Release Rates of Short-lived Fission Gases from Modern Spherical Fuel Elements with TRISO-coated Particles

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Abstract. Measurement and prediction of fission product release is important in design, licensing and operation of HTRs. Latest development status is near zero particle manufacturing defects and in-pile failures combined with very low matrix contamination levels. In-pile Release over Birth rate measurements will be described and evaluated. Quantitative predictive analyses of short-lived gas release will be provided for MTR irradiation testing with a focus on the most recent experiment with Chinese fuel from HTR-PM production. For irradiation tests with no single particle failure, models describing particle kernel release are not applicable; instead modeling of matrix contamination release is essential. Good agreement between post calculations and in-line R/B measurements has been achieved.

1. Introduction
Measurement and prediction of fission product release from the core is important in design, licensing and operation of High Temperature Reactors (HTRs). Both solid fission product release and release rates of short-lived fission gases have come down significantly during the last decades:

(i) Early HTR fuel was uncoated and direct release from the fuel was large. Reactors were built with external fission product trap trains.

(ii) The introduction of dense coating layers around the bare fuel kernel (for better handling) improved the retention significantly, but early fuel elements had high contamination levels in the particle’s outer pyrocarbon (oPyC) layer and in the fuel element matrix.

(iii) The systematic introduction of high quality TRISO particles enabled spheres with low contamination levels in the oPyC and in the fuel element matrix. At this stage, releases were dominated by the small number of manufacturing defects, at most one broken particle per three fuel spheres.

(iv) Latest development status is the near zero particle (manufacturing) defects and (in-pile) failures that go hand-in-hand with a further significant reduction of matrix contamination levels.

In-reactor R/B (= Release rate over Birth rate) measurements will be described and evaluated for irradiation tests of spherical fuel elements in the High Flux Reactor (HFR) Petten. Considerations will also be given to historic R/B measurements in operating HTRs AVR, THTR, and Fort St. Vrain. Quantitative predictive analyses of short-lived gas release will be provided for stage (iii) and (iv) irradiation tests in Material Test Reactors (MTRs). Here, the model development from stage (ii) can be
fully used although the traces of natural uranium and thorium are now lower by three or four orders of magnitudes.

With no defective and/or failed particles present, releases to be studied are from contamination of the matrix graphite (and the graphite cups in the MTR tests). They have the following components: inter-grain diffusion, grain boundary diffusion, recoil from grains into the pores, and transport in the pores the combination of gas-in-gas diffusion and Knudsen diffusion.

The modern intact HTR coated particle retains all fission gases. Therefore, the measured gas releases are, at a first approximation, related to the number of particles with defective coatings from manufacture and the number of coating failures during irradiation. This way, the failed particle fraction is determined experimentally in any fuel assembly from

\[ \Phi = \frac{(R/B)_{\text{measured}}}{(R/B)_{\text{defective particle}}} \]

where the defective-particle R/B can be from a model or from a dedicated experiment with designed-to-fail (DTF) particles.

An additional source of fissionable material might be from heavy metal contaminations in the outer pyrocarbon coatings of particles, in the matrix graphite of the fuel body and other graphite components in the irradiation setup.

Iodine is an important fission product in source term predictions [1], but iodine releases are not easily determined in MTR tests and subsequent PIE work:

- During MTR irradiations, the helium purge gas transports continuously released Kr and Xe isotopes to outside measurement stations, but condensable iodine stays within the irradiation capsule.
- Solid fission product release from spherical fuel elements is obtained from measuring the graphite cups that surround the spheres during irradiation and the inner surfaces of the steel capsule. Rig disassembly and transport of components to suitable hot cells equipped with the necessary instrumentation takes usually weeks and months by which time short-lived I-131 is already decayed.

To overcome the iodine detection problem, the MTR post-cycle xenon measurements that are in secular equilibrium with iodine in the rig can be used. In this report, it will be demonstrated how to analyze the data and how to derive quantitative R/B data for I-133 and I-135 from the irradiation tests.

2. Release of short-lived noble fission gases from operated HTRs

After the fully releasing HTR fuels in the 1960s, acceptable R/B(Kr-85m) levels were – during 70s and 80s – in the range 1–8×10^-6 as shown for Fort St. Vrain, THTR, and AVR (Fig. 1). The development of the modern spheres and compacts with clean and strong TRISO particles featuring a SiC interlayer between pyrocarbon coating layers reduced R/Bs down to the range 1×10^-10 – 3×10^-7.

Based on pre-existing particle failure prediction models, on the quality control/quality assurance (QC/QA) results from particle and fuel element manufacture including measured contamination levels in the graphitic components and operational data including breeding of fissile material from natural uranium and thorium, it has been attempted to predict release rates during Fort St. Vrain power operations [1]. While the particle failure models appear somewhat conservative, the level of R/B measurements is well bounded between the two modeling assumptions of release from contamination only and from contamination plus failed particles.

THTR fission gas release predictions worked well during the first 90 days of full power operation. Yet, after forced control rod injection exercises as ordered by the TUEV, many spheres and even coated particles were broken mechanically with subsequent increase of release rates [2]. More significantly, the release mechanisms changed completely from the usual diffusion, decay, and release sequence to knockout from crushed particles as observed by the change of slope from 0.32 down to 0.15 (see Fig. 1), where:
\[
\text{slope} = \frac{\log(R/B)}{\log(T_{1/2})}
\]

is the mean value over all short-lived fission gases and \( T_{1/2} \) is the half-life (s) of the respective short-lived fission gases.

Releases in AVR are simple to interpret: most spheres had a \( 3 \times 10^{-4} \) heavy metal contamination and with the recommended diffusions coefficients of Kr and Xe in UO\(_2\) [3], R/B values for all krypton and xenon isotopes can be easily predicted (green dashed line in Fig. 1).

R/B levels shown in Fig. 1 are typical of HTRs in the 1970s and the 1980s. With modern fuel, release rates of short-lived noble fission gases are by factors ~1000 lower.

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{Release rates of short-lived noble fission gases from operating reactors AVR, THTR and Fort St. Vrain. THTR 150 efpd (equivalent full power days) refers to the time after forced injection of control rods. The range of half-lives goes from 9 seconds to 5.25 days.}
\end{figure}

Perfect spheres with perfect TRISO particles were made by HOBEG/NUKEM from December 1978 onwards. These were then used in subsequent AVR reloads 15 and 20 with HEU (Th,U)O\(_2\) LTI TRISO and reloads 19, 21, 21-2 with LEU UO\(_2\) LTI TRISO. Since AVR still contained 50% dirty old fuel, the massive improvement of fission product releases showed up only to a limited extent.

Examples of modern clean LEU UO\(_2\) TRISO fuel from MTR irradiations tests are given in the following Figures 2 and 3.

Two irradiation proof tests, HFR-K5 and HFR-K6, were performed for the qualification of spherical fuel elements for the 200 MW(th) HTR-Modul reactor. In these tests with four spheres each, the temperature cycles characteristic of multi-pass pebble-bed reactors were simulated throughout the irradiation in HFR Petten. Selected results are shown in Fig. 2 for capsule B of HFR-K6, where the top rig A contained one sphere, the mid-rig B the spheres HFR-K6/2 and 3 (no defects, no failures), and the bottom rig C contained one sphere HFR-K6/4 with two initial manufacturing defects, but zero in-reactor failures.
Figure 2. Measured and predicted Kr-88 release rates in the temperature-cycled HTR-Modul Proof Test irradiation. The two spherical fuel elements in mid-HFR-K6 capsule B had zero initial defects and zero failed particles throughout irradiation. The predictive calculations used the Röllig 1977 model [4,5,6].

While good models for fission gas release from the various sources of contamination are existing, there are two major problems for a precise numerical prediction:

- Urgently needed are independent measurements of the contamination levels of $U_{nat}$ and Th in the matrix of the spherical fuel elements and in the binderless graphite of the surrounding cups. Even if such measurements are available, they are usually quoted as $< 1$ ppm and that is not good enough, because from our evaluation we must assume values between 1 ppb and 100 ppb.
- Second problem is that the reactor station makes precise and detailed calculations for the depletion of enriched uranium and the breeding of plutonium for the fuel in the coated particles. Since there are no defects and no failures, this fissile amount is not the real source of fission gas release rates. It is rather the natural uranium and thorium contamination in graphitic materials, for which the reactor station does not provide the necessary nucleonic predictions. An earlier attempt to overcome these problems was made by Alain Marmier [7].

In another MTR test HFR-EU1, the bottom capsule contained three German spheres from AVR reload 21-2 with the best QC/QA data ever in German sphere manufacture. Throughout irradiation, no defects nor failures were observed (Fig. 3) [8]. The top capsule of MTR test HFR-EU1 contained two Chinese spheres from the HTR-10 production demonstrating excellent irradiation behavior much better than earlier HTR-10 fuel tested in experiment VOSTOK-6 in IVV-2M [9].
One of the most successful irradiation tests of all times was HFR-PM containing high quality spherical fuel elements from the HTR-PM production. The results from operating this test [10] will be analyzed in great detail.

3. Mechanisms of fission gas release from contamination and mathematical modeling

In HTR fuel particles, fission products are retained by the dense coating layers around the fuel kernel. A very small fraction of particles may have coating defects from manufacture or the coating may have failed during irradiation. The pertinent modeling and necessary data are contained in IAEA-TECDOC- 978 [3].

Additionally, we may have heavy metal contaminations, U_{nat} and Th, in the graphite components outside the coated particles that contribute to fission product release.

3.1. Release from heavy metal contamination

The release rate of short-lived fission gases from contamination in graphitic components has been modeled since the early 1970s [4,5,11,12,13,14].

**Figure 3.** Release rates of short-lived fission gases from Petten irradiation tests HFR-EU1 (upper capsule with HTR-10 spheres (top), lower capsule with German spheres (bottom)).
Fissile material that exists or is bred from thorium and (natural) uranium traces outside of coated particles translates into a so-called effective contamination (see later Fig. 9). This contribution to releases has become important with recent irradiation tests of spherical fuel elements having zero defective particles and zero in-reactor failures during irradiation.

The contamination-generated gases escape via three routes into the HTR coolant stream:

1. Diffusion in A3-matrix graphite grains;
2. Diffusion in the amorphous component of the A3-matrix;
3. Direct recoil into pores of the A3-matrix or the graphite components;

This is shown in Fig. 4.

Diffusion in and from grains and the amorphous binder is mathematically described by the Equivalent Sphere Model predicting the ratio of steady state release rate to birth rate by

\[ \frac{R}{B} = \frac{3}{x} \left( \coth x - \frac{1}{x} \right) \]

where \( \lambda \) – decay constant [s\(^{-1}\)], \( D' \) – reduced diffusion coefficient [s\(^{-1}\)], \( Q \) – activation energy [J/mol], \( R=8.3145 \text{ J/(mol} \cdot \text{K)} \) gas constant, and \( T \) – temperature [K].

Early work on heavily contaminated THTR spherical fuel elements with HEU (Th,U)O\(_2\) HTI BISO particles had established the values as given in Table 1 [12].

**Table 1.** Parameters for the prediction of gas release rates from contamination in A3.

| Parameter                      | Krypton   | Xenon   |
|--------------------------------|-----------|---------|
| Frequency factor D'\(_0\) [s\(^{-1}\)] | 3.04×10\(^{-5}\) | 1.67×10\(^{-7}\) |
| Grain fraction                 |           |         |
| Amorphous fraction             | 1.7×10\(^{-2}\) | 1.7×10\(^{-2}\) |
| Activation energy Q [kJ/mol]   |           |         |
| Grain fraction                 | 106       | 78.6    |
| Amorphous fraction             | 54        | 54      |
| Amorphous fraction in A3       | 0.005     |         |
| Recoil fraction into pores     | 0.0002    |         |
With the amorphous fraction and recoil fraction into pores given in Table 1, the R/B ratios of the noble gas isotopes released from the heavy metal contaminations in the A3 matrix can be finally calculated with their corresponding decay constants as a function of irradiation temperature. They are given in Table 2 for some temperature values.

### Table 2. Effective total R/B values from contamination in A3 matrix according to the Röllig model [4].

| Temperature (°C) | Isotope    | 700 | 900  | 1100 | 1300 |
|-----------------|------------|-----|------|------|------|
|                 | Kr-85m     | 0.0082 | 0.0159 | 0.0291 | 0.0482 |
|                 | Kr-87      | 0.0057 | 0.0104 | 0.0177 | 0.0281 |
|                 | Kr-88      | 0.0072 | 0.0136 | 0.0242 | 0.0395 |
|                 | Xe-133     | 0.0128 | 0.0226 | 0.0364 | 0.0533 |
|                 | Xe-135     | 0.0051 | 0.0070 | 0.0090 | 0.0113 |
|                 | Xe-135m    | 0.0026 | 0.0041 | 0.0055 | 0.0067 |

Fission gas transport in the pores of the A3 matrix proceeds by two parallel processes: (i) Knudsen diffusion, and (ii) gas-in-gas diffusion, and these are determined by different material and operational parameters:

- Knudsen diffusion is predicted by temperature, molar mass of the diffusing species and the pore diameter (assumption here: 270 nm); the Kr-88 diffusion coefficient at 1000°C is $3.32 \times 10^{-6}$ m²/s, independent of pressure.
- Gas-in-gas diffusion describes the competition between the released Kr, Xe atoms and He in the pores. This makes the transport dependent on temperature, pressure, the respective atomic weights and the collision integrals.

Here, transport between the 0.1 MPa in an MTR test and the, e.g., 7 MPa in an operating HTR can make a significant difference. The combination of Knudsen and gas-to-gas diffusion processes (modeled by taking the harmonic mean of the respective diffusion coefficients much like two parallel electrical circuits) at 1000°C, for example, results in a Kr-88 diffusion coefficient of $2.33 \times 10^{-6}$ m²/s in an MTR at atmospheric pressure and $0.11 \times 10^{-6}$ m²/s in the operating HTR.

Finally, transport is further hindered by porosity and tortuosity. The A3 matrix has a total 20% porosity and a tortuosity factor of 3 is assumed.

### 3.2. Release of short-lived iodine derived from post irradiation measurements of xenon

Fuel performance is demonstrated by two dedicated measurement technologies:

- Continuous in-reactor on-line measurements of short-lived noble (rare) fission gases and relating these to a computed birth-rate resulting in “R/B” values;
- Post irradiation measurements of longer-lived solid fission product deposited in graphite cups and capsule walls.

Neither of these technologies cover iodine isotopes, because during irradiation iodine remains within the capsule and the iodine isotopes are too short-lived for post-test measurements.

There is, however, a practical possibility for experimental iodine release determination that had been applied already in ORR fuel irradiations tests in the years 1964 to 1969 and in Oak Ridge HFIR tests in the 70s [15]: when fission gas measurements are continued after irradiation cycle reactor shutdown, Xe-135 and Xe-135m releases of Xe-135 and Xe-135m can still be measured while the krypton releases stop immediately. Now these two xenon isotopes decay – because in secular equilibrium with the precursor – with the I-135 half-life (Fig. 5). And this works similarly for I-133.
Figure 5. Post-cycle measurements of Xe-135m (blue) and Xe-135 (yellow) demonstrating I-135 half-life. Results from HFR-Petten measurements after cycle #12 of the thirteen HFR-PM irradiation cycles.

The procedure is as follows: the iodine precursor of xenon is emitted from fissile sources and, as a condensable, deposits on the interior surfaces of the capsule. Upon decaying into xenon, this isobar enters the sweep gas instantaneously. This way, the amount of iodine deposited is measured by the residual xenon in the sweep gas with krypton dropping to zero at the end of the irradiation cycle and xenon isotopes Xe-133, Xe-135m, and Xe-135 decaying with the corresponding iodine half-life. Then the analysis of the xenon activities is used to derive a quantitative measure of I-133 and I-135 release rates at the end of the prior irradiation cycle.

4. Analysis of fission gas and iodine release from HTR-PM fuel irradiation testing

The HFR-PM irradiation test [10] was conducted with five HTR-PM spherical fuel elements in the High Flux Reactor in Petten. The irradiation of 13 cycles took place from August 2012 until December 2014. The rig consisted of one single capsule with five spheres, each surrounded by graphite cups (Fig. 6).
Figure 6. Arrangement of five spherical fuel elements of HTR-PM fuel production irradiated in the High Flux Reactor Petten.

The spheres were irradiated at a temperature of 1050°C up to 12% FIMA and accumulated a fast neutron fluence near $5\times10^{25}$ n/m$^2$ (E>$16$ MeV). All five spherical fuel elements had zero manufacturing defects and zero in-reactor failures as can be seen from the R/B measurements (see later Fig. 9).

Throughout, the HFR-PM irradiation experiment has been closely monitored with release rate measurements recorded every hour, also through the down-cycles between active irradiation cycles. In the absence of defective and failed particles, the release modeling has concentrated on fissile isotope breeding in the heavy metal contamination, even if this is only at the ppb level.

Starting with natural uranium, the fissile contribution is – due to build-up of Pu isotopes – linearly increasing from 3 to $5\times10^{-9}$ of the complete fissile content of the whole rig with 5 spheres (blue curve in Fig. 7).

Thorium contribution to fissile starts – of course – with zero, but is steeply rising to the $7\times10^{-9}$ fraction of total (red curve in Fig. 7). 1050°C predicted contamination based R/Bs for the Kr and Xe releases are shown in Fig. 8 as function of the isotope half-life computed with the 1978 Röllig model [4]. Notice different release mechanisms for krypton and xenon.
The applicable 1050°C numerical R/B-values are also listed in top row of Table 3. These have to be multiplied by the respective contamination levels, namely

- in the A3-matrix: 0.5 ppb $U_{\text{nat}}$ and 0.5 ppb Th, and
- in the graphite cups: 12 ppb $U_{\text{nat}}$ and 14 ppb Th,

to reproduce measured R/Bs. Figure 9 shows the superb agreement with the huge numbers of measurement data from the HFR Petten reactor station.

Two minor deviations are noticeable:

- Early R/B measurements are much smaller than predicted and we postulate that possibly the assumed A3-matrix pore structure has not yet fully opened during the early phases of the irradiation.
- Excess Kr-85m can be observed around day 190 and day 270: this was caused by a krypton contamination in the neon gas supply at the time as indicated in the Petten cycle reports.

In any case, reported HFR-PM R/Bs are unbelievably low at $5 \times 10^{-11}$ to $5 \times 10^{-9}$ (compared to $1 \times 10^{-7}$ to $1 \times 10^{-5}$ in the earlier diagrams): the sheer steady stable measurement technology at these extremely low levels is already admirable.

The dashed lines at the top of Fig. 9 show hypothetical R/B predictions to be expected from a single defect or failed particle in any of the five spherical fuel elements with 12,000 particles each. This comparison demonstrates that we are very far away indeed from any fuel failure.

The red-yellow circles at the bottom of Fig. 9 are the results of the derivation of I-135 R/B-values from the inter-cycle activity measurements of Xe 135 and Xe-135m with the error bars indicating the difference in derived values from between the two xenon isotopes. This demonstrates that xenon releases are a safe upper limit for iodine release predictions by a far distance. Interestingly, the I-135 R/B seems to remain constant throughout irradiation while all the other noble gas isotopes are increasing due to the increase of fissile content in the test, but we have no explanation for this so far.
Table 3. HFR-PM predicted R/B-values as computed by the product of the momentary fissile contribution from the existing heavy metal contamination with the temperature dependent contamination specific release rate. R/B-values from one defective or failed particle have a different temperature and half-life dependence.

| Isotope      | Xe-133 | Kr-85m | Kr-88 | Kr-87 | Xe-135 |
|--------------|--------|--------|-------|-------|--------|
| Irradiation  |        |        |       |       |        |
| time (efpd)  |        |        |       |       |        |
| 50           | 4.12×10^8 | 1.35×10^9 | 1.04×10^9 | 8.70×10^10 | 6.44×10^10 | 3.51×10^10 |
| 150          | 7.04×10^8 | 2.30×10^9 | 1.78×10^9 | 1.49×10^9  | 1.10×10^9  | 5.99×10^10 |
| 250          | 1.15×10^7 | 3.74×10^9 | 2.89×10^9 | 2.42×10^9  | 1.79×10^9  | 9.74×10^10 |
| 350          | 1.72×10^7 | 5.61×10^9 | 4.34×10^9 | 3.63×10^9  | 2.68×10^9  | 1.46×10^9  |
| R/B          | 6.34×10^6 | 1.21×10^6 | 9.59×10^7 | 6.47×10^7  | 4.45×10^7  |

Figure 9. Measured release rates of short-lived noble fission gases during the operation of the HFR-PM irradiation test in comparison to release predictions from 0.5 ppb U_{nat} and 0.5 ppb Th in the fuel element matrix and from 12 ppb U_{nat} and 14 ppb Th in the graphite cups surrounding the five spherical fuel elements. Also shown hypothetical release rates for the case of one (out of 60,000) defective or failed particle with a noticeable different half-life behavior. I-135 releases have been derived from post-cycle measurements of Xe-135 and Xe-135m.

5. Summary and conclusions
Modern HTR fuels are by factors ~1000 cleaner than those of the 1970-80 power stations, because manufacturing, quality control, test irradiation and post-irradiation technology have advanced massively during the last decades.

All the recent irradiations have shown spherical fuel elements with zero manufacturing defects and zero irradiation-induced particle failures featuring – at the same time – extremely low contamination levels, ppb rather than ppm.
Models and data exist for the comprehensive prediction of in-reactor release of short-lived fission gases also using the full set of irradiation data provided by the reactor station. There are two problems though:

(i) we need better precision $U_{nat}/\text{Th}$ trace contamination measurements in the graphitic components of the irradiation rig; and

(ii) we need the reactor station to predict the precise fissile contribution arising from the breeding of $U_{nat}/\text{Th}$. In both cases, we had to use circumstantial evidence to provide the necessary data.

A 1964 method has been used to derive iodine release rates from measured post-cycle xenon activities. The results show that our long-term conservative assumption - those noble gases provide an upper limit to iodine release - is fully correct. Furthermore, the newly determined iodine releases are actually factoring 2 to 5 lower than the corresponding measured xenon releases.

Future work should include independent measurements of $U_{nat}/\text{Th}$ contamination levels in spherical fuel elements and in the graphite cups that are used in irradiation experiments. In addition, isotope analyses of the uranium and thorium obtained in the burn-leach tests of spherical fuel elements with TRISO particles are needed. While, wide-ranging studies are performed and reported in the preparation of irradiation experiments, full scale post-test analyses that account for measured monitors and measured burnups are missing and should be more forcefully requested in future tests. This must also include post-test depletion code computations including the breeding of U-233 from the thorium contamination in matrix material and in graphite cups. Studies so far have neglected the Röllig proposed modeling of Knudsen and molecular diffusion in the pores of the A3. This model aspect has the potential to predict lower gas release rates at the higher pressures of operational HTRs, but is negligible in the evaluation of irradiation tests.

In conclusion, release rate measurements from fuel elements in MTR tests and in operating HTRs are the important indicator both for initial fuel quality and for performance during operation.

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