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Design of an On-Line, Multi-Spectrometer Fission Product Monitoring System (FPMS) to Support Advanced Gas Reactor (AGR) Fuel Testing and Qualification in the Advanced Test Reactor

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Abstract-- The US Department of Energy (DOE) is embarking on a series of tests of TRistructural ISOtropic (TRISO) coated-particle reactor fuel for the Advanced Gas Reactor (AGR). As one part of this fuel development program, a series of eight (8) fuel irradiation tests are planned for the Idaho National Laboratory's (INL's) Advanced Test Reactor (ATR). The first test in this series (AGR-1) will incorporate six separate capsules irradiated simultaneously, each containing about 51,000 TRISO-coated fuel particles supported in a graphite matrix and continuously swept with inert gas during irradiation. The effluent gas from each of the six capsules must be independently monitored in near real time and the activity of various fission gas nuclides determined and reported.

A set of seven heavily-shielded, high-purity germanium (HPGe) gamma-ray spectrometers and sodium iodide [NaI(Tl)] scintillation detector-based total radiation detectors have been designed and are being configured and tested for use during the AGR-1 experiment. The AGR-1 test specification requires that the fission product measurement system (FPMS) have sufficient sensitivity to detect the failure of a single coated fuel particle and sufficient range to allow it to "count" multiple (up to 250) successive particle failures. This paper describes the design and expected performance of the AGR-1 FPMS.

I. INTRODUCTION

The US Department of Energy (DOE) is embarking on a series of tests of coated-particle reactor fuel for the Advanced Gas Reactor (AGR) [1]. The AGR is a near-term deployment option (possibly deployable in the next 10 years) for the Very High-Temperature Reactor (VHTR) system. The VHTR appears to be the nearest term option for cogeneration of electricity and hydrogen. The primary distinguishing features

of VHTRs are the use of helium coolant, a low-power-density ceramic core capable of withstanding very high temperatures, and coated-particle fuel. As one part of this fuel development program a series of eight fuel irradiation tests are planned for the Idaho National Laboratory's (INL's) Advanced Test Reactor (ATR). One important measure of the fuel performance in these irradiation experiments is quantification of the fission gas release over the nominal 2-year duration of each irradiation.

Each of the planned fuel test experiments will incorporate a multi-capsule fuel test train inserted into an irradiation position in the ATR. The effluent gas from each of the six capsules must be independently monitored in near real time and the activity of various fission gas nuclides determined and reported. To meet this important test objective (and provide one spare), a set of 7 heavily-shielded high-purity germanium (HPGe) gamma-ray spectrometers and sodium iodide [NaI(Tl)] scintillation detector-based total radiation detectors have been designed and are being configured and tested for use during the AGR-1 experiment. This collection of radiation measurement systems constitutes the AGR-1 Fission Product Monitoring System (FPMS).

The AGR-1 test specification [2] requires that the AGR-1 FPMS have sufficient sensitivity to detect the failure of a single coated fuel particle and sufficient range to allow it to "count" multiple (up to 250) successive particle failures. (It should be noted that less than 10 actual failures per capsule are expected.) This paper describes the design and expected performance of the AGR-1 FPMS.

II. THE AGR-1 EXPERIMENT

The first test in the series of eight scheduled AGR fuel irradiation experiments is the AGR-1 test. The AGR-1 test is presently scheduled to begin irradiation in September 2006. The AGR-1 "test train" (the in-reactor portion of the experiment) incorporates 6 individual test capsules. Each

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capsule contains 12 fuel “compacts” arranged in three stacks of four compacts per stack supported in a graphite matrix. Each compact contains about 4300 TRISO-coated fuel particles, thus each capsule will contain about 51,000 fuel particles. Each individual capsule is continuously swept with an inert sweep gas during irradiation. The FPMS continuously monitors the sweep gas effluent from each capsule to provide fuel integrity data.

The test design, irradiation schedule, and the irradiation position in the ATR have been carefully chosen to mimic as closely as possible the fuel temperature, neutronics, and expected fuel burn up of the AGR environment. To reach the specified minimum fuel compact-averaged burn up of >14% FIMA (fissions per initial metal atom) will require a test duration of about two years.

One of the critical parameters to be controlled in these experiments is the fuel temperature. Temperature control is effected by varying the composition, and thus the thermal properties of the sweep gas. The gases that can be used to provide the needed thermal and reactivity control include He, Ne and ^3He . The sweep gas system also carries any released fission gases to the FPMS for measurement.

Figure 1 presents a simplified diagram of the AGR-1 gas flow and fission product monitoring system. The sweep

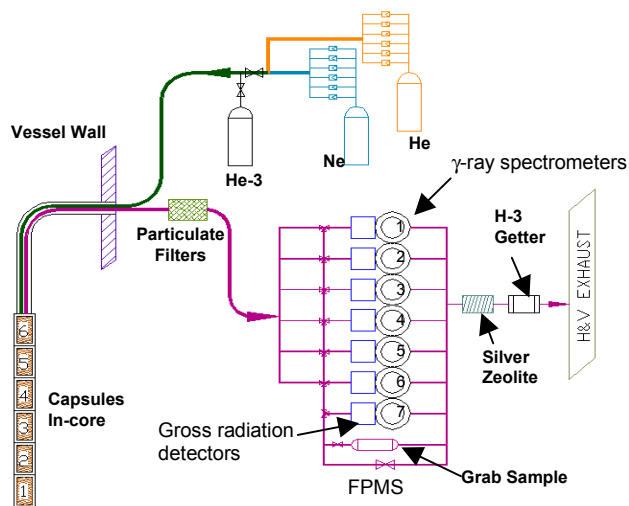


Fig. 1. A simplified diagram of the AGR-1 sweep gas flow and fission product monitoring system.

gas from each individual fuel capsule flows to a gross radiation detection system and gamma-ray spectrometer dedicated to monitoring that capsule’s effluent. To provide redundancy in the case of a detector failure, the effluent from any individual capsule or any selected combination of capsule effluents can be routed to a seventh spectrometer and gross activity monitor, or can be sampled at the grab sample station. Fission gas transport times from capsule to spectrometer are expected to be on the order of 100 to 300 seconds.

The FPMS is required to monitor the fuel performance by sensing and quantifying the increased fission gas activity in the

sweep gas following a particle failure (meaning the rupture of a particle’s TRISO coating). This activity increase must be sensed over a continuous fission gas release from any “tramp” uranium contamination of the fuel particles or compacts, and from a small number of initially defective fuel particles.

III. THE FISSION PRODUCT MONITORING SYSTEM

A. Sample routing and FPMS installation

The AGR-1 FPMS incorporates seven heavily-shielded gamma-ray spectrometers and gross gamma-ray radiation monitors – one for each of the individual capsule effluent lines and a redundant seventh unit. The gamma-ray spectrometer detectors are closed-end coaxial hyperpure germanium (HPGe) detectors with a nominal relative efficiency of 10%. The gross gamma-ray radiation monitor detectors are Ø25 mm X 25 mm NaI(Tl) detectors.

Each capsule effluent line (there are six) exits the reactor vessel through an experiment flange, routes the gas through a particulate filter (primarily for protection against irradiated particles downstream), and then enters into a shielded “cubicle” in the ATR basement separated from the spectrometer-containing cubicle by a thick concrete shielding wall. Each sample line then penetrates the wall, passes through the associated gross activity monitor, and then into the HPGe spectrometer shield where it flows through a 50 cm³ sample chamber enclosed in a beaker located at the center bottom of each shield. The sample chamber is viewed by the HPGe detector from below through a collimator.

Each HPGe detector is installed in a shield providing a nominal 10 cm of copper-lined lead surrounding the detector and sample. The shields were procured commercially. This shielding arrangement is similar to that used successfully for a multidetector system presently installed and operating in the ATR for monitoring a different experiment. Figure 2 presents a cutaway diagram of the detector, shield, and sample arrangement.

The gross activity monitors incorporate Ø25 mm X 25 mm NaI(Tl) detectors with integral photomultipliers, voltage dividers, and preamplifiers. Each NaI(Tl) detector is installed in a specially fabricated shield that protects against ambient background radiation and ensures a view of a 25-mm long segment of the sample line. The gross activity monitor shield assembly attaches to the spectrometer shield between the shield and the cubicle wall.

B. Instrumentation and control

Spectrometer and gross activity monitor electronics are in equipment racks located just outside the spectrometer cubicle door. The FPMS control computer is also located in these racks.

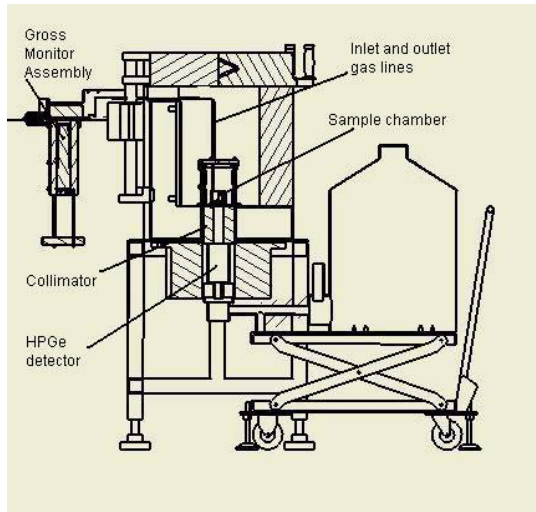


Fig. 2 Cutaway diagram of an AGR-1 spectrometer detector and shield

The spectrometer electronics consist of a main shaping amplifier and an analog-to-digital converter (ADC) feeding the histogramming memory of a Canberra Acquisition Interface Module (AIM). Each AIM provides 8192 channels of MCA memory for two ADCs. Each ADC has been modified to include an interface that, with a stable dual amplitude pulser installed on each detector, implements the technique of pulse injection with subsequent removal [3], a technology that we have found to be indispensable in unattended, on-line monitoring systems. The AIMs communicate with the control computer via Ethernet.

The NaI(Tl) gross detector electronics include a shaping amplifier providing input to the multichannel scaler (MCS) input of a Canberra Multiport II. Each Multiport II provides 4 MCS inputs. The Multiport IIs communicate with the control computer via a universal serial bus (USB).

The control computer is high-end personal computer with dual 3.2 GHz 64 bit (XENON) processors running the Windows XP operating system. The control computer communicates with the laboratory's intranet through one Ethernet interface and with the FPMS electronics through a separate Ethernet interface and the computer's USB interface. Figure 3 illustrates this configuration.

Acquisition control and data archival and analysis software have been developed to automate the acquisition and analysis tasks. The control software is designed to operate without continual operator intervention. The communications between the control program and the Canberra acquisition modules are carried out through a proprietary Canberra Virtual Data Manager (VDM). The VDM handles all of the low-level communications between the control program and the MCA hardware. The control program communicates with the VDM using the proprietary Canberra Genie 2000 Programming Library.

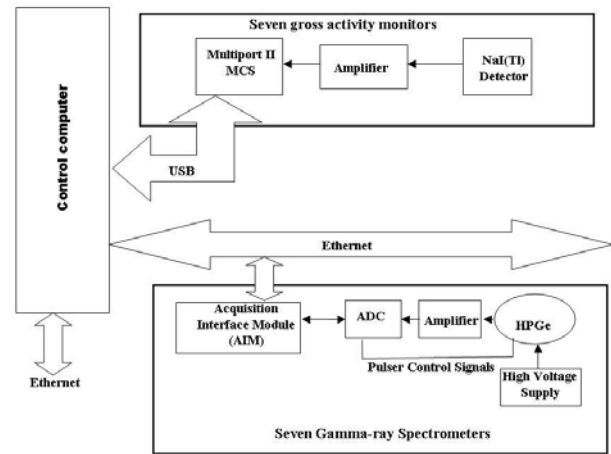


Fig. 3 Schematic depiction of the FPMS hardware. Only one set of gross monitor and spectrometer electronics are shown.

The main process thread of the software provides the control interface to the user for setting the sequencing and timing of the fission product measurements. Independent processing threads control each spectrometer and gross monitor.

Normal operation is a loop in which the acquisition runs for a preset time after which data is transmitted, stored, the acquisition restarted, and the acquired data analyzed to provide a "quick look" at sample conditions. The control interface also alerts the operator to any error conditions detected by the individual spectrometer processing threads. Acquired data stored on the control computer disk are routinely backed up to a remote disk in a different building via a connection to the laboratory network. Operators may interface to the control computer via the Remote Desktop feature of Windows XP.

IV. EVALUATION OF EXPECTED SYSTEM PERFORMANCE

A. Source term estimates

The AGR-1 FPMS is required to have sufficient sensitivity and range to detect and quantify the fission gas release from each TRISO particle failure in each capsule up to and including the unlikely prospect of 250 possible failures. To evaluate the system performance relative to this specification requires an estimate of the fission gas release expected under various test conditions.

The expected fission product inventories in each test capsule were modeled in detail using a calculational approach, termed Monte Carlo With ORIGEN (MCWO), that couples the Monte Carlo-based MCNP-4C transport code with the build-up and depletion code ORIGEN2 [4]. The detailed calculation divided each of the six test capsules into multiple "nodes". A typical ATR operating power and power tilt were presumed, as was a typical ATR repetitive operating schedule of 40 continuous days at full power followed by a 7-day shutdown period. Calculations were continued to a total exposure of 760 effective full power days (EFPD) or a total test duration of 893 days (about 2.5 years). Because of certain test train design

features (incorporation of some neutron poisons in the graphite holders and a Hafnium absorber on the core side of each capsule) at any given time during the irradiation, the maximum and minimum fission rates (and thus the resulting fission product inventories) among the six vertically arrayed capsules and among the three fuel stacks within a capsule differed by less than 30% [5]. The calculated inventories for a node near the fission rate maximum were chosen for the release estimate calculations.

Fission product release data from a TRISO-coated fuel experiment run in the ATR in 1991 elucidate the fission gas release profile expected when an irradiated particle fails. The NPR-1A experiment was fueled with an early generation of TRISO-coated particles and was run at conditions beyond the fuel performance limits; consequently, a number of particles failed during this experiment [6]. Figure 4 presents the response noted by the gross gamma-ray monitor when the first particle failed in the NPR-1A experiment.

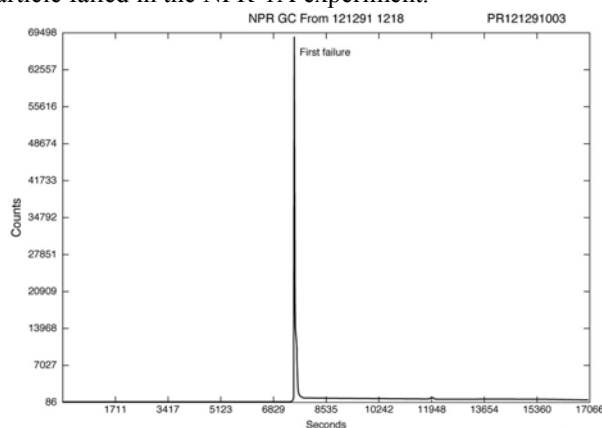


Fig. 4 Response of the gross gamma-ray monitor to the first TRISO-coated fuel particle failure during the NPR-1A experiment.

Note in Fig. 4 that there is a low, but non-zero counting rate prior to the particle failure (due to a small fraction of initial defected particles and/or “tramp” uranium contamination of the particles or compacts), the counting rate spikes rapidly at particle failure when the stored particle inventory is released, and then stabilizes in a few minutes to a steady-state rate slightly higher than that before failure. The rate after failure is elevated because of releases from the continued irradiation of the newly defected particle.

We have estimated capsule fission gas releases for a number of expected conditions during the AGR-1 experiment using a combination of a transient release model and a steady-state release model. Both models used release-to-birth ratios computed from a Booth diffusion model based on semi-empirical diffusion coefficients recommended for modern TRISO fuel particles [7]. The transient release model releases a computed fraction of the particle fission gas inventory into the flowing gas stream for a release interval of one minute. The released inventory fraction for each fission gas nuclide is determined from diffusion calculations for that nuclide. The steady state release prior to the first particle failure is that

computed for release from an amount of “tramp” uranium contamination and initially-failed particles at the allowable maximum of the fuel performance specifications. As each particle fails, its steady state release is added to the tramp uranium and prior particle releases to estimate a new total release value. Details of these models are provided in a published report [8].

B. The Expected Gross Gamma-Ray Detector Response

The response of the gross gamma-ray detector is expected to reflect the total fission gas concentration in the viewed section of the line. For irradiation periods early in the test (4 EFPD), at the mid point of the experiment (244 EFPD), and near the end of the irradiation (760 EFPD), the fission gas nuclide releases computed (as above) before, during, and after a given particle failure were corrected for dilution by the inert gas flow and for decay during transport to the detector station (nominally 2.5 minutes), and the volume viewed by the detector. These activities were then summed to estimate the activity profile viewed by the gross detector.

The gross detector response to the first particle failure, using inventory values at 244 EFPD, is presented Figure 5. Note the similarity to the NPR-1A particle failure response (Fig. 4).

In a similar manner, the expected response to the failure of a TRISO particle after 250 prior particle failures might have occurred has been computed and is presented in Figure 6. The model predicts that the 251st particle failure should be detectable by the gross detector system, even above the elevated background from the prior failures

C. The Expected Gamma-ray Spectrometer Response

The expected performance of the AGR-1 gamma-ray spectrometers was evaluated using the spectral simulation program *SYNTH* [9] and the modeled steady-state release concentrations. The modeled releases were corrected for dilution by the inert gas flow and for decay during transport to the spectrometer location. The spectrometer views a 50-cm³ volume of this flowing gas mixture. Fission gas first daughter decay progeny were also modeled as present in an amount computed from the parent concentration. The detector parameters and the modeled geometry were adjusted to provide a photopeak efficiency response equal to that measured for this spectrometer configuration. The spectral counting time was set to 10 hours – a value less than half the allowable 24 hour counting time. The system gain was set to store an energy range of 0 to 3 MeV in the modeled 8192 channels. The background prior to the first particle failure was the steady-state release from initial fuel defects at the maximum allowed by the fuel specification.

Spectra were synthesized at exposure times of 4, 284, and 760 EFPD, and in all cases the activity increase due to a particle failure was easily detectable. When these spectra were analyzed, the net single particle release activities of most fission gas nuclides were quantified with precisions (expressed as one relative standard deviation) of less than $\pm 2\%$. When spectrometry systems are operating near their detection limit

relative standard deviations of >40% are common.

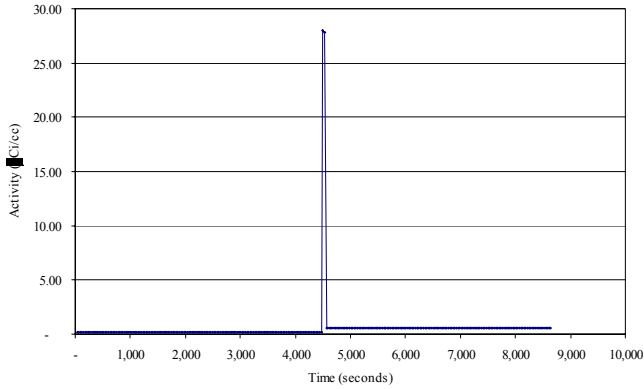


Fig. 5 The modeled total fission gas activity profile at the gross detector for the failure of the first TRISO particle at about 244EFPD. Note the similarity between the modeled results and the NPR-1A measurement presented in Fig 4

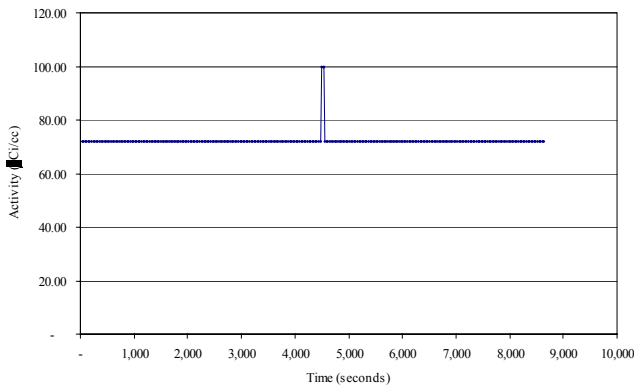


Fig. 6 Modeled fission gas activity profile at the gross detector for the failure of the 251st particle failure.

The modeled initial condition (background) spectra at 4, 284, and 760 EFPD were analyzed to determine the fission gas activities that could be expected to be detected with high (95%) confidence. The approach was to compute (following the formulation of Currie [10]) the net photopeak area increase detectable with nominally 95% confidence above the expected background (from initial defects) from the synthesized 10-hour duration spectra and their analyses. These peak areas, when converted to activities define the “system sensitivity parameters” (SSPs) presented in Table 1. Others might term these SSP values “Minimum Detectable Activities” (MDAs), but since they are not the minimum activities detectable under these conditions, but rather the activities detectable with high (95%) confidence, we avoid that designation. Table 1 also compares the SSPs (in microCuries) with the expected releases from a particle failure. Note that the fission gas releases expected from a single particle failure are several times the level that should be reliably detected.

TABLE I
EXPECTED SYSTEM SENSITIVITY PARAMETERS (SSPs) EXPRESSED AS THE ACTIVITY (μCi) DETECTABLE WITH A CONFIDENCE OF ABOUT 95% COMPARED WITH THE EXPECTED RELEASE FROM THE FIRST PARTICLE FAILURE.

Isotope	Primary Gamma-ray (keV)	SSP (μCi)	Single Particle Release (μCi)	Ratio Single Particle Release/SSP
Kr-85m	151.2	0.0017	0.033	19
Kr-87	402.6	0.033	0.12	3.5
Kr-88	2392.1	0.009	0.11	12
Kr-89	586	0.026	0.57	22
Xe-135m	526.6	0.003	1.3	507
Xe-137	455.5	0.016	2.7	177
Xe-138	258.4	0.012	0.48	39
Xe-139	218.6	0.011	1.2	110

V. SUMMARY

A fission product monitoring system (FPMS) consisting of seven NaI(Tl)-based gross gamma-ray monitors and seven HPGe-detector-based spectrometers has been designed to monitor the effluent from AGR-1 capsules irradiated in the ATR for fission gas content. Modeling results indicate that the FPMS should be capable of detecting each individual TRISO particle failure and counting up to 250 successive failures.

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