A REVIEW OF FISSION GAS RELEASE DATA WITHIN THE NEA/IAEA IFPE DATABASE

J A Turnbull (Independent Consultant), P Menut (IAEA), and E Sartori (OECD/NEA)

ABSTRACT

The paper describes the International Fuel Performance Experimental Database (IFPE Database) on nuclear fuel performance. The aim of the project is to provide in the public domain, a comprehensive and well-qualified database on Zr clad UO_2 fuel for model development and code validation. The data encompass both normal and off-normal operation and include prototypic commercial irradiations as well as experiments performed in Material Testing Reactors. To date, the Database contains some 380 individual cases, the majority of which provide data on FGR either from in-pile pressure measurements or PIE techniques including puncturing, Electron Probe Micro Analysis (EPMA) and X-ray Fluorescence (XRF) measurements. The paper outlines parameters affecting fission gas release and highlights individual datasets addressing these issues.

INTRODUCTION

Data from PIE and in-pile measurement of rod internal pressure have shown that fractional release is very dependent on fuel temperatures. Predictive models based on single gas atom diffusion through the UO_2 lattice, with a characteristic temperature dependent diffusion coefficient, have been very successful for predicting data where the fractional release was equal to or greater than around 10%. However, such models were poor at predicting low values of gas release and release values at low irradiation temperatures or short times. In particular, PIE of modestly rated power reactor fuel showed very small fission gas release (FGR) even after 3 cycle irradiation and burn-up levels approaching 30 MWd/kgUO_2. In this regime, an empirical relation of 0.7% release per 10 MWd/kgUO_2 gave satisfactory predictions for a wide range of fuel designs.

These observations lead to the concept of a 'threshold' which distinguished between irradiation histories resulting in either 'low' or 'high' fractional fission gas release, and the conditions that resulted in a translation between the two regimes. With this approach, it is possible to reconcile the data in terms of: a low temperature burn-up dependent correlation, a criterion for the start of enhanced release based on burn-up and temperature, and a high temperature diffusion based release mechanism.

The perfection of in-pile instrumentation to measure fuel temperatures and rod internal pressure during irradiation allowed investigators at the Halden Reactor Project [1] to determine the point and conditions during irradiation when enhanced FGR commenced. From measured increases in pressure they derived an empirical threshold between 'low' i.e., <1% and 'high' i.e., >1% FGR in terms of fuel centreline temperature TF (°C) and burn-up BU (MWd/kgUO2) as follows:

$$BU = 0.005 \times \exp(9800/TF).$$

Although derived from Halden experimental fuel rods, the correlation has been shown to work with equal success for a variety of fuel manufactures and designs. Recent studies have shown that the criterion can also be successfully applied to MOX fuel [2]. Theoretical explanations for the threshold have been proposed in terms of the accumulation of gas atoms on grain boundaries by diffusion tempered by irradiation re-solution, [3].

With this general framework for FGR it is the purpose of the IFPE Database to provide sufficient information to allow modellers to develop individual models describing the several processes contributing to release and also allow them to validate code predictions as a whole. At present, the Database comprises some 380 individual cases, [4] and covers most of the parameters of interest for FGR modelling. These can be broadly grouped under: Rod Design, Vendor/Manufacturing Process and Irradiation History as elaborated in the following sub-sections. These are followed by a brief description of the most appropriate datasets and the areas were they contribute.

ROD DESIGN

Under this heading may be considered the effect of fuel-clad gap size, fill gas pressure and composition, axial transport of released gases to the plenum and the difference between hollow and solid pellets. The influence of gap size comes mainly through its effect on fuel temperatures where the larger the gap, the higher the temperatures across the pellet radius. Later during irradiation, small or closed gaps formed as a result of clad creep-down and fuel swelling also restrict axial transport of released fission gases along the active fuel column. This leads to a 'poisoning' of the gap thermal conductivity and a positive feedback on FGR. This poisoning is reduced for high initial helium fill pressures, and rod pressure is a variable addressed in several programs, e.g. the High Burn-up Effects Program (HBEP).

By removing the central part of a fuel pellet it was hoped to reduce centreline temperatures and hence reduce FGR. Unfortunately this aim is confounded by the need for higher linear heat rates for hollow compared to solid pellets in order to extract the same amount of power. However, there are other perceived benefits for hollow pellets, namely in reducing pellet clad mechanical interaction (PCMI). A comparison of performance can be obtained from within the HBEP and, the Siemens RE Ginna experiment datasets.

VENDOR/MANUFACTURING PROCESS

Even with identical rod design and irradiation conditions, fuel rods supplied by different vendors will not behave identically, due to differences in pellet fabrication route and differences in clad properties, particularly clad creep-down as indicated above. The major influences on FGR come from UO_2 grain size, as this affects the diffusion distance that a gas atom must cover before being released. The effects of additions like gadolinia (Gd_2O_5) as a burnable poison or Nb_2O_5 or Cr_2O_3 as a grain growth promoter can affect both the UO_2 thermal conductivity and the gas atom diffusion coefficient. Pellet density and dimensional stability affect the thermal behaviour of the pellet and therefore indirectly the fission gas release. Regarding dimensional stability, pore size distribution and not just density is the important parameter; for the same density, a distribution of large pores is more stable against densification than a distribution of small pores and is therefore likely to show a lower FGR behaviour.

IRRADIATION HISTORY

Under this heading can be considered: normal operation, power transients, power cycling, loading scheme and discharge burn-up. These are generic to any type of reactor; in addition, reactor type can be added: PWR, BWR, CANDU, WWER etc. as each have different modes of operation. With the single exception of power cycling, the IFPE Database addresses all of these. It is hoped to add power cycling in the future, but many experiments have shown that this mode of operation does not have a specific influence on FGR. Although not addressed further in this paper, it is worth noting that the Database includes information on the escape of fission products from defected fuel under different regimes of power including power cycling where the enhanced release of ^{131}I becomes very important from a reactor safety stand point.

IFPE DATASETS

This section gives a brief description of the datasets containing important information regarding the fission gas release process and the parameters influencing it. The Database is available to anyone contacting the NEA in Paris and is supplied on a CD which includes all datafiles in ASCII format and accompanying reference documents as PDF files.

High Burn-up Effects Program

The HBEP was an international, group-sponsored program managed by Battelle North West Laboratories whose principal objective was to obtain well-characterised data on fission gas release for typical LWR fuel irradiated to high burn-up levels. The program was organised into three tasks. Under Task 2, 45 existing fuel rods, either at moderate burn-up levels or undergoing irradiation to higher burn-up levels, were identified, acquired and subjected to PIE. Some rods were also subjected to power-bumping irradiations. In Task 3, a series of fuel rods were built for irradiation in BR3 to high burn-up levels. Four design variations and three variations in operational history were used to study the effect of design and operation parameters on high burn-up FGR. This program provides a substantial amount of data on fission gas release and the variations observed with different vendors and manufacturing routes, different reactor systems and different characteristics of the fuel. In the latter case it is possible to quantify the different release between: high and low internal pressure, hollow versus solid pellets, the effect of different grain sizes and the effect of adding gadolinia to the UO_2 pellets. A total of 45 rods were considered under Task 2 and 37 rods for Task 3 all of which have been included in the database.

To investigate the relationship between fuel microstructure and local retention of fission products, three special post-irradiation examinations were performed in addition to the standard post-irradiation examinations (visual, rod puncture and gas analysis, ceramography). The special examinations included electron probe microanalysis (EPMA) and x-ray fluorescence (XRF) for radial profiles of retained fission products and scanning electron microscopy (SEM) to supplement the optical microscopy examinations of the fuel microstructure. Using these techniques, fission gas atom depletion in the ‘rim’ region was detected and measured. Also, It has been assumed that grain size must be doubled in order to obtain significant local FGR through grain boundary sweeping. No correlation between a doubling of grain size and local FGR (as measured by EPMA and XRF) was discerned for the HBEP fuels. This applied to both bumped and non-bumped fuel rods.

The Risø Transient Fission Gas Release Project and the Third Risø Fission Gas Release Project

In the Risø Transient Fission Gas Release Project (Risø II), short lengths of irradiated fuel were fitted with in-pile pressure transducers and ramped in the Risø DR3 reactor. The fuel used came from either IFA-161 irradiated in the Halden reactor or from segments irradiated in the Millstone BWR. Using this refabrication technique, it was possible to back fill the test segment with a choice of gas and gas pressure and to measure the time dependence of fission gas release by continuous monitoring of the plenum pressure. The short length of the test segment was an advantage because, depending on where along the original rod the section was taken, burn-up could be a chosen variable, and during the test the fuel experienced a single power. Some segments were tested without refabrication. Here the fuel stack was longer than in the case of the refabricated tests and hence the segments experienced a range of powers during the ramp depending on axial position in the test reactor. These 'un-opened' segments were used to confirm that refabrication did not affect the outcome of the tests. Extensive hot cell examination compared the fuel dimensions and microstructure before and after the tests.

Some 17 tests were performed and all but one (which failed) have been included in the database and provide valuable information on fission gas release during power transients at high burn-up as well as clad diametral deformation and fuel swelling as a function of ramp power and hold time. Figure 1 shows the evolution of fission gas release as a function of time during the power ramp for one of the tests using fuel from IFA-161. At each step, the fractional gas release shows a square root dependence on time which is characteristic of release by a diffusion type process. The sudden increase in release at the end of the test on decreasing power shows that towards the end of the hold time, there was contact between the fuel and the cladding, thus causing a restriction to the axial communication to the plenum where the pressure transducer was situated. This was confirmed by a comparison of diameter traces before and after the test which showed no significant diameter increase but a significant increase in permanent ridge height. The database also includes diametral profiles of retained fission products measured by EPMA and XRF. The radial position for the onset of release is clearly evident in Figure 2. It is argued that the difference between the two types of measurements on retained xenon can be taken as the gas residing on grain boundaries; i.e. the difference between the total gas content (XRF) and the gas residing only in the matrix (EPMA).

The third and final Risø Project bump tested fuel re-instrumented with both pressure transducers and fuel centreline thermocouples. The innovative technique employed for re-fabrication involved freezing the fuel rod to hold the fuel fragments in position before cutting and drilling away the centre part of the solid pellets to accommodate the new thermocouple. The fuel used in the project was from: IFA-161 irradiated in the Halden BWR between 13 and 46 MWd/kg UO_2, GE BWR fuel irradiated in Quad Cities 1 and Millstone 1 between 20 and 40 MWd/kg UO_2 and ANF PWR fuel irradiated in Biblis A to 38 MWd/kg UO_2. The data from the project are particularly valuable because of the in-pile fuel temperature and pressures measurements as well as extensive PIE. The database includes seven cases with ANF PWR fuel, six cases with GE BWR fuel and two cases using fuel from IFA-161. Within the test matrix it was demonstrated that the refabrication did not interfere with the outcome of the tests and that there was a correspondence with the results of the previous project. FGR data are available in both projects for the end of the pre-irradiation, during and after the ramp tests.

WWER Data

Representing WWER fuelled reactors, FGR data are available from 2 low burn-up un-instrumented rods from the SOFIT 1.1 program. The Database also includes data for two pre-characterised standard WWER-440 fuel assemblies: FA-198 and FA-222 manufactured by the Russian fuel vendor Elektrostal and irradiated in the Kola-3 reactor. These assemblies were the centre of a program called Blind Calculations for the WWER-440 High Burn-up Fuel Cycles Validation, initiated in Spring 1994 with the objective of testing the predictive capabilities of several Russian codes. The maximum linear heat generation rate (LHGR) of FA-198 was <31 kW/m at the beginning of life and decreased to about 14 kW/m by the beginning of the fourth cycle and 11 kW/m at the end of life. In FA-222, peak LHGR values of 21 to 26 KW/m were experienced at the beginning of the second cycle, followed by steady state operation at LHGR of 10 to 22 kW/m before gradually decreasing to around 8 kW/m at the end of life. During the whole of the irradiation, both assemblies were located remote from any control rods. Consequently, the irradiation conditions are considered representative of base load operation for WWER-440 reactors.

The database contains details of 16 rods from each assembly; these are the corner rods 1, 7, 58, 69, 120 and 126 and rods along the diagonal shown in Figure 3. As well as comprehensive pre-characterisation, the data include detailed 10 zone irradiation histories and PIE observations of dimensional changes and fission gas release. The measured gas release varied from ~0.5% for the diagonal rods to ~1.2% for the FA-198 corner rods, and 1-1.6% for the diagonal rods to 2.3-3.7% for the corner rods of the higher burn-up FA-222.

Halden Project Instrumented Fuel Assemblies

The Halden irradiated IFA-432 was commissioned by the USNRC with the objectives to measure fuel temperature response, fission gas release and mechanical interaction on BWR-type fuel rods up to high burn-ups. The assembly featured several variations in rod design parameters, including fuel type, fuel/cladding gap size, fill gas composition (He and Xe) and fuel stability. It comprised six BWR-type fuel rods with fuel centreline thermocouples at two horizontal planes. Rods were also equipped with pressure transducers and cladding extensometers. Data from five rods have been included in the database providing in-pile and limited PIE data up to 46 MWd/kgUO_2. From retained gas measurements it was found that lowest release (6 %) occurred in the small gap rod 3, whilst larger amount of release were found in the large gap rods, 24-35% in the 95 %TD rod 2 and 46% in the 92 %TD rod 5.

IFA-429 consisted of PWR type fuel rods assembled in three axially separated clusters of six rods. The eighteen-rod assembly had been designed to investigate gas absorption, fission gas release and thermal behaviour of UO_2 fuel during both steady state and a period of repeated rapid power transients. In order to achieve the objectives, the assembly was instrumented with nine vanadium neutron detectors, and one cobalt detector. Two rods were instrumented with fuel centre line thermocouples and nine rods were instrumented with null-balance gas pressure transducers monitoring rod internal gas pressure.

The Database contains power histories for seven rods as well as the measured temperature history for one of the middle cluster rods up to the burn-up level of 53 MWd/kgUO_2 and the measured internal pressure data for three upper and three lower cluster steady-state irradiated fuel rods subjected to two series of rapid power transients. Manually initiated gas pressure measurements were performed during the testing sequence allowing comparison of FGR data at three different fuel densities (91% TD, 93% TD and 95% TD), at two different grain sizes (6 and 17 microns) and at two different fuel-cladding gap sizes (200 and 360 microns). The parabolic form of release as a function of time at power is indicative of release by a diffusion controlled process.

After twelve years pre-irradiation, two fuel rods were re-instrumented with fuel centre thermocouples and reloaded as IFA-533.2 into the reactor in order to investigate fuel thermal behaviour at high burn-up. Also, four neighbouring rods were re-instrumented with pressure transducers and ramp tested in IFA-535.5 (slow) and IFA-535.6 (fast) providing useful data about FGR at two different ramp rates, Figure 4. As the irradiation history of IFA-533.2 in the first months was very similar to the history of the ramp tests, the fuel temperature and FGR data measured in the different IFAs complement each other, although the fuel-cladding gap sizes were slightly different and due to re-instrumentation the internal gas conditions were also dissimilar.

The Belgo-Nucléaire Tribulation Project

The objectives of the TRIBULATION program were twofold. It was primarily a demonstration program aimed at assessing the fuel rod behaviour at high burn-up, when an earlier transient had occurred in the power plant. The second objective was to investigate the behaviour of different fuel rod designs and manufacturers when subjected to a steady state irradiation history to high burn-up.

The first objective was met by irradiating fuel rods under steady state conditions in the BR3 reactor and under transient conditions in BR2. The effect of the transient was determined by comparing data from four identical rods tested as follows:

BR3 irradiation followed by PIE;
BR3 irradiation followed by BR2 transient then PIE;
BR3 irradiation followed by BR2 transient and re-irradiated in BR3 before PIE;
BR3 irradiation and continued BR3 irradiation to maximum burn-up before PIE.

The Database contains data from 19 cases using rods fabricated by Belgo-Nucléaire (BN) and Brown Boveri Reactor GmbH (BBR). The matrix provides good data on clad creepdown and ovality as a function of exposure as well as the effect of the different irradiation histories on fission gas release. Values of FGR ranged between 1 and 12 % depending on irradiation history.

Studsvik Projects

The 87 PWR and BWR rods included from the Studsvik ramp tests are a sub-set of the total data available on the failure propensity in power ramps by PCI and SCC. In all cases, the in-pile testing was followed by an extensive PIE program including diameter changes before and after ramping and fission gas release measurements on unfailed rods. The rods were supplied by different fuel vendors and embrace both PWR and BWR designs. The rods tested include variants such as: gadolinia doped UO_2, large grain UO_2, fuel of different density and annular pellets.

CEA/EDF/Framatome Data

Data on prototypic commercial PWR fuel performance were obtained with agreement from EDF, CEA and Framatome. The data are for 4 full length rods irradiated in EDF reactors. Rods K11 and J12 were irradiated for 2 cycles in Gravelines 3 and 5 respectively to ~24 MWd/kgU, G07 was irradiated for 3 cycles in Gravelines 3 to ~35 MWd/kg and rod H09 was irradiated in Cruas 2 for 4 cycles to ~46 MWd/kgU. These rods were well characterised prior to irradiation and subjected to extensive PIE after irradiation. During PIE measurements made included: diameter change, oxide thickness, hydrogen content of cladding, length change, fission gas release, pellet density, radial distribution of fission products and actinides and metallography. Care was taken to accurately reproduce the axial power profile when processing the data by constructing 18 axial zone power histories reflecting the difference in power at and between the grids. FGR had a minimum value of 0.2% for the 2 cycle rod K11 and a maximum value for the 4 cycle rod H09.

Sections of rods J12 and K11 were cut from span 5 and re-fabricated for ramp testing in the CEA OSIRIS reactor at Saclay. Rodlet J12-5 was conditioned to 21 kW/m before ramping to 39.5 kW/m without failure. Rodlet K11-5 was conditioned at 24 kW/m and ramped to 43.7 kW/m without failure. Subsequent PIE provided measurements of clad diameter changes, fission gas release and metallography of the fuel structure at different elevations. The FGR measured on these segments was 0.74 % and 6.3 % for J12-5 and K11-5 respectively.

The CONTACT series of experiments was a program of in-pile tests conducted in the SILOE reactor in Grenoble, France, funded jointly between CEA and Framatome. They were short rods of Zr-4 clad UO_2 pellets of typical PWR 17×17 design, irradiated under conditions designed to simulate commercial PWR conditions. Each rod was equipped with a fuel centreline thermocouple, diameter gauge, gas lines providing a flow of gas through the rod and internal pressure gauges to measure pressure drop along the fuel stack. The gas flow entrained released fission gases which were measured by a gamma detector installed in the out-of-reactor gas handling system. The experiment is unique in that the rods operated under near constant powers for the majority of their lives. CONTACT 1 operated at a constant 40 kW/m up to a burn-up of ~22 MWd/kgU whilst CONTACT 2 and 2bis operated at 25 kW/m to burn-up levels of 5.5 and 12.4 MWd/kgU respectively. The data include temperatures as a function of burn-up, clad diameter changes as a function of power and burn-up, stable (^{85}Kr) and radioactive fission gas release as a function of centre temperature and burn-up. The kinetics of stable fission gas release as measured with the long half-life ^{85}Kr are shown in Figure 5. The release of the radioactive species are useful for developing and validating models for calculating 'gap inventories' of radiologically significant species like, for example, $^{131-}$I.

Siemens PWR Rods Irradiated in the R E Ginna Reactor

Under a co-operative agreement between Siemens Power Corporation, Empire State Electric Energy Research Corporation (ESEERCO) and Rochester Gas and Electric Corporation (RG&E) 4 14×14 demountable test assemblies (DTAs) were loaded into the R E Ginna reactor in 1985 and irradiated for 4 cycles (42.5 MWd/kgU) with one LTA irradiated for a further cycle (52.1 MWd/kgU). Two of the DTAs contained 11 segmented rods each. The segmented rods consisted of four segments, the centre two of which were pre-characterised and examined in detail after irradiation. Three combinations of pellet design (annular and solid) and cladding type (Zr-4 with sponge zirconium liner, and through wall Zr-4) were used in the fabrication of the segmented rods. The fuel-to-clad gap was another design variable with values of 160, 190 or 216 μm. The objective of the program was to develop a fuel design with increased margin to failure, increased high burn-up potential and to obtain performance data up to high burn-up for use in fuel modelling. The power history during the 4-5 cycle irradiation was quite onerous, averaging 20, 30, 25, 10 and 22 kW/m in cycles 1-5 respectively.

The PIE included the usual measurements of dimensional changes, oxide thickness, fission gas release and metallography and data for 17 rodlets are included in the Database. Fission gas release was measured nondestructively at poolside by determining the ^{85}Kr concentration in the plenum using gamma spectrometry as well as by conventional puncturing and mass spectrometry. The highest FGR measured was 2.36% for a five-cycle solid pellet rodlet (55N5), while the lowest FGR was 0.83% for a five-cycle annular pellet rodlet (AZW5). These data confirmed the expectation that annular pellets would lead to lower fission gas release and substantially lower final internal pressure.

IMC Out-of-Pile Annealing Data

The next dataset is the first departure in type of data from previous additions as it comprises a set of measurements made of fission gas release during out-of-pile annealing experiments. Small samples of UO_2 fuel were extracted from CAGR fuel pins and annealed in helium at pre-defined temperatures 1500 to 1900°C for periods between 2 and 40 hours. The rate at which the final temperature was attained varied from 0.1 to 8.0 °C/s in order to determine whether or not the behaviour of intragranular bubbles, present during and after irradiation, was sensitive to changes in temperature ramp rate within this range. The fuel used for this study had a burn-up of ~17 MWd/kgU and a mean linear intercept (mli) grain size of 6 or 18 μm. Fission gas release at the end of the base irradiation was low in both cases, <0.1%, with no visible grain boundary porosity. Examples of the results are illustrated in Figure 6. The figure shows the effect of grain size for a ramp rate of 0.5°C/s followed by a hold at 1800°C for 6 hours. These experiments provide valuable data for developing models of fission gas release during rapid high temperature transients where the effect of the irradiation conditions is small compared to that of temperature and time. Note that the classical parabolic diffusion release kinetics evident at short times changes to a more linear behaviour at long times.

CONCLUSION AND ACKNOWLEDGEMENTS

It should be noted that other datasets not discussed here are included in the Database, e.g. for CANDU type reactors, and the extent of the Database is constantly being increased with the inclusion of new data as they become available. Nevertheless, it is concluded that in its current state, the database already contains much of the information required to develop and validate fission gas release models for inclusion in fuel performance codes for application to any reactor system over a wide spectrum of operational and transient conditions.

In compiling this Database, the authors wish to acknowledge the co-operation of the following participating organisations for the supply of their data and assistance in preparing the database: Risø National Laboratory, Denmark; Halden Project, Norway; Imatran Voima Oy, Finland; The Kurchatov Institute and VNIINM, Russian Federation; Battelle North West, USA; Belgo-Nucleaire, Belgium; Studsvik Nuclear AB, Sweden; CEA, EDF and Framatome, France, AECL Canada, HSE/IMC of the UK and SPC of USA.

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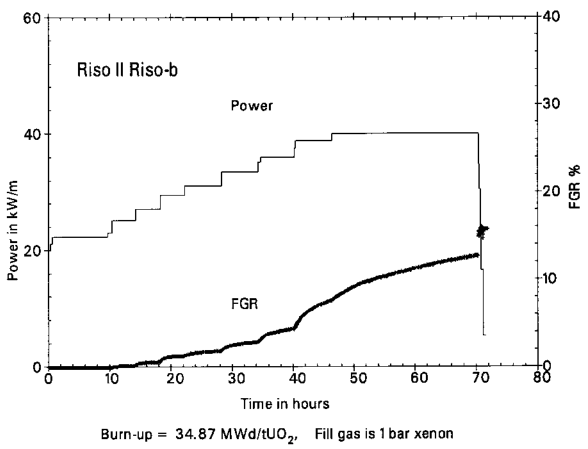

Figure 1.

Fission gas release as determined from rod internal pressure for test Risø-b (Risø Transient Fission Gas Release Project) during the power ramp in the DR3 reactor

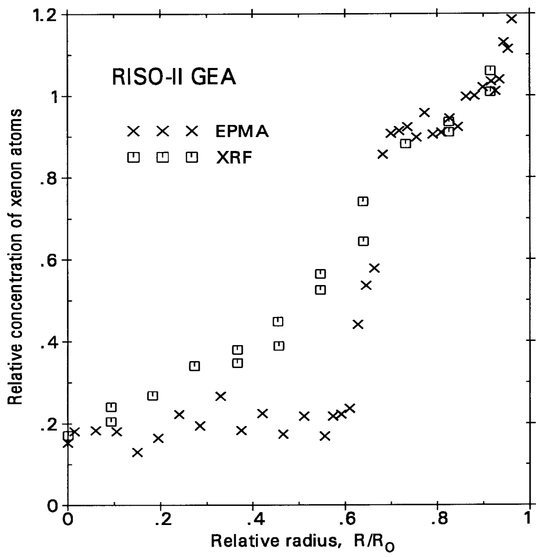

Figure 2.

Comparison of EPMA and XRF retained xenon profiles for test GEa of the Risø Transient Fission Gas Release Project

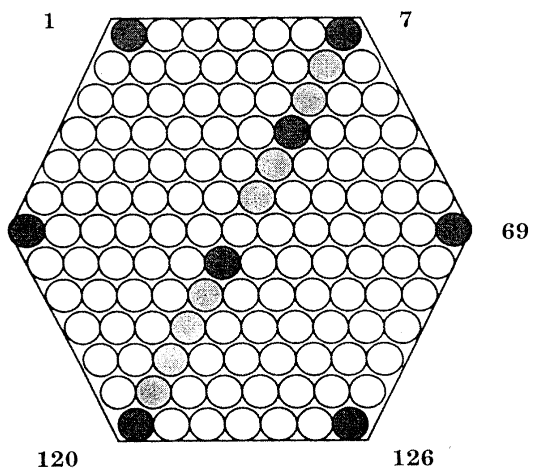

Figure 3.

Arrangement and numbering of rods in the WWER-440 assemblies FA-198 and FA-222 irradiated in the Kola-3 reactor

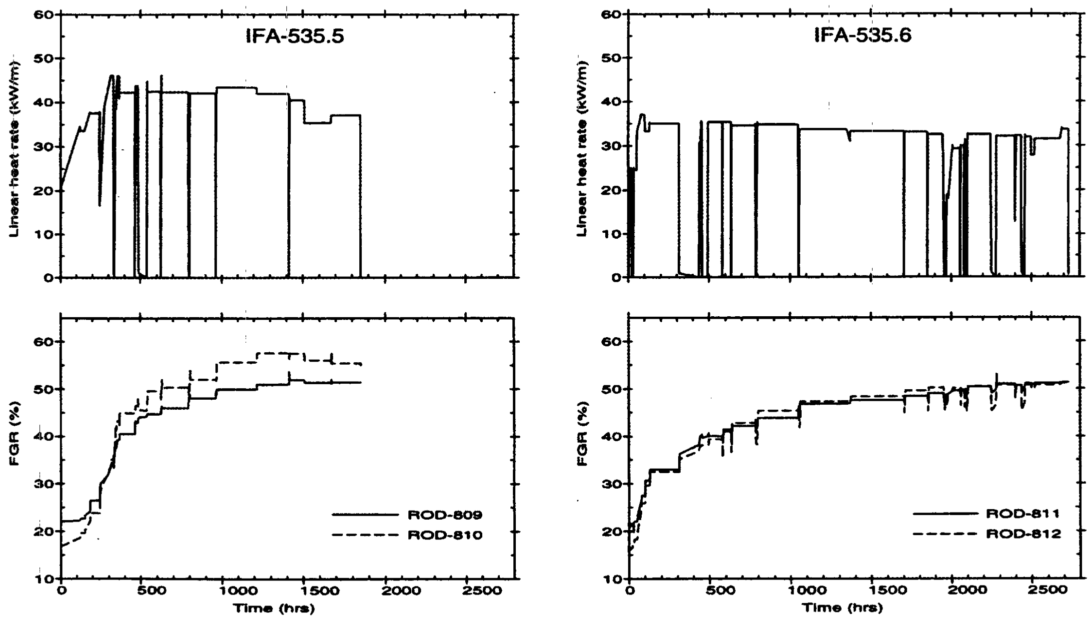

Figure 4. Fission gas release for the slow ramped IFA-535.5 and the fast ramped IFA-535.6

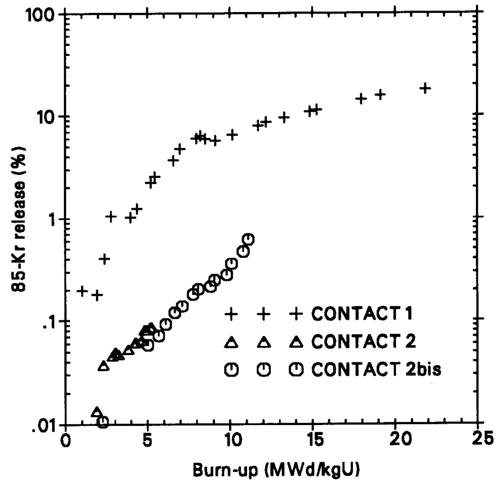

Figure 5.
^{85}Kr release as a function of burn-up for the three CONTACT experiments 1, 2 and 2bis.

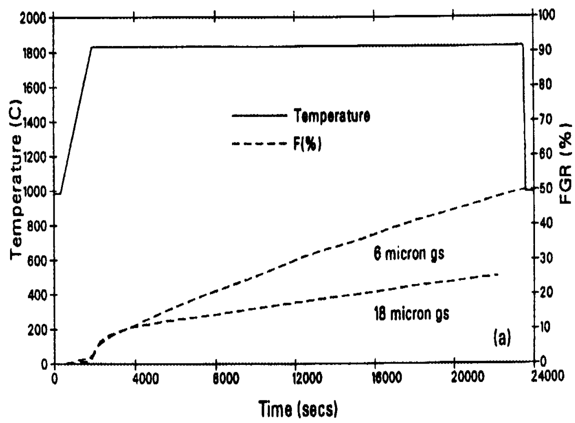

Figure 6.
Fractional release of ^{85}Kr during IMC out-of-pile annealing experiments showing the effect of different grain sizes