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THE USE OF STABLE NOBLE GASES AS A PREDICTOR OF REACTOR FUEL TYPE AND EXPOSURE

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ABSTRACT

Ensuring spent reactor fuel is not produced to provide weapons-grade plutonium is becoming a major concern as many countries resort to nuclear power as a solution to their energy problems. Proposed solutions range from the development of proliferation resistant fuel to continuous monitoring of the fuel. This paper discusses the use of the stable isotopes of the fissiogenic noble gases, xenon and krypton, for determining the burnup characteristics, fuel type, and the reactor type of the fuel from which the sample was obtained. The gases would be collected on-stack as the fuel is reprocessed, and thus confirm that the fuel is as declared.

I. INTRODUCTION

Countries possessing nuclear reprocessing plants would have the capability to secretly produce and reprocess weapons-grade plutonium. An ongoing study at Los Alamos National Laboratory indicates that the fissiogenic stable isotopes of xenon and krypton released in the reprocessing of the fuel can provide definite signatures that indicate the fuel characteristics and burnup of the fuel being reprocessed.[1] The determination is made following the analysis of on-stack collection of gases released in reprocessing. Such a determination could be made at the request of the country to ensure to others that the fuel is as declared. The determination could be made by the IAEA in fulfilling their mandate to ensure that a country is not producing weapons-grade plutonium.

This study demonstrates that enough information is contained in the ratios of these stable isotopes to determine the reactor fuel's characteristics. The noble gas production and their ratios in various worldwide reactor fuel types as a function of burnup have been calculated using reactor transport and burnup computer codes. Six different reactors were selected for analysis: BWR, PWR, Calder Hall, RBMK, CANDU, and a Savannah River Site (SRS) Production Reactor. The results from these calculations have been subject to a variety of types of statistical analysis. These analyses have shown that significant differences exist between the results obtained for each reactor and that the differences are enough to determine reactor type and fuel burnup. Plots of some of the calculational results will be presented for visual examination of the differences found in the results.

The results of the reactor physics calculations could be benchmarked and entered into a database. To utilize a benchmarked database for safeguards, the following procedure would be followed. The on-stack samples of gas from the reprocessing of the fuel in question would be analyzed using a mass spectrometer. The ratio of the isotopes obtained from the mass spectrometer analysis would be compared through reverse analysis with those ratios in the benchmarked database. This would allow for the determination of the most likely fuel type and burnup in the database based on the observed ratios.

For demonstration of the methodology, a gas sample was collected on-stack while fuel was being reconditioned at the SRS. The sample was subjected to mass spectrometer analysis. The resulting data were compared, using a Bayesian analysis, to those results obtained from a transport and burn calculation. The Bayesian analysis and results of the comparison will also be discussed in a following section.

In the next section, we begin with a discussion of the reactor physics calculations. This will be followed with a discussion of the analysis of the gas samples. The results of the reactor physics calculations and comparisons will demonstrate the validity of the methodology.

II. REACTOR CALCULATIONS

The objective of this part of the study is to provide detailed information concerning the xenon and krypton production and their ratios in various worldwide reactor types. The specifics of a reactor design have an influence on the production rates and ratios of isotopes that are produced. Six different reactors were selected for analysis: BWR, PWR, Calder Hall, RBMK, CANDU, and a SRS Production Reactor.

The calculations are made in two distinct phases: transport and burn. Los Alamos has a number of codes for these tasks, for example, the DANT[2] system for the transport calculations and CINDER[3] for the burnup. These two codes were chosen for the calculations here. Combining these processes, the reactor isotopics, and thus the xenon and krypton ratios, may be determined for any reactor type, operating scenario, and time.

A. Calculational Methods

The calculational phase begins with a pin cell model for the reactor. A neutron transport calculation is made and the resulting fluxes are used as weighting functions to homogenize the pin cell structure. For the PWR, BWR, and Calder Hall reactors, this process is straightforward because of the uniformity of the core. For the CANDU, RBMK, and the Savannah River Site Reactor, all of which contain concentric circles of fuel pins, a lattice model was developed in which material was conserved in a model with concentric fuel regions surrounding one pin cell. The lattice fluxes were used to homogenize the center pin and the nonreactive material (e.g., structure and moderator) into the homogenized core model.

Following the development of the reactor core model, a series of burn and transport calculations were made. The output fluxes from the transport calculation, which are normalized to the reactor level, are input to the burnup code CINDER. The fluxes provide a means for absolute value reaction (burnup) calculations and for weighting the cross sections. The new number densities calculated for a specific burn time are input into the transport code and the process is repeated. This continues until a desired burnup in megawatt days per metric ton of heavy metal (MWd/t) is reached.

B. Reactors

The enrichment on the BWR fuel is 1.69% enriched uranium dioxide and the total power level is 3470 MW thermal. The PWR fuel is 2.4% enriched uranium dioxide and the power level is 3410 MW thermal. The PWR and BWR are water-cooled and moderated. The RBMK fuel is 2.4% enriched uranium dioxide and the power level is 3140 MW. The RBMK is a graphite moderated, water-cooled reactor. The Calder Hall reactor fuel is natural uranium and has a power level of 225 MW. It is a graphite-moderated, carbon-dioxide-cooled reactor. The CANDU reactor is a natural uranium, heavy-water-moderated and -cooled reactor with a power level of 693 MW. The Savannah River Reactor is a heavy-water-moderated reactor and consists of both driver elements and target elements.

C. Results and Discussion of Results of Reactor Physics Calculations

The models used are quite simple and do not take into account spectral changes resulting from temperate changes, pressure changes across the core, control rods, fuel management over the life of the core, effects of different initial enrichments, and power levels. These parameters will be considered and parameter studies made to ascertain the effects on the xenon and krypton production.

The RBMK, CANDU, and Savannah River Site reactors will also be revisited and several calculations remade with a lattice physics code to ascertain the effects of the approximations made in their geometry. A second approximation was made with the RBMK reactor. Because of the RBMK reactor's large size, single transport calculations exceeded 10 hours and had difficulty converging; thus, an infinite lattice approximation was used in the transport calculations. This approximation affected the neutron spectrum, but it is not expected to be a serious defect because of the RBMK's size. However, this problem will also be revisited.

In Figures 1 and 2, as an example of the results, the ratios of $^{131}Xe/^{134}Xe$ and $^{132}Xe/^{134}Xe$ for three reactor types are plotted. The use of Xe isotopic ratios have been previously suggested[4,5] along with other ratios to determine burnup and other reactor characteristics, and the statistical analysis made here on the results confirm that this is possible. It may be noted in Figures 1 and 2 that considerable differences exist between the ratios presented. Adding more isotopes to the analysis considerably enhances the differentiation between burnup and reactor type. In the next section, an analysis of a specific gas sample is made.

Figure 1. $^{131}Xe/^{134}Xe$ Ratios as a Function of Burnup for Various Reactor Types

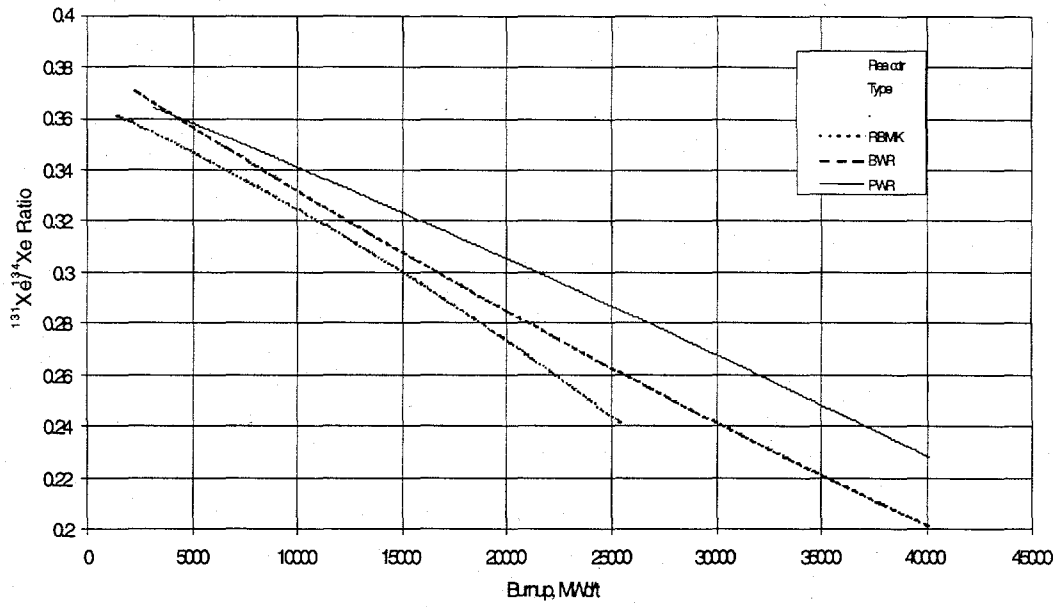
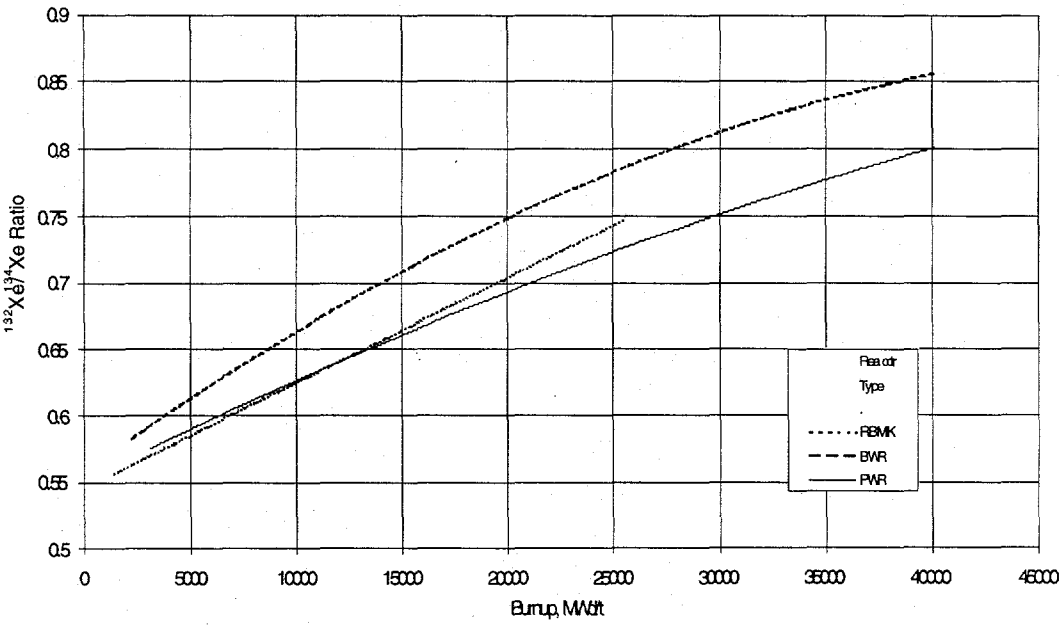


Figure 2. $^{132}Xe/^{134}Xe$ Ratios as a Function of Burnup for Various Reactor Types



III. STACK SAMPLE ANALYSIS

In the following sections, we will discuss the collection of a gas sample, its analysis, and comparison of the results with those from a transport and burn calculation. This comparison combines the results of the mass spectrometer analysis and the calculational results from reactor physics models of the SRS target fuels in a Bayesian[6,7] framework to extract the burnup from the stable xenon isotopic data. This analysis will demonstrate the validity of the methodology.

A. Sample Collection and Analysis

Eight whole-air samples were collected from a Savannah River Site reprocessing plant stack during the dissolution of 14.7 metric tons of Mk-31A depleted uranium, aluminum-clad target fuel elements. The samples were scheduled to coincide with the two additions of nitric acid ("cuts") that characterize the dissolution process.

The second sample taken at the peak of the first dissolution was analyzed three times on a microwave ion source/mass spectrometer. Analyses of air were interspersed with the sample analyses. The mass spectrometer was equipped with a single collector at the time, resulting in roughly 1% accuracy in the isotopic ratios. Results are shown in Table 1.

These data are corrected for mass fractionation using a comparison of the air samples with standard air values. These results show a marked increase over the background for the heavier isotopes, despite the low burnup of the fuel.

B. Data Analysis and Interpretation

In this section, a method for solving the "inverse problem" of inferring the fuel burnup from the xenon isotopic data is discussed. Knowledge of the burnup is critical for judging whether the fuel being reprocessed is reactor- or weapon-grade. More broadly, devising general methods for inferring facility operational parameters from environmental data is of prime importance for a wide range of nonproliferation and arms control applications.

C. Methodology

We have a data set D, which consists of the increments of the stable xenon isotopic ratios above the atmospheric background (the fourth column in Table 1) together with their measurement uncertainties (the fifth column in Table 1). The parameter we want to estimate is B, the fuel burnup. The model for the data is the following:

$$D_i = a_i \alpha B + e_i,$$

where the $\{e\}$ are Gaussian errors with known variances $\{\sigma\}$, given in Table 1. The $\{a\}$ are coefficients generated by a combined transport-decay reactor model using LANL's CINDER decay code and DANT discrete-ordinates transport code. The linearity of the model in burnup is because at small burnups the heterogeneous driver-target system has a linear response. The parameter α represents the effect of dilution within the F Canyon stack. It is a nuisance parameter; something of no interest to us, but which unavoidably enters the analysis.

Table 1. Analytical Results for Stable Xenon

Isotope	Sample 2 Average	Air Xe	Increment	Sigma
129	=1	=1	—	—
130	0.156	0.154	—	—
131	0.878	0.801	0.0770	0.001
avg. 132	1.13	1.02	0.110	0.001
134	0.580	0.395	0.185	0.001
136	0.607	0.335	0.272	0.001

The rules of probability theory relate the probability distribution function (pdf) for the burnup given the data (usually known as the posterior distribution) via the equations:

$$p(\alpha, B|D, I) \propto p(D|\alpha, B, I) \times p(\alpha, B|I)$$
$$p(B|D, I) \propto \int d\alpha p(\alpha, B|D, I).$$

The nuisance parameter representing the dilution is taken care of by integrating it out. This is a special example of a standard and useful technique known as marginalization. I represents background knowledge that is relevant to the problem. By indicating it explicitly, we can see how probability theory automatically integrates the data with any other relevant pieces of information—an essential requirement for the effective use of all the information at hand.

The problem is now reduced to evaluating the two pdfs on the right-hand side of these equations and then doing an integral. The first pdf is known as the likelihood and the assumption of Gaussian errors implies that it can be written as:

$$p(D|\alpha, B, I) = C \exp\left[-\frac{1}{2}\Sigma w_i (D_i - a_i \alpha B)^2\right],$$

where $w_i = 1/\sigma_i^2$, and the Cs that appear here and below are normalization constants.

The second pdf is known as the prior distribution. We assume that α and B are independent parameters, which allows us to factor the prior:

$$p(\alpha, B|I) = p(\alpha|I)p(B|I) = (const.)p(\alpha|I),$$

where we have assumed that the prior for the burnup is constant for B positive (and zero for B negative). This assumption amounts to saying that our prior information gives us no special knowledge about the burnup (other than the fact that it must be positive).

However, we do have prior knowledge about the stack dilution. It can be written in terms of the known stack volumetric flow rate, F, and the dissolution throughput (in tU/s), r, as:

$$\alpha = \frac{r}{F\left[^{129}Xe\right]}.$$

where $[^{129}Xe]$ is the atmospheric concentration of ^{129}Xe. (This constant is needed to convert the amounts of the heavier xenon isotopes computed by the reactor models into ratios with respect to ^{129}Xe that are the measured data.)

The numerical values of these parameters are known (see below) as part of the background information of the problem. We parametrize our knowledge of them by assigning a fractional uncertainty δ in a Gaussian prior as follows:

$$p(\alpha|I) = C \exp\left[-\frac{(\alpha - \hat{\alpha})^2}{2\hat{\alpha}^2\delta^2}\right],$$
$$\hat{\alpha} = \frac{r}{F\left[^{129}Xe\right]}.$$

Now insert these assignments into Bayes' theorem and compute the integral. There results the posterior pdf:

$$p(B|D, I) = \frac{C'}{\sigma_B} \exp\left[-\frac{(B - B_0)^2}{2\sigma_B^2}\right]$$
$$B_0 = \frac{1}{\hat{a}} \frac{\Sigma waD}{\Sigma wa^2}$$
$$\sigma_B^2 = \frac{1}{\hat{a}\Sigma wa^2} + B^2\delta^2.$$

This is the key result of the analysis. It demonstrates how the data can be effectively integrated with each other and with background information to yield the probability distribution of the burnup. The posterior is a Gaussian centered on B_0, with a spread given by σ_B.

D. Numerical Results

The production coefficients $\{a\}$ given by the reactor models are listed in Table 2. The units are such that if the burnup is given in MWd/tU, then a_iB gives the amount of isotope ^{i}Xe in the spent fuel in mol/tU.

Table 2. Production Coefficients

Isotope	Production Coefficients {a}
131	1.35E-04
132	2.06E-04
134	3.40E-04
136	5.45E-04

From the background knowledge of the November dissolution, we can estimate the throughput from the fact that 14.7 tU were dissolved in about 14 hours, yielding $r = 2.9 \times 10^{-d}$ tU/s. The stack flow rate has a nominal value of about 250 000 CFM, or about 118 m^3/s. The background concentration of ^{129}Xe is known to be about 9.3×10^{-7} mol/m^3.

The combined uncertainty in these numbers is represented by δ. To be conservative, we will assign two fairly large values to δ: 0.2, and 0.4. These values should ensure that our burnup prediction does not reflect an overly optimistic assessment of our background knowledge.

With these numerical values, the burnup inference from the isotopic data and background knowledge may be summarized as in Table 3. Note that the true burnup[8] is clearly within the inferred burnup with error bars.

Table 3. Summary of Burnup Inference

δ	Inferred Burnup (MWd/tU)	True Burnup (MWd/tU)
0.20	195 ± 39	178
0.40	195 ± 78	178

IV. CONCLUSIONS

This paper discussed use of the stable isotopes of the fissiogenic stable noble gases, Xe and Kr, for determining the burnup characteristics, fuel type, and the reactor type of the fuel from which the sample was obtained. The results of the reactor physics calculations and the Bayesian analysis of the results confirmed the validity of the methodology.

It is our firm belief that the methods of inference advocated above can be fruitfully applied to a wide range of environmental monitoring problems of nonproliferation and arms control interest.

V. REFERENCES

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