# An INDEPTH Analysis of Spent Pebbles

Ben Impson, Zeyun Wu, Braden Goddard

*Department of Mechanical and Nuclear Engineering, Virginia Commonwealth University,*
*401 West Main Street, Richmond, Virginia, 23284*
impsonba@vcu.edu, zwu@vcu.edu, bgoddard@vcu.edu

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## INTRODUCTION

Pebble Bed Reactors (PBRs) are a family of advanced high temperature reactors including gas and molten salt models, both being considered generation IV reactors. Both of these versions of PBRs are currently in development under the United States Department of Energy's missions, as well as the gas version being constructed overseas in China [1][2]. The pebbles these reactors are named for are spheres of graphite, with thousands of fuel particles embedded inside them. The pebbles then "flow" through the reactor core, in some cases being removed at the bottom of the reactor during operation and others at the end of a cycle. When first developed, these fuel particles were Bistructural-isotopic (BISO) fuel particles, with a fuel kernel surrounded by two layers of pyrolytic carbon. More modern pebbles use Tristructural-isotopic (TRISO) particles, with three protective layers around each fuel kernel, including two pyrocarbon layers and a layer of silicon carbide [3]. Both structures serve as improved cladding and fission product containment. Fig. 1 shows a picture of a cut TRISO particle with its different layers visually identifiable. TRISO fuel can survive upwards of 1800 °C, in comparison to 374 °C for pressurized water reactor Zircalloy cladding [4]. Generally, PBRs, such as the Xe-100, operate using High-assay Low-enriched Uranium (HALEU), which is enriched to between 5% and 20% $U^{235}$ weight percent. The exact enrichment of the fuel depends on factors such as if the reactor fuel is at steady state burnup or if this is the first fuel loading of the reactor.

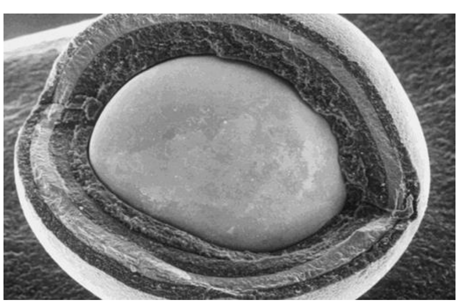

Fig 1. Cut TRISO Particle showing the different protective cladding layers [5].

In contrast to the benefits of PBRs, these reactors have a potentially significant unsolved problem, the variable isotopic composition of used pebbles. Due to the flow of the pebble through the core, the burnup each pebble experiences will vary depending on the path and speed it takes though the core. Unlike stationary fuel reactor core designs, where flux and burnup can be predicted with moderate accuracy [6][7], the burnup in a PBR will depend on the pebble and its constantly changing position. The primary method of determining the composition of used pebbles is destructive assay, in which the pebbles and constituent TRISO particles are disassembled and analyzed. This requires the destruction of a percentage of used pebbles, which is not ideal given their higher enrichments and the release of fission products that would be better contained by the pebbles.

Analysis of the radiation spectra from used pebbles would provide an alternative, nondestructive method of isotopic determination. By using a High Purity Germanium (HPGe) detector, shown in Fig. 2, the contents of each pebble could be estimated. If the burnup is determined to be low enough, the pebble could be sent through the reactor again, otherwise it would be disposed of. However, this does not provide a clear picture of the reactor's behavior or an indication of what isotopes the pebble produced or what the pebble experienced in the core.

Fig 2. Examples of different types of HPGe Detectors [8].

The Inverse Depletion Theory (INDEPTH) code is built using ORIGEN in SCALE [9]. It takes nuclide inventories, such as those generated from an MCNP6.2 depletion simulation [10] and will estimate key reactor parameters, such as initial enrichment, cooling time, burnup, and reactor type. Using a weighted Sum of Squared Errors to optimize, it runs ORIGEN through iterations of reactor parameter guesses until it finds a match to the nuclide inventories. Given isotope data, this can provide an insight into the experience of each individual pebble.

## INDEPTH ANALYSIS PROCEDURE AND RESULTS

### Generating Spent Pebble Isotopies

Using CINDER in MCNP6.2, burnup calculation can be performed to generate isotope data of spent pebbles. A single pebble in a cube of helium with reflective boundaries is used for this burnup evaluation, as seen in Fig. 3. The power level is scaled to a single pebble, based on the Xe-100 reactor model [4]. The reflective boundary causes the pebble to experience a harder neutron spectrum than is expected of pebbles in a reactor, but modeling a full reactor is computationally prohibitive at this stage in this project. The TRISO particles are placed in a lattice, with the positions randomized using URAN, a stochastic randomization method in MCNP6.2. The initial enrichment is 15.5%. The pebbles are burned for 1304 days and then allowed to cool for 30 days. The output isotope data can then be used in INDEPTH to reverse engineer the reactor conditions and compare that output against the known MCNP6.2 inputs. These simulations produce isotope inventories for every time step, however, only the final step is used with INDEPTH. A pebble in an active reactor would not be measurable while moving through the core, only upon removal.

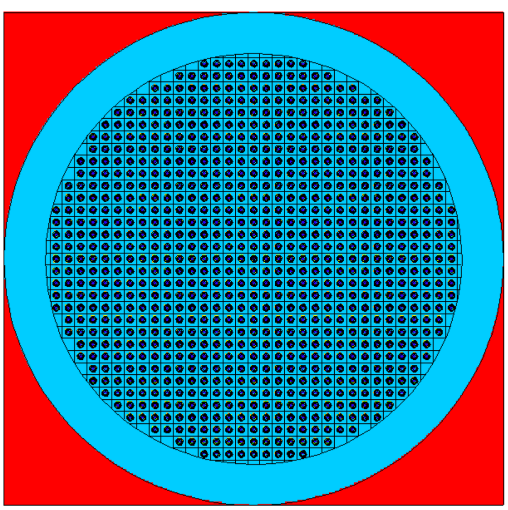

Fig 3. MCNP6.2 cross-sectional view of the pebble model. Red is helium, blue is graphite, and the small spheres are TRISO particles. The URAN randomization cannot be shown with this cross-sectional viewing software.

The most important isotopes for this analysis are gamma emitting fission products and transuranics (TRUs). Alpha and beta radiation are unlikely to be detectable outside the pebble. Curium isotopes such as $^{243}$Cm and $^{245}$Cm are useful for this, as are several americium isotopes such as $^{243}$Am and $^{244}$Am. Fission products, such as $^{137}$Cs will also be important for measuring burnup. While most examples of INDEPTH use more comprehensive lists of isotopes [9], usually including uranium and plutonium contents, that data will not be available for an individual pebble without destructive assay. By limiting the isotopes to those detectable by a HPGe detector, the effectiveness of INDEPTH in determining the reactor conditions can be emulated.

### Using Existing Pebble Bed Reactors

INDEPTH/SCALE have an existing library of reactors consisting of current and past power and research reactors. Included in the library are several high temperature gas reactors (HTGR), including the AVR, although these were operated at far lower enrichments than new PBR designs. This prevents these existing models from accurately representing the newer reactors. INDEPTH cannot converge on a solution using these reactors. In addition, SCALE PBR models that can be added to INDEPTH, such as the public model developed by D. Hartanto and colleagues [11], is of a full reactor core and thus is not suitable to compare against the nuclide composition created from the single pebble MCNP6.2 simulations.

### Creating PBR Models

The libraries INDEPTH uses can be generated by running a depletion calculation using a SCALE model. Using a single pebble SCALE model developed by Jonathan Wing [12], several libraries were generated at enrichments from 1% to 19.99%. The geometry of this SCALE model can be seen in Fig. 4.

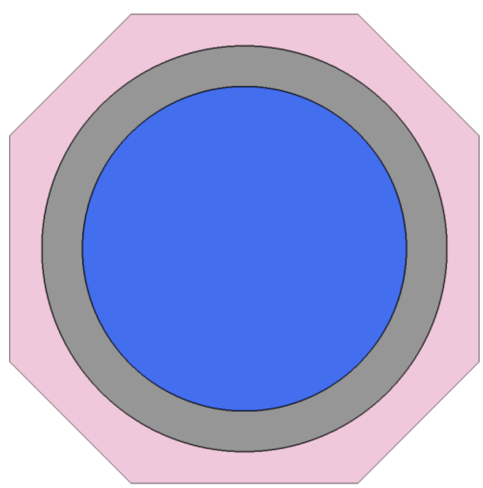

Fig 4. SCALE cross-sectional view of the pebble model. Pink is the helium, gray is the graphite, and blue is the homogeneous fuel and graphite region in the center of the pebble [12].

**Using Uranium, Plutonium, and TRUs**

While uranium and plutonium are unlikely to be used for burnup evaluation for an operating reactor, they can be used to benchmark the accuracy of an MCNP6.2 model for burnup analysis and reconstruction. Uranium, plutonium, and TRU inventories (TRUs) were tracked in this simulation. By using full nuclide inventories, INDEPTH will usually converge faster than smaller nuclide inventories. However, the issues with the neutron spectrum in the original MCNP6.2 model prevent INDEPTH from perfectly recreating the initial conditions, leaning towards shorter irradiation times, shorter cooling times, and higher enrichments. The results of all nuclides considered in this work are summarized in Table I.

TABLE I. Entire Nuclide Inventory

| Combination of Isotopes | Irradiation Time (days) | Cooling Time (years) | Enrichment (wt. %) |
|---|---|---|---|
| U, Pu, and TRUs | 1112.88 | 0.0274 | 16.871 |

Not all actinides produce meaningfully detectable radiation. Of particular interest are curium and americium. With only curium isotopes as input data, INDEPTH returns noticeably longer irradiation times and higher enrichments, with similar cooling times when compared with using americium and curium data. While unlikely to be a burnup determining isotope, including neptunium in the sample also influences the results. The important points of comparison can be found in Table II.

TABLE II. INDEPTH results using TRUs.

| Combination of Isotopes | Irradiation Time (days) | Cooling Time (years) | Enrichment (wt. %) |
|---|---|---|---|
| Cm | 1118.62 | 1.0000 | 15.500 |
| Cm, Am | 835.78 | 0.9670 | 10.506 |
| Cm, Am, Np | 1341.40 | 0.0274 | 13.974 |

Each combination orbits around the original conditions of 1304 days of irradiation, 0.082 years of cooling, and an initial enrichment of 15.5%, although none perfectly recreates them. Of the three, the combination of curium, americium, and neptunium results in the most accurate reconstruction, but still has a noticeably lower enrichment and shorter cooling time.

## FUTURE WORK

While initial results are promising, several paths need to be evaluated to improve the performance of this INDEPTH model. The models used to generate both the samples and the libraries used by INDEPTH do not perfectly reflect a fuel pebble in reality. Due to the reflective boundaries in the MCNP6.2 model, the neutron spectrum is harder than would be in a full reactor. A full reactor model would produce more accurate nuclide inventories compared to the individual pebble models. In addition, the SCALE model used to compensate the lack of a full core MCNP6.2 model used homogeneous fuel, which has been shown to produce significantly different results than heterogeneous PBR fuel [13]. Work is ongoing to develop a full core heterogeneous model of the Xe-100 reactor using the Monte Carlo code OpenMC [14].

The nuclide inventory itself will also need to be revised for detectability of gamma and neutron emitting nuclides as well as optimized for the INDEPTH program. If pebbles are to be tested immediately after removal from the reactor, then short lived isotopes should be included in the nuclide inventory. With the current data, which only had TRU information, INDEPTH tends towards lower irradiation times and longer cooling times than the known conditions. Rather than only using TRUs, including fission products would likely also improve accuracy. These results using only TRUs show that with improvements to models and isotope inventories, INDEPTH and similar programs likely could be used to determine the burnup of advanced nuclear reactors. Simulations are in progress to generate fission product data for the MCNP6.2 simulations.

The difference in flux between reactor positions is also an important issue to consider in future work regarding pebble isotopics, however is less valuable when analyzing spent pebbles. The intermediate steps of the simulation will also need modification to accurately model the motion in the core. The flux will not be the same, meaning the isotopic production will also not be the same. By varying the flux or expanding the model to a full core model will provide more accurate data. This work is currently in progress [14].

## ACKNOWLEDGMENTS

The authors would like to thank Jonathan Wing for his help with the SCALE pebble model and the creation of the SCALE libraries, as well as Brandon Grogan for his help using those libraries with INDEPTH. This work is performed with the support of U.S. Department of Energy’s Nuclear Energy University Program (NEUP) with the Award No. DE-NE0009304.

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