Radial Deconsolidation of Irradiated AGR-3/4 Compacts at Idaho National Laboratory

Philip Winston[1], John Stempien[1] and Grant Helmreich[2]

[1]*Idaho National Laboratory, Idaho Falls, USA*
[2]*Oak Ridge National Laboratory, Tennessee, USA*
Corresponding author: Philip Winston <Philip.winston@inl.gov>

The Idaho National Laboratory (INL) evaluated the concentration gradient of fission products released in the AGR-3/4 experiment by selective removal of annular volumes of the fuel compacts. Purpose-built designed-to-fail particles aligned along the axis of an experimental compact released fission products whose concentration was determined by progressive radial electrochemical dissolution of the binding matrix material. Precise dimensional measurement was achieved by video imaging of the eroded compact at each of several steps. The video measurement of the compact diameter eliminates the need to attempt manual caliper measurements that may lack precision and may damage the sample surface.

AGR-3/4 is part of a series of irradiation experiments of high-temperature gas reactor TRISO particle fuel produced and tested by the Advanced Reactor Technology group of the Idaho National Laboratory. TRISO particles tested in the previous two experiments (AGR-1 and -2) performed at a level in which release of gaseous krypton and xenon fission products was undetectable during irradiation, and during post-irradiation examination, evidence of particle coating failures was no greater than 0.03 %. To investigate fission product transport and deposition in structural graphite, AGR-3/4 was designed to include 20 designed-to-fail (DTF) particles arranged at the axis of 12.3 mm OD x 12.5 mm long compacts, each containing nominally 1872 particles. The DTF particles consist of a standard 350 um diameter TRISO uranium oxycarbide kernel surrounded by 20 um layer of pyrolytic carbon, which is not sufficient to retain fission products. A photo of a typical compact and an x-ray image highlighting the DTF orientation are shown as Figure 28.

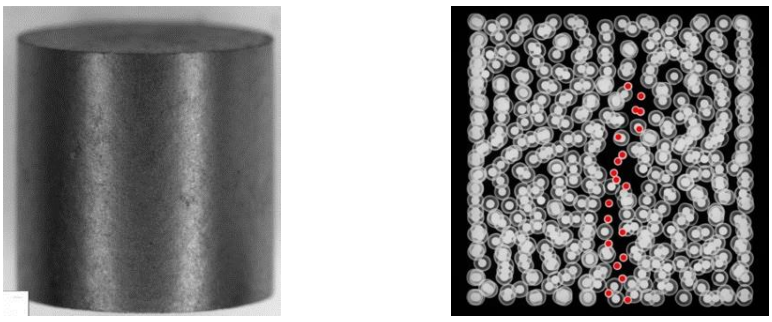

Figure 28: 12.5 mm diameter x 12.3 mm long compact (left), X-ray of compact showing DTF particles in red (right)

To investigate the gradient of fission product transport within the compacts, the normal method of complete electrolytic deconsolidation of the compact is modified and the TRISO particles are separated from the compact by rotating it against a platinum-rhodium screen anode while partially immersed in a nitric acid anode, allowing an approximately 1 mm layer of particles to be removed

from the surface of the compact during each sequential step. This work is done in an individual air atmosphere hot cell located at the INL Materials and Fuels Complex MFC-752 Analytical Laboratory. The cell floor area is approximately 3.6 m^2, is viewed through nominal 1 m^2 oil-filled leaded glass window, 60 cm thick and is serviced by 2 CRL Model L-HD manual master-slave manipulators. Because this cell already contains a shielded macroscope, an oxidation furnace and glassware for leaching of particles, the equipment for doing the deconsolidation step was designed to fit into a nominal 30 cm^3 volume.

The image processing software was developed by Grant Helmreich using Matlab® (Mathworks), and analyses individual frames from video recording of the rotating compact to measure the compact diameter at multiple points to a precision approaching 0.01 mm on a 12.3 mm diameter.

Process Description

The process of deconsolidation proceeds by electrolytically breaking down the matrix carbon, which is partially graphitized by the addition of electrical current and nitric acid. This process owes some of its heritage to Heinz Nabielek, who developed a variation of this technique to determine variations within “pebbles” from the German Pebble Bed High Temperature Gas Reactor. The description given here applies to the approach used at INL to selectively remove layers of particles for differential evaluation of the resulting deconsolidation solution. It is the result of development work at Oak Ridge National Laboratory and Idaho National Laboratory.

Mechanically, the compact is glued onto a 6 mm OD stainless-steel shaft that is driven by a 12V DC, 10 rpm gearmotor that rotates the compact against the Pt-Rh anode screen. The shaft and the compact are joined using graphite-filled epoxy (Atom Adhesives AA-CARB-61). The task of accurate joining is done by mounting the geared shaft and its manipulation handle in a machined fixture that aligns the shaft at the center axis of the compact. The 2-part epoxy is mixed outside the hot cell, and transferred into the cell in a 1 ml plastic syringe. The glue is then injected down the hollow shaft, filling the gap between the compact and the end of the shaft. After a 24-hour ambient temperature cure, the geared shaft and compact are mounted in the drive unit and positioned over a beaker of 4M HNO_3 that has a Pt-Ir cathode wire placed in it. The anode is placed in contact with the compact cylindrical surface, and lowered to the point that the compact surface just contacts the surface of the acid. The motor is actuated, and the power supply is turned on to apply a voltage of <10 VDC at a current of < 1 A. As the compact is rotated, the external surface is eroded as the matrix carbon is broken down.

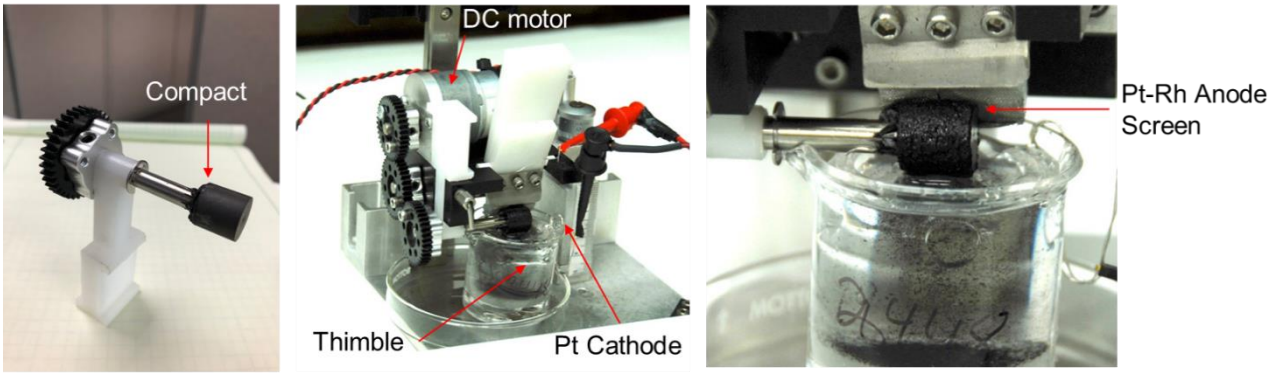

Figure 29: Compact glued on shaft with drive gear and manipulator handle (left), deconsolidation unit (center & right)

The simplified chemical intercalation process is as follows:

- Cathode (reduction):

$$HN(V)O_3 + e^- \rightarrow 2\ N(IV)O_2 + OH^*$$

- Anode (oxidation):

$$C(0) + 4\ OH^* \rightarrow C(IV)O_2 + 2\ H_2O + 4\ e^-$$

The net effect is that the matrix is broken down into carbon dioxide and nitrogen oxide as a result of the imposed current without adverse effect on the TRISO particle coatings. During initial unirradiated development of the axial deconsolidation process, it was determined that electrolytic input in excess of 10 W led to breakdown of TRISO coatings, so the process has been limited to the 10V/1A value.

The particles released from the matrix are collected in a porous-bottomed container called a “thimble”. After a prescribed period (typically 15 minutes), approximately 1 mm of the surface has been removed, and the process is stopped. The gear driven shaft and compact are raised from the acid, and a video of the rotating shaft is made through the cell window to produce image data that can be analysed to determine the precise diameter of the compact at each of the completed intervals. The beaker is replaced with one containing fresh acid and an empty thimble, and the process is repeated typically two more times, until the remaining material of the compact is the same diameter as the shaft to which it is glued. The final step is to deconsolidate the shaft diameter material by progressively electrolytically dissolving it in acid with the same cathode in a beaker, but using a Pt-Ir electrode imbedded in the glue to complete the circuit. The acid from each step is analysed by gamma ray spectrometry and strontium separation to determine fission product content. Mass spectrometry is used to detect the presence of uranium and other non-gamma emitting products released from the DTF particles.

Using the volume removed in each deconsolidated layer and data from the fission product solution analysis, it is possible to determine fission product concentrations and evaluate individual product mobility and the gradient within the compact. The compact gamma ray emitting isotope inventories were established prior to deconsolidation using precision gamma scanner located at the Hot Fuel Examination Facility.

Image Analysis

The process of video analysis of the diameter of the progressively eroded compact is used because attempting to measure by physical contact using a mechanical caliper can damage the surface of the compact, as well as break the glue joint, ending the process unless it can be precisely re-glued. The video analysis allows precise prediction of the number of particles removed in each radial deconsolidation step, as well as the volume of the matrix dissolved. Groups of 10 images from a given compact rotation selected from up to 15 minute-long videos taken at 24 fps with 1280x720 resolution were analysed to determine the compact diameter.

The video is done using a Nikon D5000 digital single-lens reflex camera with a Nikkor 80-400mm telephoto zoom lens attached. The camera is positioned at approximately 2 m from the window, with the deconsolidation equipment 30 cm from the inside of the window. The lens is used at the 400mm full zoom setting. The orientation of the camera and the general configuration of the in-cell deconsolidation equipment is shown in Figure 30.

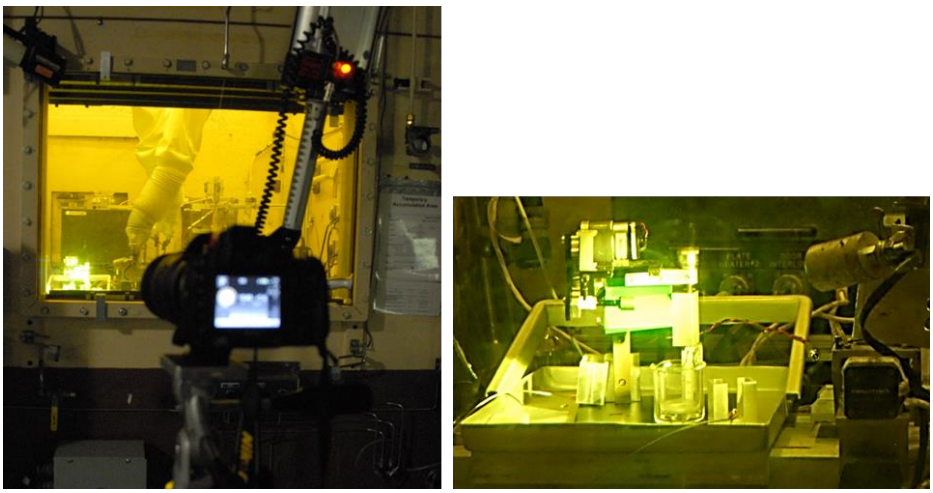

Figure 30: Camera location relative to hot cell window (left), Deconsolidation unit (right)

When the compact and shaft are raised from the acid, a small green-screen background is located behind it, allowing the image-processing software to distinguish the dark, rectangular silhouette of the compact and calculate the area and thus the compact diameter. The image analysis software finds the outline of the compact and the mounting shaft by the use of active contours, then corrects for image tilt based on the observed angle of the mounting rod. The diameter of the compact was then found by taking the average height of the masked compact in the tilt-corrected image.

All of the compacts were manually measured in the HFEF hot cell as a part of dimensional checks done on all components following disassembly of the experiment, so when each compact was mounted on its shaft, it was rotated in the drive system while recording video prior to deconsolidation. This baseline video was used to do initial lighting and alignment adjustments to ensure that the dimensions measured by image analysis were plus or minus 0.01mm.

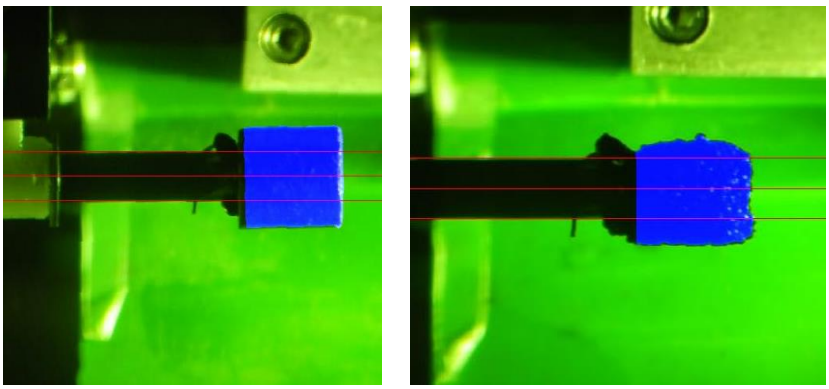

Figure 31: Compact on shaft in false color (blue) and shaft diameter (red) registration lines; Before (left), and after (right) deconsolidation

References:

Helmreich, Grant, Fred C. Montgomery, and John D. Hunn, Development of a Radial Deconsolidation Process, ORNL/TM 2015\699, December 2015.

Stempien, J. D., Radial Deconsolidation of AGR-3/4 Compacts 3-3, 12-1, and 12-3, INL/EXT 17-43182 September 2015.