Benchmark Evaluation of Dounreay Prototype Fast Reactor Minor Actinide Depletion Measurements

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Abstract. Historic measurements of actinide samples in the Dounreay Prototype Fast Reactor (PFR) are currently of interest for modern nuclear data and fast reactor simulation code validation. Samples of various higher-actinide isotopes were irradiated for 492 effective full-power days and radiochemically assayed at Oak Ridge National Laboratory (ORNL) and Japan Atomic Energy Research Institute (JAERI). Limited data were available regarding the PFR irradiation; a six-group neutron spectra was available with some power history data to support a burnup depletion analysis validation study.

Under the guidance of the Organisation for Economic Co-Operation and Development Nuclear Energy Agency (OECD NEA), the International Reactor Physics Experiment Evaluation Project (IRPhEP) and Spent Fuel Isotopic Composition (SFCOMPO) Project are collaborating to recover all measurement data pertaining to these measurements, including collaboration with the United Kingdom to obtain pertinent reactor physics design and operational history data.

These activities will (we hope!) produce internationally peer-reviewed benchmark data to support validation of minor actinide cross section data and modern neutronic simulation of fast reactors and accompanying fuel cycle activities such as transportation, recycling, storage, and criticality safety.

Key Words: Benchmark, Depletion, Minor Actinide, Prototype Fast Reactor.

1. Introduction

Measurements were historically performed under a collaborative agreement between the United Kingdom (UK) and United States of America (USA) to irradiate actinide samples in the Dounreay Prototype Fast Reactor (PFR). Samples of thorium, protactinium, uranium, neptunium, plutonium, americium, and curium were prepared at Oak Ridge National Laboratory (ORNL) as encapsulated samples and placed within fuel pins of the PFR (see FIG. 1). The fuel pins were irradiated in the PFR from July 1982 to July 1988 for 492 EPFD (effective full-power days) and then returned to ORNL for destructive radiochemical characterization [1]. Some of the samples were provided to Japan Atomic Energy Research Institute (JAERI) to validate the characterization results, as well as validate calculations based upon evaluated nuclear data [2].

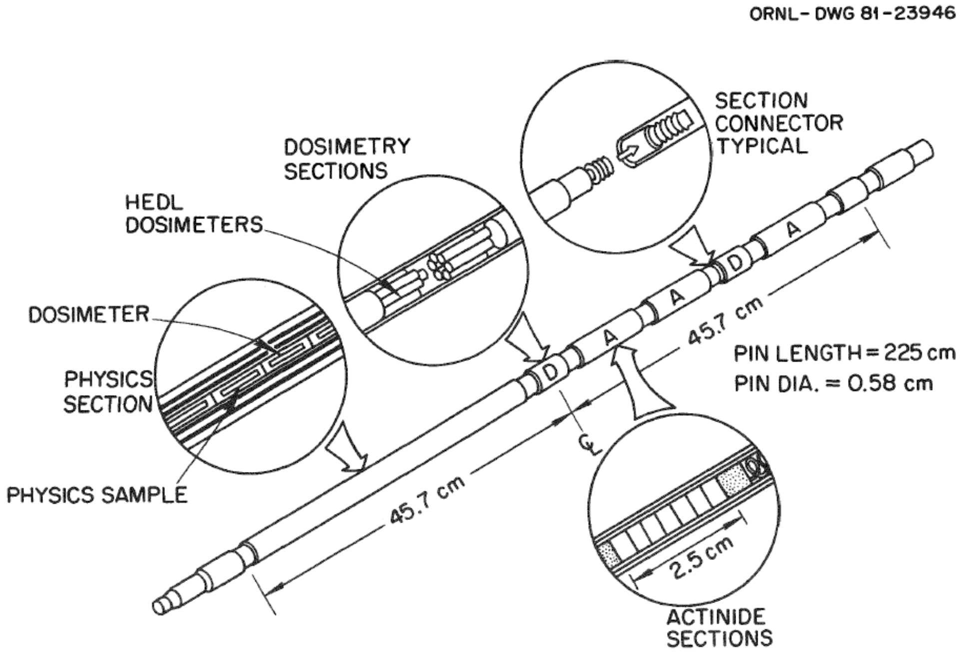

FIG. 1. Schematic of the Higher Actinides Pin Inserted in PFR for Irradiation.

These unique measurements enable development of evaluated nuclear data necessary for utilization in computational simulations of various fuel cycle activities such as reactor operations, criticality safety, storage, transportation, recycling and disposal. Some of the original interest in these measurements was to support transmutation studies of high-level radioactive waste [2] and verify the reliability of burnup calculation codes [3].

The PFR operated from 1975 to 1994 in Dounreay, along the northern coast of Scotland. It was a 630 MWt (250 MWe) power plant with PuO_2+UO_2 fuel, stainless steel cladding, and sodium coolant. Fuel pellets of 5 mm in diameter were stacked to an active core height of 0.915 m in hexagonal fuel bundles. Reactor reactivity was controlled via tantalum control rods and boron carbide shut-off rods [4]. While four actinide pins were inserted into PFR, only the fourth pin (FP-4) was irradiated to the full 492 EPFD. A basic operation history of PFR for the fourth pin sample from these tests, along with a six-group neutron energy spectra, was previously recorded [1]. A schematic of the core loading is provided in FIG. 2, with the demountable subassembly (DMSA) positions representing the 19-pin locations where the actinide sample pins could be located.

A comprehensive summary of the irradiated minor actinide sample characterization has been reported [2]. Various difficulties in performing the analytical measurements at ORNL introduced errors and lessons learned that were later incorporated into the JAERI analytical measurement techniques. Results for actinide measurements were within approximately 7 %

for both laboratories, and within approximately ±14 % for fission product measurements. Basic burnup calculations were performed and compared against the experimental data [3]. Reaction rates for fission product production, depletion, and transmutation of the actinides were calculated using fission per initial metallic atom (FIMA) values based on the production of stable fission product ^{148}Nd, a, and an effective fission product yield, to determine actinide burnup. Existing nuclear data at the time was generally found to be adequate except for large uncertainties in the fission-yield data for some of the actinides that contributed to poor estimates of FIMA due to poorly known ^{148}Nd fission-yield data. Improvement in the fission-yield data for these isotopes was recommended.

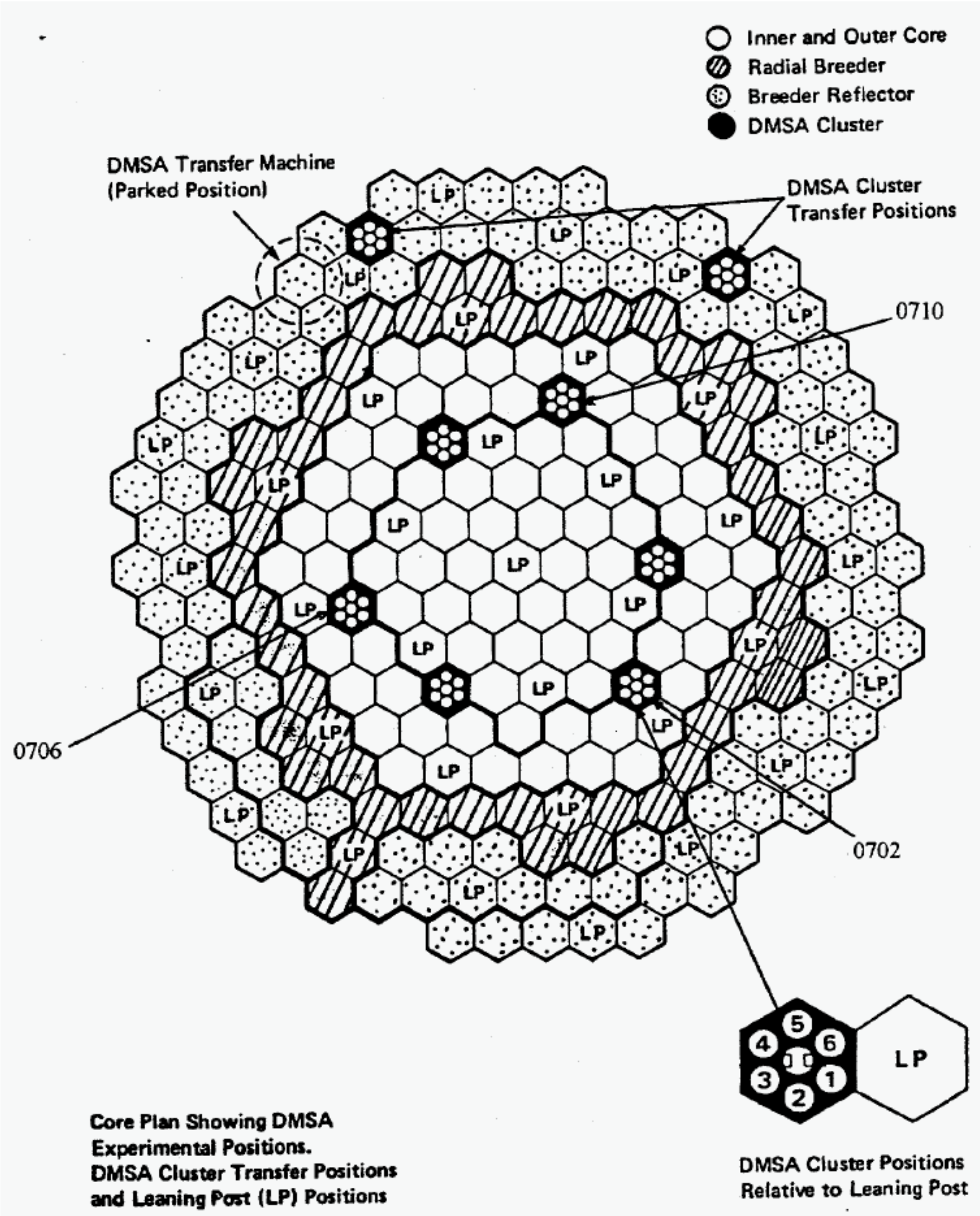

FIG. 2. Location of Demountable Subassemblies (DMSAs) in PFR [1].

The primary limitation of these previous studies was the lack of detailed PFR core and fuel design data, operational history, and neutron energy spectra data. The existing operational history and six-group spectra data was insufficient to support a comprehensive burnup analysis and validation testing of existing actinide burnup measurements. More recent studies in criticality safety have also demonstrated that existing nuclear cross section data for various actinides modelled in contemporary neutronics codes do not satisfactorily reproduce the minimum critical mass for spherical metal systems as reported in the ANS-8.15 standard [5]. Critical mass calculations of ^{241}Am and ^{243}Am under-predicted the reference values, while those for ^{244}Cm, ^{237}Np, ^{238}Pu, ^{240}Pu and ^{242}Pu were over-predicted.

There is a need for benchmark quality data to produce quality minor actinide evaluated cross section data and sustain modern computational simulations of fast reactors with their accompanying fuel-cycle activities. The application of integral benchmark data to support validation of the various components of nuclear systems data and modelling has been previously discussed [6]. The *International Handbook of Evaluated Criticality Safety Benchmark Experiments* (ICSBEP Handbook) [7] has served for many years as an extensively peer-reviewed handbook of evaluated integral benchmark data to support validation of reactor physics and nuclear criticality safety analytical methods and data, nuclear data testing, and safety analysis licensing activities. Furthermore, the benchmark experiments contained therein have enabled the development of modern-day nuclear codes and data that are built upon the experience gained in the experiment recorded within. These valuable legacy and new data support current and future nuclear programs including fuel cycle research and development, light water reactor sustainability, advanced reactor concepts, novel systems design, and global nuclear fuel assurance [6].

2. International Database Collaboration

It is desirable to continue international collaborations in development of the benchmark evaluation of these PFR burnup measurements. Under the auspices and direction of the Organisation for Economic Co-operation and Development Nuclear Energy Agency (OECD NEA) exist two database projects willing to collaborate upon this project. The Working Party on Scientific Issues of Reactor Systems (WPRS) of the OECD NEA hosts the International Reactor Physics Experiment Evaluation Project (IRPhEP), which aims to provide the nuclear community with qualified benchmark data sets collected from international reactor physics experimental data and nuclear facilities [8]. Based upon the quality and expertise employed to create the ICSBEP Handbook, the IRPhEP compiles existing experimental data into an approved standardized format, verifies the accuracy of the data to the extent possible, analyses and interprets the experiments using current state-of-the-art methodologies, and publishes the benchmark evaluation results. Benchmark evaluations are extensively peer-reviewed and, once approved, are published in the *International Handbook of Evaluated Reactor Physics Benchmark Experiments* (IRPhEP Handbook) [9]. The 2016 edition of the IRPhEP Handbook contains data from 151 experimental series representing 50 unique reactor facilities (see TABLE I) [10].

The Working Party on Nuclear Criticality Safety (WPNCS) of the OECD NEA hosts the Spent Fuel Isotopic Composition (SFCOMPO) database [11]. SFCOMPO is a relational database developed to provide experimental assay data of spent nuclear fuel for a wide selection of international reactor designs. Data included in SFCOMPO includes isotopic composition measurements, reactor operational histories and relevant design data relating to spent fuel samples. The SFCOMPO database also follows a standardised approach for the storage, retrieval and comparison of the different datasets contained therein from different post-irradiation examination (PIE) campaigns. Where possible, original experimental laboratory reports and publications are also provided. The current contents of SFCOMPO 2.0 include approximately 16,000 measurements representing 730 samples and 90 different isotopes. There are a total of 44 different reactors currently provided (see TABLE II).

Participants in the IRPhEP and SFCOMPO are actively engaged in a first-of-a-kind collaboration to develop benchmark evaluation data from the PFR minor actinide measurements. Successful retrieval and evaluation of data would support development of reactor physics benchmark data and models to be made available in the IRPhEP Handbook, and evaluated fast-spectra spent fuel isotopic data for inclusion in SFCOMPO. The integral nuclear benchmark data provided could support fast reactor design and operations and burnup calculations of actinides.

TABLE I: Available Benchmark Data in 2016 Edition of the IRPhEP Handbook* [10].

PWR (6)	**SERIES (12)**
DIMPLE	2 + 1 Draft
DUKE	1 Pending
EOLE	2
OTTOHAHN	1
SSCR	2
VENUS	2 + 3 Draft

VVER (3)	**SERIES (4)**
LR-0	3
P-Facility	1
ZR-6	1

BWR (0)	**SERIES (0)**

LMFR (9)	**SERIES (25)**
BFS-1	2
BFS-2	1
BR2	1
FFTF	1
JOYO	1
SNEAK	1
ZEBRA	3
ZPPR	11
ZPR	4

[1] Duplicate Facility

GCR (5)	**SERIES (10)**
ASTRA	1
HTR10	1
HTTR	3
PROTEUS	4
VHTRC	1

GCFR (0)	**SERIES (0)**
PROTEUS[1]	1

LWR (5)	**SERIES (25)**
CROCUS	1
DIMPLE[1]	2
IPEN(MB01)	18
KRITZ	3
TCA	3

HWR (3)	**SERIES (5)**
DCA	1
ETA	2
ZED2	2

MSR (0)	**SERIES (0)**

RBMK (1)	**SERIES (1)**
RBMK(CF)	1

Total Facilities	**Total Series**
50	151

SPACE (6)	**SERIES (12)**
ORCEF	1
SCCA	3
TOPAZ	2
UKS1M	1
ZPPR[2]	4
ZPR[2]	1

FUND (19)	**SERIES (49)**
ATR	1
BFS-1[1]	4
BFS-2[1]	1
CORAL(1)	1
FR0	3
HECTOR	2
IGR	1
KUCA	1
LAMPRE	1
MINERVE	1
NRAD	2
ORSPHERE	1
PBF	1
RA-6	1
RB	8
RHF	1
TRIGA	2
ZEBRA[1]	1
ZPR[1]	18

*PWR = Pressurized Water Reactor, VVER = Vodo-Vodynaoi Energetichesky Reactor,
BWR = Boiling Water Reactor, LMFR = Liquid Metal Fast Reactor, GCR = Gas Cooled (Thermal) Reactor,
GCFR = Gas Cooled (FAST) Reactor, LWR = Light Water Moderated Reactor,
HWR = Heavy Water Moderated Reactor, MSR = Molten Salt Reactor,
RBMK = Reaktor Bolshoy Moshchnosti Kanalniy, SPACE = Space Reactor,
and FUND = Fundamental Physics Measurements.

TABLE II: Currently Available Reactor Samples in SFCOMPO 2.0*.

AGR
Hinkley-3
Hinkley-4
Hunterston B-1

CANDU
Bruce-1
Nuclear Power Demonstration Reactor-1
Pickering A-1

MAGNOX
Bradwell-1
Hunterston A-1

BWR
Cooper-1
Dodewaard-1
Forsmark-3
Fukushima-Daiichi-3
Fukushima-Daini-1
Fukushima-Daini-2
Garigliano-1
Gundremmingen-1
Japan Power Demonstration Reactor-1
Monitcello-1
Quad Cities-1
Tsuruga-1

PWR
Beznau-1
Calvert Cliffs-1
Genkai-1
Gösgen-1
H. B. Robinson-2
Mihama-3
Neckarwestheim-2
Obrigheim-1
Ohi-1
Ohi-2
Takahama-3
Three Mile Island-1
Trino Vercellese-1
Turkey Point-3
Vandellos-2
Yankee-1

RBMK
Leningrad-1

VVER-1000
Balakovo-2
Balakovo-3
Kalinin-1
Novovoronezh-5

VVER-440
Kola-3
Novovoronezh-3
Novovoronezh-4

*AGR = Advanced Gas Cooled Reactor, CANDU = Canada Deuterium Uranium Reactor,
MAGNOX = Magnesium Alloy Graphite Moderated Gas Cooled Uranium Oxide Reactor,
BWR = Boiling Water Reactor, PWR = Pressurized Water Reactor,
RBMK = Reaktor Bolshoy Moshchnosti Kanalniy, and
VVER = Vodo-Vodynaoi Energetichesky Reactor.

3. Current Evaluation Progress

Efforts to date have primarily included the ongoing recovery of reports, drawings, and logbooks pertaining to the design and manufacture of the sample pins, as well as the preparation and subsequent radiochemical analysis of the actinide samples. ORNL prepared the actinide samples and performed the initial post-irradiation analyses with JAERI completing the follow-up comparative studies. The sample fuel pins themselves were manufactured and assembled at Hanford Engineering Development Laboratory (HEDL) prior to their irradiation in PFR. Proper characterization of the experiment design in benchmark models includes uncertainty evaluation of numerous relevant parameters such as geometric tolerances, isotopic purity, measured masses, etc. Uncertainties in the fabrication parameters and measurements need to be identified and quantified. Systematic uncertainties in measurement procedures, instrumentation calibration, and laboratory practices could similarly bias the experimental results that should be included in the benchmark parameters. Data mining for experimental details involves more than a literature search, but evaluation of existing measurement capabilities and practices during the course of this experiment such that the accuracy of the reported data are sufficiently quantified.

The OECD NEA is facilitating collaboration between Japan and the USA again with the UK to obtain detailed information regarding the irradiation program for the four fuel pins irradiated in PFR. The aim of the ongoing investigation includes identification of uncertainties relating to these measurements, estimating measurement uncertainties from fuel assay and reactor operational data, assembly of PFR reactor history, determining whether sufficient data are available to perform a suitable IRPhEP benchmark and, if necessary, identifying further work required defining an appropriate benchmark. Prior investigations into locating remaining PFR heritage data included identification of the UK National Archives, British Nuclear Fuels (BNFL), National Nuclear Laboratory (NNL) and North Highland College in Thurso as viable repositories of UK Atomic Energy Authority (UKAEA) fast reactor program reports, drawings, and/or logbooks. Considerable effort was deemed necessary to sort through archives with minimal classification and search features to establish a bibliography supporting PFR benchmark activities.

Should the efforts to investigate PFR data prove fruitful, the path forward includes initial preparation of a benchmark evaluation following the evaluation guidelines of the IRPhEP Handbook. Section 1 of an IRPhEP evaluation focuses upon the comprehensive collection of existing design and measurement data pertaining to a given reactor experiment campaign, such as the PFR minor actinide measurements. Evaluation of the data is not performed in Section 1, but collected and organized to determine further evaluation needs, identify key gaps in existing data, and facilitate evaluation of experimental uncertainties and benchmark model biases, in Section 2 and 3, respectively, of the benchmark evaluation report. Concurrently, data from these measurements will be gathered and collected into the SFCOMPO database. Successful completion of the PFR benchmark with evaluated actinide irradiation measurements would represent the first evaluation available as a reactor physics benchmark with comprehensive sample irradiation data coupled between IRPhEP and SFCOMPO. This evaluation would represent the first PFR contribution to these databases, the first fast reactor fuel sample in SFCOMPO, and successful international collaboration through the OECD NEA.

4. Conclusions

An international collaboration between Japan, the USA, and the UK has been established through the OECD NEA to evaluate the PFR minor actinide depletion measurements as benchmarks for inclusion in the IRPhEP Handbook and SFCOMPO database. These measurements proffer a unique set of measurements for the evaluation of nuclear data needed to support modern neutronic simulation of fast reactors and their accompanying fuel cycle activities. Activities to collate detailed information, including uncertainties, regarding PFR and the experiments are underway. Pending the information obtained regarding the PFR itself will determine the path forward for a successful first-of-a-kind collaboration between IRPhEP and SFCOMPO to provide evaluated benchmark data to the international community.

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6. References

[1] RAMAN, S., et al., "Dounreay PFR Irradiation History for the Joint US/UK Actinide Sample Exposures," ORNL/TM-12984, Oak Ridge National Laboratory, Oak Ridge (1995).

[2] SHINOHARA, N., et al., "Validation of Minor Actinide Cross Sections by Studying Samples Irradiated for 492 Days at the Dounreay Prototype Fast Reactor – I: Radiochemical Analysis," Nucl. Sci. Eng. **144** (2003) 115.

[3] TSUJIMOTO, K., et al., "Validation of Minor Actinide Cross Sections by Studying Samples Irradiated for 492 Days at the Dounreay Prototype Fast Reactor – II: Burnup Calculations," Nucl. Sci. Eng. **144** (2003) 129.

[4] JENSEN, S. R., ØLGAARD, P. L., Description of the Prototype Fast Reactor at Dounreay, NKS/RAK-2(95)TR-C1, Risø National Laboratory, Roskilde (1995).

[5] POPP, D., et al., "Impact of Nuclear Data Improvements on Criticality Safety Guidelines," Trans. Am. Nucl. Soc. **112** (2015) 475.

[6] PALMIOTTI, G., et al. "Applications of Integral Benchmark Data," Nucl. Sci. Eng. **178** (2014) 295.

[7] ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT NUCLEAR ENERGY AGENCY, International Handbook of Evaluated Criticality Safety Benchmark Experiments, NEA/NSC/DOC(95)/03, Paris (2016).

[8] BRIGGS, J. B., et al., "An Overview of the International Reactor Physics Experiment Evaluation Project," Nucl. Sci. Eng. **178** (2014) 269.

[9] ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT NUCLEAR ENERGY AGENCY, International Handbook of Evaluated Reactor Physics Benchmark Experiments, NEA/NSC/DOC(2006)1, Paris (2016).

[10] BESS, J. D., et al., "The 2016 Edition of the IRPhEP Handbook," Trans. Am. Nucl. Soc. **115** (2016) 1339.

[11] MICHEL-SENDIS, F., et al., "A New OECD/NEA Database of Nuclide Compositions of Spent Nuclear Fuel," Proc. PHYSOR 2014, Kyoto (2014); (CD-ROM).