



Simulated response to process abnormalities during spent nuclear fuel electrorefining



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ARTICLE INFO

Article history:

Received 15 March 2017

Received in revised form 18 July 2017

Accepted 21 July 2017

Keywords:

Nuclear fuel cycle
Pyroprocessing
Advanced safeguards
Process monitoring
Modeling
Electrorefining

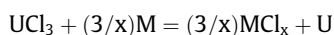
ABSTRACT

During normal operations, the only deposition on a spent nuclear fuel electrorefiner cathode consists of uranium. A computer model, ERAD, was used to simulate conditions under which the electrorefiner salt was progressively contaminated from spent nuclear fuel processing without operator intervention. The model was adjusted to reflect three scenarios: one in which salt removed with every cathode deposit was returned but the UCl_3 concentration was never adjusted, one in which the salt was not removed and UCl_3 concentration was never adjusted, and one in which the Pu/U ratio in the salt was allowed to reach a value of one before starting the first scenario. Under the base conditions simulated, uranium initially deposits alone but eventually plutonium begins to co-deposit along with uranium. Up to 350 g of plutonium was calculated to deposit along with 8.3–9.5 kg of uranium. While precise tracking of changes in the salt composition may not be possible, the model was able to identify markers for plutonium deposition by tracking changes in electrode potential. This type of marker should ultimately provide inspectors with more options for situations when they have limited access to electrorefiner data.

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1. Introduction

The recycling of spent nuclear fuel (SNF) is projected to be vital to the long-term sustainability of nuclear power. It not only minimizes the waste that requires special handling and storage, it also reduces the amount of fuel required to operate nuclear power plants, which decreases the need for extraction of uranium ore from mining. One demonstrated method that can be used to recycle SNF is pyroprocessing. Pyroprocessing involves the use of an electrorefiner (ER) with LiCl-KCl eutectic salt as the electrolyte. The electrolyte normally contains about 5 wt% UCl_3 to serve as oxidant and to support a high rate of uranium electrotransport. Metallic SNF, chopped into small segments, serves as the anode. When an electric current is applied, the uranium metal is separated from the other metals present in the SNF and is deposited on the cathode. The remaining metals either remain in the fuel (anode) basket or partition into the salt as chlorides. The metals that form chlorides are referred to as active metals and are formed via reaction with UCl_3 contained in the ER salt:



Pyroprocessing for the purpose of recycling SNF was initially researched in the 1980s when Argonne National Laboratory started developing the process for the fuel separation function of the Integral Fast Reactor (IFR) Laidler et al. (1997). It utilizes a number of high temperature process steps as shown in Fig. 1 (Garcia et al., 2017).

Electrorefining (ER) is at the center of the process and is instrumental for separating actinides from the remaining constituents in spent nuclear fuel. It utilizes a molten LiCl-KCl- UCl_3 electrolyte for electrorefining uranium or grouped actinides (U/TRU) to a cathode, while fission products either remain in an anode basket or accumulate in the molten salt that is eventually discarded. A schematic of an electrorefiner cell is given in Fig. 2.

It is technically more difficult to recover elementally pure plutonium using pyroprocessing than aqueous separation methods such as PUREX. This is based on the proximity of the free energy of actinide and rare earth chlorides in the molten salt used for electrorefining. The process has always been run in a batch mode of operation in remote equipment located in hot cells. Multi-stage separations are not practical for pyroprocessing compared to the much simpler, ambient temperature liquid extraction steps used in PUREX. Besides elemental purity, another important consideration is isotopic purity. Regardless of which processing method is used, the plutonium isotopic distribution from spent nuclear fuel is not “weapons grade” (93% Pu-239). Typical spent fuel from a

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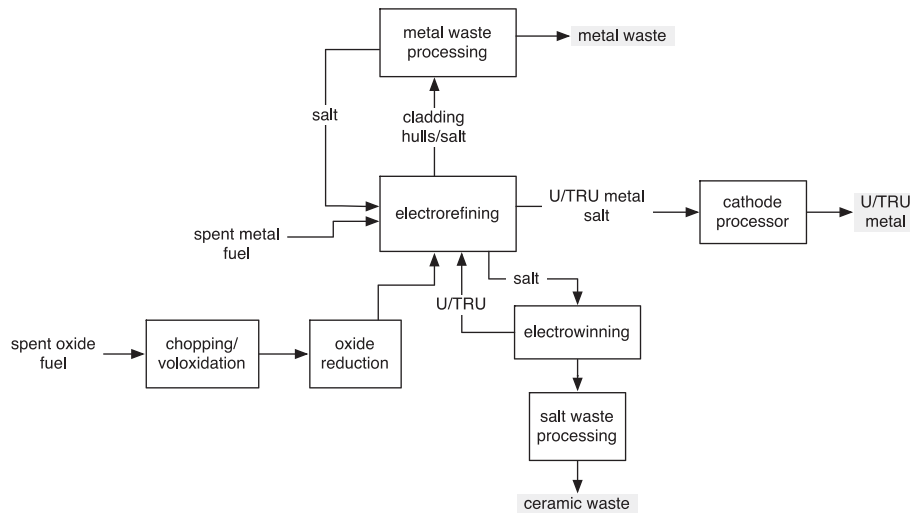


Fig. 1. Pyroprocessing flowsheet adaptable to metal or oxide fuel treatment.

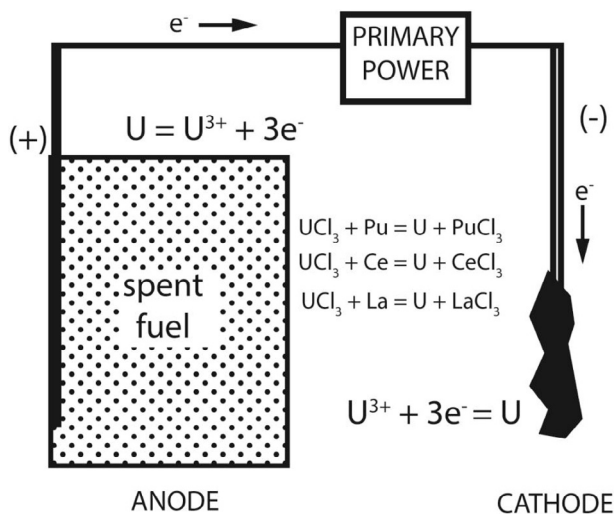


Fig. 2. Basic operation of spent fuel electrorefiners.

pressurized water reactor would contain 59% Pu-239, 21% Pu-240, 14% Pu-241, 5% Pu-242, and 1–2% Pu-238 (Carson, 1993). The implication of non-weapons grade is not that a weapon cannot be made, but rather than a good weapon cannot be made. High difficulty in weapon assembly, pre-ignition, “fizzle,” large critical mass, and low-yield are some of the problems associated with non-weapons grade plutonium (Carson, 1993). But if the objective is to prevent all destructive uses of nuclear weapons, then even poor bombs are of concern. The process chemistry differences between aqueous processing and pyroprocessing that lead to differences in elemental plutonium purity are essentially compounded by the isotopic differences between weapons-grade and reactor-grade plutonium. Pyroprocessing of spent nuclear fuel, thus, starts to appear to be a very low risk for nuclear weapons proliferation. But it should not be dismissed as a negligible risk. Once people take that attitude, then attention is moved away from it and the likelihood of detecting misuse plummets. The final conclusion stated by J. Carson Mark should be restated here. “The need for safeguards to protect against the diversion and misuse of separated plutonium applies essentially equally to all grades of plutonium,” (Carson, 1993) where we may consider it appropriate to include

other elements as tools by which the grade of the plutonium is downgraded.

Another important factor to consider when weighing the importance of applying safeguards to pyroprocessing is the issue of self-protection. Spent nuclear fuel is generally observed to be too radioactive to handle safely. It contains fission products that emit high energy gamma radiation that requires bulky/cumbersome shielding for safe handling. Referring back to Fig. 1, pyroprocessing includes two fuel treatment steps prior to electrorefining—voloxidation and oxide reduction. Each of these steps is known to cause some loss of radioactive fission products (Herrmann et al., 2007; Song et al., 2008). Cesium and strontium, for example, partition into the molten salt used for oxide reduction (Herrmann et al., 2007). Cesium, iodine, technetium, krypton, and other fission products volatilize during voloxidation (Song et al., 2008). Then the electrorefiner can further separate actinides from noble metals, alkaline metals, and alkali earth metals based on thermodynamic/electrochemical property differences. The final U or U/TRU product created by pyroprocessing will have lost much of its self-protecting characteristic.

Based on above arguments, the importance of having international safeguards in place for non-weapons states that operate pyroprocessing facilities can be considered to be well established. This refers to oversight and inspections by the International Atomic Energy Agency (IAEA) to verify user declarations and adherence to international treaties. While there currently are no commercial facilities that utilize pyroprocessing, development of effective and practical safeguards methods for this technology should be considered to be a prerequisite for startup of such a facility.

Unfortunately, the traditional international safeguards (SG) approach of nuclear material accountancy (NMA) has widely been recognized to be inadequate for pyroprocessing (Wigeland et al., 2009; Garcia et al., 2002; Kim et al., 2010). A few of the reasons behind this incompatibility are lack of input accountability, hold-up of fissile material, and inhomogeneity of the process materials (Lafreniere et al., 2015; Rappleye et al., 2015). A proposed alternative approach that could be used to replace or supplement NMA is process monitoring (PM) Garcia et al. (2017). This refers to real time, continuous sensor output used to infer conditions of the process. Neutron counting and cyclic voltammetry are two of a variety of PM techniques, which have already been studied and reported on previously (Lafreniere et al., 2015; Rappleye et al., 2015).

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