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Title:

Nuclear Monitoring by Nonradioactive Noble Gas Sampling and Analysis

Authors:

B. L. Fearey, NIS-7

C. W. Nakhleh, NIS-7

W. D. Stanbro, NIS-7

J. A. Olivares, CST-9

R. T. Perry, Jr., TSA-10

W. B. Wilson, T-2

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Nuclear Monitoring by Nonradioactive Noble Gas Sampling and Analysis

Bryan L. Fearey*, Charles W. Nakhleh, and William D. Stanbro
Nonproliferation and International Security Division

Jose A. Olivares
Chemical Science and Technology Division

R. T. Perry, Jr.
Technology and Safety Assessment Division

William B. Wilson
Theoretical Divison

Abstract

This is the final report of a two-year, Laboratory Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL). The perceived importance of measuring the xenon and krypton isotopics of nuclear activities has increased substantially in recent years. We have performed a systems analysis and theoretical simulation of the production, atmospheric dispersion, and isotopic abundances of noble-gas fission products, addressing several questions of interest, including: the relative isotopic variation as a function of nuclear fuel composition, reactor operational history, reactor type, distance from stack, and ambient meteorological conditions. Of particular importance in this analysis was the question of back-calculating process parameters of interest given noble-gas isotopic data. An analysis of the effect of measurement uncertainties was also performed. The results of these analyses indicate that this monitoring concept should be experimentally feasible.

Background and Research Objectives

The importance of detecting undeclared and verifying declared nuclear activities has increased substantially. The ability to precisely and accurately measure xenon and krypton isotopics has been suggested as a method to detect such activities, especially at distance. Although several preliminary and survey studies[1-4] of the production, and subsequent dilution, of noble gas fission isotopics and tritium production gases had been performed prior to this project, we completed the first complete systems analysis and theoretical simulation of this problem. Questions of particular interest to this problem include the relative isotopic variation as a function of nuclear fuel composition, neutron flux, burn-up

* Principal Investigator, E-mail: bfearey@lanl.gov

duration, and fuel-rod position for different types of nuclear reactors. Other important questions involve the definition of the measurement precisions and accuracies required to extract operational parameters from not only nuclear reactors but also from clandestine reprocessing activities as a function of both dilution and distance. Additional applications include detection of illicit tritium production and recovery, gas leakage from nuclear weapons activities, and environmental and intelligence monitoring as well as other classified needs.

The ability to define such criteria is particularly important to establish current and future measurement requirements of state-of-the-art instrumentation as well as appropriate application and deployment scenarios. Such next-generation instrumentation is currently under preliminary development within the Space and Atmospheric Sciences and the Isotope Sciences Groups at Los Alamos. These mass spectrometers range from highly portable, reasonably precise, and efficient systems to complex laboratory-based systems with high efficiency and ultra-high precision and accuracy. This project will provide substantial guidance for instrumental system requirements and configurations. An additional benefit from this study will be the creation of a simulation analysis capability for measured isotopic ratios and abundances of xenon and krypton for the evaluation of nuclear activities.

A brief discussion of how the xenon and krypton isotopics could be used to determine various parameters from a nuclear proliferant follows. Xenon and krypton isotopes are produced in a nuclear reactor by fission of ^{235}U. The isotope ratios of fission-product noble gases are determined by fission yields and subsequent burn-up in the reactor neutron flux. The burn-up of ^{135}Xe by neutron capture to the closed neutron shell of ^{136}Xe is an important reaction in reactor physics and is the cause of the "xenon poisoning" phenomenon. The Xe isotopes important for fuel monitoring are ^{136}Xe, ^{133}Xe, ^{132}Xe, and ^{131}Xe with respect to ^{134}Xe. Analytical solutions (clearly a first approximation) indicate that these isotopic ratios can be used to determine the burn-up time, the effective neutron absorption cross-section, the reactor flux, and (by inference) the position of a particular fuel element within a reactor.

Importance to LANL's Science and Technology Base and National R&D Needs

This research project addresses several relevant aspects of the "Proliferation Detection and Assessment" program development focus area as well as the "Global Nuclear Materials Management" focus area. In addition, this project addresses the LANL tactical goals of the *Plutonium Legacy* (i.e., technology assessment and monitoring of plutonium production and reprocessing), *Modeling, Simulation and High-Performance Computing* (i.e., specialized nuclear computations), and *Great Science* (i.e., integration of several complex sciences for new analysis needs). This project is also strongly aligned with two important core competencies: *Non- and Counter- Proliferation*, and *Nuclear Materials Management.* Finally, this LDRD project clearly provides significant information to aid in *Reducing the Nuclear Danger* throughout the world.

Scientific Approach and Accomplishments

We first used a variety of codes to calculate the krypton and xenon inventories in a variety of spent fuel types. The ORIGEN2 code package was used to generate initial estimates. For more precise calculations on LWR fuels, the DANDE package was used. DANDE combines neutron tranport (Sn) and diffusion codes with cross section (TRANSX) and burnup (CINDER-3) codes to model irradiated materials, especially reactor fuels. Cross sections (for major materials) processed in TRANSX for the correct temperature and resonance self-shielding are passed to the DANT Sn code and to CINDER-3, which uses the TRANSX cross sections (for major materials) with collapsed library cross sections (for other materials) and DANT fluxes—returning new nuclide inventories to the other codes for cross-section processing and neutron transport. The library of CINDER-3 uses 154-group ENDF/B-V cross sections and contains fewer than 300 nuclides—those important to neutron absorption and, for long cooling times, aggregate decay properties of LWR fuel.

Recently, we have been working on the modeling of MOX fuel, requiring the reconstitution of fuel between successive cycles. This work is continuing.

We have now examined LWR fuel calculations with DANT and the newer CINDER'90, having a library of 63-group cross sections and 3400 nuclides with data of more recent evaluations, expecting a benefit from the use of code and data of more recent origin. These codes are not yet joined by DANDE, and interface of cross-section data to

CINDER'90 and inventory from CINDER'90 are accomplished with difficulty. The 69-group fluxes calculated with DANT must be collapsed to the 63-group structure of CINDER'90 externally. Input of problem-specific cross sections to CINDER'90 has not been accomplished. Initial results were disappointing, with a 20-25% disagreement in fission power calculated in the two codes. Differences due to group structure, resonance self shielding, Doppler broadening, and data source have been examined for attribution of the large disagreement. Comparisons are continuing.

Once these calculations were performed, we then used gaussian dispersion theory to model the atmospheric diffusion of stable noble-gas fission products from the stack of a reprocessing plant, using the US Environmental Protection Agency's ISC2 code and the declassified parameters of the PUREX plant at Hanford. This enabled us to model xenon/krypton isotopics at distances ranging up to 10 km.

Finally, the inverse problem of backcalculating the burnup and reactor type from the isotopic data was simulated using the model data and a maximum-likelihood inversion procedure.

We have reported these results in the literature (see Publication). The most important point is that using stable xenon/krypton isotopic data in the vicinity of a reprocessing plant appears to be able to yield several pieces of relevant information, particularly the burnup and type of the fuel being reprocessed.

The next important step is experimental benchmarking of these calculations. Continuation of this work is being funded by DOE for FY 1997, and experiments at the Savannah River Site are currently being undertaken to experimentally test the conclusions of the LDRD project.

Publication

Nakhleh, C.W., Stanbro, W.D., Hand, L.N., Perry, R.T., Wilson, W.B., Fearey, B.L., "Noble-Gas Atmospheric Monitoring for International Safeguards at Reprocessing Plants," to appear in *Science and Global Security*.

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