RADIOACTIVE FLOW CHARACTERIZATION FOR REAL-TIME DETECTION SYSTEMS IN UREX+ NUCLEAR FUEL REPROCESSING

A Thesis

by

THOMAS RUSSELL HOGELIN

Submitted to the Office of Graduate Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

December 2010

Major Subject: Nuclear Engineering

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Approved by:

Chair of Committee,	Sean M. McDeavitt
Committee Members,	William Charlton
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ABSTRACT

Radioactive Flow Characterization for Real-Time Detection Systems in UREX+ Nuclear Fuel Reprocessing.

(December 2010)

Thomas Russell Hogelin, B.M.E., Auburn University

Chair of Advisory Committee: Dr. Sean M. McDeavitt

The reprocessing of used nuclear fuel requires the dissolution and separation of numerous radioisotopes that are present as fission products in the fuel. The leading technology option in the U.S. for reprocessing is a sequence of processing methods known as UREX+ (Uranium Extraction+). However, an industrial scale facility implementing this separation procedure will require the establishment of safeguards and security systems to ensure the protection of the separated materials. A number of technologies have been developed for meeting the measurement demands for such a facility. This project focuses on the design of a gamma detection system for taking measurements of the flow streams of such a reprocessing facility.

An experimental apparatus was constructed capable of pumping water spiked with soluble radioisotopes under various flow conditions through a stainless steel coil around a sodium iodide (NaI) detector system. Experiments were conducted to characterize the impact of flow rate, pipe air voids, geometry, and radioactivity dilution level on activity measurements and gamma energy spectra. Two coil geometries were

used for these experiments, using 0.5 in stainless steel pipe wound into a coil with a 6 inch diameter; the first coil was 5.5 revolutions tall and the second coil was 9.5 revolutions tall. The isotopes dissolved in the flowing water were produced at the Texas A&M Nuclear Science Center via neutron activation of chromium, gold, cerium, and ytterbium nitrate salts. After activation, the salts were dissolved in distilled water and inserted into the radioactive flow assembly for quantitative measurements. Flow rate variations from 100 to 2000 ml/min were used and activity dilution levels for the experiments conducted were between 0.02 and 1.6 μCi/liter. Detection of system transients was observed to improve with decreasing flow rate. The detection limits observed for this system were 0.02 μCi/liter over background, 0.5% total activity change in a pre-spiked system, and a dilution change of 2% of the coil volume.

MCNP (Monte Carlo N-Particle Transport) models were constructed to simulate the results and were used to extend the results to other geometries and piping materials as well as simulate actual UREX stream material in the system. The stainless steel piping for the flow around the detector was found to attenuate key identifying gamma peaks on the low end of the energy spectrum. For the proposed schedule 40 stainless steel pipe for an actual reprocessing facility, gamma rays below 100 keV in energy would be reduced to less than half their initial intensities. The exact ideal detection set up is largely activity and flow stream dependant. However, the characteristics best suited for flow stream detection are: 1) minimize volume around detector, 2) low flow rate for long count times, and 3) low attenuation piping material such as glass.

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NOMENCLATURE

AFCI	Advanced Fuel Cycle Initiative
CCD-PEG	Chlorinated Cobalt Dicarbollide-Polyethylene Glycol
FPEX	Fission Product Extraction
HKED	Hybrid K-Edge/X-ray Fluorescence Densitometry
HPGe	High Purity Germanium
HRGS	High Resolution Gamma Spectroscopy
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
IDGS	Isotope Dilution Gamma-ray Spectrometry
IDMS	Isotope Dilution Mass Spectrometry
KED	K-Edge Denitometry
KMP	Key Measurement Point
LSDS	Lead Slowing-Down Spectroscopy
LWR	Light Water Reactor
MBA	Material Balance Area
MCA	Multi-Channel Analyzer
MCNP	Monte Carlo N Particle
MIP	Multi-Isotope Process
MOX	Mixed Oxide fuel
NaI	Sodium Iodide
NRC	Nuclear Regulatory Commission
NRF	Nuclear Resonance Fluorescence
NSC	Nuclear Science Center
ORIGEN	Oak Ridge Isotope Generation and Depletion Code
PUREX	Plutonium-Uranium Extraction
SEID	Standard Error of the Inventory Difference
TALSPEAK	Trivalent Actinide Lanthanide Separations by Phosphorus- Reagent Extraction from Aqueous Complexes

TARIS	Thermal Atomization Resonance Ionization Spectroscopy
TBP	Tributyl Phosphate
TIMS	Thermal Ionization Mass Spectrometry
TMFD	Tensioned Metastable Fluid Detector
TRU	Transuranic
TRUEX	Transuranic Extraction
UREX	Uranium Extraction
XRF	X-Ray Fluorescence

Elements

Am	Americium
Au	Gold
Ba	Barium
Ce	Cerium
Cm	Curium
Cr	Chromium
Cs	Cesium
Pu	Plutonium
Rb	Rubidium
Sr	Strontium
Tc	Technetium
U	Uranium
Yb	Ytterbium

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