ORNL/TM-2023/2969
Revision 0

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OAK RIDGE National Laboratory

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Nuclear Energy and Fuel Cycle Division

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CONTENTS

REVISION LOG

Revision	Date	Affected Pages	Revision Description
0	06/27/2023	All	Initial issue

LIST OF FIGURES

LIST OF TABLES

ABBREVIATIONS

AGR	Advanced Gas Reactor (Fuel Development and Qualification Program)
AGR-1	first Advanced Gas Reactor program irradiation experiment
AGR-2	second Advanced Gas Reactor program irradiation experiment
AGR-3/4	third and fourth Advanced Gas Reactor program irradiation experiments
ATR	Advanced Test Reactor
CRM	certified reference material
DTF	designed-to-fail
FIMA	fissions per initial metal atom
GUM	Guide to the Expression of Uncertainty in Measurement
HPIC	high-pressure ion chromatography
ID	identification
IFEL	Irradiated Fuels Examination Laboratory
IMGA	Irradiated Microsphere Gamma Analyzer
INL	Idaho National Laboratory
JMOCUP	Jim Sterbentz's MCNP-ORIGEN2 coupled utility program
LBL	leach-burn-leach
MCNP	Monte Carlo N-Particle Transport
NBLPO	New Brunswick Laboratory Program Office
ORIGEN	Oak Ridge Isotope Generation and Depletion
ORNL	Oak Ridge National Laboratory
PIE	post-irradiation examination
RMAL	Radioactive Materials Analytical Laboratory
TA_{max}	time-average maximum (temperature)
TA_{min}	time-average minimum (temperature)
TAVA	time-average, volume-average (temperature)
TRISO	tristructural-isotropic (coated particles)
UCO	uranium carbide/uranium oxide mixture (fuel kernels)
UO_2	uranium dioxide (fuel kernels)
Z	atomic number

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1. INTRODUCTION AND BACKGROUND*

The Advanced Gas Reactor (AGR) Fuel Development and Qualification Program third and fourth irradiation experiments (AGR-3/4), originally planned as separate tests, were combined in one test train for irradiation in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL). The irradiation test began on December 14, 2011, and ended on April 12, 2014 (Collin 2016). The originally planned AGR-3 and AGR-4 irradiation experiments were both focused on obtaining data on fission product transport to support the improvement of modeling. The AGR-3 experimental plan was focused on gaseous and metallic fission product release from the kernels and diffusion in the coatings during irradiation and post-irradiation safety testing. The AGR-4 experimental plan was focused on diffusivities and sorptivities in the compact matrix and reactor graphite (Petti et al. 2005). These two goals were combined in the AGR-3/4 irradiation, which consisted of 12 independently monitored capsules that each contained four AGR-3/4 compacts in a single stack surrounded by an inner ring of matrix or graphite and an outer ring of graphite. Two capsule types were used: a standard capsule and a so-called *fuel body*, in which the outer graphite ring included a floor and cap that fully enclosed the fuel (Stempien et al. 2018a). The fuel body design supported post-irradiation safety testing of the intact fuel and ring assembly to provide data on fission product transport and release from the matrix and graphite at accident temperatures (Demkowicz 2017).

The key feature of the AGR-3/4 compacts was the inclusion of 20 so-called *designed-to-fail* (DTF) fuel particles distributed along the centerline of each compact. A summary of the AGR-3/4 irradiation test compact fabrication campaign has been published by Hunn et al. (2012). The DTF fuel particles and surrounding so-called *driver fuel* particles were fabricated at Oak Ridge National Laboratory (ORNL) using a lab-scale fluidized-bed chemical vapor deposition coating system with a chamber inner diameter of 50 mm. Kernels came from a single composite (G73V-20-69303) manufactured by BWX Technologies, which was upgraded by manual sorting to remove debris and irregularly shaped kernels and renamed *LEU03*. The kernels contained a mixture of uranium carbide and uranium oxide phases (UCO) with an enrichment of 19.7%, and they were similar to the kernels used in the first AGR program irradiation experiment (AGR-1). Kernel diameter was nominally 350 μm, with a measured average value of 357.3 μm and a standard deviation of 1.6 μm (Kercher and Hunn 2006). The AGR-3/4 driver fuel particles were standard tristructural-isotropic (TRISO)-coated particles similar to the TRISO particles used in the AGR-1 experiment (Lowden 2006). Four coater batches were upgraded and combined into one composite (LEU03-09T). The TRISO particle batches and composite were thoroughly characterized and found to pass all specifications (Hunn and Lowden 2007). The DTF particles were produced in a single batch (LEU03-07DTF) after an extensive development effort to ensure that the 20 μm pyrolytic carbon coating met the specified properties for a coating expected to fail during irradiation without failing prematurely during compact heat treatment up to 1800°C (Kercher et al. 2011). Overcoating and pressing the AGR-3/4 DTF and driver fuel particles into the final cylindrical compact fuel form (nominally 0.5 in. in both diameter and length) was accomplished using similar methods developed for the fabrication of compacts made for the second AGR program irradiation experiment (AGR-2), with the exception of new techniques for overcoating and distributing the DTF particles along the centerline of the AGR-3/4 fuel compact. Overcoated DTF particles were imaged with x-ray radiography to verify that each overcoated particle contained exactly one DTF particle, and the overcoated particles were hand counted to ensure that each compact held exactly 20 DTF particles. Radiographs of 2.5 mm sections cut from the center of four AGR-3/4 compacts are shown in Figure 1-1, in which the compacts were sectioned so the radiography would show the DTF particles. The DTF particles were evenly distributed in the bottom three-quarters of the compact and within approximately 1.3 mm of the compact centerline. The partial collapse of the linear

* This introduction section is a revised version of a similar section in a previous AGR-3/4 postirradiation examination report (Hunn et al. 2020) and is included herein for contextual information and the definition of common terminology used throughout this report.

stack of overcoated DTF particles was likely due to the overcoated DTF and surrounding driver fuel particles redistributing slightly after removal of the tube used to hold the overcoated DTF in position during the addition of the overcoated driver fuel particles. Thorough characterization of AGR-3/4 fuel compact composite determined that it conformed to all specifications (Hunn, Trammell, and Montgomery 2011). In addition to 20 DTF particles, each AGR-3/4 compact held an average of 1898 driver fuel particles based on a count of the particles deconsolidated from 18 randomly-selected compacts.

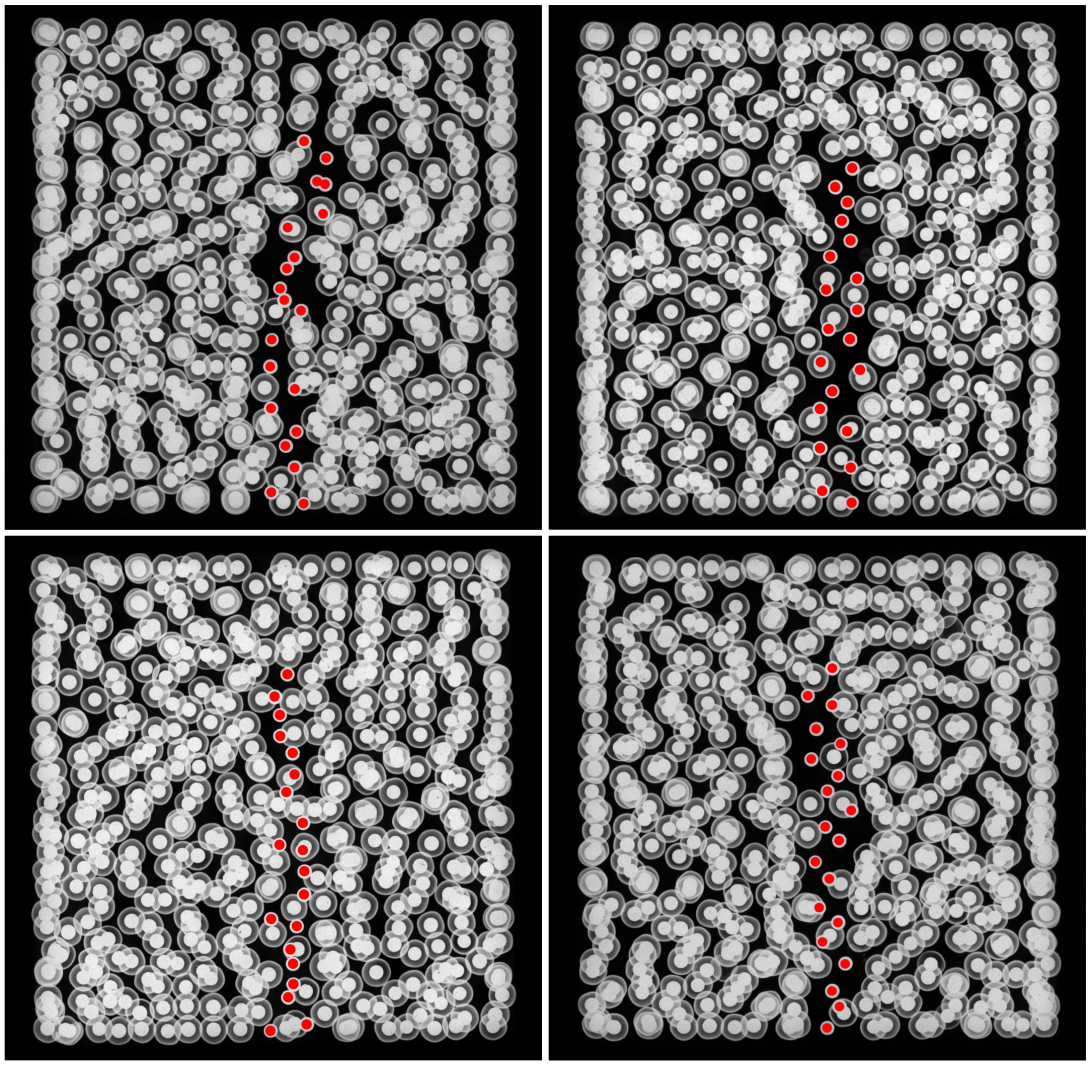

Figure 1-1. X-ray radiographs of 2.5 mm sections from four AGR-3/4 compacts, with DTF marked in red.

Post-irradiation examination (PIE) of the AGR-3/4 compacts included radial deconsolidation of individual compacts to segment the compacts into separate collections of particles and matrix debris from concentric cylindrical volumes, each of which was subjected to leach-burn-leach (LBL) analysis to quantify the presence of actinides and fission products released by the driver fuel and DTF particles. Typically, two or three cylindrical rings of driver fuel particles and matrix were removed, leaving a cylindrical core encompassing the DTF particles. The core section was then axially deconsolidated. Initial development and testing of the radial deconsolidation process were accomplished using unirradiated AGR-3/4 compacts (Helmreich, Montgomery, and Hunn 2015), and then the concept was modified for the deconsolidation of irradiated AGR-3/4 compacts in the INL hot cells (Stempien 2017). Radial deconsolidation of irradiated AGR-3/4 compacts began at ORNL in 2020. A total of eight AGR-3/4

compacts were deconsolidated at ORNL, and particles from two of these compacts: Compact 1-4 (Hunn et al. 2020) and Compact 7-4 (Helmreich et al. 2021) were used for burnup measurements. Detailed information on the radial deconsolidation process and equipment can be obtained from the referenced reports.

The AGR-3/4 compacts were irradiated in the INL ATR for 369.1 effective full power days (Collin 2016). The compacts experienced average calculated fluences of fast neutrons with energies $E_n > 0.18$ MeV of 1.19×10^{25}–5.32×10^{25} n/m^2 (Sterbentz 2015). The calculated time-average, volume-average (TAVA) compact temperatures ranged between 832°C and 1376°C (Hawkes 2016). Daily depletion calculations were performed using the Monte Carlo N-Particle Transport (MCNP) code (x-5 Monte Carlo Team 2003) and the Oak Ridge Isotope Generation and Depletion (ORIGIN2) code (Croff 1983; Ludwig and Croff 2002) via Jim Sterbentz's MCNP-ORIGEN2 coupled utility program (JMOCUP) and related software extraction modules. Description of JMOCUP and documentation of software validation are included in ECAR-958 (Sterbentz 2013). Compact average burnups based on the daily depletion calculations ranged from 4.85% to 15.27% fissions per initial metal atom (FIMA).

As reported herein, average burnup measurements have been completed on random samples of particles deconsolidated from two AGR-3/4 compacts for comparison with the average burnup values that were estimated via physics depletion calculations. Table 1-1 lists these two compacts with information on the estimated average burnup in percent FIMA, the fast neutron fluence, and the compact temperatures during irradiation in terms of the TAVA, time-average minimum (TA_{min}), and time-average maximum (TA_{max}). Both compacts were irradiated in standard (i.e., not fuel body) capsules. AGR-3/4 Compact 1-4 was from the capsule located at the bottom of the test train and experienced burnup and temperatures near the lower end of the range for compacts in the AGR-3/4 irradiation test. AGR-3/4 Compact 7-4 experienced irradiation burnup and temperatures near the upper end of the range (Stempien et al. 2018b).

Table 1-1. Irradiation and safety test parameters for AGR-3/4 compacts analyzed in this study.

Compact ID [a]	Fabrication ID number [b]	Estimated burnup [c] (% FIMA)	Fast fluence [c] (n/m^2)	Temperature [d] (°C)		
				TAVA	TA_{min}	TA_{max}
AGR-3/4 Compact 1-4	Z012	6.85	2.10×10^{25}	929	866	972
AGR-3/4 Compact 7-4	Z111	14.90	5.24×10^{25}	1319	1206	1397

[a] The compact identification (ID) denotes the compact's location in the irradiation test train: *capsule-level* (Collin 2016).
[b] Each compact in the fabrication lot (LEU03-10T-OP2/LEU03-07DTF-OP1)-Z had a unique compact ID number from 001 to 175, and physical properties data are available and referenced by compact ID number (Hunn, Trammell, and Montgomery, 2011).
[c] Compact average burnups and fast neutron fluences ($E_n > 0.18$ MeV) as reported in INL/ECAR 2753 (Sterbentz 2015).
[d] Compact TAVA, TA_{min}, and TA_{max} temperatures are based on thermal calculations (Hawkes 2016).

2. EXPERIMENTAL PROCEDURE*

After irradiation, radial deconsolidation was performed using the methods described in detail in Hunn et al. (2020); and LBL and gamma scanning of individual particles were performed using the same methods that were developed for destructive PIE of as-irradiated AGR-1 compacts (PIE) described in detail in Hunn et al. (2013). The AGR-3/4 PIE included the following steps that generated the particle archives from which samples were riffled for burnup analysis. Each compact was electrolytically deconsolidated to free the TRISO particles from the surrounding matrix, and the particles and matrix debris were leached in hot HNO_3. After leaching, the particles were separated from the matrix debris with a sieve and were surveyed with the ORNL Irradiated Microsphere Gamma Analyzer (IMGA). If the IMGA survey identified any particles with low Cs or Ce inventory, they were selectively removed from the compact population for further analysis. The other particles were recomposited and riffled with a chute splitter to randomly separate out approximately 10% as a TRISO particle archive. The remaining approximate 90% of the compact population was subjected to particle burn-leach analysis (the matrix debris was subjected to a separate matrix burn-leach analysis). The particle burn-leach analysis included heating the particles in air at 750°C, which removed the outer pyrolytic carbon layer, as well as any other exposed carbon. After the burn, particles were leached again in hot HNO_3. After the final acid leach, particles were rinsed with water, dried, and collected as a particle burn-leach archive.

Particles were riffled from the particle burn-leach archive with a chute splitter to randomly separate out particles for burnup analysis. For each compact subjected to burnup analysis, the fraction of particles that were selectively removed based on the IMGA survey was less than 0.5% of the compact population. Therefore, the removal of these particles did not significantly affect the random sampling of the particles used for burnup analysis. Burnup analysis has been completed on a 198-particle sample from AGR-3/4 Compact 1-4, shown in Figure 2-1, and a 200-particle sample from Compact 7-4, shown in Figure 2-2.

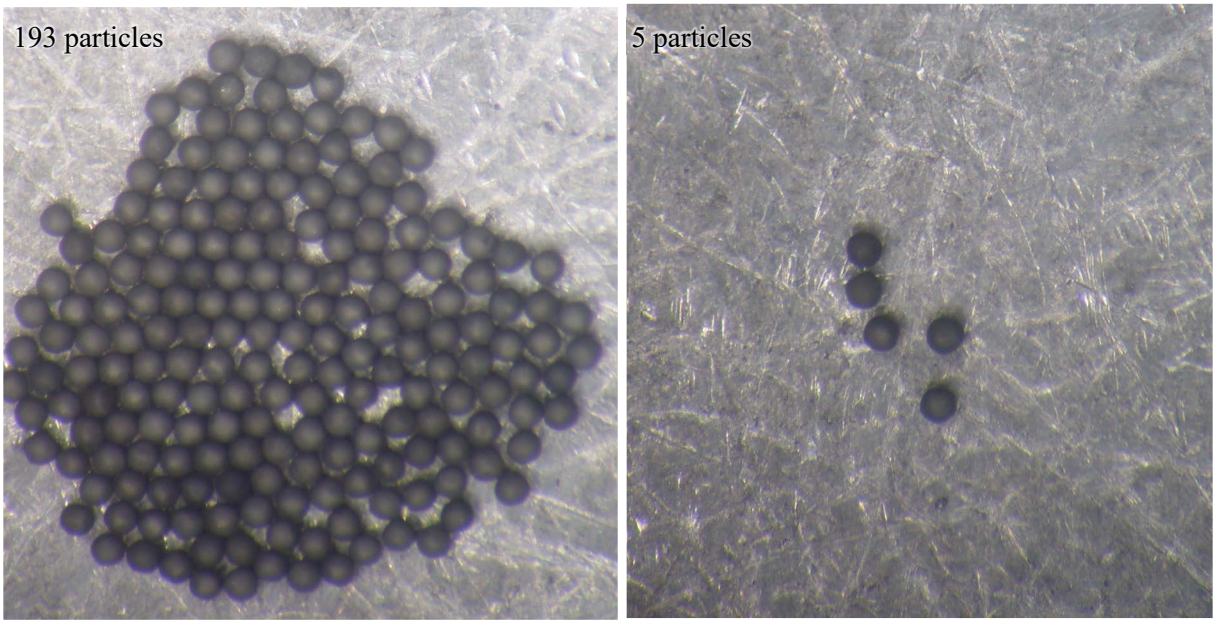

Figure 2-1. Images used to count particle samples from AGR-3/4 Compact 1-4 (arbitrary scaling).

* This experimental procedure section is a revised version of a similar section in a previous AGR-2 PIE report (Hunn et al. 2019) and is included in this report for contextual information and the definition of common terminology used throughout this report.

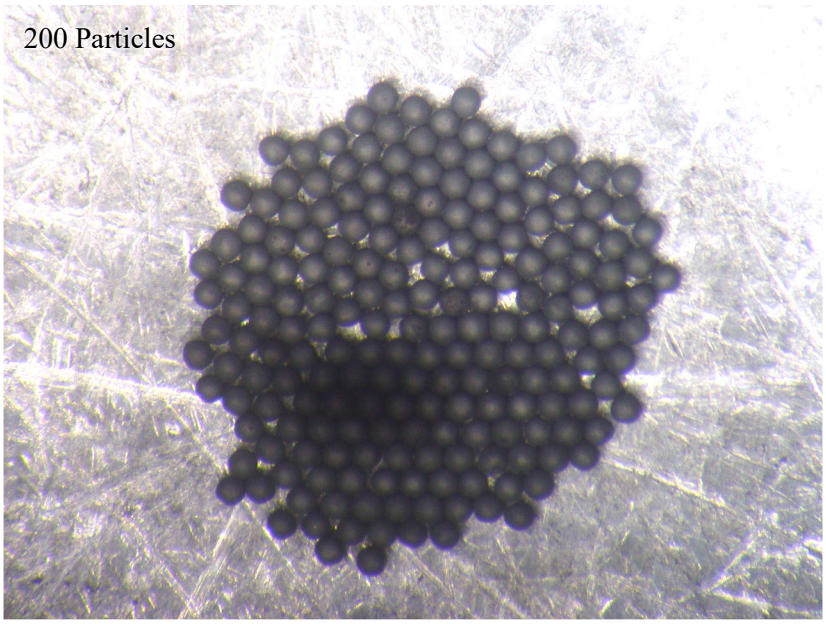

Figure 2-2. Image used to count particle samples from AGR-3/4 Compact 7-4 (arbitrary scaling).

Details of the equipment and procedures used in the Irradiated Fuels Examination Laboratory (IFEL) to produce solutions for burnup analysis are available in Section 2 of ORNL report number ORNL/TM-2018/931 (Montgomery et al. 2018) and are described only briefly herein. Each particle sample was sealed in a low-density polyethylene bottle and pulverized in a Spex SamplePrep 8000M mill to expose the kernels. After pulverization was complete, the sealed polyethylene bottle was placed inside a quartz Soxhlet thimble, and the loaded thimble was placed into a quartz flask. The bottle and its contents were burned in air by slowly ramping from room temperature to 750°C and holding at 750°C for 8 h. This burn process reduced the bottle and carbon in the coating debris to residual ash and converted most of the kernel material to a more acid-soluble oxide form. Dissolution of the oxidized residue was accomplished with HNO_3 and a Soxhlet extractor. After weighing the solutions from the first and second extractions, 30–50 mL aliquots were removed for transfer from the IFEL to the Radioactive Materials Analytical Laboratory (RMAL) for chemical analysis to determine the amount of each of the isotopes of U, Pu, and Nd listed in Table 2-1. Along with the isotopic analysis, total assays of U, Pu, and Nd were made using a modified, low-level Davies–Gray titration for U and isotope dilution mass spectrometry techniques for Pu and Nd.

Table 2-1. Isotopes analyzed relevant to ASTM E321-96 (2012)

Element	Analyzed isotopes
U	233, 234, 235, 236, 238
Pu	238, 239, 240, 241, 242
Nd	142, 143, 144, 145, 146, 148, 150

The analyses performed in the RMAL are described in Section 3 of ORNL report number ORNL/TM-2018/931 (Montgomery et al. 2018). The high-pressure ion chromatography (HPIC) elution profile used to isolate Nd in the AGR-2 Compact 2-2-2 and Compact 6-4-2 analyses (Montgomery et al. 2018) was changed for the analyses of AGR-2 Compacts 3-3-1 and 3-3-2 (Hunn et al. 2019), and this revised profile (Table 2-2) was also used for the AGR-3/4 Compact 1-4 and Compact 7-4 analyses. An increase in the diglycolic acid concentration accelerated the Nd elution, which occurred at approximately 600 s after the start of the HPIC elution process using 0.1 M diglycolic acid vs. approximately 750 s after the start of the

process using 0.006 M diglycolic acid (Montgomery et al. 2018, Figure 8). An HCl wash using Fisher Scientific Optima high-purity 2 M HCl (<1 parts per trillion Nd) and a final water rinse were added to the end of the elution profile to aid in the removal of excess U left in the separation column. This modification did not affect the elution of Nd, which was complete before the HCl rinse segment began.

Table 2-2. Elution profile script for the elemental separation of Nd from all isobaric interferences

Segment type	Segment duration (s)	Segment end time (s)	Deionized water (%)	2 M hydrochloric acid (%)	0.1 M diglycolic acid (%)	0.15 M oxalic acid (%)
Start	0	0	100	0	0	0
Ramp	300	300	40	0	0	60
Hold	240	540	40	0	0	60
Ramp	0	540	20	0	0	80
Ramp	540	1080	51	0	23	26
Ramp	120	1200	0	0	100	0
Hold	300	1500	0	0	100	0
Ramp	0	1500	0	100	0	0
Hold	120	1620	0	100	0	0
Ramp	120	1740	100	0	0	0
Hold	60	1800	100	0	0	0

Note: The listed eluent fractions are the target fractions at the end of each segment. Changes in eluent fractions during a ramp segment were made at a constant rate so that the fractions would reach the target fractions at the end of the segment. Eluent fractions were constant during each hold segment.
Note: Total flow rate for each step was 1 mL/min.

Davies–Gray titration was performed with slight modifications to ASTM C1267-17, *Standard Test Method for Uranium by Iron (II) Reduction in Phosphoric Acid Followed by Chromium (VI) Titration in the Presence of Vanadium*. The standard procedure was developed using aliquots containing 20–50 mg U. Because of restrictions on the radiological dose rates at the RMAL, the standard Davies–Gray titration procedure was modified slightly to give accurate U concentrations on aliquots containing only 5–10 mg U. To preserve the accuracy and precision of weighing titrant quantities, the standard titrant concentration was diluted threefold. The dichromate titrant was standardized at a 5–10 mg concentration using two independently prepared solutions of New Brunswick Laboratory Program Office (NBLPO) certified reference material (CRM) 112-A as control samples. Each of the two independently prepared control samples was titrated four times at U quantities that bracketed the quantities in the TRISO fuel. During analysis, a third preparation of NBLPO CRM 112-A control standard was titrated as a quality control. Because the samples may contain appreciable amounts of Si, which could occlude U during titration, researchers observed that the sample was completely dissolved after fuming and before titrations. No Si precipitates were present. This procedure was modified, and titrations were performed in triplicate, to provide uncertainties in the U analysis of <0.5% by using verified Guide to the Expression of Uncertainty in Measurement (GUM) uncertainty calculations.

3. DETERMINATION OF BURNUP*

In ORNL report number ORNL/TM-2018/931, Montgomery et al. (2018) described a method based on ASTM E321-96 (2012) that was used to calculate average burnup for AGR-2 Compact 2-2-2 and Compact 6-4-2. A modification of this method was later applied to further AGR-2 compacts in ORNL report number ORNL/TM-2019/1315 by Hunn et al. (2019). Equation 1 is the basic equation in ASTM E321-96 (2012) used to determine the burnup in percent FIMA, F_T, where F' is the number of atoms in the fuel that underwent fission, and U' and Pu' are the atoms of U and Pu, respectively, remaining after irradiation.

$$F_T = [F'/(U' + Pu' + F')] \times 100. \quad (1)$$

The ASTM E321-96 (2012) method relies on the measurement of ^{148}Nd to determine the number of fissioned atoms via Equation 2, where $FY_E(^{148}\text{Nd})$ is the effective cumulative fission yield of ^{148}Nd.

$$F' = \frac{\text{cumulative } ^{148}\text{Nd atoms from fission}}{FY_E(^{148}\text{Nd})} \quad (2)$$

3.1 CALCULATION OF EFFECTIVE CUMULATIVE FISSION YIELD

In the AGR–3/4 irradiation, the dominant contributors to fission production of ^{Z}Nd for an atomic number (Z) of 143, 144, 145, 146, 148, and 150 were three thermal neutron fission processes ($^{235}U_{th}$, $^{239}Pu_{th}$, and $^{241}Pu_{th}$) and one fast neutron fission process ($^{238}U_{fast}$). The value of $FY_E(^{Z}\text{Nd})$ can be estimated as the weighted sum of the cumulative fission yields, $FY(^{Z}\text{Nd})$, from these four dominant fission processes. Table 3-1 and Table 3-2 show the values used to estimate $FY_E(^{148}\text{Nd})$ in AGR-3/4 Compact 1-4 and Compact 7-4, respectively. Ignoring the other fission processes introduced a systematic error in $FY_E(^{148}\text{Nd})$ of less than 0.0006%. The estimated fraction each fission process contributed to the total number of fissions was based on the fraction each related heavy metal isotope contributed to the total number of fissions.

Table 3-1. Calculation of effective cumulative fission yield for ^{148}Nd in AGR-3/4 Compact 1-4

Fission process	$FY(^{148}\text{Nd})$ [a] (%)	Weighting factor [b] (%)	$FY_E(^{148}\text{Nd})$ contribution (%)	Total $FY_E(^{148}\text{Nd})$ (%)
$^{235}U_{th}$	1.674 ± 0.006	91.374 ± 0.914	1.5292 ± 0.0162	
$^{238}U_{fast}$	2.113 ± 0.015	0.516 ± 0.005	0.0109 ± 0.0001	
$^{239}Pu_{th}$	1.642 ± 0.008	7.792 ± 0.078	0.1280 ± 0.0014	1.674 ± 0.016
$^{241}Pu_{th}$	1.932 ± 0.014	0.288 ± 0.003	0.0056 ± 0.0001	
Other	<2.000	0.0295 ± 0.0003	<0.0006	

[a] Cumulative fission yields from the Evaluated Nuclear Data File ENDF/B-VII.1 (Chadwick et al. 2011) were downloaded from www.nndc.bnl.gov.
[b] The percentage each heavy metal isotope contributed to the total number of fissions was based on data extracted from the AGR-3/4 daily depletion simulation (Sterbentz 2015). An uncertainty of 1% was assumed for the weighting factors.

* This results section is a revised version of a similar section in a previous AGR-2 PIE report (Hunn et al. 2019) and is updated in this report with values for the new compacts that were analyzed.

Table 3-2. Calculation of effective cumulative fission yield for ^{148}Nd in AGR-3/4 Compact 7-4

Fission process	$FY(^{148}Nd)$ [a] (%)	Weighting factor [b] (%)	$FY_E(^{148}Nd)$ contribution (%)	Total $FY_E(^{148}Nd)$ (%)
$^{235}U_{th}$	1.674 ± 0.006	82.032 ± 0.820	1.3728 ± 0.0145	
$^{238}U_{fast}$	2.113 ± 0.015	0.579 ± 0.006	0.0122 ± 0.0001	
$^{239}Pu_{th}$	1.642 ± 0.008	15.003 ± 0.150	0.2464 ± 0.0028	1.676 ± 0.015
$^{241}Pu_{th}$	1.932 ± 0.014	2.308 ± 0.023	0.0446 ± 0.0005	
Other	<2.000	0.078 ± 0.0003	<0.0016	

[a] Cumulative fission yields from the Evaluated Nuclear Data File ENDF/B-VII.1 (Chadwick et al. 2011) were downloaded from www.nndc.bnl.gov.
[b] The percentage each heavy metal isotope contributed to the total number of fissions was based on data extracted from the AGR-3/4 daily depletion simulation (Sterbentz 2015). An uncertainty of 1% was assumed for the weighting factors.

Table 3-3 is a summary of the estimated effective cumulative fission yields for all the relevant stable and long-lived Nd isotopes that were measured for use in the burnup calculations. These were calculated in the same manner as described for the $FY_E(^{148}Nd)$ values presented in Table 3-1 and Table 3-2.

Table 3-3. Summary of effective cumulative fission yields for the stable and long-lived Nd isotopes

Isotope	AGR-3/4 Compact 1-4 (%)	AGR-3/4 Compact 7-4 (%)
$FY_E(^{143}Nd)$	5.823 ± 0.058	5.680 ± 0.052
$FY_E(^{144}Nd)$	5.352 ± 0.053	5.196 ± 0.048
$FY_E(^{145}Nd)$	3.856 ± 0.038	3.772 ± 0.034
$FY_E(^{146}Nd)$	2.956 ± 0.029	2.911 ± 0.026
$FY_E(^{148}Nd)$	1.674 ± 0.016	1.676 ± 0.015
$FY_E(^{150}Nd)$	0.682 ± 0.007	0.716 ± 0.006

3.2 CORRECTION FOR (n,γ) NEUTRON CAPTURE REACTIONS AND NATURAL CONTAMINATION

Two corrections are recommended in ASTM E321-96 (2012) to determine the number of ^{148}Nd atoms from fission used in Equation 2 from the number of ^{148}Nd atoms measured in the sample. One correction is to account for the presence of natural Nd based on the measured value of ^{142}Nd, and the other correction is to account for the production of ^{148}Nd by the $^{147}Nd(n,\gamma)^{148}Nd$ neutron capture reaction. These corrections were applied in ORNL report number ORNL/TM-2018/931 to determine the number of ^{148}Nd atoms from fission in the particle samples from AGR-2 Compacts 2-2-2 and 6-4-2 (Montgomery et al. 2018). Because the table of factors to correct for the $^{147}Nd(n,\gamma)^{148}Nd$ neutron capture reaction based on the neutron flux and fluence provided in ASTM E321-96 (2012) did not cover the irradiation conditions for AGR-2 Compacts 2-2-2 and 6-4-2, a $^{147}Nd(n,\gamma)^{148}Nd$ correction factor was estimated from Figure 3 in a paper by Suyama and Mochizuki (2005). It was recommended by Montgomery et al. in ORNL report number ORNL/TM-2018/931 that a more direct estimation of the neutron capture correction factors could be obtained by extracting information on the fraction of each isotope that was formed and lost during the AGR-2 irradiation test from the daily depletion simulation. It was also recommended that data for the other Nd isotopes should be considered for burnup determination to take advantage of the availability and accuracy of these measured values obtained as a result of the online, direct-injection, isotope-dilution, high-pressure ion chromatography–inductively coupled plasma mass spectrometry technique used to measure ^{148}Nd. These recommendations were applied in ORNL/TM-2019/1315 (Hunn et al. 2019) to recalculate burnup in AGR-2 Compacts 2-2-2 and 6-4-2, as well as complete a new series of analyses on

AGR-2 Compacts 3-3-1 and 3-3-2. The estimation of the neutron capture correction factors from the daily depletion simulations and expansion of the burnup analysis to include other measured Nd isotopes was effective for AGR-2 samples, and the same approach has been used herein to determine average burnup for AGR-3/4 Compacts 1-4 and 7-4. Table 3-4 lists the neutron capture correction factors that were applied to the measured concentration of each stable and long-lived Nd isotope ^{Z}Nd to account for the $^{Z-1}$Nd(n,γ)^{Z}Nd and ^{Z}Nd(n,γ)$^{Z+1}$Nd reactions. These factors were extracted from the AGR-3/4 daily depletion simulation data (Sterbentz 2015).

Table 3-4. Correction factors for (n,γ) neutron capture reactions

Isotope	AGR-3/4 Compact 1-4	AGR-3/4 Compact 7/4
^{142}Nd	1.0042 ± 0.0100	1.0126 ± 0.0101
^{143}Nd	1.1192 ± 0.0112	1.4030 ± 0.0140
^{144}Nd	0.8259 ± 0.0083	0.6483 ± 0.0065
^{145}Nd	1.0357 ± 0.0104	1.1066 ± 0.0111
^{146}Nd	0.9625 ± 0.0096	0.9013 ± 0.0090
^{148}Nd	0.9883 ± 0.0099	0.9695 ± 0.0097
^{150}Nd	1.0019 ± 0.0100	1.0051 ± 0.0101

Note: An uncertainty of 1% was assumed for the correction factors.

It was determined during the second AGR-2 burnup analyses reported in ORNL/TM-2019/1315 that a correction for natural contamination based on the measured ^{142}Nd should not be applied because the ASTM E321-96 (2012) assumption that the measured ^{142}Nd is representative of the natural contamination in the sample is not accurate for the fuel and irradiation conditions used in the AGR irradiation tests. Table 3-5 shows the amount of ^{142}Nd determined by the AGR-3/4 daily depletion calculations (Sterbentz 2015) compared with the average measured ^{142}Nd in the samples from each compact. The calculated amount of ^{142}Nd was 91% of the measured value in the Compact 1-4 sample and 88% of the measured value in the Compact 7-4 sample. Therefore, using the entire measured values for ^{142}Nd to subtract possible contributions from natural contamination is not warranted for these compacts. Further work would be required to adjust for the amount of ^{142}Nd generated by irradiation before applying a correction for natural contamination based on the ^{142}Nd in the irradiated fuel.

Table 3-5. Average calculated moles of ^{142}Nd compared with measured moles of ^{142}Nd

Value	AGR-3/4 Compact 1-4	AGR-3/4 Compact 7-4
JMOCUP calculated moles ^{142}Nd per compact	3.65×10^{-8}	2.20×10^{-7}
Measured number of particles per sample	198	200
Average number of particles per compact	1,918	1,918
JMOCUP calculated moles ^{142}Nd per sample [a]	3.77×10^{-9}	2.29×10^{-8}
Average measured moles per sample	4.15×10^{-9}	2.60×10^{-8}
Ratio of measured/calculated ^{142}Nd	0.907	0.881

[a] The JMOCUP calculated values for the moles ^{142}Nd per sample were estimated from the JMOCUP moles ^{142}Nd per compact multiplied by the number of particles per sample and divided by the average number of particles per compact.

3.3 CALCULATION OF BURNUP

Table 3-6 shows the burnup values determined using the as-measured number of atoms of ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, and ^{150}Nd versus using the number of atoms corrected for the generation and consumption of ^{Z}Nd via the neutron capture reactions $^{Z-1}$Nd(n,γ)^{Z}Nd and ^{Z}Nd(n,γ)$^{Z+1}$Nd. The impact of applying the neutron capture corrections is illustrated by the convergence of the burnup values calculated for each Nd isotope. With the exception of ^{144}Nd for reasons discussed below, the burnup values derived from the other Nd isotopes in Table 3-6 converge to within their propagated uncertainties. The burnup values for each compact determined using just the ^{148}Nd isotope (as prescribed in ASTM E321-96) and averaged across ^{143}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, and ^{150}Nd are given in Table 3-7, along with the calculated burnup from the AGR-3/4 daily depletion calculations obtained using JMOCUP. The averaged burnup values were nearly identical to those obtained from ^{148}Nd alone; however, the additional statistical data reduced the stochastic uncertainty by a factor of two. Comparison of the measured burnups with those calculated by Sterbentz (2015) showed good agreement.

Table 3-6. Burnup based on fission production of ^{Z}Nd in Compact 1-4 and Compact 7-4

^{Z}Nd	Correction	AGR-3/4 Compact 1-4 (% FIMA)	AGR-3/4 Compact 7-4 (% FIMA)
^{143}Nd	none (as-measured)	6.15 ± 0.07	10.91 ± 0.12
	(n,γ) reactions	6.83 ± 0.08	14.66 ± 0.18
^{144}Nd	none (as-measured)	7.54 ± 0.08	18.15 ± 0.20
	(n,γ) reactions	6.31 ± 0.08	12.57 ± 0.15
^{145}Nd	none (as-measured)	6.57 ± 0.07	13.54 ± 0.15
	(n,γ) reactions	6.79 ± 0.08	14.77 ± 0.18
^{146}Nd	none (as-measured)	7.13 ± 0.08	16.14 ± 0.17
	(n,γ) reactions	6.88 ± 0.09	14.79 ± 0.18
^{148}Nd	none (as-measured)	6.90 ± 0.08	15.14 ± 0.16
	(n,γ) reactions	6.83 ± 0.08	14.75 ± 0.18
^{150}Nd	none (as-measured)	6.73 ± 0.10	14.71 ± 0.22
	(n,γ) reactions	6.74 ± 0.11	14.77 ± 0.23

Table 3-7. Burnup results after correction for neutron capture reactions compared with JMOCUP daily depletion calculations

AGR-3/4 Compact	Data Included	Measured from Nd (% FIMA)	JMOCUP [a] (% FIMA)	Difference (%)
Compact 1-4	^{148}Nd	6.83 ± 0.08	6.85	-0.3
	^{143}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd	6.81 ± 0.04		-0.6
Compact 7-4	^{148}Nd	14.75 ± 0.18	14.90	-1.0
	^{143}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd	14.75 ± 0.09		-1.0

The deviation in the burnup values derived from the measured ^{144}Nd after application of the neutron capture correction factors in Table 3-4 versus the values derived from the other isotopes of Nd reported in Table 3-6 has also been noted in similar calculations performed for AGR-2 compacts (Hunn 2019). This consistently low result for ^{144}Nd is linked to the relatively long half-life of ^{144}Ce in the beta decay chain leading to ^{144}Nd: ^{144}Ba→^{144}La→^{144}Ce→^{144}Pr→^{144}Nd. While most of the isotopes in this chain have relatively short half-lives, ^{144}Ce has a half-life of 284.9 days. Because a significant fraction of the ^{144}Ce

decay was incomplete at the time of measurement, the measured amount of ^{144}Nd does not represent the cumulative ^{144}Nd from fission, which is needed to apply Equation 2 for the determination of the total number of atoms in the fuel that underwent fission (F'). Therefore, further corrections must be applied to the measured amount of ^{144}Nd before it can be used to derive burnup. Corrections based on the residual inventory of ^{144}Ce at the time of measurement are currently being explored to account for the ^{144}Nd inventory produced via beta decay of ^{144}Ce between the end of irradiation and the time of measurement as well as the amount of remaining ^{144}Ce. With this information, a better value for the cumulative ^{144}Nd from fission can be obtained by summing the (n,γ)-corrected amount of ^{144}Nd at the end of irradiation and the ^{144}Ce at the end of irradiation.

4. CONCLUSIONS

Burnup measurements have been completed on samples from AGR-3/4 Compacts 1-4 and 7-4 using methods previously developed for AGR-2. These methods were based on ASTM E321-96 (2012), which uses ^{148}Nd as a burnup indicator with subtractions for naturally occurring Nd based on the measured ^{142}Nd and corrections for additional ^{148}Nd production from neutron capture reactions. However, as was previously seen in burnup measurements for AGR-2, the calculated ^{142}Nd inventory generated by the irradiation represented a significant fraction of the measured inventory in both AGR-3/4 samples. Therefore, the measured ^{142}Nd inventory was not used to correct for natural Nd contributions in the compacts that were analyzed in this study, and no alternate natural contamination correction was applied. The error introduced by neglecting this correction was likely insignificant in comparison with the uncertainty of the analysis. In contrast, the application of correction factors to account for the generation and depletion of ^{Z}Nd via neutron capture reactions $^{Z-1}$Nd(n,γ)^{Z}Nd and ^{Z}Nd(n,γ)$^{Z+1}$Nd provided a significant adjustment to most of the measured values, especially for the higher burnup Compact 7-4.

As was done for the AGR-2 compacts (Hunn et al. 2019), multiple Nd isotopes were used for burnup determination. The online, direct-injection, isotope-dilution, high-pressure ion chromatography–inductively coupled plasma mass spectrometry technique used to measure ^{148}Nd also provided data for ^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd, and ^{150}Nd. Ultimately, five Nd isotopes (^{143}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, and ^{150}Nd) were used to determine the burnup, with the primary benefit being reduction of stochastic error. The measured burnup for AGR-3/4 Compact 1-4 of 6.81% ± 0.04% FIMA and AGR-3/4 Compact 7-4 of 14.75% ± 0.09% FIMA agreed well with the values obtained from the daily depletion calculations using the JMOCUP code (Sterbentz 2015).

5. REFERENCES

ASTM C1267-17. 2017. *Standard Test Method for Uranium by Iron (II) Reduction in Phosphoric Acid Followed by Chromium (VI) Titration in the Presence of Vanadium*. West Conshohocken: ASTM International.

ASTM E321-96. 2012. *Standard Test Method for Atom Percent Fission in Uranium and Plutonium Fuel (Neodymium-148 Method)*. West Conshohocken: ASTM International.

Chadwick, M. B., et al. 2011. "ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields and Decay Data." *Nuclear Data Sheets* 112: 2887–2996. Data downloaded from www.nndc.bnl.gov.

Collin, B. P. 2016. *AGR-3/4 Irradiation Test Final As-Run Report.* INL/EXT-15-35550, Revision 1. Idaho Falls: Idaho National Laboratory.

Croff, A. G. 1983. "ORIGEN2: A Versatile Computer Code for Calculating the Nuclide Compositions and Characteristics of Nuclear Materials." *Nucl. Tech.* 62: 335–352.

Demkowicz, P. A. 2017. *AGR-3/4 Phase 2 Post-Irradiation Examination Plan.* INL/PLN-5382, Revision 0. Idaho Falls: Idaho National Laboratory.

Hawkes, G. L. 2016. *AGR-3/4 Daily As-Run Thermal Analyses.* INL/ECAR-2807, Revision 1. Idaho Falls: Idaho National Laboratory.

Helmreich, G. W., F. C. Montgomery, and J. D. Hunn. 2015. *Development of a Radial Deconsolidation Method*. ORNL/TM-2015/699, Revision 0. Oak Ridge: Oak Ridge National Laboratory.

Helmreich, G. W., J. D. Hunn, F. C. Montgomery, and D. J. Skitt. 2021. *Radial Deconsolidation and Leach-Burn-Leach of AGR-3/4 Compacts 1-4 and 10-4*. ORNL/TM-2021/2178, Revision 0. Oak Ridge: Oak Ridge National Laboratory.

Hunn, J. D., and R. A. Lowden. 2007. *Data Compilation for AGR-3/4 Driver Coated Particle Composite LEU03-09T*. ORNL/TM-2007/019, Revision 0. Oak Ridge: Oak Ridge National Laboratory.

Hunn, J. D., M. P. Trammell, and F. C. Montgomery. 2011. *Data Compilation for AGR-3/4 Designed-to-Fail (DTF) Fuel Compact Lot (LEU03-10T-OP2/LEU03-07DTF-OP1)-Z*. ORNL/TM-2011/124, Revision 0. Oak Ridge: Oak Ridge National Laboratory.

Hunn, J. D., R. A. Lowden, J. H. Miller, B. C. Jolly, M. P. Trammell, A. K. Kercher, F. C. Montgomery, and C. M. Silva. 2012. "Fabrication and Characterization of Driver Fuel Particles, Designed-to-Fail Fuel Particles, and Fuel Compacts for the US AGR-3/4 Irradiation Test." *Proc. 6th International Topical Meeting on High Temperature Reactor Technology (HTR-2012)*. Tokyo, October 28–November 1, 2012. Also published in *Nucl. Eng. Des.* 271: 123–130.

Hunn, J. D., R. N. Morris, C. A. Baldwin, F. C. Montgomery, C. M. Silva, and T. J. Gerczak. 2013. *AGR-1 Irradiated Compact 4-4-2 PIE Report*. ORNL/TM-2013/236, Revision 0. Oak Ridge: Oak Ridge National Laboratory.

Hunn, J. D., F. C. Montgomery, T. J. Keever, B. D. Roach, and R. H. Ilgner. 2019. *Determination of Average Burnup in AGR-2 Compacts 3-3-1, 3-3-2, 2-2-2, and 6-4-2.* ORNL/TM-2019/1315, Revision 0. Oak Ridge: Oak Ridge National Laboratory.

Hunn, J. D., F. C. Montgomery, D. J. Skitt, and G. W. Helmreich. 2020. *Radial Deconsolidation and Leach-Burn-Leach of AGR-3/4 Compacts 1-4 and 10-4.* ORNL/TM-2020/1707, Revision 0. Oak Ridge: Oak Ridge National Laboratory.

Kercher, A. K., and J. D. Hunn. 2006. *Results from ORNL Characterization of Nominal 350 µm LEUCO Kernels (LEU03) from the BWXT G73V-20-69303 Composite*. ORNL/TM-2006/552, Revision 0. Oak Ridge: Oak Ridge National Laboratory.

Kercher, A. K., B. C. Jolly, F. C. Montgomery, C. M. Silva, and J. D. Hunn. 2011. *Data Compilation for AGR-3/4 Designed-to-Fail (DTF) Fuel Particle Batch LEU03-07DTF*. ORNL/TM-2011/109, Revision 0. Oak Ridge: Oak Ridge National Laboratory.

Lowden, R. A. 2006. *Fabrication of Baseline and Variant Particle Fuel for AGR-1*. ORNL/CF-2006/02, Revision 0. Oak Ridge: Oak Ridge National Laboratory.

Ludwig, S. B., and A. G. Croff. 2002. *ORIGEN2.2—Isotope Generation and Depletion Code Matrix Exponential Method*. Oak Ridge: Oak Ridge National Laboratory.

Montgomery, F. C., J. D. Hunn, T. J. Keever, B. D. Roach, R. H. Ilgner, E. K. Fenske, and J. M. Giaquinto. 2018. *Measurement of Average Burnup in TRISO-Coated Particles from AGR-2 UCO Compacts 2-2-2 and 6-4-2*. ORNL/TM-2018/931, Revision 0. Oak Ridge: Oak Ridge National Laboratory.

Petti, D. A., R. R. Hobbins, J. M. Kendall, and J. J. Saurwein. 2005. *Technical Program Plan for the Advanced Gas Reactor Fuel Development and Qualification Program*. INL/EXT-05-00465, Revision 1. Idaho Falls: Idaho National Laboratory.

Stempien, J. D. 2017. *Radial Deconsolidation and Leach-Burn-Leach of AGR-3/4 Compact 3-3, 12-1, and 12-3.* INL/EXT-17-43182, Revision 0. Idaho Falls: Idaho National Laboratory.

Stempien, J. D., P. A. Demkowicz, J. M. Harp, and P. L. Watson. 2018a. *AGR-3/4 Experiment Preliminary Mass Balance.* INL/EXT-18-46049, Revision 0. Idaho Falls: Idaho National Laboratory.

Stempien, J. D., P. A. Demkowicz, E. L. Reber, and C. L. Christensen. 2018b. "Preliminary Results from the First Round of Post-Irradiation Heating Tests of Fuel Compacts from the AGR-3/4 Irradiation." *Proc. 9th International Topical Meeting on High Temperature Reactor Technology (HTR-2018)*. Warsaw, October 8–10, 2018.

Sterbentz, J. W. 2013. *JMOCUP As-Run Daily Depletion Calculation for the AGR-1 Experiment in the ATR B-10 Position*. ECAR-958, Revision 2. Idaho Falls: Idaho National Laboratory.

Sterbentz, J. W. 2015. *JMOCUP As-Run Daily Depletion Calculation for the AGR-3/4 TRISO Particle Experiment in the ATR Northeast Flux Trap*. ECAR-2753, Revision 1. Idaho Falls: Idaho National Laboratory.

Suyama, K., and H. Mochizuki. 2005. "Effect of Neutron Induced Reactions of Neodymium-147 and 148 on Burnup Evaluation." *J. Nucl. Science and Technology* 42, no. 7: 661–669.

X-5 Monte Carlo Team. 2003. *MCNP—A General Monte Carlo N-Particle Transport Code.* Version 5, Volume I (LA-UR-03-1987) and Volume II (LA-CP-03-0245). Los Alamos: Los Alamos National Laboratory.