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ISOTOPE CORRELATIONS AND MEASUREMENTS IN THE UNITED STATES

by

P. J. Persiani

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I. INTRODUCTION

The changes in the isotopic composition with burnup are a measure of the irradiation history of the fuel in a power reactor. The solution of the system of differential equations required to describe the operating irradiation history is complex and depends on a detailed knowledge of core configuration, fuel management programs, spatial neutron flux distribution, spectra and cross section data. However, experimental data on LWR reprocessed fuel indicates that there may exist relationships between isotopic concentrations which have a predictable, functional behavior over a broad range of reactor operating conditions and burnup.

These observed functional relationships when coupled with reactor analysis and computational methods may be utilized primarily, in a verification mode, in two important phases of power fuel cycle system: 1) Fuel Management and Operating Optimization of Power Systems, and 2) International Safeguards and Management of Nuclear Material.

1. Fuel Management and Operating Optimization of Power Systems

The isotope correlation measurements of the inventory input to the reprocessing plant allows maintaining a more detailed operational materials flow control throughout the fuel cycle. The economics and operational performance of operating power reactors are interrelated through thermal and neutronic parameters which determine the static and dynamic behavior of reactor systems.

2. International Safeguards and Management of Nuclear Materials

Isotope correlations and measurement techniques have the potential as a safeguards accountability measure in the dissolver stage of the nuclear fuel cycle. Throughout the major portion of the fuel cycle, the fissile accountability is basically in the form of item accountability (fuel powder containers, pellets, elements, and assemblies). The measurement of the material balance input and product in the reprocessing phase of the cycle is the primary direct determination that a possible loss or diversion of material has occurred. The materials balance accountability gap which exists between the fabrication plant output and the input to the reprocessing plant can be minimized by the utilization of Isotope Correlation Techniques (ICT) at the dissolver stage of the processing plant. The safeguards significance of the ICT is that the input accountability would allow a level of verification of the fabricators' uranium content specification, the irradiation history, and the subsequent spent fuel assembly flow to the reprocessing plant.

II. RESEARCH AND DEVELOPMENT OF ISOTOPE CORRELATIONS AND MEASUREMENTS TECHNIQUES

1. Isotope Correlations

The emphasis in the development of Isotope Correlations and Measurements Techniques have been in the area of international safeguards where material accountancy is of fundamental importance.

The isotope correlation functions which are currently being considered as most effective for this purpose, have been under continuous study by many national and international laboratories and agencies.

The isotopic correlations suggested have been grouped into four categories:

a. Pu/U correlations,
b. Pu isotopic correlations,
c. U isotopic correlations,
d. U and Pu mixed isotopic correlations.

Some of the more simple relationships involving combinations of isotopic concentrations that exhibit a reasonably monotonic behavior over a broad range of reactor conditions and burnup are such functions: Pu/U vs. depletion ^{235}U, Pu/U vs. (100 - ^{239}Pu), Pu/U vs. $^{239}Pu \times {}^{242}Pu/{}^{240}Pu^2$, and ^{236}U vs. ^{235}U. Of the many preliminary functionals that have been suggested, the most effective function are those having a linear behavior. These linear relationships being independent of reactor operating conditions and burnup, effect a means of verifying the input to a reprocessing plant, and methods for establishing internal consistency of input analytical measurements, and a level of verification on initial isotopic concentrations prior to burnup.

The suggested functionals include fisson product isotope correlations in addition to the major and minor isotopes of uranium and plutonium. However, some studies on fission product correlations indicate that most of these correlations were sensitive to the neutron spectrum and may have limited applicability in the ICT program. Consequently, the initial technical assessment has been confined to correlations involving only the major and minor isotopes of uranium and plutonium. The de-emphasis on fission product correlations is also based on minimizing the performance requirements of magnet design for those measurement systems dependent on mass spectrometry. Limiting the mass scanning range to within 10% of the heavy mass isotopes may lead to a more operationally optimized and economic magnet system design.

The major effort in the study of isotope correlations has been carried out in the U.S. by Battelle Pacific Northwest Laboratories. The functional correlations where studied for reprocessed batches from: the U.S. and European pressurized and boiling water reactors, the U.K. graphite moderated reactors, and the Canadian CANDU power systems. The data base for more extensive correlation studies is somewhat limited because of the termination of reprocessing in the U.S. However, there have been suggestions to plan theoretical studies utilizing reactor models and burnup codes, where the functional relationships may be investigated and assessed over a broad range of reactor operating conditions and burnup parameters. It is also suggested that this theoretical study by supplemented by an experimental program of measurements limited to

fuel-pellet and/or fuel-element scale as a means of confirming the analytically determined functions.

2. Measurement Systems

The selection of the more safeguards effective functionals will depend not only on the level of reliability for verification, but also on the capability and difficulty of developing measurement methods. The performance characteristics of existing and proposed measurement techniques cover the general areas: (1) simultaneous multicomponent analysis techniques, (2) ion-cyclotron-resonance mass spectrometry, (3) x-ray fluorescence or densitometry with high flux monochromatic x-ray sources and high dispersion spectrometers (4) synchrotron radiation, and (5) active neutron interrogation.

A somewhat modest and dispersed effort has been supported at various U.S. laboratories and institutions in developing and assessing measurement technique capabilities and inherent limitations in terms of total systems, operational mode, sample preparation requirements and consequent effect on dissolver solution representation, and accuracy and precision estimates.

Scoping assessments of measurement systems have included not only the capability of isotopic measurements but also the potential that a measurement system can be developed for on-line or near-real-time assay of the dissolver solution.

Most of the systems are capable of measuring the elemental ratio, Pu/U. However, only a limited number of systems appear to have the capability of determining the isotopic correlation functions of interest to the ICT program. The x-ray fluorescence densitometry with energy dispersion spectrometry detection system and neutron slowing-down time spectrometry have limited capability for specific isotopic correlation functions.

The measurement systems involving mass spectrometry may be the most difficult to satisfy the timeliness accountancy aspect of the ICT program. On the other hand, the resin bead technique of sample preparation is a developed system and is competitive with the standard classical mass spectrometry currently used in the assay of dissolver solutions. The potential advantage of the resin bead technique is the reduced time period required in establishing the isotopic assay.

The Inductively Coupled Plasma (ICP) system has the potential as a simultaneous multicomponent measurement system for direct on-line assay at the dissolver stage of reprocessing. The applicability of this system would require a feasibility study to explore detecting isotopic hyper-fine-structure lines. The neutron interrogation method utilizing the measurement of isotopic resonances by the neutron transmission method needs to be demonstrated for the specifics of the ICT program.

The application of the measurement techniques under actual operating conditions would be a primary objective of the development program.

The major laboratories involved in developing techniques utilizing mass spectrometry have been New Brunswick Laboratory at Argonne, Illinois and the Oak Ridge National Laboratory where the resin bead technique has been advanced. X-ray fluorescence densitometry with wavelength dispersive systems

and gamma-ray energy dispersive systems are under development at Los Alamos Scientific Laboratory, and Monsanto Research Corporation, with some effort being supported at Lawrence Livermore Laboratory. A modest level of support at Ames Laboratory, Iowa State University, has been in scoping the feasibility of utilizing the Inductively Coupled Plasma (ICP) systems for isotopic analysis of the dissolver solution.