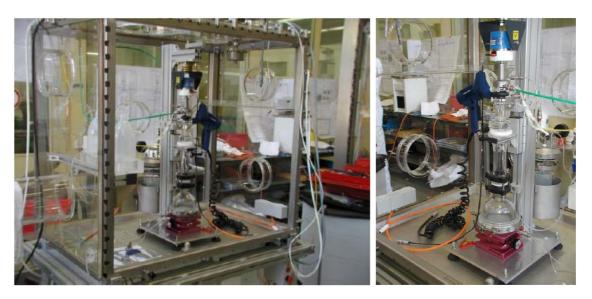
# <u> 3.4 WP4 – Fuel Manufacturing</u>

<u>Contribution to Task 1</u> : CEA (1, WP leader), ITU (2) <u>Contribution to Task 2</u> : CEA (1), FRA-ANP (6) <u>Contribution to Task 3</u> : CEA (1), FRA-ANP (6) <u>Contribution to Task 4</u> : JRC-IE (3), CEA (1), FZJ (2) <u>Contribution to Task 5</u> : FRA-ANP (6)

For reminding, the tasks to be done in the HTR-F project have ended at the end of 2002 excepted for task F-3 concerning innovative coatings. The status described below concerns the HTR-F1 project.

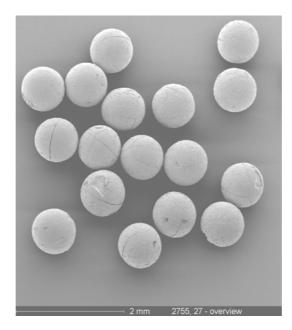
## **R&D** of manufacturing of Pu-based kernels (ITU - Task F1-1)

Modifications to the glovebox for the production of  $(U,Pu)O_2$  kernels have been made to improve the droplet production means (see figure). Cold tests are now underway to optimise the ammonia gasification unit. In parallel to these developments, dissolution of  $PuO_2$  for the tests has been initiated.



*GloveBox with the*  $(U,Pu)O_2$  *kernel production device* 

The internal gelation procedure has also been investigated. This task is being performed in replacement of the production of the ThO2 kernels and has been approved by all HTR-F/F1 partners. Tests have been made on Yttria Stabilised Zirconia (YSZ) in a simple laboratory device. Initial tests showed much particle breakage, but further adjustment of the parameters gave the results shown in Figure below. The method is flexible, an especially desirable attribute for small-scale production of compounds with different compositions. Its main disadvantage is the use of an organic heat transfer material, which could raise waste disposal costs in industrial production.



Sintered YSZ (Run 46) produced by the internal gelation route

# **Review of characterization methods (CEA+FZJ - Task F1-2)**

A review of different methods of control available to particle characterization will be done (control of kernels and coatings). This review will be based on European experience but also on international experience.

A list of characterization methods written by specified parameters (see table from the deliverable 18) used in the different countries involved in HTR projects (which are China, Japan, Germany, USA and what we are already doing in the framework of HTR-F&F1 in Europe) has been done and presented in the 6<sup>th</sup> WP4 meeting.

UO₂	PyC buffer	І & Е РуС	SiC			
500 ± 40 μm	95 ± 20 μm	40 ± 10 <i>µ</i> m	35 ± 7 <i>μ</i> m			
$\geq$ 10,4 g/cm <sup>3</sup>	$\leq$ 1,05 g/cm <sup>3</sup>	$1,9^{+0,1}_{-0,05}$ g/cm <sup>3</sup>	$\geq$ 3,18 g/cm <sup>3</sup>			
$D_{max}/D_{min} \leq 1.1$		BAF ≤ 1,06	100% beta			
defective SiC coating $\leq$ 5. 10 <sup>-6</sup>						
Uranium contamination $\leq 10^{-7}$						
O/U ratio, impurities, <sup>235</sup> U enrichment for the kernel						

 Table 1 : Guidelines for specification of HTR fuel particles (from deliverable 18)

The O/U ratio was measured by ThermoGravimetric Analysis, the Uranium enrichment by Mass spectrometer analysis, the metrology (kernel diameter and sphericity or layer thickness) by

optical (or X-Ray) particle size analysis, the impurities by ICP-MS or emission spectrometer analysis, the SiC structure by X-Ray Diffraction. Some criteria seem more difficult to be measured (because of important work of preparation) such as the density of each layer which was done by sink-float method or some characterization methods are still raising questions such as the anisotropy degree of dense PyC. This latter parameter was always measured by an optical method (and after converted in a BAF figure) but with an experimental principle slightly different for all countries, so it is not so obvious to compare the BAF or OPTAF value of each manufacturer.

Concerning, the measure of uranium contamination and of the defective SiC coating, the burnleach test is the reference (destructive) method used in Germany, Japan, USA and China whereas the Russian used also a method based on a weak irradiation.

## **Innovative coatings (CEA+FRA - Task F1-3 and F-3)**

The task F-3 concerns a **preliminary study** on **alternative materials** than SiC for coating. For instance, ZrC coating will be evaluated. A review of the existing knowledge will be done (only paper work). The task F1-3 concerns tests on innovative coatings. The tests will be done on non-radioactive simulating materials such as  $ZrO_2$  (or  $HfO_2$ )stabilized kernels.

The text here-after resumes the literature review of processes and thermodynamical considerations for innovative ZrC coating which was the task HTR-F-3 and the status of task HTR-F1-3 which is the experimental work done in 2003.

#### Status of Task F-3

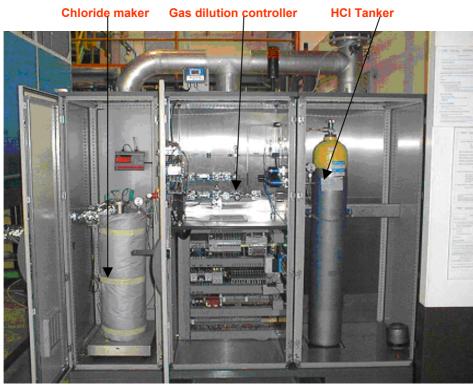
This study takes part of an European R&D project on coating technology of nuclear "fuel" particles used in high temperature gas-cooled reactors. In the aim of reaching higher temperatures, we propose to replace the silicon carbide layer by zirconium carbide, which acts as a barrier to the diffusion of metallic fission products. Coatings are made by chemical vapour deposition (CVD) in fluidised bed.

After a bibliographical comparison of ZrC and SiC properties, a thermochemical analysis has been carried out to compare iodide, chloride and bromide which are three potential precursors of Zr in deposit process. Finally the choice of a chloride process has been done and an installation has been put in place. A note on this literature review is available but not issued, and will be included in the deliverable corresponding to this task.

#### Status of task F1-3

In 2003, an installation has been realized to perform ZrCl4 synthesis (see figure) in order to manufacture ZrC CVD coating.

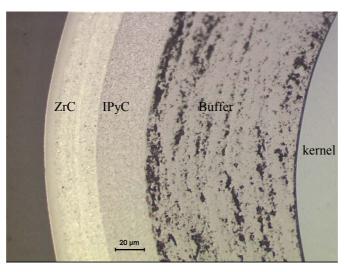
The chlorinator and the management of gases are implanted in a cupboard under depression with a chloride detection. A line of neutral gases insures the cleaning of the HCl tubes, the piloting of the pneumatic valves and the dilution of the chloride. The temperature of chlorination is adjustable between the ambient and 450 °C. The dosage of gases is automatically controlled.



Chlorination installation

The first trials showed problems of chloride crystallisation in certain cold parts of the installation. Then appeared a problem of excessive temperature ascent in chlorinator reactor during deposit phase.

Finally, a deposit of about 35  $\mu$ m (see figure) was realised in agreement with the specifications. The process coating has to be still optimised. Indeed, composition variations were observed in the deposit as well as porosities.



Cross section of a particle with a ZrC deposit.

The perspectives 2004 will concern the modification of the chlorinator and the mastery of the precursors gases distribution.

# 4. List of deliverables

Task No	Del n°	Nature	Lead partner	Deliverable Title		Foreseen Delivery Date	Updated delivery date	Date of issue	
	WP1								
F-1-1	1	Da	JRC/IE	Past experience JRC/IE	+24	2002/10	2003/03	2003/06	
F-1-2	2	Da	FZJ	Past experience FZJ	+24	2002/10	2003/03	2003/06	
F-1-3	3	Re	FZJ	Synthesis report	+36	2003/09	2004/03		
F-1-4	4	Da	CEA	Past experience CEA	+24	2002/10	2003/03	2003/03	
F-1-5	5	Da	BNFL	Past experience BNFL	+24	2002/10	2003/03		
				WP2					
F-2-1	6	Re	FZJ	Specifications HFR-EU1	+12	2001/10		2001/10	
F-2-2	7	Re	JRC/IE	Design safety report	+12	2001/10	2004/6		
F-2-3	8	Eq	JRC/IE	Experimental device	+24	2002/10	2004/09		
	7bis	Re	JRC/IE	EU1bis Design safety report	+12		2003/12		
	8bis	Eq	JRC/IE	EU1bis Experimental device	+24		2004/04		
F-2-4	9	Eq	ITU	Installation of KUFA facility	+24	2002/10		2002/11	
F-2-5	10	Eq	ITU	Commissioning of KUFA	+36	2003/09		2003/12	
	-			WP3	-				
F-3-1	11	Re	CEA	Collection of data and models	+24	2002/10	2002/12	2002/12	
F-3-2	12	Re	CEA	Selection of data & models	+30	2003/04		2003/12	
F-3-3	13	Re	CEA	First calculations results	+36	2003/09		2003/12	
				WP4					
F-4-1	14	Re	CEA+ITU	Manufacturing tests of kernels	+24	2002/10		2002/10	
F-4-1	15	Re	CEA+ITU	Review on Kernels fabrication processes	+18	2002/04		2002/04	
F-4-2	15	Re		Report on Coating experience of particles	+24	2002/10		2002/11	
F-4-3	16	Re		X-Ray Radiography quality control method	+24	2002/10		2002/09	
F-4-3	17	Re		Fuel particles specification	+12	2001/10		2001/10	

The list of deliverables for the HTR-F contract and their status at the time being is the following :

Task No	Del n°	Nature	Lead partner	Deliverable Title		Foreseen Delivery Date	Updated delivery date	Date of issue
	WP1							
F1-1	1	Re	CEA	Final report	+36	2004/11		
				WP2	-	•	•	
F1-2.1	2.1	Re	JRC/IE	Final irradiation report	+24	2003/11	2006	
F1-2.2	2.2	Re	NRG	ND PIE report on pebbles	+36	2004/11	2006	
F1-2.3	2.3	Re	NRG	Destructive PIE report on the irradiated pebbles	+36	2004/11	2006	
F1-2.4	2.4	Re	JRC/ITU	KUFA testing report	+36	2004/11		
				WP3	-	•	•	
F1-3.1	3.1	Re	FZJ	Collection of input data	+24	2003/11	2004/03	
F1-3.2	3.2	Re	CEA	Pre-calculation report on HFR-EU1 test	+30	2004/05		
F1-3.3	3.3	Re	CEA-FRA- BNFL	First qualification report of the code	+36	2004/11		
	WP4							
F1-4.1	4.1	Re	JRC/ITU	Report on U and Pu based kernel fabrication	+36	2004/11		
F1-4.2	4.2	Re	CEA-FZJ	Report on particle characterisation control	+36	2004/11		
F1-4.3	4.3	Re	CEA	Report on studies on innovative coatings	+36	2004/11		

The list of deliverables for the HTR-F1 contract and their status at the time being is the following :

# 5. Dissemination and use of the results

#### Significant aspects of the current areas of use of results

The European HTR-Technology Network (HTR-TN) provides already a European platform for effective dissemination and use of the results. The projects have been oriented to the main objective to provide the basic technology elements for an industrial deployment of HTR technology about the year 2010. The HTR-TN programme will also support European organisations to enter into commercial HTR projects in other countries as GT-MHR in Russia, PBMR in South Africa and NGNP in USA. Follow-up projects can soon be expected in Japan and China after having collected enough operational experience from HTTR and HTR-10. Close collaboration of European organisations in the HTTR and HTR-10 projects is a precondition for future involvement in industrial projects in these countries.

#### Potential for dissemination and use

The potential benefit of the HTR-technology deployment can be rather significant in terms of sustainable employment and 'return-of-invest' into the development efforts. More than 5 billion EUR have already been spent into the European HTR development, in the past. This know-how has to be recovered with high priority but time is fading away if this process is only continued with the low capacities in the actual HTR programmes.

The endeavour of HTR-TN to recover and advance past know-how is associated with an enormous benefit as compared to the cost but still cannot avoid that vast knowledge will get lost and has to be re-elaborated with much higher efforts.

The HTR-F&F1 projects contribute to this process in the field of fabrication, qualification and modelling of fuel technologies.

Although a real value analysis cannot be performed with regard to the long period and additional efforts till industrial deployment of HTR, the significant present scientific/ technological outcome of the project has been reached by rather reasonable R&D investments.

#### Patentable results

According to the present rules, the partners which originate the results are the owner of this specific know-how and keep the right for commercial deployment. Commonly elaborated know-how will be marketed jointly. The protection of the know-how is in the responsibility of the related organisation. The use of know-how stemming from the project is free within the consortium for R&D purposes on a 'Need-to-Know-Basis'.

Patents have not yet been applied. Licenses will be given on normal market conditions for use of fabrication processes and analytical tools outside the project. The results of the project is archived in an electronic form on the SINTER-Network Internet site. The access is restricted to partners and collaborative organisations.

#### **Contacts with potential users**

The European HTR-Technology Network already includes more than 20 partner organisations which approach external users of the information stemming from the projects on different and complementary levels. Participation in international conferences and e.g. in the International Generation IV Forum (GIF) also provide a broad dissemination. The consortium (HTR-F&F1) agreed to dissiminate the information through HTR-TN.

The targeted audience will extend beyond HTR-TN into the supplier market for components and systems with continuing industrial response for HTR technology.

The public, licensing authorities and political decision makers are also important recipients for paving the way towards industrial deployment and improved public acceptance.

#### **Publications and conferences**

In 2003, at ICAPP'03, two presentations of the results of the project has been made in the High Temperature Gas Reactor Technical session, Fuel and Fuel Cycle sub-session

"European programme on high-temperature reactor fuel technology" Proceedings of ICAPP'03 – Cordoba, Spain, May 4-7, 2003 – Paper 3020

"First results from HTR-F project : modelling of fission product diffusion in high-temperature gas reactor fuel" Proceedings of ICAPP'03 – Cordoba, Spain, May 4-7, 2003 – Paper 3017