Assessing a Comprehensive Material Control and Accounting Approach for Safeguarding Pebble-fueled Reactors

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Abstract

Pebble-fueled reactors (PFRs) are a category of nuclear reactors that will require accurate nuclear material accounting and control (NMAC) measures to implement effective international safeguards. Current concepts for NMAC measures place heavy emphasis on measuring burnup of individual fuel pebbles to help discern pebble characteristics such as fissile material quantities. Well-characterized data from a burnup measurement system (BUMS) could help correlate to masses of fissile material via burnup analysis codes but this correlation may not be precise due to pebbles experiencing a wide range of neutron fluxes and irradiation times while traversing the reactor core via different paths. This variability in pebble trajectory and duration could lead to two pebbles exhibiting similar fuel burnup profiles yet having different fissile material content within.

The work presented herein focuses on the justification and development of a complementary nuclear material control (NMC) approach that could function in concert with a burnup measurement system (BUMS) to effectively implement a combined NMAC approach for the eventual application of international safeguards. This study first analyzed variations in fuel pebble burnup profiles for two types of PFR designs to better understand the variability in fuel transmutation which is based on pebble trajectories through the reactor core. Second, this study is evaluating and experimentally verifying the applicability of candidate NMC technologies as a complementary measure to pebble burnup measurements. The NMC approach that is discussed herein accounts for the limitations of individual pebble identification and considers pebbles to be classified by types and accounted for in such a manner using extrinsic and/or intrinsic features to each pebble. An NMAC system is then discussed for eventual incorporation in future safeguarded PFRs but further work is needed for full private industry implementation.

Introduction

A comprehensive nuclear material accounting and control (NMAC) approach should be investigated and considered for future pebble-fueled reactors (PFRs). In conjunction with the on-going research in nuclear material accounting (NMA) technologies, the effort described herein focuses on a development and assessment of nuclear material control (NMC) techniques. The techniques pursued are to categorize fuel pebbles based on initial uranium enrichments, time of introduction in the reactor core, and other specifications using extrinsic and intrinsic features of the pebble. This paper describes the results of this work thus far.

Pebble-fueled Reactor (PFR) Designs

Pebble bed reactors (PBRs), referred to as PFRs in this study, belong to the class of Very High-Temperature Reactors (VHTR) which use Tri-structural Isotropic (TRISO) particle fuel embedded in a graphite pebble (as shown in Figure 1). [1] A PFR uses several hundred-thousand fuel pebbles depending on the rated power of a particular reactor design. These pebbles cycle through a reactor core to generate the required heat for electricity production.

Despite different operational fluids (like helium), the overall structure of PFRs is similar: pebbles continually circulate through the reactor vessel where the nuclear fission heat produced in the pebble is removed to operate the energy conversion element of the nuclear reactor facility. The pebbles are extracted individually, assessed if they contain enough fissile material to be re-inserted into the reactor vessel, or, if not, are discharged from the system. Designers are assessing the ability of fuel burnup measurements to dictate how many passes a pebble can flow through a reactor core before the amount of fissile material content depletes below utility.

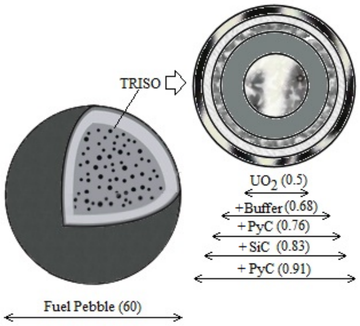


Figure 1. Schematic of a fuel pebble embedded with TRISO microspheres (dimensions in parenthesis are in mm)

Figure 2 is a schematic of the operation of a PFR. As shown in figure 2, pebbles are introduced into the reactor vessel on the top and irradiated during normal operations. Over time, pebbles traverse downward via gravity and are extracted into streams of individual pebbles through a reducer or singulizer. Each pebble migrates through a separator, which aims to extract partial pebbles into a damaged pebble storage bin for eventual removal from the fuel handling system. Whole, intact pebbles enter a buffer area where they are held until most short-lived radioactive fission products decay enough (ranging between 10 to 100 hours) where a burnup measurement system (BUMS) acquires a gamma radiation spectroscopic measurement of specific nuclides. The BUMS provides the operator insight into determining whether that particular pebble can be returned to the reactor vessel for continued fission reactions or if the pebble should be discharged from the reactor system for eventual transfer into spent fuel storage.

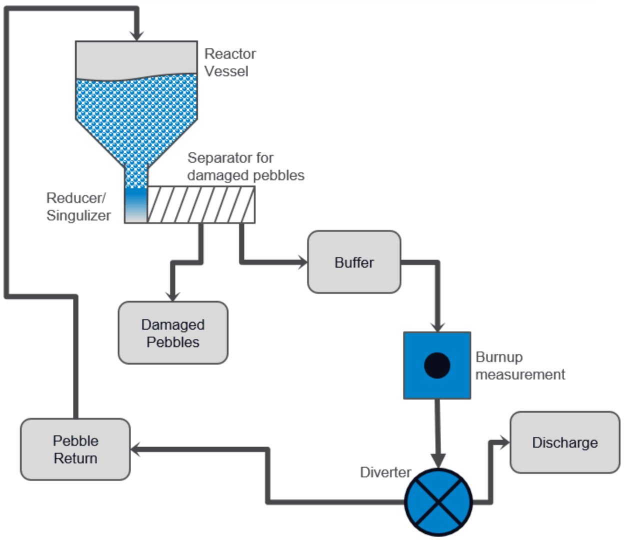


Figure 2. Schematic of PFR

Each fuel pebble contains between 1 to 10g of low enriched uranium (LEU, i.e., less than 20 wt.% ^{235}U enrichment), depending on the PFR design. In designs discussed in Kovacic et al., fresh fuel pebbles with a total mass of 200g (consisting of uranium and graphite) contain between 7 and 9g of LEU (under 1g of ^{235}U) if it is enriched to 9.6% ^{235}U. [2] After 80-90 gigawatt-day per metric ton of uranium (GWD/MTU), that fuel pebble would contain less than 0.12g of plutonium and less than 8.2g of uranium enriched to 3.8% ^{235}U. With these quantities per pebble, an International Atomic Energy Agency (IAEA)-defined significant quantity of uranium in fresh fuel pebbles would be 87,000 pebbles or in 240,000 irradiated pebbles and, for a significant quantity of plutonium, would amount to 67,000 irradiated fuel pebbles. Assuming on the order of magnitude of 400,000 pebbles circulating through a PFR, utilizing the plutonium-based number, this implies a benefit of defining 6-7 types of pebble batches in order to track a smaller number of pebbles during operation.

A key consideration for a practical NMAC approach would be to evaluate an accountancy approach by item (per pebble) or by bulk (containers of pebbles) but, ultimately, combine both into some amalgamation of item and bulk accounting. This hybridized approach is the foundation for the research work described in this paper.

Nuclear Material Accounting

Applying safeguards monitoring measures to PFRs is a challenge. Previous experience and current studies have not produced an effective and efficient safeguards approach. Some designers have confidence in the ability to account for fissile content in pebbles by correlating certain measured nuclides' gamma radiation photo-peak ratios to estimated burnup profiles. [3] Correlating burnup profiles for estimating a pebble's fissile content could assist in meeting an eventual customer's international safeguards requirements. However, this requires the knowledge of initial uranium enrichment, fabricated configuration, structural integrity, time of irradiation, and reactor neutron flux to which each pebble is irradiated. Understanding that pebbles are not all identical nor irradiated identically, a comprehensive method should exist that combines the proposed BUMS with a manner in which pebbles can be uniquely identified. When combined with burnup measurements, individually identified pebbles could provide highly useful data such as time of irradiation or initial enrichment which could offer a more accurate calculation of fissile content. Though ideal, the concept of individually identifying each pebble within a PFR is a monumental challenge due to the number of pebbles residing within a reactor system and because of the hostile environment within which the pebbles occupy: high temperatures, a dynamic system with constantly moving parts, hundreds of thousands of other pebbles abrasing against one another, a molten salt cooling medium in some designs, and a high radiation area. Some designs still consider uniquely identifying individual pebbles, which is very difficult. As favorable as it would be to have the ability for eventual customers of these reactor types able to account each unique fuel item, the reality of the situation is more difficult.

Therefore, this paper focuses on the consideration of a compromise between a material accounting method relying on incomplete data (i.e., only burnup measurements) currently being pursued in the industry and a robust material control method (i.e., item accounting individual pebbles in a PFR system) which is infeasible. Herein, the study delved into the justification of why burnup measurements alone are not sufficient and continues to propose another approach for an effective safeguards implementation strategy – one that accepts the limits of item accountancy and instead, aims for batch accountancy by identifying useful characteristics of each pebble in an attempt to complement the existing burnup measurement system.

Burnup Measurement Simulations

Development of fuel pebble and burnup simulations were performed using MCNP6.2 and its embedded CINDER90 isotope generation and depletion module. Simulations were carried out for variations of the HTR-PM (high-temperature reactor-pebble bed modular-Chinese design) with varying ^{235}U enrichments of 8.9 wt.%, 13.35 wt.%, and 17.8 wt.% to respective fuel burnups of 84.19, 89.15, and 89.15 GWD/MTU. [4] [5] The simulations were required to gather the gamma radiation source term in pebbles of varying ^{235}U enrichments needed for creating gamma-ray spectra for varying ^{235}U enrichment and fuel burnups. Another pebble model was also created based on a U.S. designed PB-FHR (Fluoride cooled high-temperature pebble bed reactor) for three ^{235}U enrichments of 8.9 wt.%, 17.8 wt.%, and 19.9 wt.% and the same burnups as above. The mass of uranium in the HTR-PM pebble is 7 g, whereas the PB-FHR consists of 1.39 g of uranium. Due to the latter's lesser quantity of fissile material, some deviation in burnup simulations occurred. Both pebbles were modeled in MCNP in the body centered cubic (BCC) configuration with reflective boundary condition. Differences in power (0.0012 MWth and 0.0005 MWth, respectively) and neutron energy spectra were necessary due to the uranium enrichment differences as well as the amount of moderating material (i.e., graphite) present (Figure 3).

The source term for the respective fuel burnups were used to produce the respective lists of gamma radiation source strengths with their corresponding gamma radiation branching ratio inputs to an MCNP model of an HPGe detector. This effort produced the gamma spectra for various burnups from various ^{235}U enrichments. Figure 4 shows how different enrichment and burnup levels can exhibit similar gamma spectra and conveys the difficulty of discerning pebble characteristics from a gamma spectroscopy.

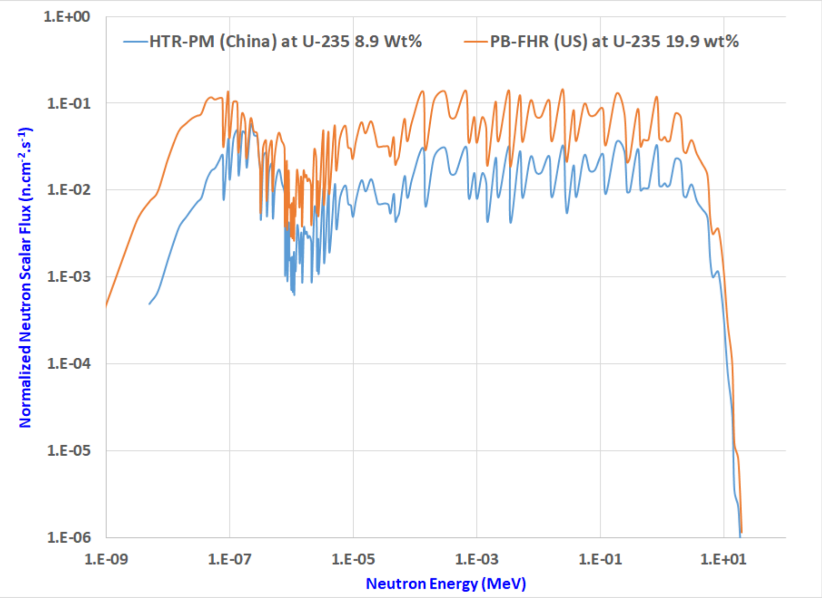


Figure 3. Neutron spectra comparison of HTR-PM and PB-FHR fuel pebbles.

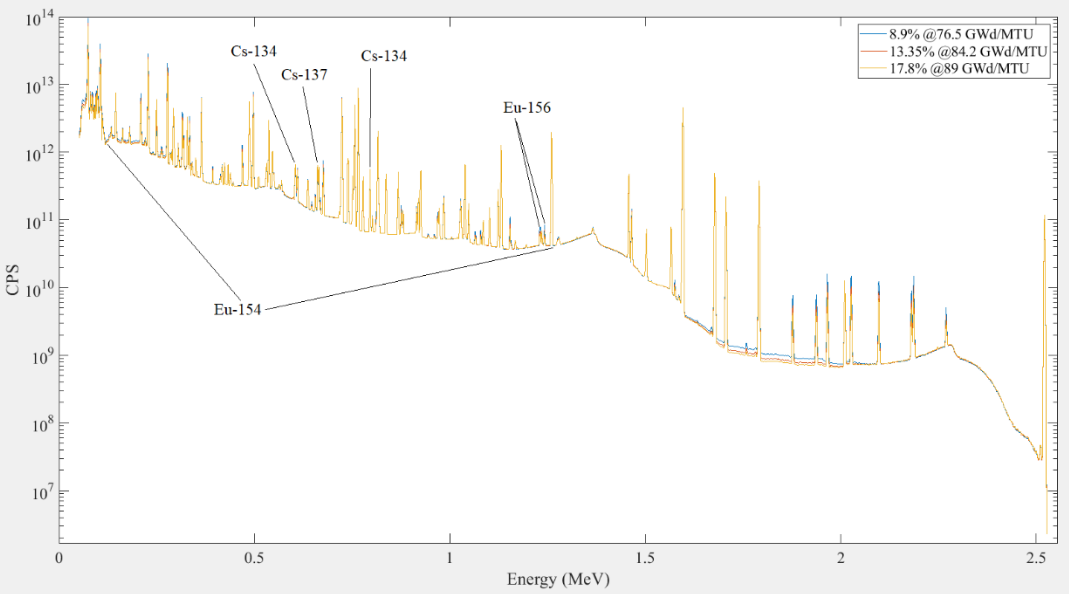


Figure 4. Gamma radiation spectra of HTR-PM pebbles at different fuel burnups and ^{235}U enrichments showing the challenge to differentiate ^{235}U enrichment using radioisotope signatures.

Relying on gamma radiation signals from key fission products (^{134}Cs, ^{137}Cs, ^{154}Eu, and ^{156}Eu), measurable activities were simulated with corresponding burnup measurements to yield Figure 5 (specifically for ^{134}Cs for both pebble designs). With the ability of measuring activity of a spent fuel pebble, it would be challenging to determine initial enrichment or burnup of that pebble.

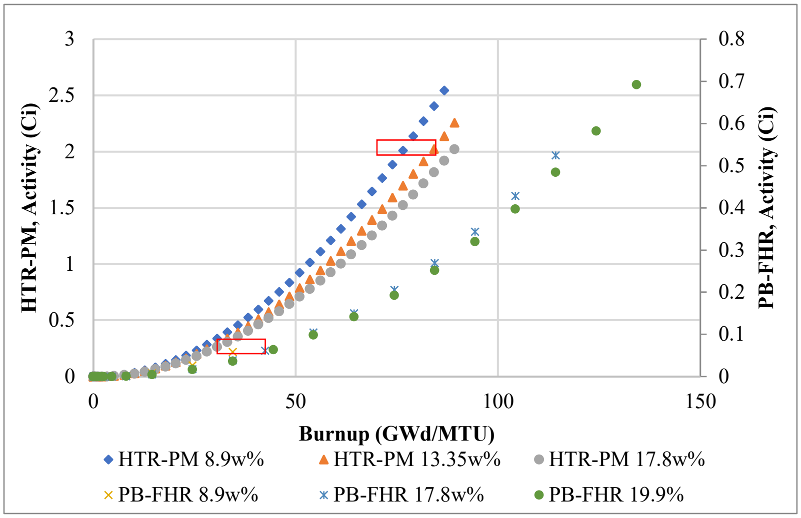


Figure 5. HTR-PM and PB-FHR radioactivities of ^{134}Cs.

Examination of photo-peaks, such as those for ^{134}Cs, cannot be used for differentiating between uranium enrichments because the photo-peak intensities are not unique. The same photo-peak intensity could be obtained in the gamma radiation spectra for various combinations of initial uranium enrichment and fuel burnup. In addition, a photo-peak other than the conventional such as a high-energy gamma radiation photo-peak of ^{156}Eu was also examined, which also proved to be not useful for differentiating between pebbles of different uranium enrichments and fuel burnups. This study also showed that the use of isotopic ratios, such as ^{134}Cs/^{137}Cs, ^{154}Eu/^{137}Cs for fuel burnup determination is challenging for pebbles irradiated with different initial uranium enrichments (for the same reason stated in the case of using individual gamma radiation photo-peaks of ^{134}Cs, ^{137}Cs, ^{154}Eu, and ^{156}Eu). Overall, measuring radioactivities (or radioactivity ratios) is simply insufficient for informing an operator of a pebble's burnup – other information such as pebble type through NMC techniques would be useful.

Nuclear Material Control

Item accounting of reactor fuel elements is part of implementing safeguards measures in a nuclear reactor facility. However, in PFR designs, hundreds of thousands of pebbles traverse the reactor core continuously and thus, reliable item accounting of individual fuel elements is not possible. Without unique identifiers for each fuel pebble, monitoring individual pebbles in a PFR is highly unfeasible. Burnup measurements for fuel monitoring cannot accurately be correlated to fissile material content in fuel pebbles without more detailed information of individual pebbles.

Developing an accounting system for PFRs requires an approach that falls between item and bulk accounting – a system devised to allow for important characteristics to be known of each pebble so that when combined with results from a BUMS, the operator and inspectorate can have a better understanding of the fissile material content within. Unique characteristics that would be useful include time of irradiation which can be assumed for an averaged neutron flux (this depends on the path within the core a pebble traverses) with a given date of insertion into the reactor vessel, types of pebbles (neutron absorbing, moderating, and/or fuel pebbles), and the initial uranium enrichment of said pebble. The latter characteristic is important as reactor designers are increasingly contemplating a range of uranium enrichments in their fuel. Though pebble type can be easily determined by gamma spectroscopy— non-fuel pebbles will have a starkly different gamma signature than fuel pebbles— the date of pebble insertion to better estimate duration of irradiation will be a challenge to determine without specifically marking pebbles in some way.

An initial concept for an effective material control approach consisted of incorporating identifiable features into the outer pebble layer of graphite. As shown in Figure 1, pebble designs commonly consist of an outer layer of graphite ranging between 3-5 mm in thickness. Preliminary work was completed in evaluating the ability to distinguish varying averaged spatial distributions of impregnated inert microspheres within the outer graphite matrix with ultrasound imaging. [6] Microsphere materials and sizes were chosen for their high thermal conductivity and insignificant neutron absorption, and it was projected that they would only have a minimal impact (less than 1%) on the PFR neutronics and operation. [7] The imaged spatial density of the microspheres was considered as a way to define pebble types – i.e., fuel pebbles with a particular ^{235}U enrichment. For example, if a reactor design relied on two initial uranium enrichments of fuel, the developed imaging system should be able to different two spatial densities of microspheres.

Despite the assurance of neutronically inert materials for the microspheres, designers lacked interest in accommodating their fuel fabrication processes. This lack of designer's interest and a stated interest to use imaging techniques for identifying voids in the graphite matrix for operational and safety needs, led the research to consider using already-incorporated engineered surface markings and voids within the outer graphite layer. [8] This concept was studied in the German AVR design to help identify kinds of fuel pebbles optically using engineered ridges along and near the equator of each pebble (as shown in Figure 7). [9] Scanning for inhomogeneities like this would serve a dual purpose: enhancing the ability of identifying (and thereby monitoring pebbles for international safeguards needs) and being able to detect sublevel imperfections or other dislocations that could inform operators of the structural integrity of said pebbles.

Figure 6. Surrogate fuel pebble with embedded microspheres

Three imaging systems were evaluated and experimentally validated for identifying these features: optical imaging via ultrasound technology which requires submerging a pebble in a liquid medium to ensure a strong coupling between the scanning medium and the pebble surface, thermal imaging intended to utilize the internal heat signature of an individual pebble, and electromagnetic imaging using eddy current sensors to evaluate surface and subsurface inhomogeneities of the graphite matrix. Ultrasound imaging was evaluated with the original concept of embedded microspheres. Having water or other transparent liquid serving as the necessary coupling medium proved unfeasible and was therefore, abandoned during the validation process. Investigating the feasibilities of both thermal and eddy current imaging continued.

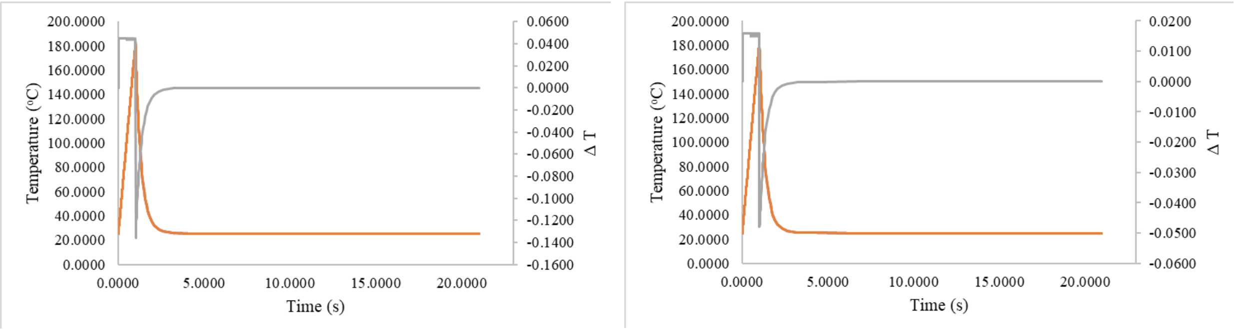
Figure 7. Ridges along equators of a fuel pebble

Thermal Imaging

Infra-red thermography (IRT) was investigated to image substructures in graphite pebbles. Just as the microspheres, the concept was to use the distribution of these substructures to uniquely identify individual pebbles (akin to the AVR equatorial grooves exhibited in Figure 7 above). Furthermore, there is promise for IRT to help PFR operators monitor structural integrity of pebbles due to their exposure to high burnup, elevated temperatures, and otherwise hostile environments.

The basic principles of IRT in this application can be explained by considering a graphite pebble which has a set of randomly distributed substructures. The substructures in this case can be subsurface voids or embedded particles, such as zirconia microspheres. In an actual IRT experimental setting, the pebbles were shined with a low power laser for a very short period (~0.1-1 seconds). The heat generated on the surface then diffused into the pebble. However, the presence of substructure interferes with the heat flow, leading to a temperature contrast on the surface. This temperature contrast was a function of the size, shape, location, and properties of the substructure. Therefore, the temperature contrast provided input about the substructure, and for a randomly sized and placed substructure features in each pebble, the temperature contrast can be used to uniquely identify pebbles.

The feasibility of IRT in detecting substructures by using finite element analysis via the commercial software ABAQUS was preliminarily evaluated. The sample studied was the coating of a pebble, made of graphite, with a thickness of 4 mm. The modeled substructure was a spherical, 1-mm in diameter, void placed at a distance of 0.1 and 0.5mm from the surface of the structure where heat was to be applied. The surface was radiated with a uniform heat flux (to emulate heat generation within a fuel pebble) and the temperature on the surface was obtained by solving the continuum heat transfer equations in 3D.



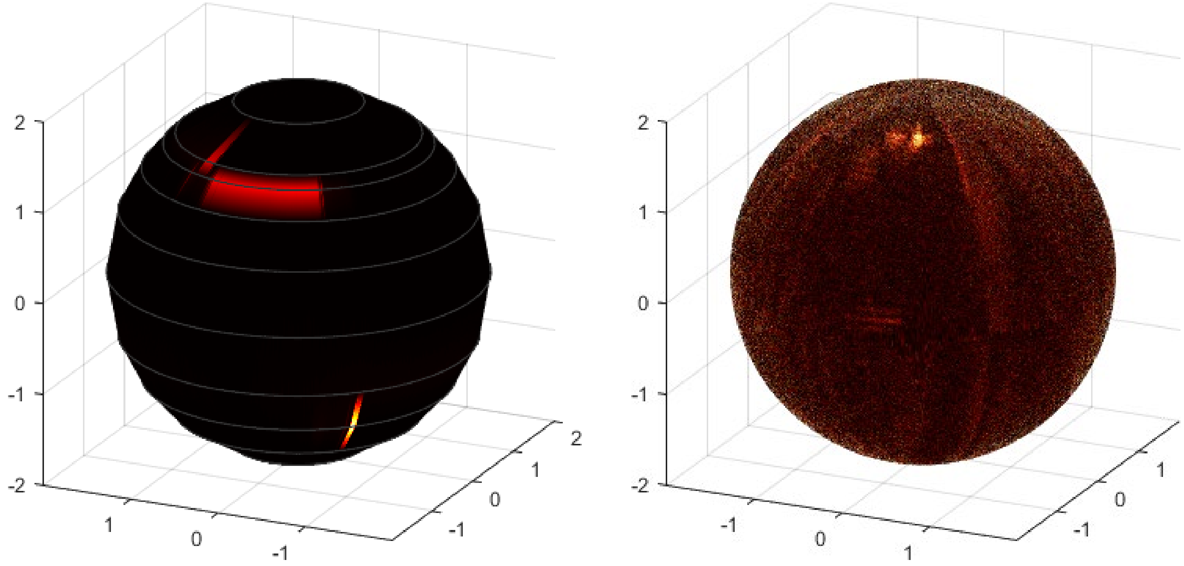
0.1 mm from surface (bottom) 0.5 mm from surface (bottom)

Figure 8. Temperature (orange) vs. time graphs vs. Δ T (grey) and structure visualization

An initial temperature of both samples was established at 25ºC (initial condition), and the surface closest to the void underwent a heat flux of $100000\frac{mW}{mm^2K}$ for 1 second (based on carbon to simulate a graphite matrix). The sample was then cooled down to 25ºC in ~ 20 seconds. All other surfaces of the block were subjected to adiabatic boundary conditions. The surface temperatures at two points of each sample were selected: a point on the surface right above the void and a point far away from it. The assumption is that the temperature of these two surface points would be the most and the least affected, respectively, by the presence of such a void. Hence, a measure of the contrast will be the difference between the temperature of these two points (ΔT). The highest temperature contrast conveyed below was achieved with the void was 0.1 mm from the surface: 0.14°C.

Electromagnetic Imaging

Electromagnetic, i.e., eddy current (EC), imaging is a mature non-destructive examination (NDE) technology that is fast and economical and well-studied. [10] [11] The successful imaging of EC scanning relies on a medium's conductivity. Physical interruptions, such as defects or dislocations in the material, cause changes in the received signal, which can be interpreted by the operator as inhomogeneities to be identified and recorded. Penetration with EC imaging is best with a highly conductive material such as copper; however, graphite, as the external layer of the fuel pebbles of interest having limited conductivity, requires a higher frequency EC coil. Initial hand-made scans with two different types of EC coils (one pencil, single point probe and one flexible tape, multi-point probe) were conducted by Argonne National Laboratory's Sensors, Instrumentation, and NDE (SINDE) group and dislocations were identified that could be used for identifying inhomogeneities that can be further assessed as extrinsic or intrinsic features. Figure 9 displays scan results with a pencil probe and the flex probe projected onto a spherical visualization.



(a) (b)

Figure 9. (a) pencil probe pebble projection and (b) flex tape probe pebble projection

NMAC for Safeguarding PFRs

The ability to categorize and identify pebble types in PFRs eases the burden of monitoring and tracking pebbles throughout the duration of the reactor's life. It also adds a strong material control mechanism that works in concert with a BUMS for fuel handing and a burnup analysis code (i.e., a material accountability-measure vital for applied safeguards of the reactor) for material quantification. If this system can be incorporated into the overall pebble extraction system, the potential reduction in ex-core time for pebbles is significant. Furthermore, the discussed technologies for effective NMC have varying levels of detecting dislocations beyond engineered features – some are able to detect subsurface dislocations that could arise from the fuel fabrication process or from irradiation. These additional performance features are of interest to reactor designers who have stated concerns on the structural integrity of pebble fuels. Regardless of the specific design, PFRs rely on pebbles passing through the reactor core multiple times – the exact number, though estimated, will only be known after operational experience is gained. The NMC technologies discussed herein could benefit future operators in being able to monitor pebble health and can lead to their inclusion in eventual final deployed designs. Implementing NMC techniques for international safeguards purposes can benefit from designers' interest of monitoring pebble integrity. It is important to convey this joint benefit which meets two disparate needs for operating such a reactor design.

Recalling Figure 2, an NMC technique applied at the buffer stage could help identify useful characteristics such as the pebble's initial uranium enrichment and/or when the pebble was first introduced into the system quickly. When combined with the eventual burnup measurement taken at the next step of the system, a more accurate quantity of fissile mass can be estimated for each pebble (and, by extension, eventually the entire reactor system). For example, if a gamma spectrum is acquired and used for quantifying fissile content in each pebble (e.g., for Pebble #1 through Pebble #100) within the system displayed in Figure 2, there is not a way to monitor if Pebble #37 (as an example) is counted more than once. Therefore, some form of pebble tracking is necessary to ensure reduced quantification errors.

Conclusions

Figures 4 and 5 convey the difficulty in using burnup measurements for unique identification because gamma spectra per pebble can be indistinguishable between some pebbles. Without an ability to identify and monitor each individual pebble, one cannot reliably account for all fissile content for safeguards requirements. Hence, the overall system would benefit from the discretization of the numerous pebbles into a more manageable classification system where groups or types of pebbles can be identified to make material accountancy more achievable and precise. Just as the implementation of effective and efficient international safeguards measures relies on defining compartmentalized material balance areas for more accurate reporting, quantifying fissile content in batches of pebble types would assist an operator be more precise in their material reporting for meeting their safeguards requirements which, in turn, could also benefit their operational efficiency by shortening the amount of time a pebble resides outside the reactor core. IRT and EC imaging technologies assessed as part of this work help show the potential for these to serve as an effective pebble monitoring technique for material control measures as well as for defect monitoring for operators. With more study, a potential dual benefit application for these imaging techniques can help justify implementation in commercial scale PFR facilities in the future.

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