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Real-time monitoring of actinides in chemical nuclear fuel reprocessing plants

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A B S T R A C T

Spent nuclear fuel chemical reprocessing plants process several 10^3 kg of spent nuclear fuel, especially Pu isotopes, which present significant potential for terrorism (Note: ~ 8 kg Pu constitutes a threat level quantity). This requires detecting actinides in transit, but also, to ensure they are not diverted. Present-day sensors disallow real-time monitoring leading to significantly non-optimal operations.

The tensioned metastable fluid detector (TMFD) sensor technology has been developed by researchers at Purdue University in partnership with Texas A&M University, and various national laboratories (sponsored in part by several United States federal agencies and private enterprise). It is based on nano-to-macro scale interactions of radiation with molecules of fluids that are in a state of *tensioned* metastability. Developed are lab-scale prototypes for adapting to chemical reprocessing plants providing real-time directionality to within $\sim 10^\circ$ – 20° , with $\sim 90\%$ efficiency to detect neutrons (from eV to MeV) and alpha emitting nuclides energies to within 1–5 keV recoil resolution, and sensitivities to ultra-trace levels (e.g., to 10^{-15} g/cc Pu). TMFD systems are robust, portable and offer $100\times$ lower cost potential compared with present-day systems (e.g., NE-213 based neutron–gamma liquid scintillator based systems). A multi-physics design framework has been developed, and validated. This paper highlights state-of-art developments and adaptations of TMFDs for in situ, real-time monitoring of U, Pu, Am and Cm actinides from the sensitive (in the past virtually impossible to monitor) front-end wherein radioactively hot spent nuclear fuel is chopped and dissolved, to throughout the subsequent stages in a chemical nuclear fuel chemical process.

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1. Introduction

The U.S. Department of Energy (DoE) has developed advanced methods for reprocessing spent nuclear fuel in commercial reprocessing plants which, however, also present the most challenging area within the nuclear fuel cycle to safeguards (Durst et al., 2007). The majority of this development was accomplished under the Advanced Fuel Cycle Initiative (AFCI), building on the strong legacy of process development R&D over the past 50 years. The emergence of the new Global Nuclear Energy Partnership (GNEP), which aims to continue and expand the development of AFCI processing methods, has

elevated the U.S. and worldwide commitment to advanced fuel cycles. These advanced processing methods must be scaled up and engineered for real-scale implementation.

The most prominent processing method under development is named UREX+ depicted schematically in Fig. 1. The name actually refers to a family of processing methods that begin with the Uranium Extraction (UREX) process and incorporate a variety of other methods to separate uranium, selected fission products, and the transuranic (TRU) isotopes from dissolved spent nuclear fuel (SNF). It is pointed that UREX+ is similar to the well-known PUREX process currently used worldwide (e.g., at Sellafield, Great Britain; La Hague in France; and Rokasho in Japan) in that multiple chemical separation processes are used to remove the major sources of

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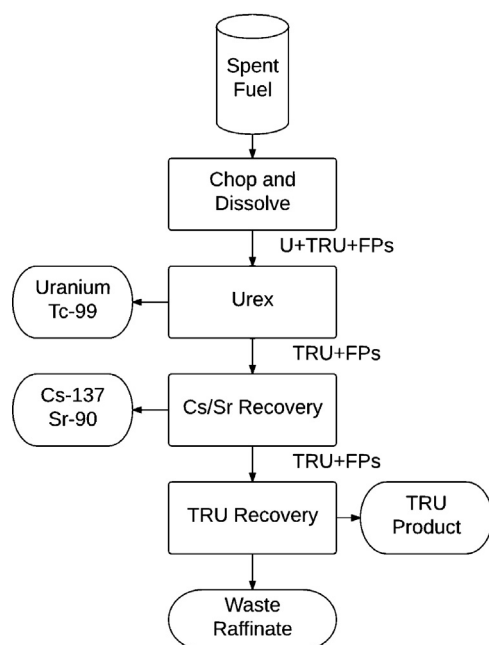


Fig. 1 – High-level process flow diagram for the UREX+ aqueous separations plan.

radioactivity; with specific goals to recycle U and Pu into the fuel cycle. As GNEP/DoE moves toward the intended implantation of UREX+ on a real scale over the next 20 years (e.g., over 1000 tons of fuel per day in the future to accommodate the worldwide SNF from ~1000 operating reactors each with an inventory of ~100 T of UO_2 and, additionally, also for the reprocessing of the legacy inventory of SNF) it is a valuable time to consider issues such as safeguards strategies and materials control and accountability methods. Monitoring of higher actinides during aqueous separations is a critical research area for the U.S. GNEP/AFCI program. A key deficiency of paramount importance (Cipiti, 2005; Bean, 2007) in such monitoring for material accountability is the lack of real-time assessments to detect the diversion of TRU elements such as Pu. This is especially relevant for ^{239}Pu ; a single fuel assembly can contain close to 7 kg (close to the quantity required for a Nagasaki-type nuclear explosive). By providing on-line materials accountability for the processes, covert diversion of the materials streams becomes much more difficult. Maintaining control and knowledge of such special nuclear materials (SNMs) is presently conducted via time consuming off-site assessments which greatly affect the throughput and efficiency of a reprocessing plant which must reprocess hundreds to thousands of SNF fuel assemblies a year.

1.1. Present-day techniques for SNM monitoring and technology gaps

Currently, alpha emitter detection requires time-consuming off-site laboratory based methods and most on-line neutron detection systems are readily saturated in the extreme gamma fields associated with the copious quantities of fission products like Cs-137. As noted from Fig. 1 the first step at any reprocessing facility is to chop the SNFs and dissolve the accountable materials in an acidic solution upon which the solution is transferred to an accountability tank; samples are used then to crudely determine the sum total of initial nuclear material inventory (Bean, 2007) – Note: determination of the initial SNM inventory is critical to maintaining

material accountability throughout the process and to ensure lack of diversion. Unfortunately, till now, near real time accountability (NRTA) of transuranic actinides has remained elusive. Although techniques for measuring near real time for bulk quantities, e.g., the volume of dissolved fuel and flow rates have been developed, it is the NRTA issues related to on-line measurement of the elemental and isotopic concentrations that has not been possible to accomplish with conventional detection methods (e.g., with K-Edge densitometry; X-ray fluorescence; Hybrid K-Edge/X-ray fluorescence densitometry; mass spectrometry; high resolution gamma spectrometry; isotope dilution gamma spectroscopy; constant coulomb coulometry; titrimetry; gravimetry; spectrophotometry; calorimetry). Cipiti (2005) provides a good summary of present-day approaches along with their relative merits and shortcomings; overall, it may be concluded that such approaches in general do not offer NRTA capability for isotopic assessments and importantly, do not allow one to determine SNM isotopic inventories in situ.

A transformational detection methodology that permits on-line assessment of the U and Pu type actinides even at the front end itself (and over following later stages) and also one that is adaptable with complementary present-day systems for later stages would represent a major, timely boost to reprocessing operational and safeguards-related goals. This article describes a framework and methodology that achieves such a goal using the technology of tension metastable fluid detector (TMFD) sensor system.

As introduction to the multitude of issues to overcome for effective NRTA of SNM isotopic inventory we first start by examining a typical inventory of actinides in spent nuclear fuel (SNF) as it relates to chemical reprocessing.

1.2. Spent nuclear fuel composition, issues and challenges

Spent nuclear fuel (SNF) from a typical light water reactor (LWR) contains a large collection of fission products with isotopes that span the periodic table from Fe-72 to Er-167 (plus a minor amount of tritium from tertiary fissions). In addition, SNF contains radioactive activation products and transuranic (TRU) actinide elements (i.e., Pu, Np, Am and Cm). While the majority of the fission products are gamma-beta emitters, it is the alpha-emitting uranium and TRU isotopes that form the basis of significant concern from safeguards and security viewpoints. Table 1 depicts the inventory of uranium and TRU elements in representative spent fuel assemblies from a pressurized water reactor (PWR). All of the uranium and TRU isotopes emit alpha particles but only some of them generate significant quantities of neutrons from spontaneous fission (SF).

The data in Table 1 indicate both a commonality and a differentiator between U and Pu isotopes in SNF. The commonality is that both isotope groups exhibit alpha particle emissions with energies defined by the individual isotopes that vary between ~4MeV and ~6MeV. The differentiator arises from differences in neutron emissions due to spontaneous fission. Uranium has a maximum emission rate in Table 1 of $\sim 10^4$ n/s/MTU for ^{238}U a value which, upon dilution and spread out over space in piping (e.g., to over 1m^2 of surface area) can make this difficult to passively decipher over cosmic background neutron fluxes. On the other hand, some of the TRU isotopes like ^{244}Cm can emit $\sim 10^9$ n/s/MTU; which constitute a readily measurable quantity (diluted or

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