

HTR Fuel Design, Qualification and Analyses at PBMR

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Abstract

This paper presents an overview of the safety and design requirements of PBMR fuel, design and performance analyses performed, analyses models and software being developed, and the current program to qualify PBMR fuel for use in the demonstration power plant. PBMR fuel design is based on the German reference fuel design, and will be utilised inside the operating envelope of the original German fuel qualification program. Fuel design, safety functions of the fuel, phenomena that influence fuel performance and fission product release and the design criteria derived from these functions and phenomena are described. Fuel qualification and validation of analyses methods are achieved by evaluations of previous experimental irradiation data and a fuel qualification programme for PBMR type fuel. The performed and planned validation and qualification efforts are presented with some results and issues discussed. The fuel performance analyses methods and legacy software products inherited from the German fuel program are being further developed at PBMR. New models and software are being developed as new requirements such as Monte Carlo design analyses become necessary.

KEYWORDS: *Fission product release, fuel, design, performance, qualification, HTR, PBMR, spherical fuel elements, verification, validation*

1. Introduction

TRISO-coated fuel particles imbedded in graphite form the basis of fuel elements utilised in most high temperature reactor designs. Whether block- or spherical fuel elements are used, unique design and analyses issues must be overcome. PBMR fuel design is based on German Proof Test fuel for the HTR-MODUL (High Temperature Reactor – Modular) (1). This fuel design was extensively tested and evaluated in Germany and the Netherlands, most notable are the fuel irradiation tests HFR-K5 and –K6 performed in the HFR (High Flux Reactor) at Petten.

Approximately 450 000 fuel spheres are required for a critical assembly. Fuel spheres enter the top of the core through three fuelling points in the top reflector, from where they flow down through the annular core formed by the centre reflector and the inner side reflector, and are extracted at the bottom of the core through three defuel cones into defuel chutes. Burn-up is measured and spheres that do not exceed the permissible burn-up are returned to the reactor core for another pass (2). This annular core geometry is a unique feature of the 400 MW_t PBMR core design.

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Fuel design criteria were selected to ensure that all safety requirements are met for normal operation and all design base accident conditions. In order to demonstrate that manufactured PBMR fuel will meet these safety requirements it is necessary to prove acceptable fuel performance for all operating conditions and design base accidents.

Expected fuel performance is based on an analytical consideration of fuel failure mechanisms identified from previous experimental irradiation data, and a fuel qualification programme for PBMR fuel.

Analysis of the performance of the fuel against each of the design criteria are used to demonstrate that the design criteria will be satisfied for all anticipated fuel requirements. Fuel performance analyses are based on the normal operating envelope:

- Maximum core temperature of 1068 °C.
- Maximum burn-up of 97 500 MWd/t (10.1% FIMA)
- Maximum fast neutron dose ($E > 0.1$ MeV) of $2.72 \times 10^{25} \text{ m}^{-2}$
- Maximum fuel sphere power of 2.76 kW

Design base accident conditions achieve the following maximum fuel temperatures:

- Depressurised loss of forced coolant: 1620 °C
- Pressurised loss of forced coolant: 1320 °C

Fuel performance models, software and methods are continuously developed, verified and validated under PBMR quality assurance programme.

2. Fuel Design

The product specification for PBMR fuel is based on the specification for German fuel spheres produced for the High-Temperature Reactor (HTR) 500 and HTR-Modul Proof Tests. This fuel type is generally regarded as state of the art for German pebble fuel production. It is designed for optimal performance under normal operating conditions, and to withstand all design based accident conditions. General descriptions of the fuel spheres are widely available in literature ((1) and (3)), and the PBMR specific description is described below.

Fuel spheres are manufactured from graphite matrix material, in which the TRISO-coated particles are imbedded. The outer 5 mm layer is matrix material only. The graphite matrix material functions as a good heat transfer medium and stabilizes the coated particles in the sphere. Good thermal contact is achieved between the coated particles and matrix material, so that low temperature gradients occur in the fuel sphere. The outer fuel free zone protects the coated particles from damage from outside direct mechanical effects such as abrasion and shock. It further acts as a barrier layer against chemical corrosion in the case of water or air ingress in the core.

The TRISO-particle consists of a spherical UO_2 -kernel, 500 micron in diameter, surrounded by four coating layers. UO_2 has a high melting point (~ 2880 °C), therefore retaining its integrity under all reactor conditions. The released oxygen binds with fission products to form immobile oxides. The majority of fission products are retained in the kernel this way. The kernel produces almost all the power of the reactor through nuclear fission. It also acts as a retention barrier of gaseous fission products, thereby reducing the internal pressure of the coated particle. Fission products that do not form stable oxides are released from the kernel through a diffusion process. All fission products are therefore retained or their release reduced by the UO_2 kernel.

The kernel is surrounded by a 95 micron low density pyrocarbon layer, known as the buffer layer. This layer acts as a sacrificial layer, allowing the kernel to swell under irradiation, and providing void volume for fission gases released from the kernel. The rest of the layers are therefore protected from recoiling fission products and excessive internal pressure by the buffer layer.

The next layer is made up of dense pyrocarbon, 40 micron thick and known as the inner pyrocarbon (PyC) layer. It forms an impenetrable barrier to gaseous fission products, and slows down the transport of metallic fission products to the silicon carbide (SiC) layer. During manufacture it provides a smooth surface for the SiC to adhere to, and protect the kernel from chlorine in the form of hydrochloric acid during SiC deposition.

The SiC layer is the primary fission product barrier, being 35 micron thick. It retains all gaseous and metallic fission products to a very high extent, with the exception of silver and strontium. It provides the structural support required to contain the internal gas pressure in the coated particle.

The final layer is again dense pyrocarbon, 40 micron thick and known as the outer pyrocarbon (PyC) layer. It is under compressive stress, putting positive pressure on the SiC, helping to contain internal gas pressures. It protects the SiC layer during manufacture from chemical and mechanical damage.

To prevent coated particles touching each other in the matrix material, which may lead to failures during the pressing stage, each coated particle is over coated with a layer of matrix material graphite before being mixed with the bulk matrix material. Fuel spheres are pressed and machined to form perfect spheres, 60 mm in diameter

2.1 Safety Functions

Spherical fuel elements are the first and most important safety feature of the PBMR design. The fundamental safety functions and the required design criteria to achieve these safety functions are summarised below:

- Confinement of radioactive materials and control of operational discharges, as well as limitation of accidental releases. This function is assigned principally to the fuel and is maintained by the other two fundamental safety functions.
- Control of reactivity through negative temperature coefficients and core design.
- Removal of heat from the core by core materials and core design

The fundamental safety functions are achieved by ensuring that high quality fuel is used, operating temperatures and fuel burn-up is within fuel specification and chemical attack on the fuel is prevented.

2.2 Design Criteria

Design criteria are derived from the fundamental safety functions and from the functions that the fuel must perform in the environment in which it will be used. The main criteria can be described as follows:

Fuel integrity must always be maintained throughout the whole lifecycle of the fuel sphere. Fuel spheres must be able to withstand transport and handling stresses during shipping and handling, and remain intact under all expected and design reactor conditions. Finally fuel must contain fission products for the entire interim storage period, and eventually long term disposal. Coated particles failure fractions must remain low enough not to cause any significant

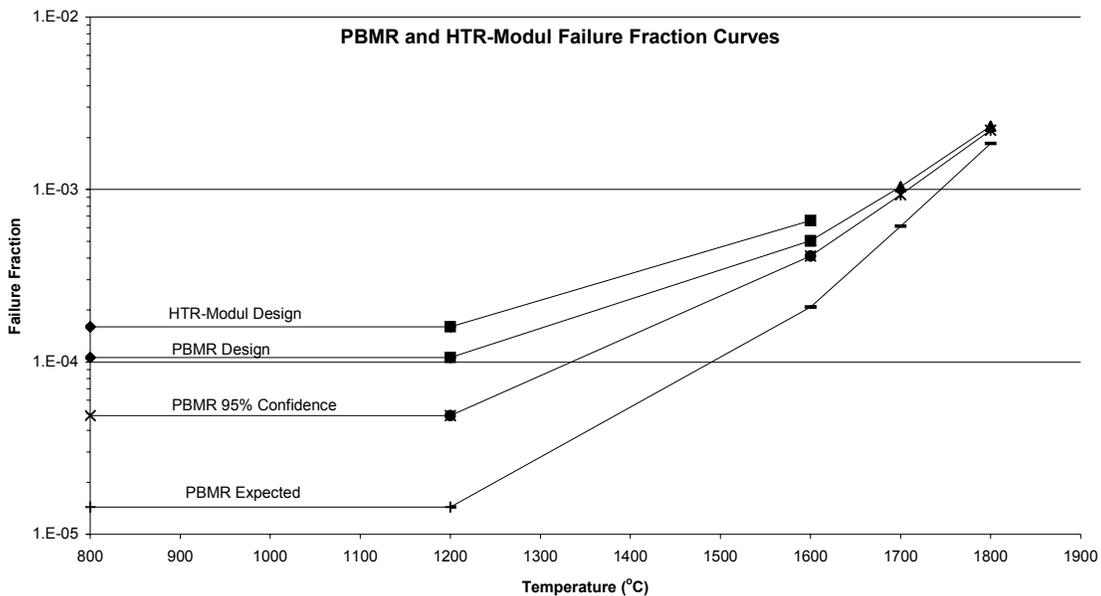
radiological risk to operating personnel and the general public. It is therefore imperative that chemical attack on fuel and core structures is prevented. Fuel integrity is easiest observed by monitoring gaseous fission product release from fuel in main coolant. The most important design criteria and fuel performance analyses performed to assess each criterion is discussed below.

2.2.1 Manufactured and Operational Fuel Failures

Free uranium is defined as all uranium not enclosed within an intact silicon carbide layer, and includes enriched uranium in damaged coated particles, enriched uranium contamination on the outer surface of the outer PyC layer from the coating process, and natural uranium contamination in matrix graphite from natural graphite used in its manufacture. The free uranium fraction value should be kept as low as reasonably achievable (ALARA) to minimize fission product release during normal operation and design base events. Additional failures during reactor operation will have a significant effect on fission product release, because the manufactured free uranium fraction is so low. Operational caused failure should therefore be as low as possible. Statistical analyses of German production fuel were used to determine PBMR manufactured fuel failure fractions. A one-sided upper 95% confidence level of 6×10^{-5} was determined. It is expected that this value could be adjusted downward when burn-leach results for PBMR fuel become available.

Irradiation induced failure data were obtained from all relevant irradiation tests performed under controlled irradiation conditions on German fuel types containing LEU-TRISO coated particles. In all applicable tests no coated particle failure occurred during irradiation. Failure fractions due to temperature transients as would be expected during loss of forced coolant events were derived from German post-irradiation heating tests. Failure fraction vs. heating temperature curves were constructed using all applicable and available results at temperatures of 1600 °C, 1700 °C and 1800 °C. Fig. 1 shows the failure fraction curves derived for PBMR together with expected and design curves derived in Germany for HTR-Modul.

Figure 1: PBMR and HTR-module Failure Fraction vs. Temperature Curves



2.2.2 Fission Product Diffusion through Intact Coating Layers

Fission products are retained by all components of a fuel sphere. As such, the fuel sphere, and specifically the TRISO-particle are nearly impermeable to fission products (with the exception of silver). At high temperatures however, fission products diffuses slowly through all coating layers. Coating layers are therefore designed in such a way that fission products can not diffuse through all the layers in the time that a fuel sphere spends in the reactor core, under the temperature it will experience. The only exception is silver, which is not contained completely by the fuel.

The time it takes for a specific fission product to penetrate through a coating layer by the process of diffusion is known as the breakthrough time. It is dependent only on the reduced diffusion constant, which is a function of temperature. Breakthrough times are directly derived from transport theory and calculated by numerical diffusion codes. It is ideally suited to provide conservative estimates of the coating layer thickness required to prevent fission product release. Breakthrough times for different fission products and different retention layers at a temperature of 1 130 °C are listed in Tab. 1. From the table it can be seen why 1 130 °C has been selected as the maximum operating fuel temperature. Strontium is about to start breaking through at the end of the irradiation life of the fuel sphere. Silver has already broken through, but the low fission yields from uranium fission limits the total activity released to the main power system. Plutonium fissions however, have higher silver yields. High burn-up, low enriched fuel has a large plutonium fission fraction, which increases the silver inventory significantly. The released silver plates out on cooler regions of the main power system, specifically on the power turbine, and add another limitation on the reactor power and maximum fuel temperatures reached.

Table 1: Breakthrough Times for Single Layers at 1 130 °C

Layer	Thickness	Breakthrough Time at 1 100 °C (d)			
		Noble Gas	Cs-137	Sr-90	Ag-110m
PyC	80 µm	29 039	36.1	0.116	1.26
SiC	35 µm	1.84 x 10 ⁸	1 060	84.4	66.3
Matrix Graphite	5 mm	-	1.46	920	0.121

The breakthrough times during design based accident conditions were calculated with the computer diffusion code GETTER. As a conservative approach fuel elements were irradiated to 9.85% FIMA under normal reactor operating conditions. The fuel elements were then heated to the target temperature, simulating a post irradiation heat-up test. Fractional releases from the coated fuel particles and through fuel elements were calculated with GETTER, and break through curves were plotted. From these curves expected breakthrough times can be estimated. For each investigated nuclide design (best conservative values) transport parameters and worst irradiation conditions (maximum fast fluence) were used. In Fig. 2 and 3 the Cs-137 and Sr-90 breakthrough times at various temperatures through a coated particle are presented. Fig. 3 shows that strontium release from the PBMR core during postulated accident events where maximum fuel temperatures exceed 1700 °C can not be ignored, and must be included in fission product release analyses.

Figure 2: Cs-137 breakthrough

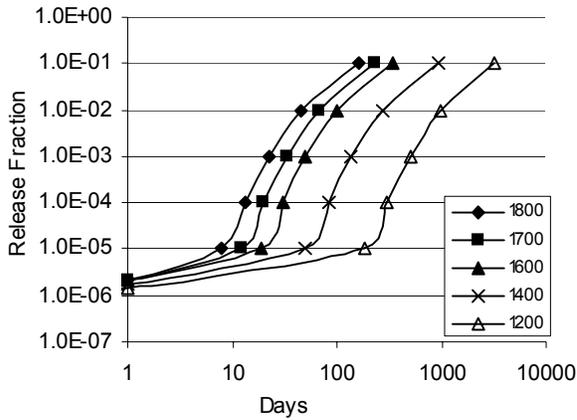
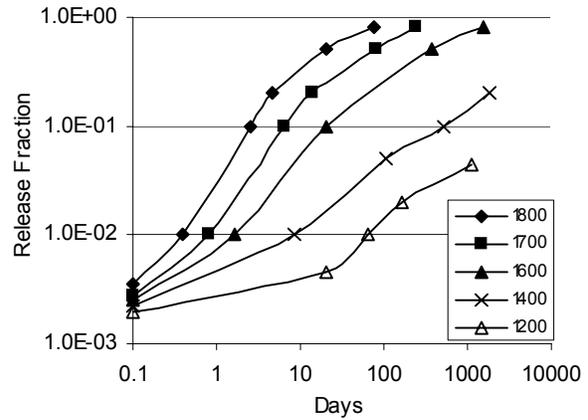


Figure 3: Sr-90 breakthrough



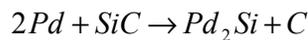
2.2.3 Kernel Migration

In block type fuel under certain irradiation conditions it was observed that the kernel migrated inside the buffer layer. This effect, known as the amoeba effect, always occur up the temperature gradient. Kernel migration is caused by the removal of carbon from pyrocarbon layers at the hot end of the kernel and deposition of the removed carbon at the cold end of the kernel thus forcing the kernel to migrate away from the cold end towards the hot end of a coated particle situated in a temperature gradient. In extreme cases kernels may reach the inner pyrocarbon and SiC layers, allowing fission products to interact with the SiC layer, causing it to fail.

For spherical fuel, an evaluation of experimental kernel migration data for UO₂ kernels collected from diverse sources was performed. Under worst possible conditions where a fuel sphere remains stationary for its whole life time in the hottest positions in the core, the kernel migration will only be 36 micron. This means that the kernel will not leave the buffer layer region, and will have no interaction with coating layers.

2.2.4 SiC Layer Degradation

Corrosion of the SiC coating layers was observed during ceramographic examination of irradiated coated particles containing low enriched uranium. The metallic fission product palladium was identified as the chemical element reacting with SiC through the chemical reaction:



This corrosion could reduce the SiC layer integrity, leading to fission product release form the particle. Investigations by Tiegs (4) showed that palladium penetrations up to 63% into the SiC layer led to no increased release of Cs-137, and conservatively concluded that penetrations up to 50% do not reduce SiC retention abilities. Using this conservative failure criterion and data from several sources, a maximum average core failure fraction of 2.3×10^{-7} was calculated for the current PBMR core design, which is about a factor 260 less than the free uranium fraction for fresh fuel. Therefore no significant increases in fission product releases from failed particles are expected during normal operation or during the first 100 hours of a loss of forced coolant event.

3. Qualification of Fuel

From the beginning of the PBMR project it was decided to base the reactor core design around German state of the art LEU-TRISO fuel. An extensive experimental performance base was developed in Germany in the 1980's, culminating in the HTR-Modul Proof Tests in the High Flux Reactor in Petten and for bulk testing (AVR 21-2 reload) in the AVR. It is consequently not necessary to repeat all the irradiation tests from which the operating envelope for the German LEU-TRISO fuel types were derived, provided that it could be demonstrated that PBMR fuel requirements fall within the envelope of the German LEU-TRISO fuel performance base. If this were true, it need only be demonstrated that the irradiation performance of PBMR fuel will meet PBMR-specific fuel requirements, and to perform such irradiation and heating tests that are required to provide a statistically viable performance base for PBMR fuel.

PBMR fuel qualification is therefore two-fold, firstly to ensure that manufactured fuel is equivalent to state-of-the-art German fuel, manufactured by Hobeg in 1988. Using the same fuel specification, applying quality control for PBMR fuel to the same parameters as those of the reference fuel, using the same process steps and using direct materials that comply with similar specifications to those used for the reference fuel, will ensure equivalence between PBMR fuel spheres and the reference fuel as far as manufacture is concerned.

Secondly, testing of PBMR fuel will be undertaken to confirm that its performance characteristics match those of the German reference fuel and the validity of the data for PBMR application will be confirmed. The fuel irradiation programme for PBMR fuel consists of a partial burn-up test to a burn-up of 5% FIMA, a proof test to a burn-up of 11.3% FIMA, a test on machined graphite sphere samples, and a test using pressed graphite sphere samples. A pre-irradiation test programme to provide an independent assessment of unirradiated properties of PBMR fuel spheres and coated particles will supplement the irradiation programme. Following irradiation, an extended post-irradiation programme including heating tests will be performed.

3.1 Irradiation testing

The test reactor presently considered for the PBMR irradiation programme is the IVV-2M reactor located at Zarechny in the Russian Federation. The IVV-2M reactor consists of a water moderated reactor core with a nominal power output of 15 MW, surrounded by a beryllium reflector. The unperturbed thermal and fast ($E > 0.1$ MeV) neutron fluxes in the core centre are approximately 5×10^{14} and $2 \times 10^{14} \text{ cm}^{-2} \cdot \text{s}^{-1}$ respectively. The reactor is operated in 300 h cycles with shutdowns of approximately two days between cycles, and is shut down twice yearly for refuelling. Fuel sphere irradiation tests are performed in irradiation rigs that can accommodate four full-sized fuel spheres per rig. During irradiation and subsequent heating tests fission gas release will be continuously measured. Metallic fission product release that occurred during irradiation will be measured in the irradiation capsules that surrounded the fuel spheres in the irradiation rig after irradiation, and in the heating oven circuits during heating tests. Coated particle failures that occur during irradiation and heating tests can be quantified by means of increased fission gas release.

The first irradiation test will achieve burn-ups of 5 % fissions per initial metal atom (FIMA) and fast neutron doses of $1.7 \times 10^{21} \text{ n} \cdot \text{cm}^{-2}$. It will be used to qualify start-up fuel, in order to licence reactor operation in the first demonstration phase. The four fuel spheres will be subjected to post irradiation examination (PIE) and heating tests to demonstrate that fuel loaded into the

demonstration module can safely be irradiated to the particular burn-up value achieved in the irradiation test, and that it would perform satisfactorily under Design Basis Accident (DBA) conditions. Irradiation will be performed at 1200 °C centre fuel temperature. All four irradiated fuel spheres will be subjected to heating tests simulating DBA transient temperatures, first at 1600 °C for 100 h and then at 1800 °C for 100 h. One heated fuel sphere will be deconsolidated to provide coated particles for ceramography and fission product distribution measurements.

The second irradiation test will prove the ability of the fuel to reach final burn-ups of 11.3% FIMA and neutron doses of $3.6 \times 10^{21} \text{ cm}^{-2}$. Twelve fuel spheres will be irradiated, after which one fuel sphere will be deconsolidated immediately. Five fuel spheres will undergo post-irradiation heat-up testing to 1600 °C, and the rest to 1800 °C for 100 hours. One fuel sphere from each temperature test will be deconsolidated to provide coated particles for ceramography and fission product distribution measurements

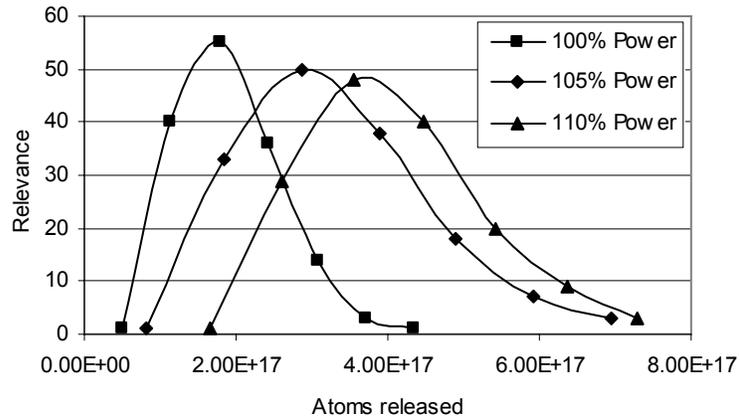
4. Fuel Performance and Fission Product Release Analyses

Fission product releases from coated fuel particles and spherical fuel elements used in modern high temperature gas cooled reactors is one of the first source terms used in describing the safety of planned nuclear plants during normal and accident conditions. Fission products are primarily released through diffusion through fuel materials according to Ficks' laws of diffusion (5). Since the layers of the coated particle provide an almost impenetrable barrier to diffusion (except silver and strontium), fission product sources are mainly failed particles and the heavy metal (uranium and thorium) contamination in the matrix material. Fuel failure probabilities are calculated by legacy software products (PANAMA) and PBMR developed models. Heavy metal contamination in the fuel materials is dependent on the quality of the fuel materials and manufacturing processes used by the manufacturing plant. Fission product diffusion from these sources, as well as from intact particles, is described by the differential equations of Ficks' laws, which are solved by computer software programs (eg. GETTER and FRESCO). For the special case of short-lived fission products, which are primarily gaseous fission products (noble gases, bromine and iodine), the Booth equation is utilised by the computer software program NOBLEG (6). These software programs are all legacy computer codes from the German fuel and reactor development program. They have been further developed at PBMR to fulfil specific PBMR design and licensing analyses requirements.

All foreseeable reactor operation conditions, occurrences and accident events can be modelled and analysed. Models and methods are continuously developed, verified and validated to calculate expected results based on the best available fuel data and reactor parameters, as well design values based on uncertainty ranges of all fuel data and reactor parameters. To perform design analyses, the software product FIPREX was developed by PBMR to perform Monte Carlo uncertainty analyses with the existing software programs. Fig. 4 presents a Monte Carlo uncertainty analyses performed on I-131 release from the PBMR core structure during a DLOFC event. The calculation was performed with FIPREX-GETTER, the fuel and reactor parameters selected from a Weibull distribution based on the uncertainty ranges of each parameter. The analysis was performed three times, with reactor power set to 100%, 105% and to 110%. The increased power rating causes higher stresses on the fuel, the operating envelope is pushed further, fuel and reactor parameters become more uncertain, and the distribution widens. This

figure also shows that the current 400 MW design is near the limit of the current fuel design envelope. Only 10% percent increase in reactor power and the design radioactivity release more than doubles.

Figure 4: I-131 total release from core during DLOFC for different power ratings



All calculation models, input parameters and values, software and methods must be verified and validated to the requirements laid down by the licensing authorities. Evaluations of German fuel element irradiation tests performed at the HFR at Petten currently form the bases of PBMR fuel analyses software validation. The gaseous fission product release code NOBLEG is validated using experimental results obtained during the HFR-K5 and HFR-K6 irradiation experiments conducted under relevant PBMR reactor conditions. The metallic fission product release code GETTER is partly validated by the post irradiation examination of HFR-K3. Gaseous fission product release measurements conducted during irradiation is used to validate noble gas transport in fuel materials. Fuel element deconsolidation and analyses after post irradiation heat-up tests evaluates fuel performance during accident conditions. Pertinent results and conclusion of evaluations completed are described and discussed in (7). Verification and validation of the software, calculation models and all fuel parameters are ongoing through analyses of all PBMR fuel qualification test and experiments. Special irradiation tests are planned to resolve specific issues such as silver transport in SiC and testing of alternative fuel materials.

In the long term a client specific software program is envisioned, that will perform all reactor control and fuel performance analyses for commercial PBMR units. An integrated system is foreseen that will determine reactor conditions for each operational mode and state, and provide quick and accurate prediction of radiological impacts on plant personnel during operation and to the public after specific accident events.

4. Conclusion

The PBMR company will in the following years licence and build the first commercial high temperature nuclear reactor. Central to the inherent safety claims of the design, is its fuel's ability to withstand all operational and accident conditions. The development of the PBMR fuel plant and fuel test and qualification facilities are at an advanced stage.

The current program has focused on the German fuel development, manufacture and qualification program. Evaluation of data from this program simplifies fuel design and qualification significantly. The design criteria have been derived and a fuel performance analysis of each criterion has been performed based on German fuel program data.

The future program revolves around assuring the quality of manufactured fuel, and proving that manufactured fuel can maintain its integrity under irradiation loads and temperatures during expected operational conditions and designed accident events.

Software and calculation models are continuously developed to predict fuel behaviour and specifically fission product release. It is important that all available irradiation tests and experiments are evaluated with current software to identify future software and analyses requirements.

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