Annals of Nuclear Energy 106 (2017) 221–234

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Analysis of new measurements of Calvert Cliffs spent fuel samples using SCALE 6.2 ☆

J. Hu *, J.M. Giaquinto, I.C. Gauld, G. Ilas, T.J. Keever

Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831-6172, United States

ARTICLE INFO

Article history:
Received 12 January 2017
Received in revised form 6 April 2017
Accepted 7 April 2017

Keywords:
Destructive assay, (DA)
Radiochemical analysis, (RCA)
Spent fuel
Isotopic composition
Burnup
Validation
Calvert Cliffs
SCALE

ABSTRACT

High quality experimental data for isotopic compositions in irradiated fuel are important to spent fuel applications, including nuclear safeguards, spent fuel storage, transportation, and final disposal. The importance of these data has been increasingly recognized in recent years, particularly as countries like Finland and Sweden plan to open the world's first two spent fuel geological repositories in 2020s, while other countries, including the United States, are considering extended dry fuel storage options. Destructive and nondestructive measurements of a spent fuel rod segment from a Combustion Engineering 14 × 14 fuel assembly of the Calvert Cliffs Unit 1 nuclear reactor have been recently performed at Oak Ridge National Laboratory (ORNL). These ORNL measurements included two samples selected from adjacent axial locations of a fuel rod with initial enrichment of 3.038 wt% ^{235}U, which achieved burnups close to 43.5 GWd/MTU. More than 50 different isotopes of 16 elements were measured using high precision measurement methods. Various investigations have assessed the quality of the new ORNL measurement data, including comparison to previous measurements and to calculation results. Previous measurement data for samples from the same fuel rod measured at ORNL are available from experiments performed at Pacific Northwest National Laboratory in the United States and the Khoplin Radium Institute in Russia. Detailed assembly models were developed using the newly released SCALE 6.2 code package to simulate depletion and decay of the measured fuel samples. Results from this work show that the new ORNL measurements provide a good quality radiochemical assay data set for spent fuel with relatively high burnup and long cooling time, and they can serve as good benchmark data for nuclear burnup code validation and spent fuel studies.

Published by Elsevier Ltd.

1. Introduction

High quality experimental data for isotope compositions in irradiated fuel are important to spent fuel applications, including interim and long-term storage, transportation, and final disposal. The importance of these data has been increasingly recognized as countries like Finland and Sweden plan to open the world's first two spent fuel geological repositories in the next decade, and other countries, including the United States, are considering extended dry fuel storage options. The isotope compositions in spent fuel define the fuel's characteristics, such as residual decay heat and radioactivity. Quantification of uncertainties in isotope compositions is important since these uncertainties can directly impact safety margins of a given spent fuel facility. Destructive assay (DA), also known as radiochemical assay (RCA), is the gold standard method to measure spent fuel isotope compositions; nondestructive assay (NDA) techniques only measure a limited number of observables (mainly neutron and gamma radiations). Therefore, NDA techniques cannot be directly used to quantify all isotopes important to spent fuel applications. DA is a complex procedure that includes cutting fuel samples, dissolving the fuel, dilution, sample preparation, chemical separation, and mass spectrometry. For high precision measurements, the solution must be spiked using known enriched isotopic standards. Errors in any of the measurement steps can lead to errors in measurement results that would be difficult to identify. Therefore, it is important to verify the quality of the measurement data before they can be used for safety purposes.

☆ This manuscript has been authored by UT-Battelle, LLC, under Contract No. DE-AC05000R22725 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for the United States Government purposes. The Department of Energy will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (http://energy.gov/downloads/doe-public-access-plan).

* Corresponding author.
E-mail address: huj1@ornl.gov (J. Hu).

http://dx.doi.org/10.1016/j.anucene.2017.04.005
0306-4549/Published by Elsevier Ltd.

In this work, two fuel-pellet scale spent fuel samples from the same fuel rod were used, which was extracted from a fuel assembly that had been irradiated in the Calvert Cliffs Unit 1 (CC-1) pressurized water reactor (PWR). The samples were measured using DA at Oak Ridge National Laboratory (ORNL) between 2010 and 2011. A large selection of isotopes (51) was considered in this experiment. A similar DA measurement campaign was recently performed at ORNL on 5 spent fuel samples from the Three Mile Island–1 reactor; these measurements were analyzed using SCALE 6.1.3 and documented in (Gauld et al., 2016). In addition to DA for the two CC-1 samples, gamma spectrometry was performed on the fuel rod segment from which these samples were obtained. Computer models were developed to simulate the burnup and decay of the measured spent fuel samples using the latest release of SCALE in 2016, version 6.2 (Rearden and Jessee, 2016). The new CC-1 experimental data provides an additional benchmark to validate SCALE 6.2 and compare the accuracy of this new version to version 6.1.3.

Under the Approved Testing Material (ATM) program sponsored by the US Department of Energy (DOE) Office of Civilian Radioactive Waste Management (OCRWM), studies on spent fuel were conducted in the 1980s to 1990s to investigate fuel and cladding characteristics, radioisotope inventory, and redistribution of fission products (Guenther et al., 1991). Under the ATM program, CC-1 fuel samples (ATM-104) from the same fuel rod measured at ORNL were previously measured at Pacific Northwest National Laboratory (PNNL) in the US and at Khoplin Radium Institute (KRI) in Russia in 1987 and 1995, respectively (Radulescu et al., 2010). Those measurement data served as a benchmark for validating irradiated fuel data used in criticality calculations and were used for the Organisation for Economic Co-operation and Development/Nuclear Energy Agency burnup credit criticality safety calculation benchmark (Ilas et al., 2008). Those independent measurements also provide an important benchmark to cross-check and evaluate the new ORNL measurement data. Compared to the previous measurements, the ORNL measurements included a few more isotopes (e.g., ^{244}Cm). More rigorous treatments for the lanthanides were taken in the ORNL measurements, including incorporation of isotope dilution techniques and high pressure separations for faster and more efficient separations, which resulted in cleaner element fractions. In addition, these measurements used isotope dilution for a larger suite of elements than the previous measurements.

2. Description of the measured fuel samples

The CC-1 reactor is a PWR with a generating capacity of 900 MWe operated in the US since 1975. Under the ATM -104 spent fuel program, three CC-1 assemblies were analyzed at PNNL (Guenther et al., 1991). Examinations conducted on the fuel included gamma scanning, fission gas analyses, ceramography, metallography of the cladding, electron probe microanalyses, analytical transmission electron microscopy, fuel burnup measurements, and radiochemical analyses of the fuel and cladding (Guenther et al., 1991). DA measurements were performed for nine fuel samples taken from three different fuel rods, with one rod from each of the three considered assemblies. For the work described in this paper, three samples from rod MKP109 of assembly D047 are of particular interest because the two samples measured at ORNL were also taken from this fuel rod.

Assembly D047 is a Combustion Engineering (CE) 14 × 14 design with 176 fuel rods and 5 large guide tubes for the assembly control cluster. Fig. 1 shows the schematic of the assembly design with 7 intermediate grid spacers, as well as one at the bottom and another at the top. The assembly layout illustrated in Fig. 2 highlights the location of rod MKP109. The guide tubes are much larger

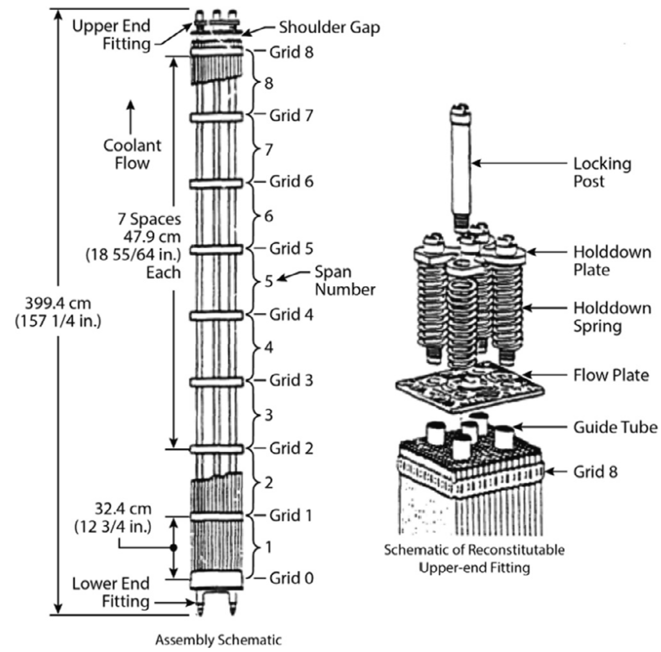

Fig. 1. Schematic of Combustion Engineering 14 × 14 fuel assembly (Ref. Guenther et al., 1991, Fig. 3.1).

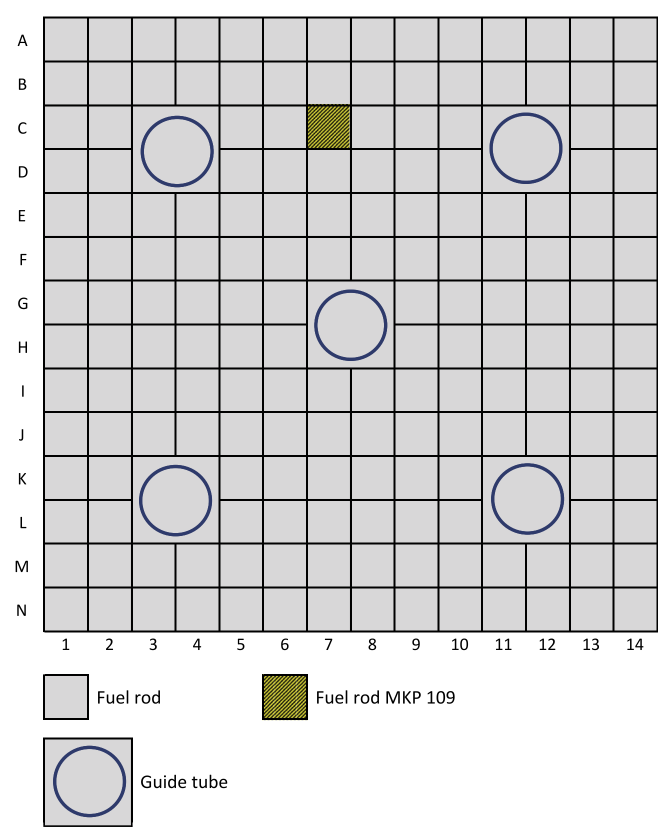

Fig. 2. Configuration of assembly D047 from CC-1 (Ref. Guenther et al., 1991, Fig. 3.3).

than those of other PWR assembly types, with each guide tube occupying the space of four fuel rods. Fig. 3 shows the fuel rod and pellet dimensions of assembly D047. The length of the measured rod is 373 cm, and the active fuel length is 347.2 cm. Table 1 summarizes the assembly design and operating data (Radulescu