

Nuclear Material Control and Accountancy Approach for Pebble Fueled Reactors using a Novel Pebble-Type Identification and Classification Technology

**Prepared for
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Nuclear Material Control and Accountancy Approach for Pebble Fueled Reactors using a Novel Pebble-Type Identification and Classification Technology

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1 Introduction

In FY21, Argonne National Laboratory (ANL) with researchers at Texas A&M University (TAMU) designed and engineered a prototypical device for accounting types of irradiated pebbles (for example with different ^{235}U enrichments or pure graphite) in Pebble-Fueled Reactors (PFRs). Through engagements with reactor designers, a need arose to assist in identifying and categorizing types of pebbles as a complementary nuclear material control method that would synergize with designers' use of fuel burnup measurements for material accountancy needs. As part of an overall nuclear material control approach, a concept of pebble batch accounting was investigated using extrinsic non-radiological features to identify intrinsic characteristics. This concept of batch accounting led to the ability of identifying types of pebbles based on characteristics such as initial ^{235}U enrichment of pebble batches or dates of introduction into the reactor core. Identification was achieved by embedding the 5-mm thick graphite periphery of pebbles with 2-mm diameter inert Yttria-Stabilized Zirconia (YSZ) microspheres to achieve an averaged volumetric density (i.e., common spacing between microspheres) unique to that type of pebble. With an ultrasound imaging system in proximity with each pebble, the YSZ microspheres in the pebble proved visible and their spacing became the unique feature upon which pebble type categorization could occur. At the culmination of FY21, ANL intended on delivering and installing the prototype at TAMU for initial testing but, due to the on-going pandemic, this was postponed until FY22.

2 Background

2.1 History of Pebble fueled reactors

The pebble-fueled HTGR (High-Temperature Gas-cooled Reactor) design was pioneered by the Federal Republic of Germany (FRG) in the 1960s. The Arbeitsgemeinschaft Versuchsreaktor (AVR) pebble-fueled HTGR operated from 1967 to 1988 in West Germany. The AVR was an experimental reactor, which was operated as a testing facility for pebble-fueled HTGRs. The 45 megawatts-thermal (MWt) AVR was able to demonstrate that a reactor fueled by small fuel pebbles, and cooled by gas, could be safely operated. Over its lifetime, the AVR was home to tests that primarily focused on the qualification of pebble fuel. Varying combinations of uranium and thorium and fuel sizes were tested under a wide range of operating conditions to determine optimum combinations for safety and economics.¹

Using experience gained with the operation of the AVR, the FRG designed and built the Thorium High Temperature Reactor (THTR) that served as the link between the experimental AVR facility and commercial scale facilities. Although the THTR only operated from 1985 to 1988, the over 16,000 hours of operational experience laid the foundation for the pebble-fueled HTGR designs that are being pursued today by the People's Republic of China (PRC) and the Republic of South Africa (RSA).^{2,3} The PRC began its pebble-fueled HTGR program in the 1992 with the approval to build a reactor at Tsinghua University's Institute of Nuclear Energy Technology site outside of Beijing. Completed in 2000, the HTR-10, a 10 MWt pebble-fueled HTGR, has been used by the

¹ E. ZIERMANN, "Review of 21 years of power operation at the AVR experimental nuclear power station in Jülich," *Nucl. Eng. Des.*, **121**, 135-142 (1990).

² R. BÄUMER, I. KALINOWSKI, E. RÖHLER, J. SCHÖNING, and W. WACHHOLZ, "Construction and operating experience with 300-MW THTR nuclear power plant," *Nucl. Eng. Des.*, **121**, 155-166 (1990).

³ D. SCHWARZ, R. BÄUMER, "THTR operating experience," *Nucl. Eng. Des.*, **109**, 199-205 (1988).

PRC as a research facility. Much like the AVR, the HTR-10 has come to be a testing ground for the PRC in HTGR technology including testing of pebble fuel and verification of inherent safety features associated with pebble-fueled HTGRs.⁴

In 1995, the RSA was investigating ways to increase electrical generating capacity in anticipation of increased demand. At the time, a coal-fueled power plant would have taken almost 10 years to construct and be located near coal fields in the central part of the country. Deemed not economically viable, the government was interested in pursuing a means of electricity generation that would require lower capital costs, have a construction time on the order of 18 to 24 months, and could be located in coastal regions or remote areas. Conducting a feasibility study on modular, high temperature, helium-cooled reactor design options led the RSA to pursue the pebble-fueled HTGR design.⁵ Building on the experience from the AVR and THTR, the RSA began to design a new facility based on proven technologies. Initially designed to generate 100 megawatts-electric (MWe), this design came to be known as the Pebble Bed Modular Reactor (PBMR). Since its initial consideration and despite its abandonment as a viable next-generation option for RSA, lessons learned from the PBMR concept and its material control and accounting research have been applied to numerous designs being currently designed and developed around the world – two of particular interest by private companies in the US: X-Energy Ltd. and Kairos Power.

2.2 Pebble Fueled Reactor designs

Pebble bed reactors, otherwise referred to as pebble fueled reactors (PFR), belong to the class of Very High-Temperature Reactors (VHTR) and use Tri-structural Isotropic (TRISO) particle fuels embedded in a sphere or a pebble.⁶ A PFR uses several hundred-thousands of fuel pebbles depending on the rated power of a particular reactor design. Each fuel pebble contains between 5 and 10g of Low Enriched Uranium (LEU), depending on the PFR design.^{7,8,9} In later designs, each pebble can have a total mass of 200g with the contained uranium enriched to 9.6% ²³⁵U. This amounts to between 7 and 9g of LEU (and under 1g of ²³⁵U) before irradiation. With this configuration, after irradiation (between 80 and 90 GWD/MT), fuel pebbles would contain less than 0.12g of plutonium and less than 8.2g of uranium enriched to 3.8% ²³⁵U.¹⁰

PFRs use collections of pebbles (commonly inside vessels – hence, the origin of pebble ‘beds’) where the collective heat generated from hundreds of thousands of pebbles is transferred to an

⁴ “Evaluation of high temperature gas cooled reactor performance: Benchmark analysis related to initial testing of the HTTR and HTR-10,” IAEA-TECDOC-1382, International Atomic Energy Agency (2003).

⁵ M. FOX and E. MULDER, “Pebble Bed Modular Reactor – South Africa. Design and Development of Gas Cooled Reactors with Closed Cycle Gas Turbines,” IAEA-TECDOC-899, International Atomic Energy Agency, 251-256 (1996).

⁶ USDOE and the GIF (2002). “A Technology Roadmap for Generation IV Nuclear Energy Systems,” U.S. DOE Nuclear Energy Research Advisory Committee and the Generation IV International Forum Report No.GIF-002-00, December.

⁷ Z. WU, D. LIN, D. ZHONG. “The design features of the HTR-10,” Nuclear Engineering and Design, 218, 25–32 (2002).

⁸ IAEA. “Evaluation of high temperature gas cooled reactor performance,” International Atomic Energy Agency Report No. IAEA-TECDOC-1694 (2013).

⁹ D.L. MOSES. “Nuclear safeguards considerations for pebble bed reactors (PBRs),” Nuclear Engineering Design, 251, 216-221, (2012).

¹⁰ Kovacic, D., Gibbs, P., Scott, L. *Model MC&A Plan for Pebble Bed Reactors*. ORNL Technical Report ORNL/SPR-2019/1329. March 2019.

operational fluid medium via an inert gas (for example, helium). Some current designs being developed in the U.S. include other media such as molten fluoride salt. Despite different operational fluids, the overall structure of PFRs are similar: pebbles continually circulate through the reactor vessel where heat is removed to operate the energy conversion element of the nuclear reactor facility. The pebbles are extracted individually and assessed whether they still contain enough ^{235}U fuel to be re-inserted into the reactor vessel or to be discharged signifying the end of their utility. The manner by which designers are evaluating the presence of ^{235}U is through a designed fuel Burnup Measurement System (BUMS) which is incorporated (refer to Fig. 1) into the process and helps the operators operate the reactor efficiently and effectively.

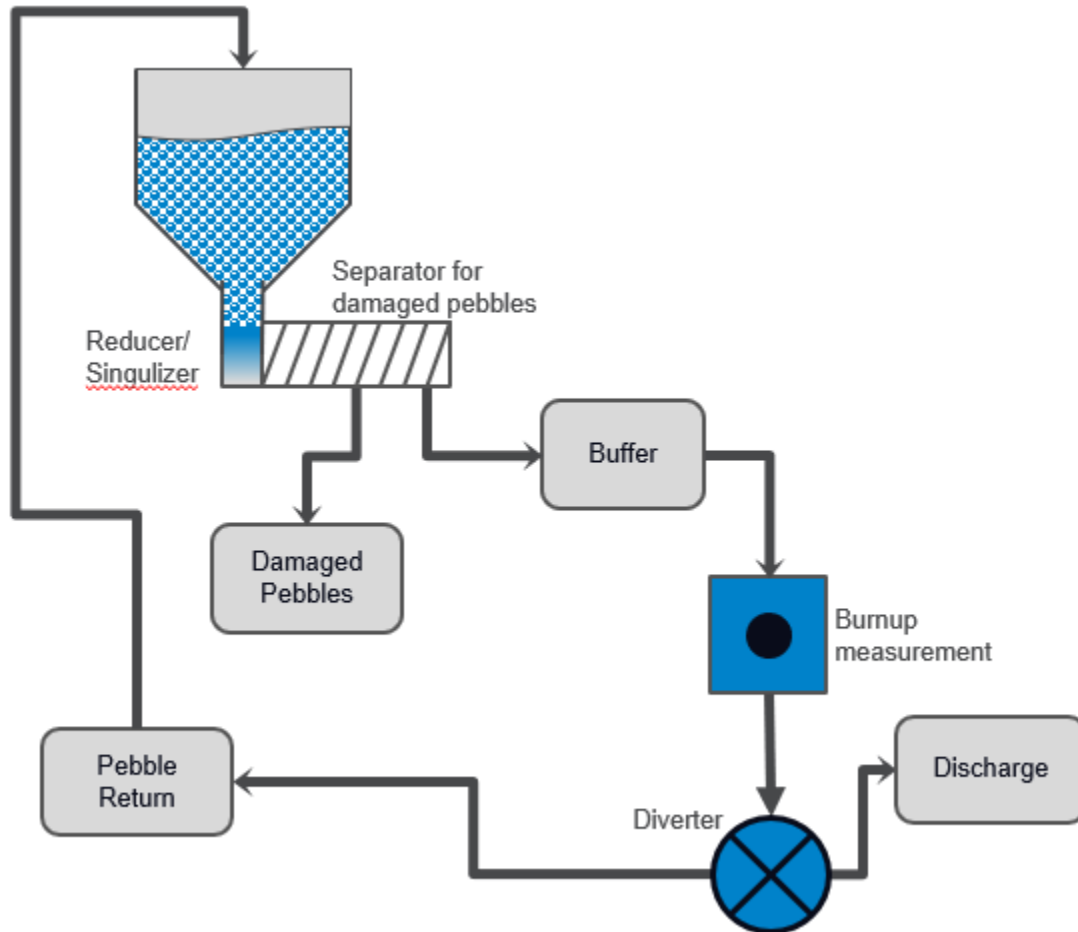


Figure 1. Schematic of a PFR

Figure 1 is a schematic of the operation of a PFR. As shown, pebbles are introduced into the reactor vessel 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 or otherwise, non-whole pebbles into a damaged bin for eventual discharge. Whole intact pebbles enter a buffer area where they are held until most short-lived radioactive fission products diminish enough (ranging between 10 to 100 hours)¹¹ in order for the BUMS to acquire an accurate gamma scan. The BUMS (a gamma

¹¹ Personal conversation with industry experts. April 2021.

spectroscopic analysis on the order of minutes) provides the operator insight into determining whether that particular pebble can be returned to the reactor vessel for continued heat generation or if the pebble should be discharged from the reactor system for eventual transfer into spent fuel storage.

Within each discrete fuel pebble, the total amount of fissile material is low: sub-gram quantities of ^{235}U and plutonium before and after irradiation per pebble. With these low quantities, Material Control and Accounting (MC&A) approaches as defined by appropriate regulatory authorities must reflect various characteristics of the pebble fuel (e.g., the fissile material content within each pebble, the portability of individual pebbles, the quantity of pebbles during transport and storage, the categorization of pebbles by type, etc.). A key consideration for a proper MC&A approach would be to define the accountancy approach by item (per pebble) or by bulk (containers of pebbles). There is a possibility that an effective ultimate pebble MC&A approach can be some amalgamation of both item and bulk accounting. This hybridized or batch approach is the foundation for the research work described in this report.

2.3 An MC&A Approach for PFRs

Proper MC&A of a PFR would naturally include operational features already essential in effective operation of such a facility. Defining item control areas (similar to what is currently done for light water reactor facilities) from the fresh fuel storage, reactor, and spent fuel storage areas continues to make sense. However, with pebble fuel and the ability to distinctly count each pebble allows for a strong material control approach to be used in the second item control area defined around the reactor itself. Knowing the initial amount prior to initial operations, the number of pebbles in the reactor vessel would define the *inventory*. Pebbles being fed into the vessel after operations have begun or re-inserted after BUMS would be counted individually as *additions*. Approaching discharge near the bottom of the vessel, the flow of pebbles is reduced and pebbles are singularized where they are evaluated for discharge as damaged or continue through to the BUMS. Technologically, each pebble passing through the singularizer is tallied as a *removal* for gross counting purposes within the vessel. Maintaining operational knowledge of the *active inventory* is conceptually simple ($\text{active inventory} = \text{inventory} + \text{additions} - \text{removals}$) yet potentially practically challenging during PFR operations.

In terms of material control, the total fissile material content can conceptually be maintained through this process. However, with the variation and diversity of pebble types (based on duration inside the reactor vessel, initial ^{235}U enrichment, subjected neutron flux based on the trajectory within vessel, etc.), material accounting measures that are planned as part of the process (BUMS based on radiation measurements using nondestructive assay techniques like gamma spectrum analysis) can be used to help supplement a comprehensive MC&A approach. Any effective approach should incorporate both elements.

The work conducted for this project consisted of designing, developing, and eventually incorporating a pebble accounting system based on engineered features added into the outer layer of individual pebbles. More specifically, this concept relies on the spatial distribution of embedded neutronically-inert microspheres (defined as YSZ previously in this report) into the exterior 5-mm thick graphite layer of each pebble. Through experimentation, it was settled the spatial density would serve as the unique identifier between types of pebbles instead of individual pebbles. The

YSZ microspheres would ideally be introduced into the fuel fabrication process which entails engaging with pebble fuel designers and fabricators (intended for FY22).

3 System Description

In most PFR designs, the TRISO fuel particles contained in the pebbles have the same ^{235}U enrichment. As seen in Figure 2 (adapted from IAEA-TECDOC-978), some recent pebble designs consist of a spherical graphite matrix with embedded fuel kernels of fissile uranium surrounded by a simple graphite shell with an exterior diameter of 60mm.¹²

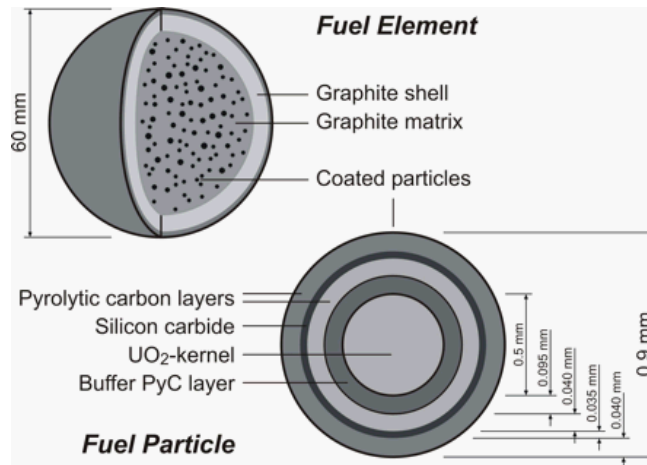


Figure 2. A sphere embedded with TRISO particle fuel with cross-section showing individual fuel particles inside.

Some PFR designs plan to use a mixture of one or two kinds of pebbles, each kind containing TRISO fuel particles with a different ^{235}U enrichment but with the same pebble dimensions. In addition to the fuel pebbles, most of the large PFR designs will use pure graphite pebbles, without uranium, for the purpose of providing neutron moderation. There would be neutron absorbing pebbles as well in some PFR designs. These different types of fuel pebbles continuously flow through the reactor and come out of the reactor vessel at its bottom. Depending on the refueling scheme envisaged for the particular PFR design, the pebbles coming out of the reactor will be re-inserted into the reactor vessel at its top. When the fuel pebbles have reached the desired burnup after multiple recirculation in the reactor core or found to be damaged, they need to be removed and stored. Potentially, other kinds of pebbles (moderator and neutron absorber pebbles) are reinserted into the reactor depending on the refueling scheme and based on the integrity of these pebbles.

The novel technique at the core of this project consists of impregnating YSZ microspheres of diameter 2 mm in the 5 mm graphite shell that forms the outer layer of the pebble fuel element (see Figure 2). The YSZ microspheres have high thermal conductivity and insignificant neutron absorption and hence will have only a minimal impact (less than 1%) on the PFR neutronics and operation. These YSZ microspheres were impregnated at different spatial densities (unique for

¹² IAEA-TECDOC-978 *Fuel Performance and Fission Product Behaviour in Gas Cooled Reactors.* (https://www-pub.iaea.org/MTCD/Publications/PDF/te_978_prn.pdf), 1997.

each pebble type), which will be utilized to identify and categorize pebbles using an Ultrasound Scanning (US) system. The YSZ impregnations were carried out on surrogate pebble materials due to the unavailability of the real pebbles to the investigators. Ultrasound scans and image analyses were initially conducted with flat surface surrogates made out of equivalent graphite material impregnated with YSZ microspheres. Subsequent studies were conducted with spherical surrogates impregnated with YSZ microspheres. A pebble sorting system was designed and assembled for operational testing (which was delayed due to the ongoing pandemic).

3.1 Pebble Preparation

Prior to engaging with private industry fuel fabricators, researchers at TAMU used the best-available open-source information to replicate the graphite shell material used on the periphery of pebble fuel elements. TAMU researchers then successfully embedded the YSZ microspheres into both planar and spherical samples of the replicated graphite shell material.

3.1.1 Planar Surface Preparation

Flat or planar surfaces were utilized first to simplify the learning process. A mold cavity was prepared on a steel sheet using high temperature rubber tape for refining the microsphere impregnation into the graphite shell material. Subsequently, Durez 34306 Phenolic resin, phenol combined with formaldehyde (PF), was poured on the mold cavity up to about 2 mm. Toray's Torayca® yarn, a high-performance carbon fiber made of Poly Acrylo Nitrile (PAN) was then used as a supporting mat for distributing the YSZ microspheres on it. Glue was sprayed on the carbon fiber mat before the microspheres were distributed.



Figure 3. Images of a flat-surface graphitic surrogate impregnated with 2 mm diameter YSZ microspheres with an interstitial spacing of 3 mm.

Figure 3 exhibits square and triangular lattice structure for the microspheres on a flat surface. For the uniform distribution of the YSZ microspheres on the carbon fiber mat, a 3D printed mesh of acrylonitrile butadiene styrene with openings of 2.2 mm, slightly more than the 2 mm-diameter YSZ microspheres, was used. After uniformly distributing the YSZ microspheres on the carbon fiber mat, one more 2 mm layer of PF resin was poured on top, and the thus prepared sample was covered with metal plates. The 3D mesh used for uniformly distributing the YSZ microspheres was removed before the second layer of PF resin was poured. The sample was then subjected to a

pressure of 0.8 MPa and a temperature of 150° C. A Differential Scanning Calorimetry (DSC) measurement revealed that the sample was completely cured at 150° C in 10 minutes. Using the aforementioned process, two kinds of flat-surface graphitic surrogates impregnated with 2mm YSZ microspheres were prepared with interstitial spacings of 3 mm and 6 mm between the microspheres.

3.1.2 *Spherical Surface Preparation*

A 2-mm PF resin layer was first applied to a steel sphere of diameter 50 mm and partially cured at 100° C for six minutes. After partial curing, the PF resin coated steel sphere was sprayed with glue and then the 2 mm YSZ microspheres were attached uniformly to it using tweezers. The final step was to add a second layer of PF resin coating on top and curing it at 150° C for eight minutes. Two such steel sphere samples were prepared, one with 15 and the other with 30 YSZ microspheres. Figure 4 shows the completed spherical graphitic surrogates of pebbles impregnated with YSZ microspheres.



Figure 4. Completed spherical graphitic surrogates of pebbles embedded with 15 YSZ microspheres (left) and 30 YSZ microspheres (right).

A modified second method of preparing the spherical graphitic surrogates of pebbles was also carried out. In this method a composite sphere sample was prepared with the PF resin. There were YSZ microspheres attached to these samples to serve as a control variable for this project. The spheres were drilled to have a threaded hole so that the sphere can be positioned (suspended) on the bottom half of the sphere mold. The sphere mold was coated with mold release spray carefully in the spherical surface of both mold components and then applied to the entire mold. The purpose of the release agent is for the PF resin not to get attached to the mold after curing, so that the ball with the cured resin can be released. The YSZ microspheres were attached to the ball as follows. The PF resin was heated to 60°C to decrease the viscosity temporarily. The steel sphere was coated in a thin layer of PF resin and was partially cured in the oven for one hour at 150°C. Then, ~ 20 YSZ microspheres were added on the surface of the partially cured steel sphere. The epoxy sphere with YSZ microspheres was secured on the screw of the bottom half of the sphere mold. The steel sphere with YSZ microspheres attached on it was placed inside the bottom half of the mold. The PF resin was injected into the sphere mold's bottom half by using a syringe. The two halves of the

sphere mold are then fastened together. PF resin was then injected through the top half of the sphere mold by using a syringe to coat the top of epoxy sphere with PF resin. The mold was then shaken to make sure the resin was able to coat all the epoxy resin sphere's surface. The sphere mold with epoxy sphere with PF resin was cured in the oven for one hour and 30 minutes (the oven was set to 150°C). The sphere mold then was taken out of the oven to cool. The cured sample was removed from the sphere mold. Figure 4 shows the spherical graphitic surrogate embedded with YSZ microspheres.

3.1.2.1 Refined Process for Spherical Surface Preparation

In order to fabricate pebbles with better smoothness using phenol formaldehyde (PF) resin coating, specialized aluminum molds were designed and machined. The mold houses and secures a base pebble (as the core of the pebble) at its geometrical center with the aid of a tether (screw at the bottom of the mold), allows for pouring the PF resin uniformly around the core and cure it in place. The setup was tested and the results point to considerably enhanced smoothness of the coating. The following materials were used to make samples with no YSZ microspheres: epoxy spheres, release film, release spray, sphere mold, phenol formaldehyde (PF) resin, syringe, aluminum foil, an oven, and hot plate. Two composite sphere samples were prepared with PF resin. There were no YSZ microspheres attached to these samples to serve as a control variable for this project. The spheres were drilled to have a threaded hole so that the sphere can be positioned on the bottom half of the sphere mold. The oven was set to 150°C. For the first epoxy sphere sample, the sphere mold was coated with release film carefully in the spherical surface of both mold components and then applied to the entire mold (Figure 5a). Then, the PF resin was poured on an aluminum foil tray and placed on hot plate and heated for three minutes at 60°C to decrease the viscosity. The epoxy sphere was secured on the screw of the bottom half of the sphere mold. The PF resin was injected into the sphere mold's bottom half by using a syringe (Figure 5b). The two halves of the sphere mold were then fastened together by using an Allen wrench on four screws on the top half of the mold (Figure 6a). PF resin was then injected through the top half of the sphere mold by using a syringe to coat the top of the epoxy sphere with PF resin (Figure 6b). The mold was then shaken to make sure the resin was able to coat all the epoxy resin sphere's surface. The sphere mold with epoxy sphere and PF resin were cured in the oven for one hour and 30 minutes. The sphere mold then was taken out of the oven to cool. The cured sample was removed from the sphere mold.

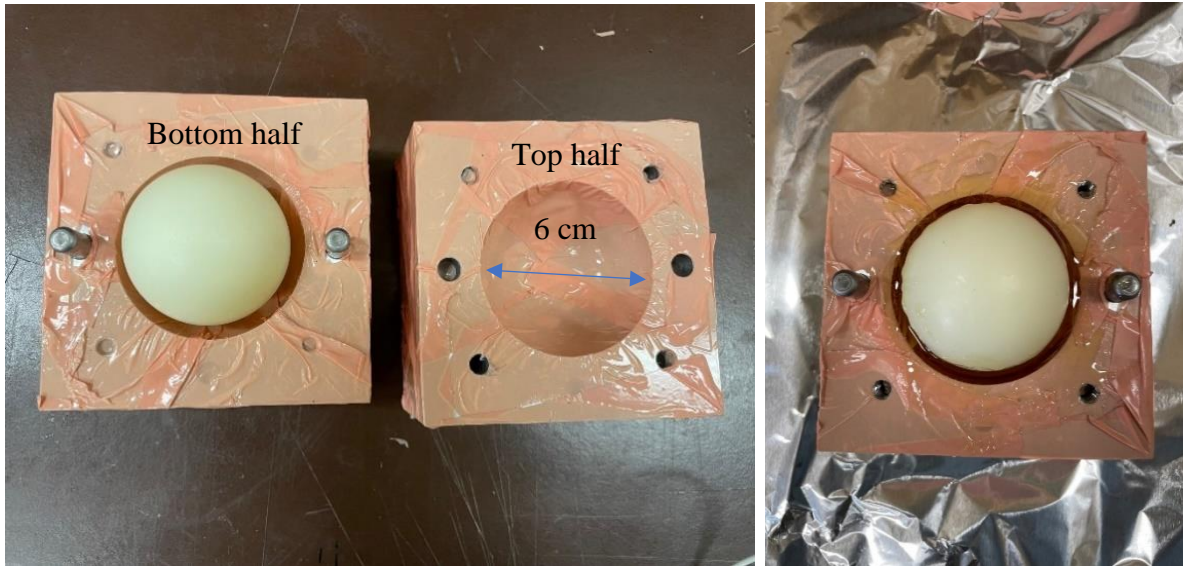


Figure 5. a) Release film is applied to the mold's spherical surface; b) PF resin injected in bottom half of sphere mold.

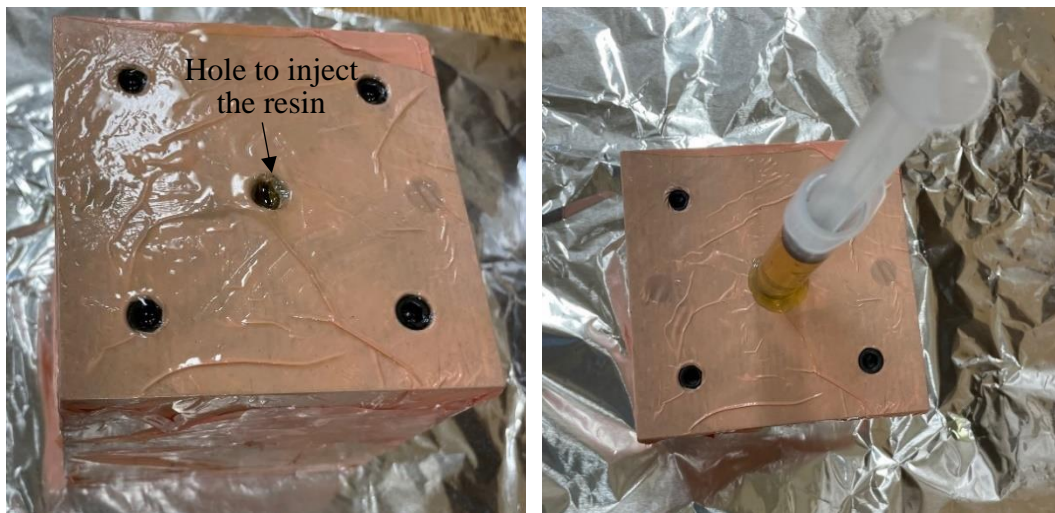


Figure 6. a) Two halves of the sphere mold closed and secured with Allen wrench for four screws; b) Second sample of cured steel sphere with carbon fiber strip and PF resin being injected into mold.

This procedure was repeated to prepare a second cured epoxy sphere sample by not using release film on the sphere mold. Instead, resin release spray was used to coat both halves of the sphere mold (Figure 7). The resulting cured steel sphere sample prepared with release film is seen in Figure 8, and the resulting cured steel sphere sample prepared with release spray is seen in Figure 9.

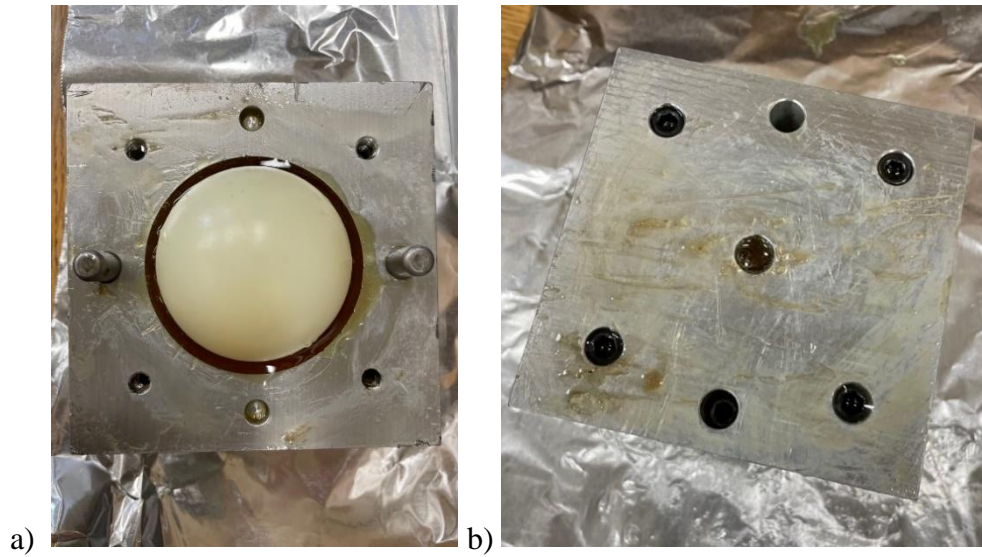


Figure 7. a) Epoxy sphere submerged in resin on bottom half of sphere mold for second sample with release spray. b) Closed sphere mold with release spray for second sample after injecting resin for top half of epoxy sphere.

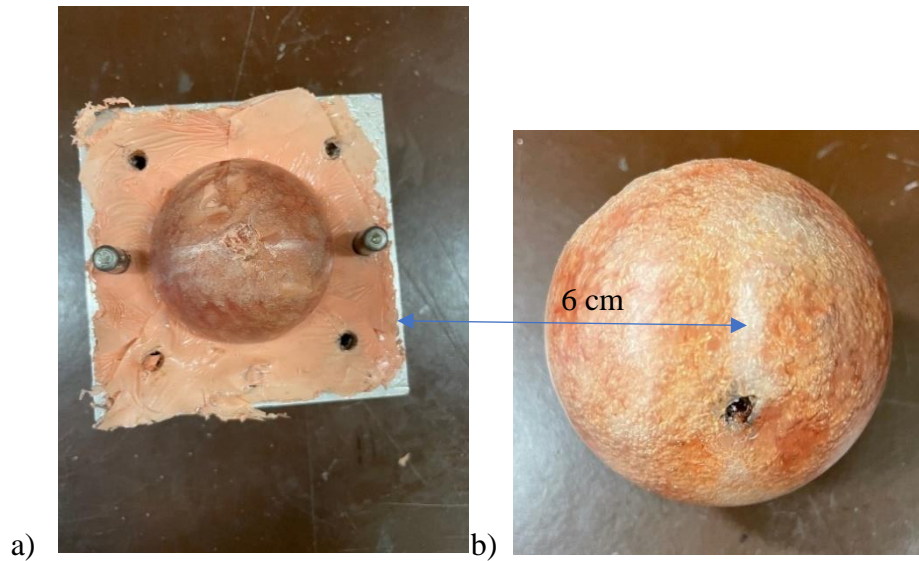


Figure 8. a) Top half of first sample of cured epoxy sphere made with release film. b) Bottom half of first sample of cured epoxy sphere with release film.

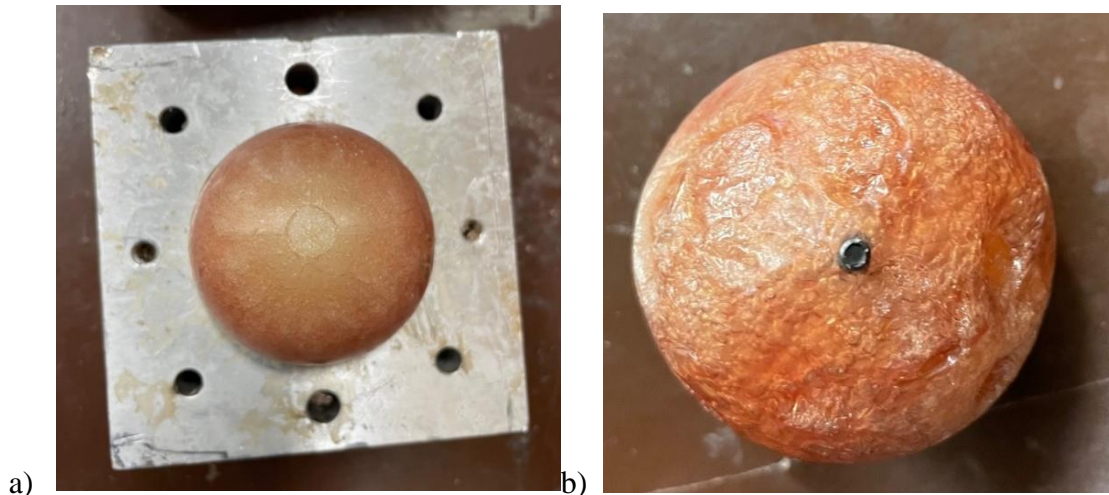


Figure 9. a) Top half of second sample of cured steel sphere made with release spray. b) Bottom half of second sample of cured epoxy sphere with release spray.

For the first cured pebble made without release film, the PF resin seeped on top of the release film and formed a layer on the release film around the epoxy sphere (Figure 8). For the second cured pebble made without release film, the top of the epoxy sphere had a smooth surface of PF resin. The bottom half of the sample was not as smooth (Figure 9). To make a cured pebble with a smooth finish of PF resin on both halves, the resin can be injected on the bottom half of the sphere mold before attaching the sphere on the screw. Once a smooth surface is established on top and bottom parts of an epoxy sphere, then the procedure will eventually be repeated using steel spheres for more accurate experimental testing (weight differences between sphere materials can have an unintended effect on the operation of the pebble sorting system in subsection 3.3).

3.2 Ultrasound scanning of graphitic surrogates embedded with YSZ microspheres

An Ultrasonix Sonix Touch commercial-off-the-shelf (COTS) ultrasound scanning (US) system was used for imaging both the flat-surface and spherical-surface graphitic surrogate samples. Ultrasound B-mode¹³ imaging data were acquired using a motorized linear probe held by a metal stand. An aqueous ultrasound gel pad (Aquaflex, Parker Laboratories, NJ, U.S.) was placed between the transducer probe and the surface of each sample. Experiments were performed with the sample immersed in water to ensure continuous coupling between the probe and the samples. 3D data were then recorded while the transducer probe was steered along perpendicular scan directions.

B-mode images were thresholded using Otsu's method¹⁴ followed by morphological reconstruction to localize the sample surface. Imaging artefacts near the sample boundaries were suppressed to improve the accuracy of surface detection. The volume of interest (VOI) was then determined as a volume with constant thickness relative to the top surface of the reconstructed geometry. This thickness was measured in practice from the sample. Within this VOI, voxel values

¹³ B-Mode is a two-dimensional ultrasound image display composed of bright dots representing the ultrasound echoes.

¹⁴ In image processing, Otsu's method, named after Nobuyuki Otsu, is an algorithm used to perform automatic image thresholding. Thresholding is the simplest method of segmenting images used to turn a gray-scale image into a binary image.

were populated to represent the histogram of the volumetric image data. Of note, the superficial region (in terms of image depth relative to the sample surface) was not considered when generating histograms due to the confounding effect of specular reflection on detecting the microspheres. Quantification was performed by extracting the voxel value at which a global peak was observed in acquired histograms.

3.2.1.1 *Ultrasound scanning of flat-surface graphitic surrogates*

Two types of flat-surface graphitic surrogate samples were scanned using the US system. In sample-1, the spacing between the 2 mm YSZ microspheres was approximately 3 mm, and for sample-2, the spacing was 6 mm. Figure 10 shows both samples with microsphere location marked with asterisks for the reader's ease.

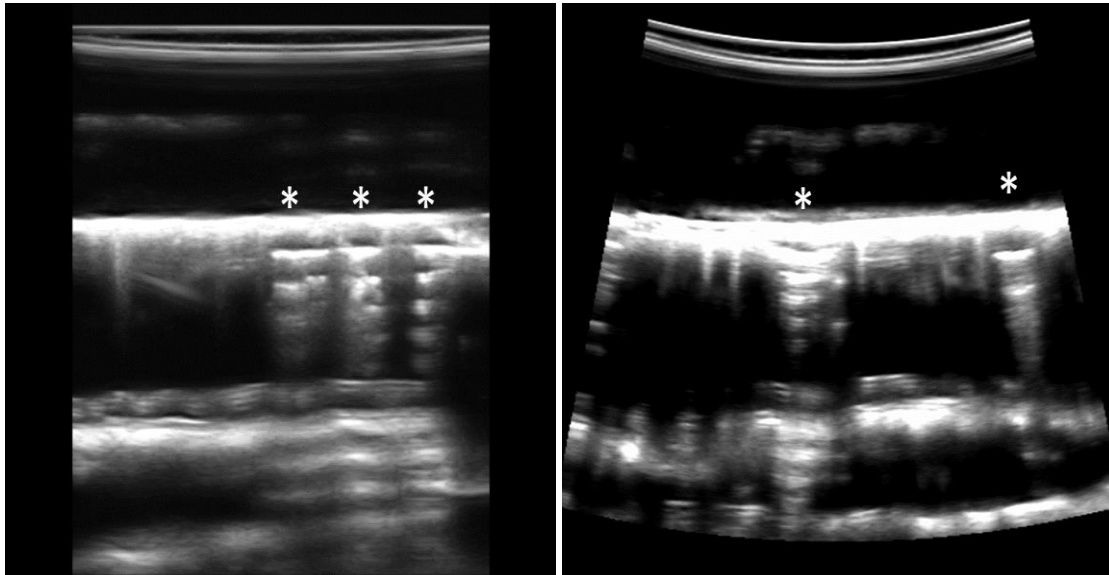


Figure 10. B-mode US images of flat-surface graphitic surrogate sample-1 and sample-2 impregnated with 2 mm diameter YSZ microspheres with an interstitial spacing of 3 mm and 6 mm. The asterisk annotations are to depict the identification possibility.

Analysis of the US images for both samples showed that the sample-2 with lower microsphere concentration (*i.e.*, with 6 mm spacing) peaks at higher intensity than the histogram obtained from the sample-1 with higher microsphere concentration (*i.e.*, with 3 mm spacing). The spacing between the reverberation artefacts for sample-1 and sample-2 (marked in Figure 10 with asterisks) appears to be well correlated and identifiable to the underlying YSZ microsphere distributions.

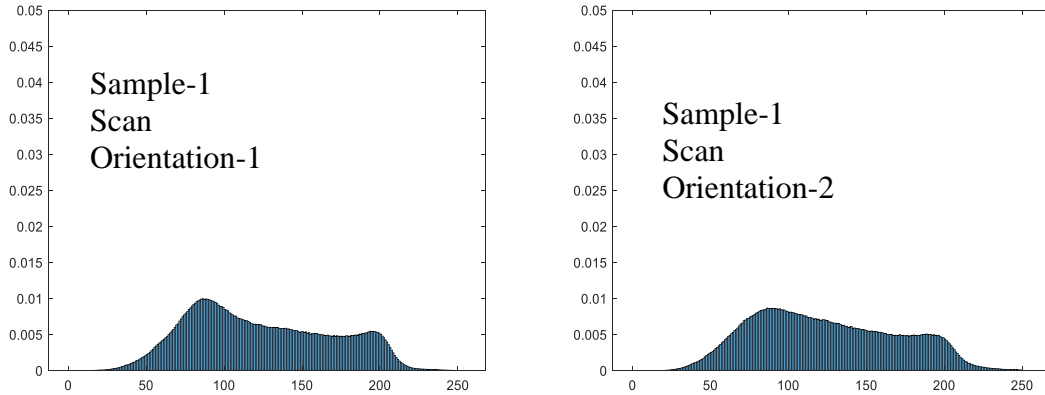


Figure 11. Histogram distribution of intensities (x-axis) obtained for the US scan of the flat-surface sample-1.

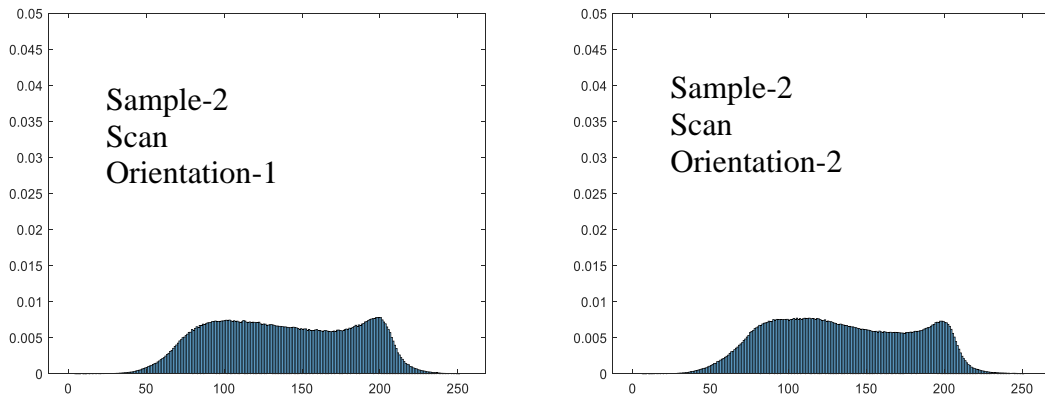


Figure 12. Histogram distribution of intensities (x-axis) obtained for the US scan of the flat-surface sample-2.

The histogram distributions obtained for the images of sample 1 and sample 2 were bimodal (Figures 11 and 12). Therefore, two histogram peaks for each sample were estimated. The voxel values where each histogram peaks from the interior of the two samples scanned using two orthogonal orientations of the US probe are listed in Table I.

Table I. Voxel values for each histogram peak from the US images of two samples using two probe orientations.

	Orientation-1		Orientation-2	
	Sample-1	Sample-2	Sample- 1	Sample-2
Intensity of peak 1	87	104	87	114
Intensity of peak 2	194	199	189	198

3.2.1.2 Ultrasound scanning of spherical graphitic surrogates

For each of the two spherical surrogates, four volume datasets were collected at two perpendicular orientations of the ultrasound transducer with two repetitions by using a motorized linear probe (A), i.e., a volumetric probe of the 3D imaging system. The same experiments were also performed by employing a linear array probe (B), where the 3D volumes were reconstructed by using the multiple 2D images taken at different positions. This second set of the experiments better emulate the testing conditions that will be implemented in the future prototype. During the data collection, the space between the sample and the transducer was filled with water. A schematic of the set-up is shown in Figure 13.

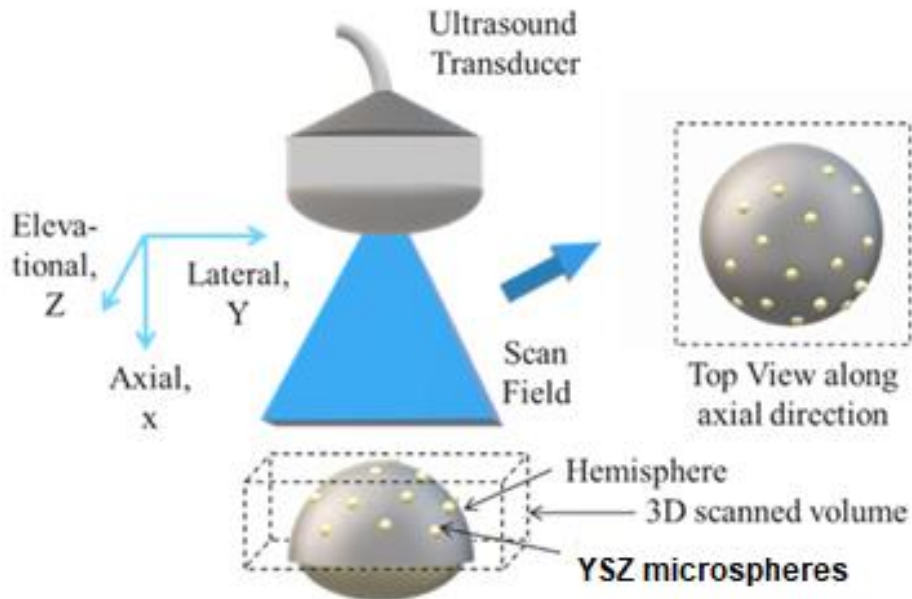


Figure 13. Schematic of the US data acquisition experiments with spherical surrogates.

In order to analyze the 3D US data (i.e., B-mode) for the YSZ microsphere impregnated spherical surrogates collected by probes (A) and (B), the datasets were pre-processed by selecting the interrogative 3D volume (shown in Figure 14) that mainly included the top hemisphere and a reduced quantity of YSZ microspheres. The amplitude of the voxels within the selected volume were normalized and histogram analysis detected differences between the two spherical surrogates. The imaging data was conducted on the penultimate spherical samples developed by the pebble preparation team (i.e., the setup shown in Figure 14 were the latest scanned samples while the pebbles from the refined process described in subsection 3.1.2.1 were not scanned in time for this report).

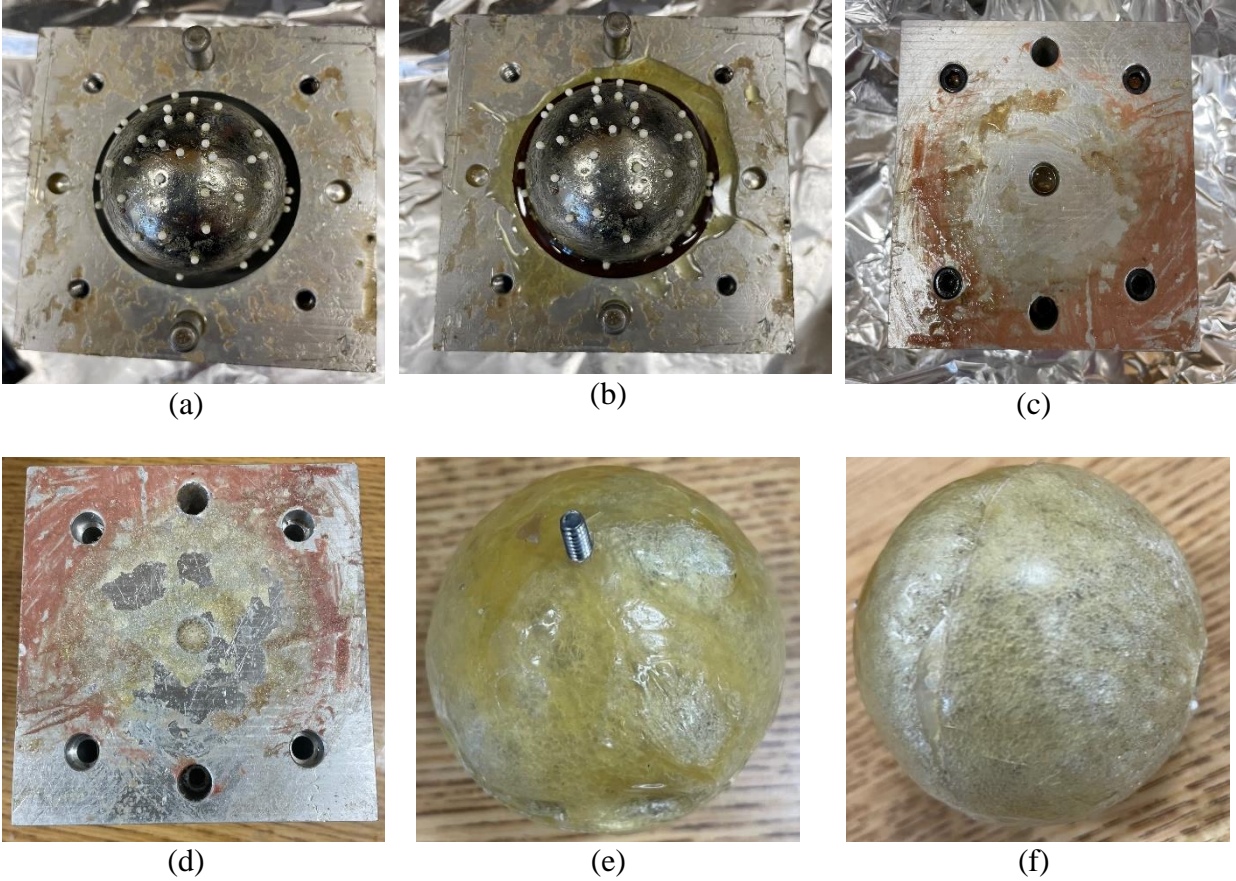


Figure 14. **Top** (Left to Right): (a) curved sphere with YSZ microspheres screwed onto bottom half of sphere mold with diameter 5cm; (b) PF resin injected into the bottom half of the sphere mold; (c) two halves of the sphere mold are closed and secured with Allen wrench for four screws and resin injected into top half of the sphere mold. **Bottom** (Left to Right): (d) sphere mold with samples taken out of the oven; (e) bottom view of cured sphere with YSZ microspheres; (f) side view of cured sphere with YSZ microspheres.

This analysis was performed in a slightly different way than for the flat-surface sample cases. For the histogram analysis, the frequency or the number of occurrences of the amplitude values at uniform intervals (i.e., bins) over the whole normalized value range (0 to 1) was calculated. The bin interval was as low as 0.005 to get a smooth distribution. Then, the percentage of the total number of **voxels** that were present in the upper half region of the histograms (**0.5-1**) was measured. The results obtained are shown in Tables II and III.

Table II: Histogram results for the four datasets of two samples (for probe (A) of 3D imaging)

Sample	Hemisphere 1 with 10-15 YSZ microspheres	Hemisphere 2 with 25-30 YSZ microspheres
Mean % of Voxels (upper half of the histogram)	3.99	5.62

Table III: Histogram results for the four datasets of two samples (for probe (B) of 3D imaging)

Sample	Hemisphere 1 with 10-15 YSZ microspheres	Hemisphere 2 with 25-30 YSZ microspheres
Mean % of Voxels (upper half of the histogram)	1.80	2.16

Data shown in Tables II and III were obtained from the histogram data (Figures 15 and 16) generated from the experimental US scan and image analysis. Histogram shows marked difference between 10-15 YSZ microspheres embedded sample and 25-30 YSZ microspheres embedded sample.

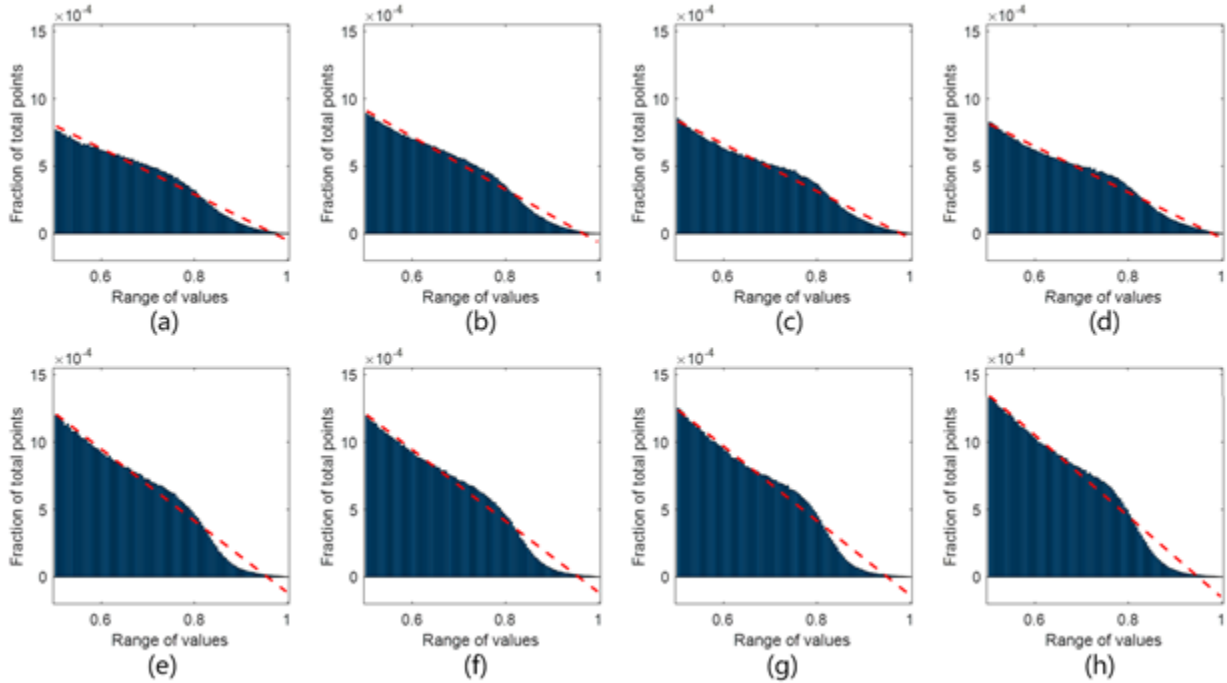


Figure 15. Histograms for the upper half intensity values along with fitted lines. The first row corresponds to the 4 data sets obtained from the pebble with 10-15 microspheres. The second row corresponds to the 4 data sets obtained from the pebble with 25-30 microspheres. Both rows refer to the data acquired using probe (A), i.e., the volumetric probe.

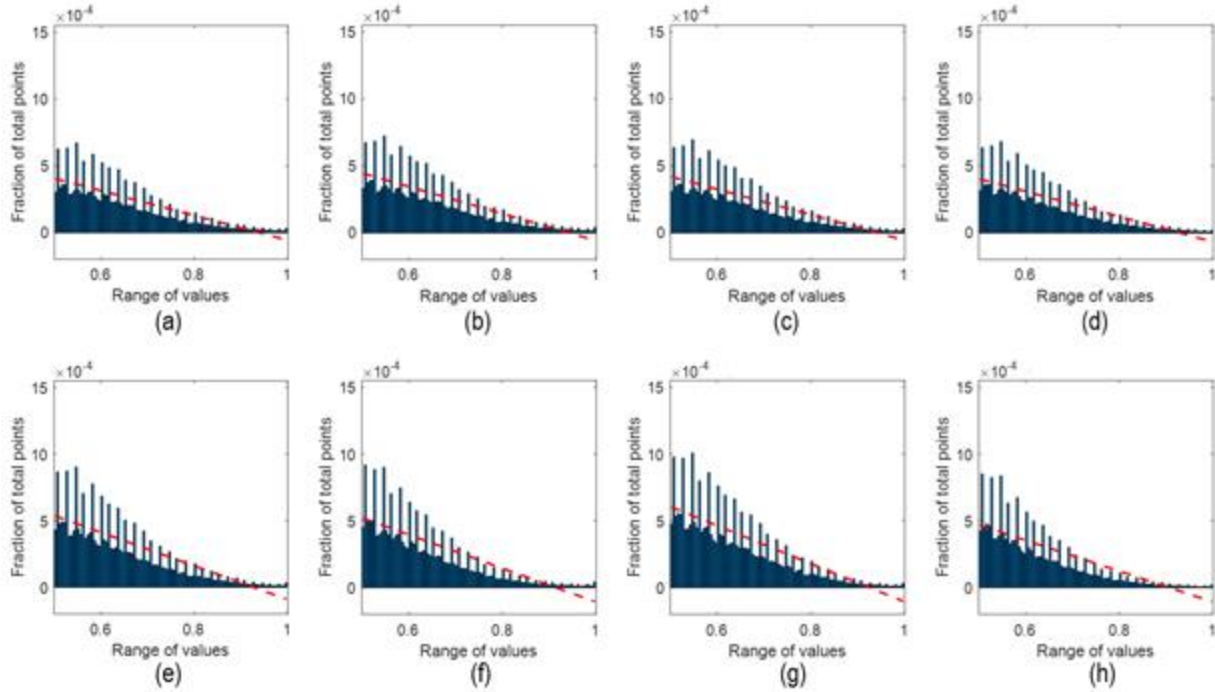


Figure 16. Histograms for the upper half intensity values along with fitted lines. The first row corresponds to the 4 data sets obtained from the pebble with 10-15 microspheres. The second row corresponds to the 4 data sets obtained from the pebble with 25-30 microspheres. Both rows refer to the data acquired using probe (B), i.e., the linear array probe.

The scoping ultrasound scanning and image analysis conducted of flat-surface and spherical-surface surrogates suggest that the spatial density differences of impregnated YSZ microspheres can be differentiated using histogram analysis per the data provided in Tables I, II, and III. Maximum distinguishability is seen while using the volumetric probe. The difference between the percentages for probe (A) and probe (B) is the result of different US system-defined parameters such as probe frequency, number of probe elements, and the quantization levels in the output image. Higher percentage of amplitude values at the upper part of the intensity histograms for sample with more YSZ microspheres was observed – which is consistent with the theoretical expectations. The difference of percentages is dependent on the distribution as well as the number of YSZ microspheres over the sample surface. In addition, rough surface geometry and the placement of the transducer over the spherical sample are critical to get accurate estimates. Nevertheless, the histogram findings were found to be consistent for several scan datasets. In conclusion the study showed that the US method of identifying kinds of pebbles in a PFR is promising. The two advantages of the novel nuclear material control technique developed are: (1) classification of pebbles with extrinsic feature in addition to the radiation-based fuel burnup measurement, (2) no cooling time needed for the pebbles, and (3) decreases the pebble time ex-core while waiting on the decision to reinsert it into the reactor or send to storage.

3.3 Pebble sorting mechanism prototype

This task involved designing a prototype (proof-of-concept) pebble scanning system capable of moving single nuclear fuel pebbles through a device that scans the outer surface of the pebbles. The design of this system will integrate the US system. The design goals for the pebble sorting

system were to design and build a system that would control movement of the pebbles and scan as much of the outer surface of the pebbles one at a time using a COTS ultrasound machine. The ultrasound machine requires that the scanning process take place in water. Other factors influencing the design of the system were an aggressive timeline and limited budget. As a result, an approach was taken to design the system as simply as possible in order to reduce cost, accelerate development time, and minimize unknowns and risk as much as possible. The following specific dimensions were provided:

1. Pebble outside diameter: 2.36" (60mm)
2. Pebble weight: not to exceed 500g
3. Ultrasound scan rotation: 360° or more

3.3.1 Concept development

Early concepts first explored involved using off-the-shelf industrial motion control systems. Numerous motion control companies have flexible pre-packaged systems designed to control the movement of parts and components typically for use in manufacturing and production facilities, as well as repetitive testing applications. These systems can incorporate pneumatics, electro-pneumatics, and pure electrical motion control devices, and commonly have feedback sensors to monitor movement, force, speed, velocity, etc. These devices typically come with software to tune and adjust all parameters of the system, as well as document all movements. The downsides of using these systems are the cost and lead times associated with these types of systems, as well as the added complexity of the systems.

After much investigating and speaking to multiple technical representatives, it was concluded that the best solution was to design a very basic and simple system utilizing a minimum of movements and parts. This approach would result in an inexpensive, reliable system that could be fabricated within the budget and time allowed. To accomplish this economic approach, a system was designed that could be built from mostly off-the-shelf components, with a minimum of custom fabricated parts.

The design was reduced to just the minimum of components needed to accomplish the basic movements, defined as:

1. Feed a single pebble into a defined location underwater
2. Hold the pebble at that location while inducing rotation in the pebble to perform the ultrasonic scan
3. Once rotated a minimum of 360°, move the pebble out of the way
4. Drop the next pebble into the same defined location and repeat.

It was then determined that these required movements could be achieved with only two steps. The first would be to feed individual pebbles down a chute to the location underwater. The next is to rotate the pebble and drop it out of the way. It was decided that using rotational movements instead of linear movements was far easier and cheaper since gear reduction motor assemblies were readily available.

The key to achieving these movements was the design of a 'Y' tube assembly, which is shown in Figure 15. The vertical branch of the 'Y' is where the US probe will be located, and the other is the chute where the pebbles travel to the rotation location, as shown in Figure 15.

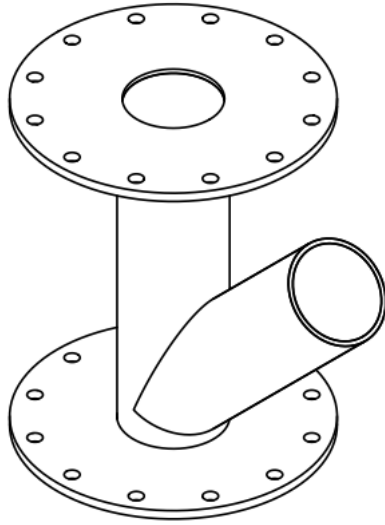


Figure 15. 'Y' Tube

Common, consumer-rated ABS plastic flanges were created to be attached to the top and bottom of the Y tube. These flanges allowed simple support of the Y and also support a power transmission shaft, which will be shown later.

The next step was to design the two rotating assemblies to control both the movement of the pebbles into the 'Y' and out of the 'Y'. The controlled feed of pebbles into the 'Y' is accomplished with a paddle wheel type of device, with the rotation being driven by a DC gear reduction motor. This paddle wheel is shown in Figure 16. This wheel is designed to transport a pebble from the vertical feed tube to the inlet of the sloped tube of the 'Y'. The wheel is designed to hold a pebble in every other slot, this was done to allow for controlled delivery of a single pebble at a time.

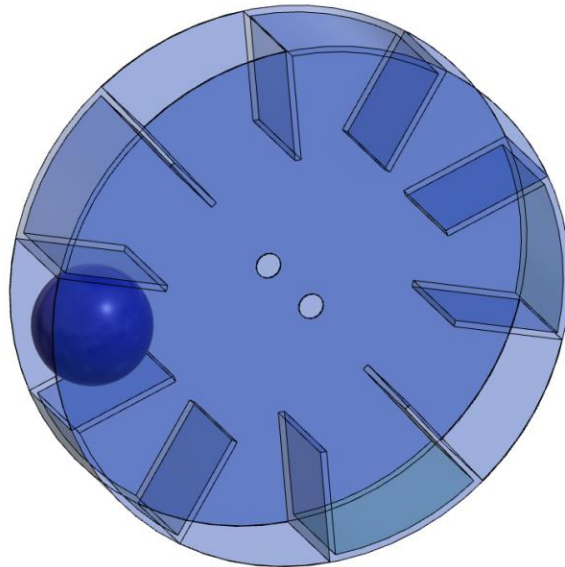


Figure 16. Paddle Wheel Assembly

Once a pebble slides down the tube, it will rest at the bottom of the ‘Y’ tube. The rotation of the pebble once in position is performed with the use of a serrated disk with an elongated hole in it. The serrations maximize friction to ensure that the rotating surface causes rotation in the pebble. The hole allows the pebble to drop out once a complete scan is performed. This disk (shown in Figure 17) is attached to a shaft and powered by another DC gear reduction motor.

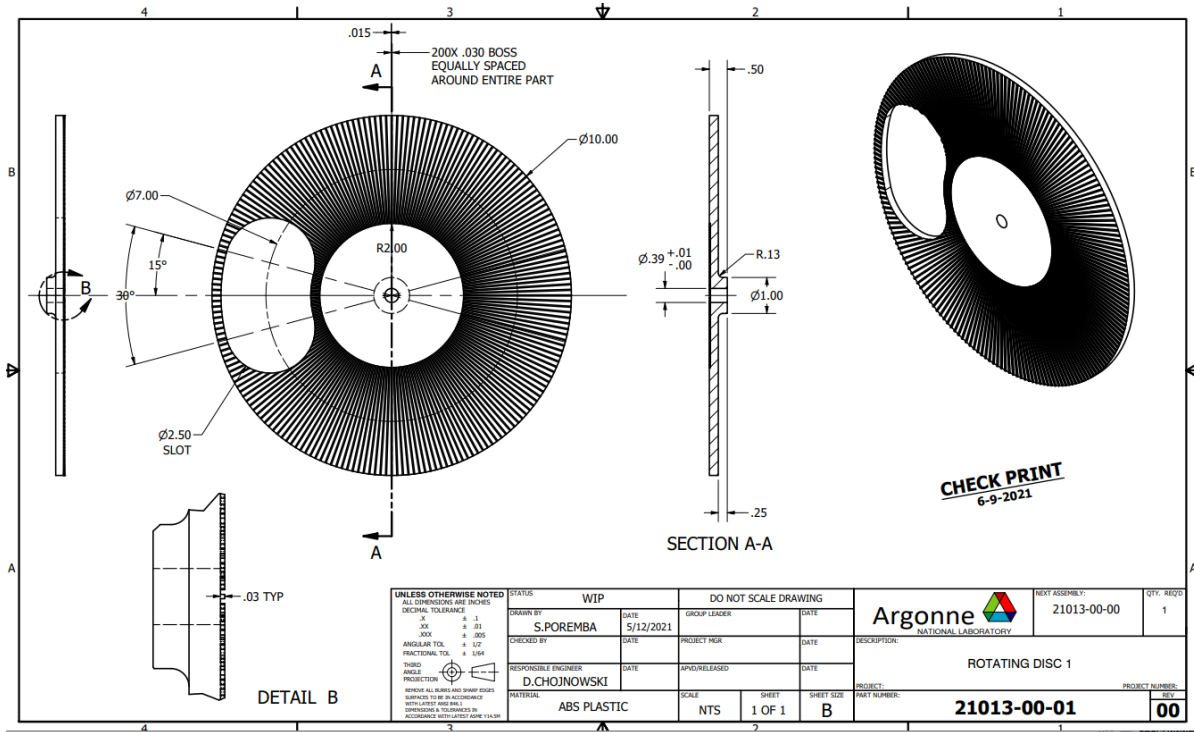


Figure 17. Drawing for rotation disk with hole

3.3.2 System Design

The disk and wheel shown above were integrated with the ‘Y’ tube fitting to control the motion of the pebbles. This final design of the key components is shown below in Figure 18.

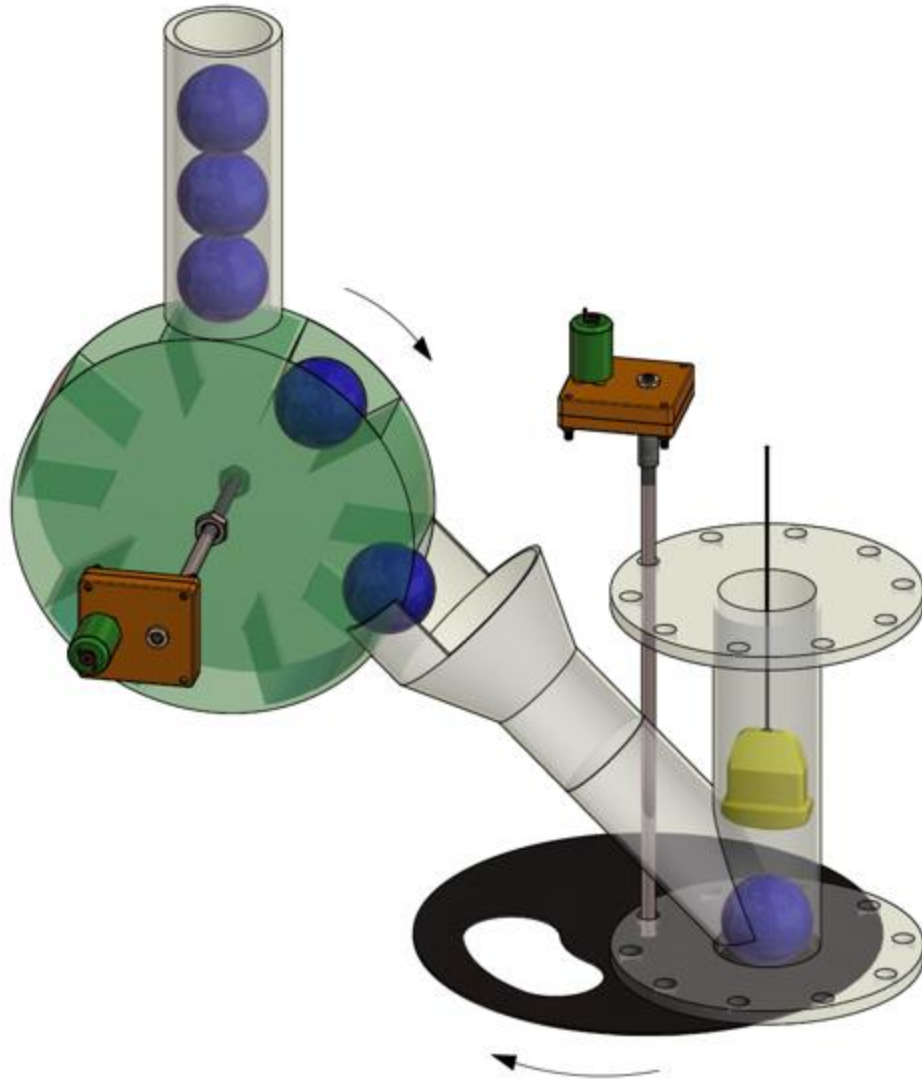


Figure 18. Assembly showing pebble feed wheel, polycarbonate ‘Y’, and rotation disk, along with drive motors

Figure 18 illustrates how the overall system works. The paddle wheel rotates slowly, dropping pebbles one at a time down the ‘Y’ tube. Once the pebble is in position at the bottom of the tube, the rotating disk induces rotation of the pebble, while the US system performs a scan of the pebble surface. The two gear reduction motors are speed adjustable and must be synchronized and adjusted so that a new pebble drops down the tube just after the previous one drops through the hole. The ultrasound scanner is shown as the yellow device located in the vertical tube section. This entire assembly needed a support structure, and also required that the pebbles and ultrasonic scanner are immersed in water. Therefore, a Unistrut channel frame was constructed, and this frame was designed to sit on top of an off-the-shelf 15-gallon aquarium tank. This tank has clear glass sides that allow for viewing of the pebbles while rotating and scanning occurs. This assembly mounted to the tank is shown in Figure 19.

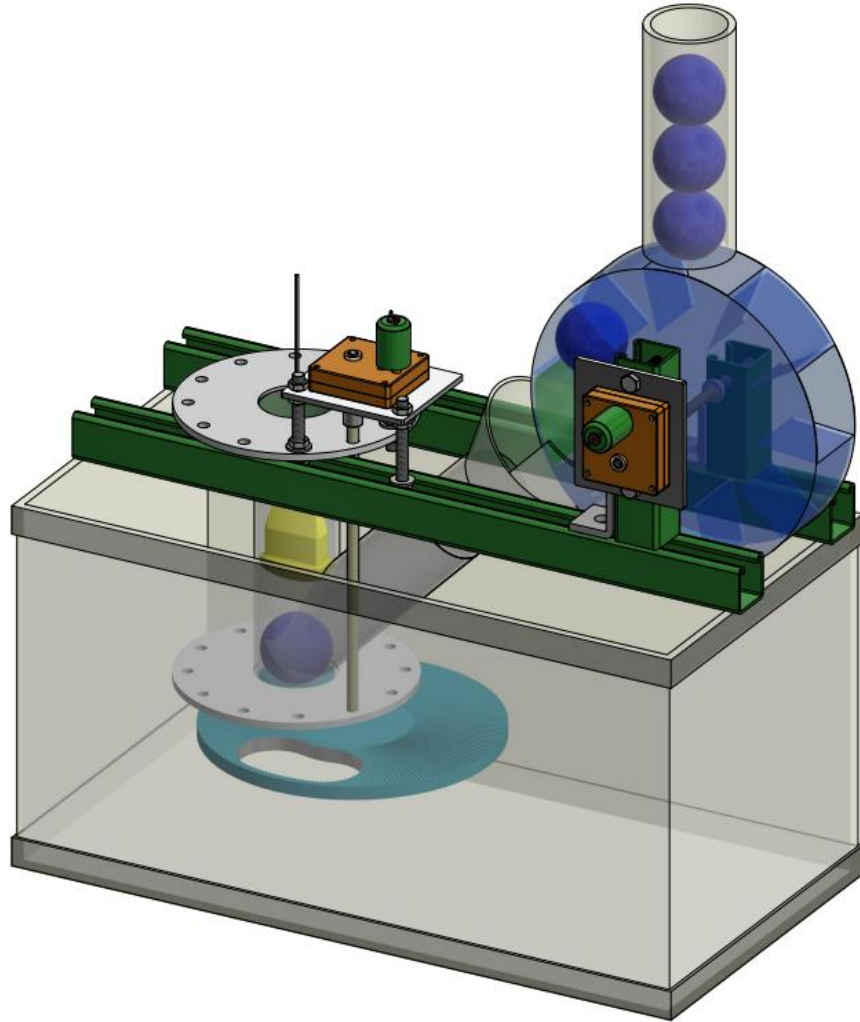


Figure 19. Pebble handling assembly shown mounted to water tank

Figure 20 shows a side view of the assembly mounted to the tank is shown, indicating the water level required in the tank. This water level is required to allow the US system to effectively scan the pebbles.

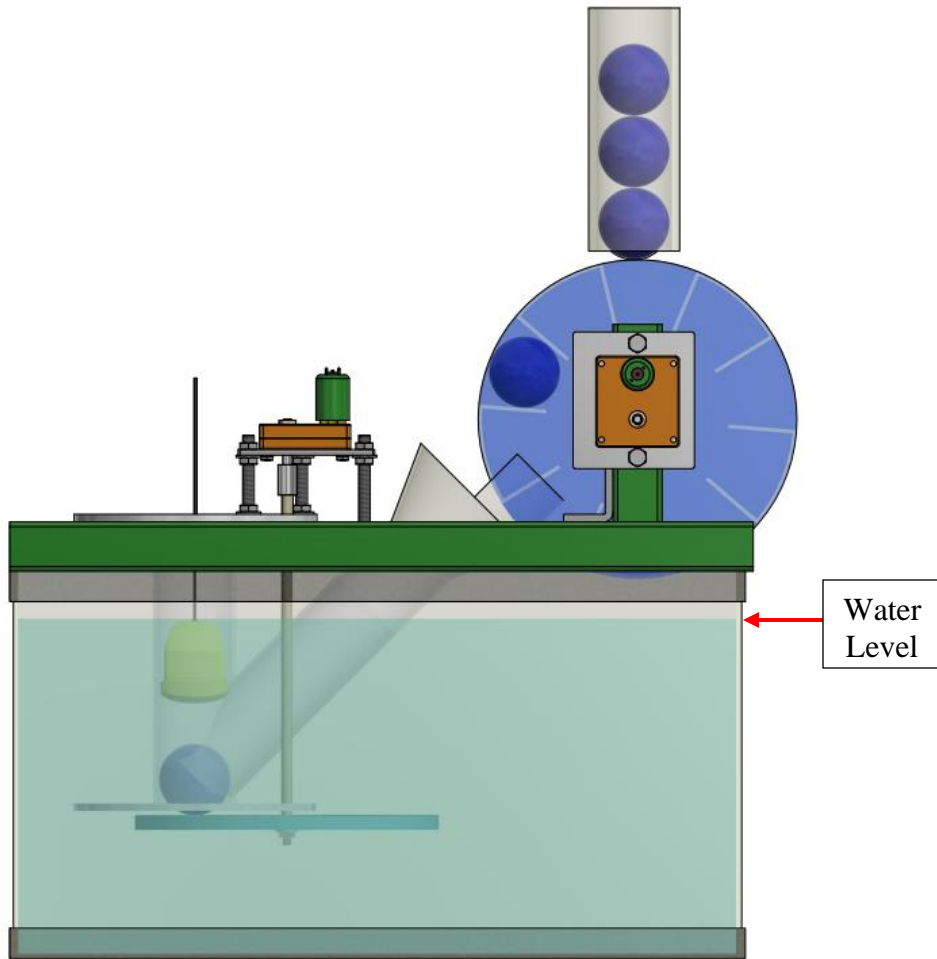


Figure 20. Pebble handling system shown with required water level in tank

3.3.3 Component Procurement and Fabrication

The pervasiveness of the COVID-19 virus and its effects on supply and delivery of parts and labor had an impact on progress achieved this fiscal year. Lead times for anything other than stock parts were extremely long. As a result, the decision was made to build this system in two stages, in order to minimize risk and ensure a functional prototype by the deadline. The first phase would omit the paddle wheel feature and include all other features. In addition, availability of key components was investigated early on during the design, and these readily available parts were designed into the system. This technique was employed to ensure availability and budget requirements were met. A local parts distributor (McMaster-Carr) was used as a source for many parts. Several off-the-shelf parts were modified slightly by our on-site machine shop. The custom designed composite parts that could not be fabricated out of standard stock materials were designed to meet the size limitations of an on-site 3-D ABS plastic printer. This printer can quickly generate ABS plastic parts that have impressive structural capabilities for just the cost of the bulk material.

The tube ‘Y’ was the most complicated part to fabricate since this had to be custom made. Initially clear Acrylic tubing was ordered, but once received it was determined that dimensionally it was not round, and the machinists were reluctant to work with it due to its likelihood to crack. As a result, Polycarbonate tubing was ordered instead. This material was dimensionally more consistent, and machining was successful. The fabrication drawing of the Y is shown in Figure 21.

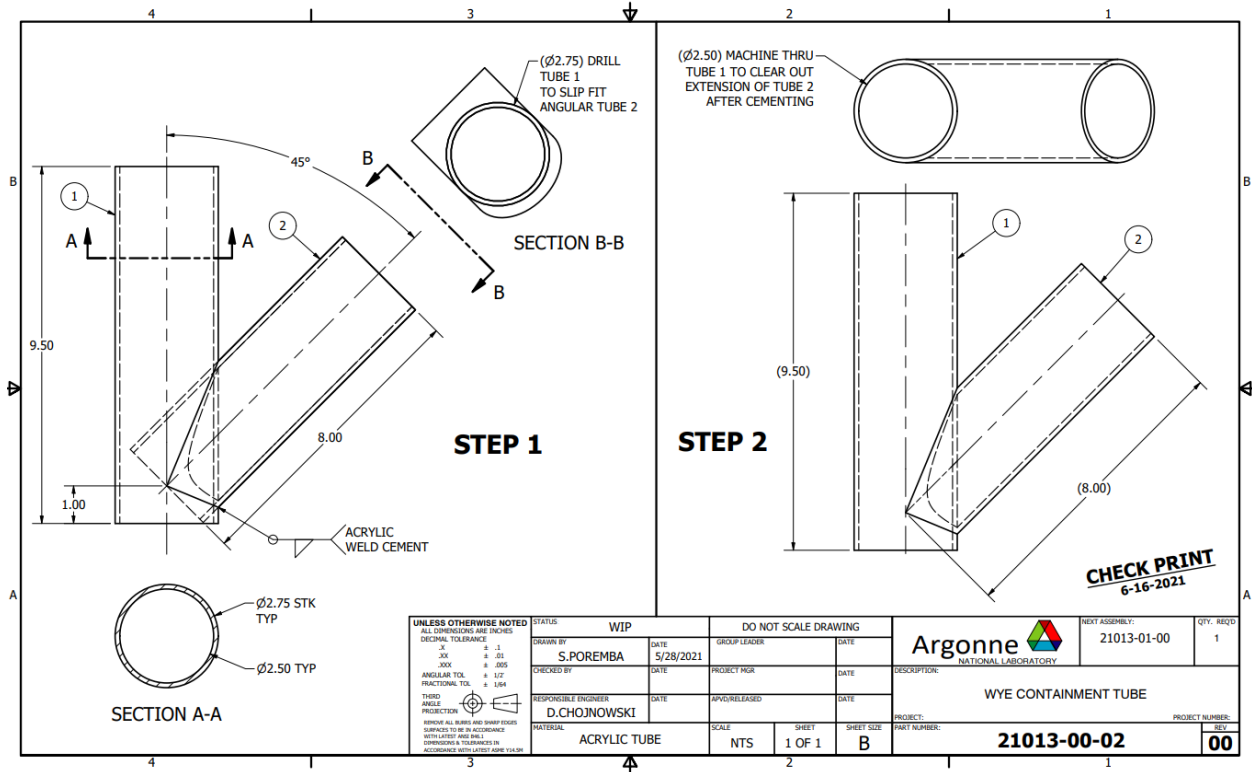


Figure 21. Fabrication drawing for polycarbonate tube Y assembly.

Off-the-shelf parts were ordered as much as possible, such as the gear reduction motors, shown in Figure 22. The system is designed to use two of the same motors for simplicity. These motors operate on 12 V DC, and have variable speed of up to 50 RPM, which is needed to synchronize the pebble wheel and rotation disk. The gear reduction drive gives these motors an impressive torque of 160 in-oz., which is required to turn both the paddle wheel and the rotation disk.

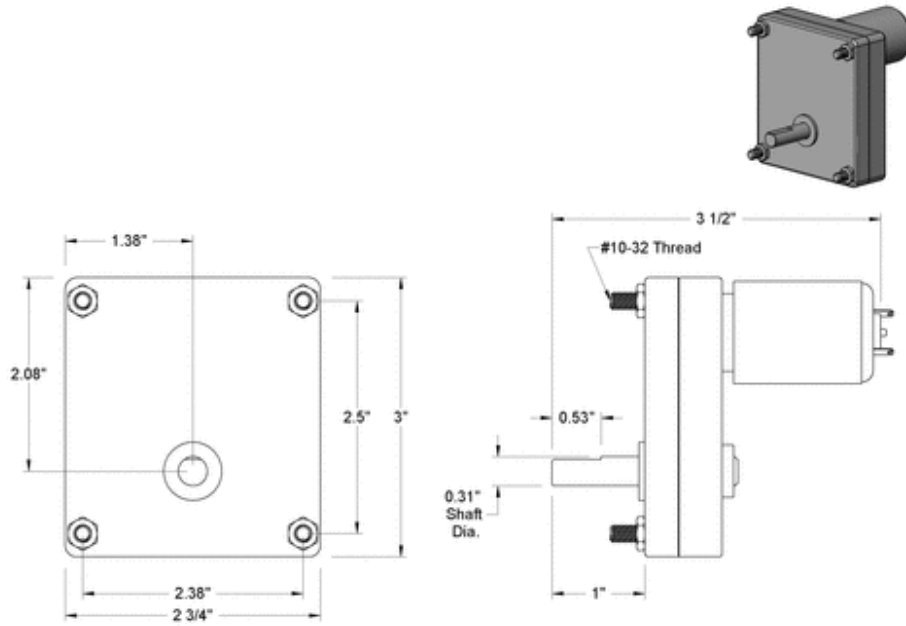


Figure 22. Gear reduction DC motor

A motor mounting plate was needed to support the motor and connect it to the Unistrut frame on top of the tank. The plate was designed to be fabricated out of standard 3/16" aluminum stock. A drawing of the plate design is shown in Figure 23.

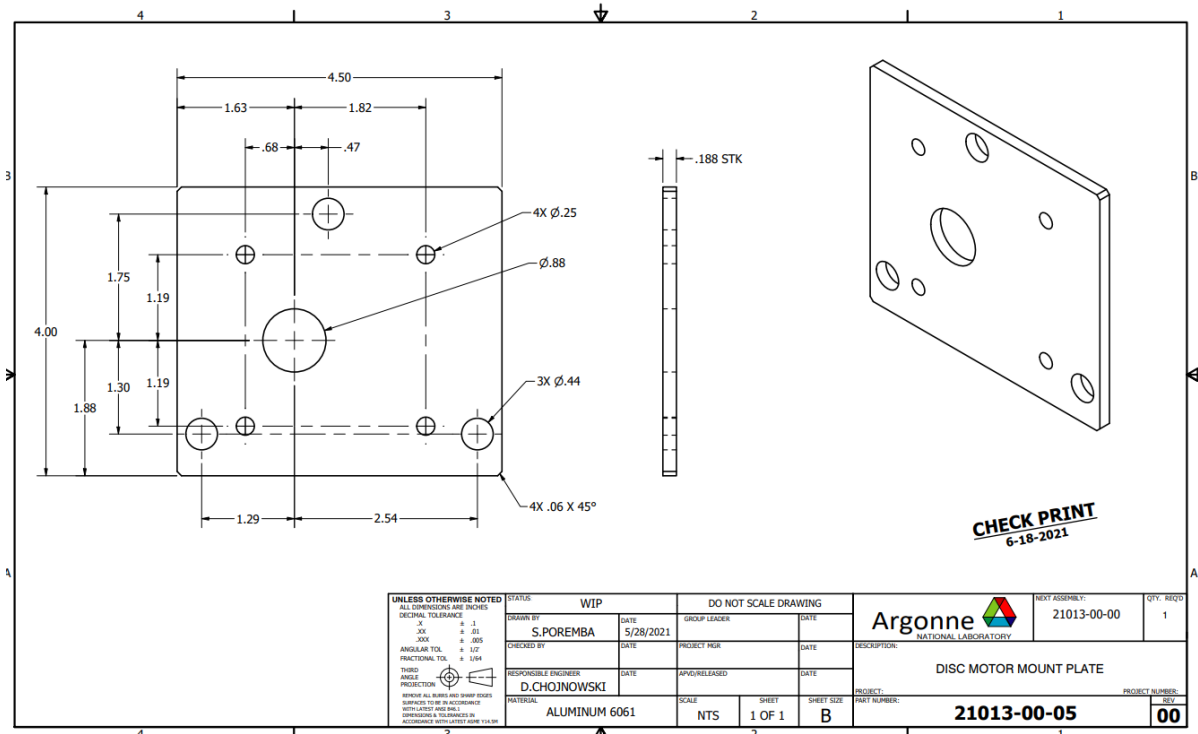


Figure 23. Aluminum motor mounting plate

A triple output DC power supply was ordered to power the DC motors. This power supply is made by BK Precision, and has an output range from 0-32 VDC and is capable of delivering up to 3.0 A. This multichannel supply is necessary to power the two DC motors independently. A photo of the DC power supply unit is shown in Figure 24.

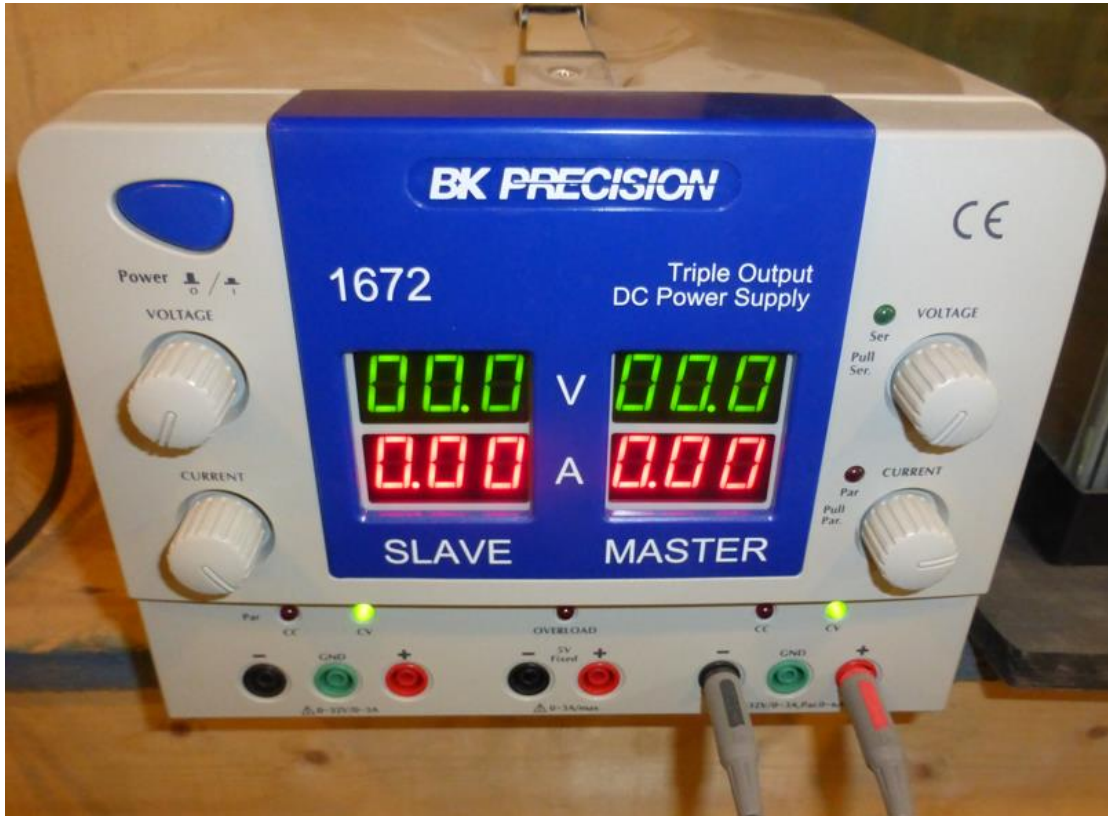


Figure 24. Variable output multichannel DC power supply

3.3.4 Assembly

Assembly of the system was mostly straightforward, with the exception of the ‘Y’ tube and flanges. The tube components were cemented together with special adhesive designed for use on polycarbonates and ABS-polycarbonate joints. An alignment plug was fabricated out of Teflon to aid in assembly and hold the two pieces of polycarbonate aligned during adhesive cure. This was critical, since any misalignment could create a discontinuity inside the tubing and impede smooth travel of the pebbles. The Teflon material was chosen so that the adhesive would not bond to it, and it could be removed afterwards. A threaded rod pull system was fabricated to aid in pulling the alignment plug from the ‘Y’ after adhesive cure was complete. The polycarbonate ‘Y’ is shown during the adhesive cure process in Figure 25.



Figure 25. Assembly of Y tube using adhesive and alignment plug pull device

The assembly of the flanges was the next step, which required careful indexing of the flanges with respect to each other. This was required in order to allow the transmission shaft for the rotation disk to pass through a set of holes in the flanges. An assembly drawing for the 'Y' flange is shown in Figure 26.

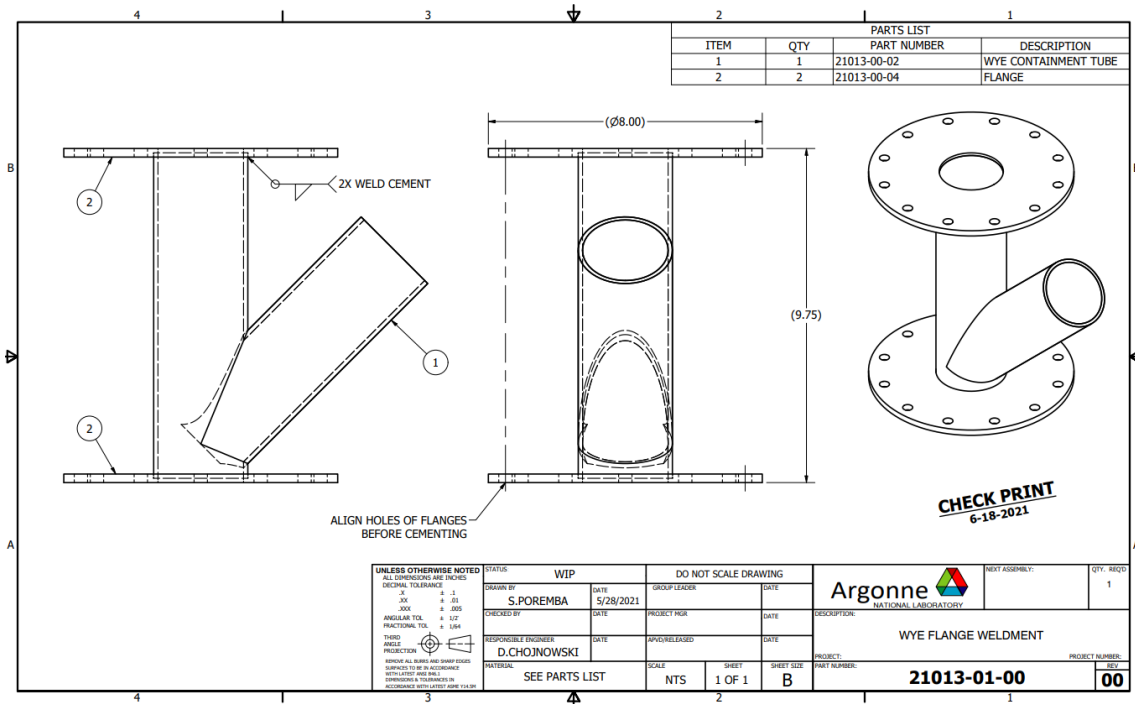


Figure 26. 'Y' flange assembly drawing

After the ‘Y’ tube assembly was completed, the Unistrut frame support was built. This was built using double angle, so that mounting nuts could be used either on top or below the strut and sized to sit on top of the plastic frame of the water tank. ‘L’ angles were attached to the bottom of the strut, to keep the strut members tightly in place. A short crossmember was also cut and installed for strength and rigidity.

Once this strut assembly was completed and mounted on top of the tank, the ‘Y’ assembly was attached, as well as the rotation disk and transmission shaft. Next, the motor was mounted on its plate, and attached to the strut with threaded rods. As mentioned earlier, the paddle wheel component was postponed until the next phase of the project due to time and budgetary constraints. The assembly at the completion of Phase 1 is shown in Figure 27.



Figure 27. Pebble Scanning System during assembly

Initial operational testing was conducted with the assembly with surrogate pebbles provided by TAMU. These pebbles (as shown in Figure 4) did not accurately exhibit the eventual design specifications of actual fuel pebbles used in PFRs. In an attempt to gauge functionality of the pebble sorting assembly, COTS steel bearing balls were acquired (Figure 28) to serve as pebbles.



Figure 28. 60mm-diameter steel bearing balls from McMaster-Carr

Though the outer dimensions matched for testing the system, two physical characteristics differed greatly when compared to actual fuel pebbles for which the system was designed: the mass and the surface roughness. The density of the steel bearing balls resulted in masses of 2,000g each ball – compared to the total mass of a fuel pebble being 200g. Furthermore, considering the steel balls were meant to serve as ball bearings, the surface roughness of the polished surface made for frictionless rotating. With the rotation disk (Figure 17) of the pebble sorting assembly relying on friction to promote rotation of each pebble, it was deemed the steel ball bearings would not provide valuable data in testing this component. After engaging with the TAMU team, next generation pebbles were provided for testing (as shown in Figures 8 and 9) which better matched all physical dimensions including mass, size, and surface roughness.

4 Discussion

The original plan for this project was to uniquely identify and track each individual pebble. Based on an earlier MC&A approach for PFRs, the concept embraced by this project employed tracking engineered characteristics for each fuel pebble using an optical system that would uniquely identify pebbles using orientation of embedded microspheres (Figure 29 shows initial scans from the original study).¹⁵ With a constructed database of each uniquely identified pebble from initial insertion, a subset of discharged pebbles upon exit would be scanned in the same manner to gain confidence that those exiting were those entering the reactor vessel. With unique identification possible, Continuity of Knowledge (CoK) on a statistically significant percentage of pebbles traversing through pebble-fueled reactors is theoretically possible.

¹⁵ Gitau, E.T., Charlton, W.S. *Use of a Microsphere Fingerprint for Identity Verification of Fuel Pebbles in a Pebble-fueled HTGR*. Journal of Nuclear Materials Management (Winter 2012, Volume XL, No. 2), pg. 19.

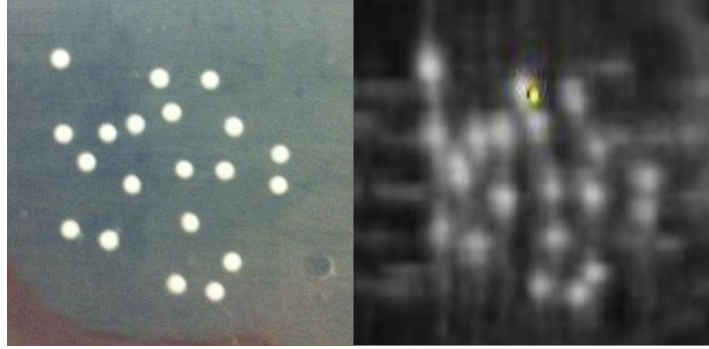


Figure 29. Comparison of initial image and ultrasound image of microspheres used to approximate a resolution for the imaging system.

The team was able to embed microspheres into graphite material replicating the graphite shell of pebble fuel elements and proved the feasibility of doing so in a controlled manner. Instead of random placement, structured microsphere placement allowed more precise experimental results to gauge the ability of optically imaging varying surface spatial densities. The limiting factor in imaging the microspheres rested on the US system being able to reliably identify hundreds of thousands of unique pebbles in a short time. When reported to industry experts, a secondary concept was considered: identifying groupings of pebbles to assist in reactor operations where types of pebbles could be identified and classified into batches. This concept still supports fresh fuel loading activities by distinguishing fuel pebbles from moderator pebbles – a recently stated objective identified by IAEA researchers.¹⁶

4.1 Potential MC&A Approach Implementation

It was theorized that types of pebbles could be assigned averaged spatial densities for the microspheres based on a weight percent of microspheres within the 5-mm outer layer graphite shell. Though later steps will require engagement with fuel pebble fabricators, the concept was proven to be feasible and potentially beneficial to an overall MC&A approach for these types of reactors. For example, some PFR designs include fuel pebbles consisting of more than one enrichment (differing weight percent of ²³⁵U). To distinguish pebbles of varying enrichment levels, different densities of embedded microspheres can be used. The aforementioned US system was verified as capable to image microspheres that have an average pitch of 3mm and 6mm. Also studied was a 12 mm pitch. It was theorized that averaged spacing between microspheres could be used to identify at least two different types of pebbles with pitches of 6 mm and 12 mm. Assuming a higher enriched type of fuel pebble is used at reactor startup and a lower enriched type of fuel pebble is designed for use in operating such a reactor, the operator would be able to identify when all the higher enriched pebbles used during reactor startup have been fully extracted. Moreover, with the ability of fabricating pebbles with more microsphere densities, other categorizations of fuel pebbles could be considered as well. For instance, pebbles inserted into the reactor vessel in a given year could have an average spatial spacing of 6 mm while the following year could be identified with 12 mm-spatial spacing. This would allow the operator to quickly assess upon initial discharge whether or not to reinsert the pebble or allow it to be assessed by the BUMS. This concept could essentially shorten the ex-core cooling time (ranging between 10 to 100 hours) before BUMS and thus, increase operational efficiency of the reactor system.

¹⁶ Lee, J, Doo, J.Y., Whitlock, J. *Safeguards by Design (SBD) for Small Modular Reactors (SMRs)*. Proceedings of the INMM & ESARDA Joint Virtual Annual Meeting, August/September, 2021.

Some reactor designs rely on moderator pebbles as well. Though those fuel pebble types could easily be identified via the BUMS, there would not be a method to identify them prior to BUMS and thus would still have to endure the ex-core cooling time before entering the BUMS. This MC&A approach with microspheres could simplify the process even if the absence of microspheres were used as a pebble type that would not have to rely on radiation measurements (and hence, require no cooling time). The ultrasound scan could occur immediately upon extraction and pebbles without fissile content would easily be identified as moderator pebbles by their lack of microspheres. The actual time spent for scanning individual pebbles is on the order of minutes so the resulting ex-core time each pebble would spend is substantially reduced.

The ability of categorizing and being able to 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 the BUMS (a material accountability-measure vital for applied safeguards of the reactor). If this system is able to be incorporated into the overall pebble extraction system, the potential reduction in ex-core time for pebbles is significant.

The system as designed provides an opportunity to identify and categorize batches of fuel pebbles based on specific fuel fabricator-defined features. Testing of the prototype device described in subsection 3.3 did not occur this fiscal year due to the pandemic resurgence in Q4 of FY21. However, the functionality of individual components such as the pebble sorting system, the ultrasound imaging system, and the microsphere embedding process were independently confirmed to operate as designed. With the inability to experimentally confirm vulnerabilities in the laboratory with all elements compiled, the system is anticipated to function as designed. The first major limitation foreseen is with the imaging system. Thus far, the best resolution has resulted from when the YSZ microspheres were 6mm and 12mm apart. This signifies that, at this juncture, the system can confidently distinguish between three categories of pebbles: no microspheres, microspheres with 6mm spacing, and microspheres with 12mm spacing. With better ultrasound systems and varied microsphere specifications (i.e. diameters), more variability (and hence more categories or types of pebbles) can be explored.

4.2 Current Project Status

This project has been conducted during the confines of a global pandemic with prohibited travel and limited time spent on-site (either in laboratories or offices). Despite these limitations, the eventual MC&A system that was developed proved effective enough to entice private industry professionals to engage with the research team and express continued interest on the project. The research team was able to test individual components that will be ultimately assembled into a comprehensive system for monitoring fuel pebbles in future PFRs. Microsphere impregnation processes were devised and tested to best replicate the commercial fuel fabrication process based on openly available specifications. Ideally, the need to replicate pebble surfaces will subside once the team is able to engage directly with interested commercial fuel fabricators and collaborate with them to acquire surrogate pebbles for further testing in FY22. The US system was successfully tested to determine whether it is capable to scan and identify various densities of microspheres embedded within graphite material replicating the graphite shell of pebble fuel elements (ranging in curvature, graphite concentration, and material density). Optical imaging is the primary element of the overall system and has so far proven able to be effective and quick. Scanning time per

surface occurred in minutes and can be theoretically completed upon immediate extraction from the core thus reducing a pebble's ex-core time. Operationally, the team must still incorporate the US system into the pebble sorting prototype for automatically scanning pebbles within a PFR.

As of September 30, 2021, this project has resulted in the independent design and development of three components of an overall pebble accounting system. Unfortunately, the final step of assembling these components into one comprehensive system will not be achieved until early FY22. When assembled and initial data is acquired, an addendum will be issued to include with this report for the benefit of the sponsors. Per PicsNE, \$354,360 of the original \$475,000 funding has been spent. The team continues to develop the system and will endeavor to complete assigned FY21 tasks in FY22. The team also expresses its gratitude to the sponsors for continuing financial support into FY22 to facilitate engagement with private industry, continued development, and design refinements of the system. The ultimate objective of this project is to get the private industry interested in incorporating such a system as part of regular operations in PFRs to support operators in meeting their regulatory requirements for nuclear material control and accounting.

5 Conclusions

Overall, individual components of the comprehensive system have proven to function as designed. Based on design specifications agreed upon by the cadre of researchers at Argonne and TAMU, the US system, the microsphere impregnation process, and the pebble sorting system operate as designed. The team will continue to install and provide initial test results of the assembled system in early FY22. Beyond, the project aims to engage closely with private industry on securing surrogate pebble fuel, collaborating with fuel fabricators in impregnating pebbles with the inert YSZ microspheres, and refining the overall system's design to best meet the design specifications and the needs of operators in countries in which these reactors would eventually be deployed.



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