Use of curium neutron flux from head-end pyroprocessing subsystems for the High Reliability Safeguards methodology

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A B S T R A C T

The deployment of nuclear energy systems (NESs) is expanding around the world. Nations are investing in NESs as a means to establish energy independence, grow national economies, and address climate change. Transitioning to the advanced nuclear fuel cycle can meet growing energy demands and ensure resource sustainability. However, nuclear facilities in all phases of the advanced fuel cycle must be 'safeguardable,' where safety, safeguards, and security are integrated into a practical design strategy. To this end, the High Reliability Safeguards (HRS) approach is a continually developing safeguardability methodology that applies intrinsic design features and employs a risk-informed approach for systems assessment that is safeguards-motivated. Currently, a commercial pyroprocessing facility is used as the example system. This paper presents a modeling study that investigates the neutron flux associated with processed materials. The intent of these studies is to determine if the neutron flux will affect facility design, and subsequently, safeguardability. The results presented in this paper are for the head-end subsystems in a pyroprocessing facility. The collective results from these studies will then be used to further develop the HRS methodology.

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1. Introduction

The deployment of nuclear energy systems (NESs) is expanding around the world. Nations are investing in NESs as a means to establish energy independence, to grow national economies, and to address climate change (GIF, 2002; IAEA, 2013). In the future, the sustainable use of nuclear energy will require transition to advanced nuclear energy systems. These will employ remotely handled facilities in which batch-type processing will occur in hot cells. Safeguardability of these facilities must be demonstrated for successful licensing and operation. To this end, the High Reliability Safeguards (HRS) approach has been established (Borrelli, 2013; Borrelli, 2013, 2014a,b). HRS is a continually developing methodology that applies intrinsic design features in order to enhance proliferation resistance and physical protection and employs a risk-informed approach for systems assessment that is safeguards-motivated. The intent is to integrate proliferation resistance and physical protection measures as equally weighted with safety and physical security. There is considerable latitude in the development of safeguardability methodologies as no safeguards goals have yet been formalized by the International Atomic Energy Agency (IAEA) for the advanced fuel cycle (PRPPWG, 2011; Kim et al., 2010).

The HRS methodology exhibits two primary branches: (1) a set of functional components as part of a design strategy and (2) a systems assessment based on a risk-informed approach. This paper falls within the context of (1). A commercial pyroprocessing facility is the subject of current study. In addition to sustainability, an advanced NES is also beneficial to back-end management of the contemporary fuel cycle. Advanced reactors; e.g., a Generation IV, sodium fast reactor (SFR), can consume existing used fuel inventories from contemporary, Generation II and III light water reactors. However, in order to do this, pyroprocessing will be needed in order to convert the used fuel from the current ceramic form to the metal form that is needed to fabricate the fuel elements for the advanced reactor. Therefore, safeguardability of a commercial-scale pyroprocessing facility will be critical in assuring the overall safeguardability of the advanced fuel cycle. This paper follows from Borrelli (2013), which modeled the neutron flux associated with the fuel fabrication process in pyroprocessing. This study models the remaining subsystems, applying the same methodology and input data set. The intent is to model the neutron flux from commercial-scale pyroprocessing materials, observe the magnitude of the flux for each subsystem, and analyze the results in terms of how this might affect the facility design, safety, and security, and whether the neutron flux could potentially be used to enhance

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safeguardability. The neutron flux associated with each subsystem will affect is modeled for a hypothetical facility design. The results presented in this paper are for the head-end subsystems in the pyroprocessing facility. The collective results from these studies will then be used to further develop the HRS methodology.

2. Background and motivation

2.1. Pyroprocessing overview

Pyroprocessing treats used UO_2 ceramic fuel and fabricates a metal fuel alloy comprised of uranium, transuranic elements (TRU), rare earth (REFP) fission products, and zirconium. There are four subsystems relevant to proliferation risk: (1) electroreduction, (2) electrorefining, (3) electrowinning, and (4) metal fuel fabrication. Materials are pyrophoric and must be processed in hot cells with an inert atmosphere. This paper will focus on (1)–(3). The processing materials in these stages share similarities in that metal is anodically dissolved in different types of salts. Results for (4) are reported in Borrelli (2013). The manner in which used fuel is treated by pyroprocessing to produce the metal alloy for use in an advanced reactor, the materials used at each processing stage, and the form of TRU-bearing materials, is discussed extensively in Borrelli (2014a). A generalized pyroprocessing system is shown in Fig. 1.

Used fuel assemblies are first mechanically chopped and decladded. Voloxidation converts UO_2 to a U_3O_8 powder. This powder is then converted to a metal by electroreduction and anodic dissolution in salt. At this stage, the high-heat radionuclides Cs and Sr are removed. Electrorefining, also by salt dissolution, then extracts uranium metal. The TRU remains in this salt and then obtained as a metal on a liquid, cadmium cathode by electrowinning. Metal slugs are finally fabricated containing an alloy of U, TRU, zirconium, and a small weight fraction of REFP.

2.2. Use of Cm neutron flux within the context of the HRS methodology

The design of a pyroprocessing facility will be different from contemporary PUREX facilities due to intrinsic material properties and the manner by which materials are processed. Because none of the TRU elements are chemically separated during pyroprocessing, the detection of Cm neutrons due to spontaneous fission then indicates the presence of Pu (Borrelli, 2014a). This is a well-established technique with regards to PUREX safeguards and could offer considerable benefits to safeguardability of a pyroprocessing facility primarily in terms of potential material transfers and cell cleaning for routine repairs, maintenance, and accident remediation (Borrelli, 2013).

3. Model

3.1. Input data

Radionuclide inventory was obtained by ORIGEN2.2 simulation for a fresh fuel composition of 1 MTU 4.5% ^{235}U, 55 GWD/MTU burnup, cooled for 10 years (KAERI, 2010). This was based on prior assessment of an advanced fuel concept for the Republic of Korea (ROK) (Borrelli, 2013; Borrelli, 2013, 2014a,b). Activation products were neglected. The default libraries for a ^{235}U enriched UO_2 pressurized water reactor (PWR) for cross sections, decay, and photons were applied (Croff, 1980). There were no additional geometry assumptions.

For this fuel composition, there are 87 g of Cm in the used fuel, containing 79 g of ^{244}Cm. The total Pu mass is 1.1×10^4 g. The ratio of ^{244}Cm/Pu is 0.0071. The specific neutron emission rate for ^{244}Cm

Table 1
The 'processing states' modeled for the three subsystems of interest are: (1a) electroreduced metal dissolved in LiCl-Li$_2$O salt, (1b) electroreduced metal, (2) uranium and TRU metal dissolved in LiCl-KCl salt, (3a) TRU metal dissolved in the same salt after U extraction, and (3b) TRU metal and liquid cadmium, after extraction from the salt. Commercial batch sizes are shown. The Cm and Pu content was determined by ORIGEN simulation for a fresh fuel composition of 1 MTU 4.5% ^{235}U, 55 GWD/MTU burnup, cooled for 10 years. Density was determined by the principle of additive volumes.

State	Batch size (kg)	Density (g/cc)	^{244}Cm (g)	Pu (g)
1a: ERD metal + salt	70	2.03	1.6	231
1b: ERD metal	20	18.35	1.6	231
2: U + TRU + salt	70	2.33	1.6	231
3a: TRU + salt	38.1	1.71	1.6	231
3b: TRU + Cd	1.1	9.70	1.6	231

is 1.1×10^7 neutrons/s/g with a neutron per fission yield of 2.76 for spontaneous fission. These values are consistent with outside sources (Rinard and Menlove, 1996). The neutron emission rate for ^{244}Cm for this fresh fuel composition is 8.8×10^8 neutrons/s, and this contributes to 98.4% of the total neutron emission rate in the used fuel at the ten-year cooling time (Borrelli, 2013).

A material flow for the pyroprocessing facility was then calculated, by performing a mass balance with assumed process losses at each processing stage, defined previously in Section 2.1. The material flow is explained in detail in Borrelli (2013). The estimated process losses are shown in Figure 1. The neutron rate from (α, n) reactions and neutron multiplication is currently neglected due to the extremely high neutron rate from ^{244}Cm for typical LWR used fuel (Rinard and Menlove, 1996). Additionally, the metal fuel alloy will have a high purity, and therefore, these (α, n) reactions are neglected as well (Ensslin et al., 1998).

3.2. Processing states

Knowledge of the location and form of TRU in the system is important. For the three subsystems (1)–(3) studied in this paper, five 'processing states' of interest were selected for study. These are: (1a) electroreduced metal dissolved in LiCl-Li$_2$O salt, (1b) electroreduced metal, (2) uranium and TRU metal dissolved in LiCl-KCl salt, (3a) TRU metal dissolved in the same salt after U extraction, and (3b) TRU metal and liquid cadmium, after extraction from the salt. These processing states are numbered to correspond to the subsystems labeled in Section 2.1. Table 1 contains the commercial batch sizes for each processing state, including the ^{244}Cm and Pu content. The density for each state is included and is explained subsequently.

3.3. Modeling scenarios

Although the Cm and Pu content is the same for each state, the form is not; i.e., dissolved in different salt type or bare metal. It is important to confirm whether the form of the processing state will affect the magnitude of the flux. If the flux associated with each processing state is substantially different, then this difference could be used to enhance the safeguardability of the facility by identifying each process. The flux was modeled for the commercial batch sizes for each processing state given in Table 1.

In addition, the flux is modeled for the 'conceptual held-up material.' As defined in Borrelli (2013), this is a representation of a lower threshold for a detectable mass in each subsystem. During normal processing, there will be material that is held-up in the equipment, for any of the subsystems for many initiating events, both normal and off-normal. Quantification of all the SNM in the facility will therefore be needed both for both IAEA requirements and State Materials Accounting and Control, as well as to restore continuity of knowledge in the event of an accident. For final