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Spent fuel interrogation using delayed fast neutron spectrum at Missouri University of Science and Technology Reactor

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ABSTRACT

Interrogation of nuclear fuel and Plutonium (Pu) and Uranium (U) discrimination was performed using Missouri University of Science and Technology Reactor (MSTR) fuel by non-destructive (NDA) method. Post-irradiated delayed fast neutron spectra were obtained for two pairs of burnt and fresh fuels. Burnup and ^{239}Pu conversions were calculated based on neutron emission intensity ratios. After 100 kW high power runs, all fuel elements showed three distinct regions of neutron spectra; a distinct low energy peak followed by intermediate energy region without distinct peak but a wide hump, followed by a high energy peak with a long tail. At 10 kW low powers, intermediate energy hump and low energy peak seems to merge together while the high energy peak still remains distinct. Based on data from 10 kW power runs, the burnup values of F1 and F2 fuel elements were estimated to be 149 MWD/T and 196 MWD/T, respectively. ^{239}Pu conversion since 1992 for low enriched (19.75%) fuel elements was calculated as 0.24 g for F1 and 0.32 g for F2. Results based on high power runs of 100 kW provided comparable burnup of 217 MWD/T for F2. However the results for F1 were approximately 10 times higher perhaps due to unique burnup history and consequently high poison buildup. These experimental burnup results compare well with the reactor burnup calculation as reported to the Nuclear Regulatory Commission (NRC).

1. Introduction

The nondestructive analysis (NDA) is an important technique for monitoring spent fuel for burnup credit calculations, accountancy of special nuclear material, safeguard and ensuring adherence to proliferation control programs. NDA is a well-established technique and can utilize either fission product gamma or the delayed neutrons. The primary objective of this work is to develop a technique capable of determining Plutonium burnup and (Pu) content of the original fuel using delayed fast neutron based non-destructive method. Missouri University of Science and Technology Research Reactor (MSTR) was utilized for validation of the technique. Willman and co-workers reported NDA method for discriminating mixed oxide (MOX) fuel from low enriched uranium (LEU) fuel using gamma signature of fission products (Willman et al, 2006a). Dennis and Usman (Dennis and Usman, 2006) reported a similar technique evaluating the feasibility of using gamma ray spectroscopy for MOX fuel discrimination. Subsequently they identified ^{106}Ru isotope as their isotope of choice (Dennis and Usman, 2010) for burnup indicator and MOX fuel discriminator using gamma ray spectroscopy at MSTR. The gamma ray based NDA method is the only commercially available technique for measuring the composition of Plutonium and Uranium (U) in the spent fuel (Riffard et al, 2006). The technique has the drawback that it relies on the lowest energy direct peaks from Pu and U. The emitted gamma rays can be lost in the background since these isotopes emit low energy photons which make discrimination of Pu content from U difficult especially in MOX fuel analysis. However, Willman and coworkers were able to accomplish discrimination of MOX fuel and Uranium fuel using nondestructive gamma spectroscopy by obtaining ^{134}Cs/^{154}Eu ratio experimentally (Willman et al, 2006a; Willman et al, 2006b). Akyurek and coworkers compiled a list of candidate isotopes for Pu–U discrimination for various applications (Akyurek et al, 2014). Nondestructive analysis can also be developed based on neutron measurements. Zhao (Zhao, 2004) investigated the feasibility of using neutron measurement techniques for pebble bed reactor fuels for online burnup measurement. In fact, a technique

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based on the combination of the delayed neutron and gamma technique was developed to measure relative fission rate ratios between spent fuel and new uranium fuel (Perret and Jordan, 2011). Delayed thermal neutron measurement technique is also being developed at Paul Scherrer Institut in Switzerland where the goal is re-irradiation at zero power and mapping the delayed neutron post irradiation to characterize the interfaces between burnt and fresh fuel assemblies using BF_3 detector as reported by Jordan and Perret (Jordan and Perret, 2011). Due to the detector being used at Paul Scherrer Institut their analysis relay on thermal neutron measurements which would introduce undesired additional uncertainty because of moderation process.

With recent Megatons to Megawatts program, the U.S. nuclear power industry is planning to introduce MOX fuel in the present fleet of nuclear reactors (Shaw Arevaervices, 2008). According to the agreement 35 metric tons (MT) of weapon-grade plutonium will be converted into MOX fuel (United States Enrichment Corporation 13 April, 2013). Duke Energy utilized AREVA's MOX fuel assemblies for their Catawba Nuclear Station in 2005 starting a new era of nuclear fuel cycle in U.S. Regardless of how the U.S. nuclear fuel cycle evolves, better tools and techniques for measuring burnup and monitoring spent fuel will be required. Burnup measurement is important for various applications such as; burnup credit validation and verification, pre-transportation compliance certification, monitoring spent fuel for Special Nuclear Material (SNM) control, accountability, monitoring during storage for historic burnup analysis and possibly online burnup measurement for next generation reactors.

As a tool for technique development and validation, MSTR can be used for burnup analysis and discrimination of Pu and U content using nondestructive neutron measurements with LEU fuel. This research focused on developing a technique to be able to discriminate MOX(Pu) from LEU fuel using delayed fast neutron measurements. Bubble Technologies (BTI) Portable Spectroscopic Neutron Probe (N-Probe) detector (Ing et al, 2007) was used in this study to collect fast delayed neutron spectra from various fuel elements.

2. Description of MSTR reactor

Fig. 1 shows MSTR which is a pool-type reactor licensed by NRC to operate at 200 kW thermal power. MSTR has been in operation for more than 50 years. It is the first nuclear reactor in the state of Missouri, which serves as an excellent facility for research and training.

The reactor pool contains 121,133 L of high purity water in a pool which is 9.14 m deep, 2.74 m wide and 5.79 m long. The reactor core is movable in the pool and there are two modes of operation; the T mode and the W mode. Reactor is said to be in the "T" mode when the core is moved close to the thermal column providing improve thermalization. On the other hand, when the reactor core is pushed away from the graphite it is said to be in "W": or water mode. The reactor core was refueled to LEU fuels in 1992 (Straka, 1998) and the LEU fuels have been in use since then. The reactor core consists of 15 fuel elements and 4 control rods. Fig. 2 shows a mock fuel element and the configuration of fuel plates in each fuel element. Each fuel element consists of 9 fuel plates and each plate contained 12.5 g of enriched ^{235}U at the beginning of its life.

The fuel is in the form of Uranium Silicide-Aluminum (LEU U_3Si_2–Al) with 19.75 ± 0.2 wt% enriched ^{235}U. The silicon content of U_3Si_2 is 7.5 ± 0.4 wt%. MSTR current fuel configuration (called T121 to specify that it is close to the thermal column) is shown in Fig. 3. The beam port, which was used in our experiments, is located near the fuel element "F11". Table 1 shows the burnup and conversion history of the reactor core fuel elements as reported to NRC. During the last 23 years of operation with LEU, the core went through many core re-configurations and consequently the burnup of various fuel elements are significantly different. Some new and used fuel elements are still available in the reactor pool storage and can be used as Pu free elements. These new fuel elements can be used to compare with irradiated U element to develop burnup measurement and discrimination technique. Old fuel is expected to have higher quantities of Pu content and hence the difference in post irradiation delayed neutron measurement can be related to Pu contents of the fuel.

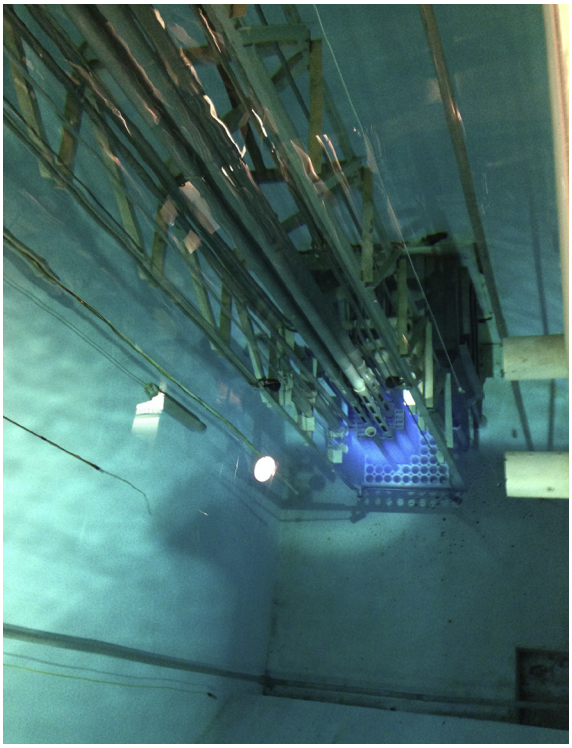

Fig. 1. Picture of MSTR Reactor pool and core with Cherenkov radiation (The blue glow). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1 shows approximate burnup history of MSTR fuel. The fuel is shuffled periodically and the most irradiated element is assigned burnup credit in the units of g of ^{235}U. It is important to recognize the approximate nature of burnup credit assignment to various fuel elements. Periodically, the most burnt element is assigned with total core burnup in grams. While this information is rather crude, in the absence of anything better it is being used as the approximate burnup of a specific element. Over the years, burnup value (in grams) has been assigned to various fuel elements in the core. This randomly assigned burnup approximation does not represent the actual burnup value in a particular fuel element. Therefore, a better burnup calculation or measurement is needed for the reactor core. Table 1 also shows the approximate conversion of ^{238}U to ^{239}Pu based on the total kW-hr of reactor run time.

3. Theory of delayed neutrons

Fission produces a large number of isotopes many of them being neutron rich. These neutron rich isotopes are the source of delayed neutrons. Decay of neutron rich isotopes and production of delayed neutrons play significant role in the control of nuclear reactors even though they comprise only about 1% of the total neutrons released from fission. There are over 250 well known precursors responsible