ANL/TD/CP-95328

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By:

S. E. Aumeier and J. H. Forsmann

Technology Development Division
Engineering Division
Argonne National Laboratory-West
P. O. Box 2528
Idaho Falls, ID 83403-2528

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To be presented
at
American Nuclear Society Third Topical Meeting
DOE Spent Fuel and Fissile Materials Management
September 8-11, 1998
Charleston, South Carolina

* Work supported by the U. S. Department Energy, Office of Nuclear Energy, Science and Technology, and the Office of Environmental Management, under contract W-31-109-Eng-38.

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S. E. Aumeier and J. H. Forsmann
Argonne National Laboratory - West
P.O. Box 2528
Idaho Falls, Idaho 83403

Abstract

The ability to nondestructively determine the presence and quantity of fissile and fertile nuclei in various matrices is important in several nuclear applications including international and domestics safeguards, radioactive waste characterization and nuclear facility operations. Material irradiation followed by delayed neutron counting is a well known and useful nondestructive assay technique used to determine the fissile-effective content of assay samples. Previous studies have demonstrated the feasibility of using Kalman filters to unfold individual isotopic contributions to delayed neutron measurements resulting from the assay of mixes of uranium and plutonium isotopes. However, the studies in question used simulated measurement data and idealized parameters. We present the results of the Kalman filter analysis of several measurements of U/Pu mixes taken using Argonne National Laboratory's delayed neutron nondestructive assay device. The results demonstrate the use of Kalman filters as a signal processing tool to determine the fissile and fertile isotopic content of an assay sample from the aggregate delayed neutron response following neutron irradiation.

I. Introduction

The nondestructive assay (NDA) of samples containing fissionable isotopes using neutron irradiation followed by delayed neutron (DN) counting is a well known procedure with demonstrated utility in a variety of applications.[1,2,3] Typically, such an assay involves irradiation of the fissile material bearing sample followed by integration of the delayed neutron response over some specified counting time. This gross delayed neutron response is then related to the equivalent fissile mass of the assay sample via a predetermined linear calibration curve or calibration standard. Although such a procedure may be capable of yielding an accurate estimate of the total fissile mass or mass of a dominant fissionable isotope contained in an assay sample, it does not provide a direct estimate of individual fissionable isotope content. However, there is isotope specific information in the temporal delayed neutron signal due to differences in the delayed neutron precursor yields resulting from the fission of different isotopes.[1]

Determination of individual fissionable isotope concentrations in an assay sample may be of interest in many applications, particularly when the assay sample contains significant quantities of both ^{235}U and ^{239}Pu. Recently, two analysis techniques were proposed[4] to extract isotope specific information from an aggregate temporal delayed neutron signal. The techniques, Kalman filtering and genetic algorithms, operate on the temporal delayed neutron response instead of the integrated delayed neutron signal. Using simulated delayed neutron data, studies have shown[4] that both techniques are capable of unfolding individual isotopic contributions to an aggregate delayed neutron signal and thus are capable of estimating isotopic masses. We present the preliminary results of the analysis of Kalman filter performance in determining individual U/Pu isotope concentrations in mixed-isotope samples assayed at Argonne National Laboratory-West (ANL-W).

II. The Kalman Filter

Kalman filtering is a well known and widely applied signal processing technique[5,6,7] used to obtain best estimates of model parameters given known dynamical system models, observation-state relationships, and noisy observation data. When noise in system observations is known to be zero-mean and Gaussian, the Kalman filter yields a minimum variance, unbiased estimate of system state parameters. The development of the Kalman filter algorithm has been presented in numerous signal processing and probability texts, e.g., References 5 through 7, and is summarized in Figure 1 and Table I.

The Kalman filter provides an ideal means with which to analyze delayed neutron signals for determination of isotopic masses. We consider the assay of a sample containing 4 fissionable isotopes, ^{238}U, ^{235}U, ^{239}Pu, and ^{240}Pu. For brevity, we ignore cosmic ray induced background or any activation that may corrupt a measurement. It is assumed the

Table I: Summary of Terms for Kalman Filter Algorithm

Symbol	Description
x	State Vector
$\bar{x}_k$	Best Estimate of State Vector at Time t_k Based on Observations to Time t_{k-1}
$\hat{x}_k$	Best Estimate of System State at time t_k Based on Observations to Time t_k
y_k	Observation at Time t_k
P_k	Covariance of State Estimate $\hat{x}_k$
$\bar{P}_k$	Covariance of State Estimate $\bar{x}_k$
H_k	Observation-State Mapping Matrix
$\Phi(t_k, t_{k-1})$	State Mapping Matrix for Mapping From $t_{k-1} \rightarrow t_k$
K_k	Kalman Gain Based on Observations to Time t_k
ξ_k	Observation/Calculation Residual (Observation - Calculation)
$G(x)$	Observation-State Relationship

assay sample is irradiated, the irradiating source turned off and delayed neutron counting begun following a short delay time. The irradiating source is monitored separately. The process of irradiation followed by delayed neutron counting is carried out for N cycles. The data is assumed taken in list mode and processed (integrated) with a desired dwell time. The system state x is taken to be the cumulative delayed neutron count $c(t)$ expected in time δ about time t. For 4 isotopes, 6 temporal groups, and constant irradiation cycles we have:

$$c(t) = \sum_{j=1}^{4} m_j \sigma_j \sum_{i=1}^{6} \frac{A_{ji}}{\lambda_{ji}} \beta_{ji} (1 - \exp(-\lambda_{ji}\tau)) \times (1 - \exp(-\lambda_{ji}\delta)) \exp(-\lambda_{ji} t) \times \sum_{n=1}^{N} \sum_{l=1}^{n} \exp(-\lambda_{ji}[n-l]\theta) \times \sum_{m=1}^{M} \exp(-\lambda_{ji}[M-m]\tau) \quad (1)$$

where

m_j = mass of isotope j

σ_j = fission cross section of isotope j

λ_{ji} = decay constant, isotope j and temporal group i

β_{ji} = group yield, isotope j and temporal group i

A_{ji} = constant to account for flux, detection efficiency, etc.

t = time since end of irradiation

τ = channel dwell time (irradiation monitor)

δ = channel dwell time (delayed neutron counts)

Θ = total cycle time (delays + irradiation time + counting time)

n = cycle number

N = total number of cycles

m = source monitor intervals

l = elapsed cycle index

M = total number of source monitor intervals.

Equation (1) is derived from the well known equation for radioactive decay, e.g., see reference 8. For small δ, Equation (1) is then easily differentiated to yield the dynamic system state model $F(x,t)$ for delayed neutron emission:

$$F(x,t) \equiv \dot{c}(t) = \sum_{j=1}^{4} m_j \sigma_j \sum_{i=1}^{6} -A_{ji}\beta_{ji} \times (1 - \exp(-\lambda_{ji}\tau))(1 - \exp(-\lambda_{ji}\delta)) \times \sum_{n=1}^{N} \sum_{l=1}^{n} \exp(-\lambda_{ji}[n-l]\theta) \times \sum_{m=1}^{M} \exp(-\lambda_{ji}[M-m]\tau). \quad (2)$$

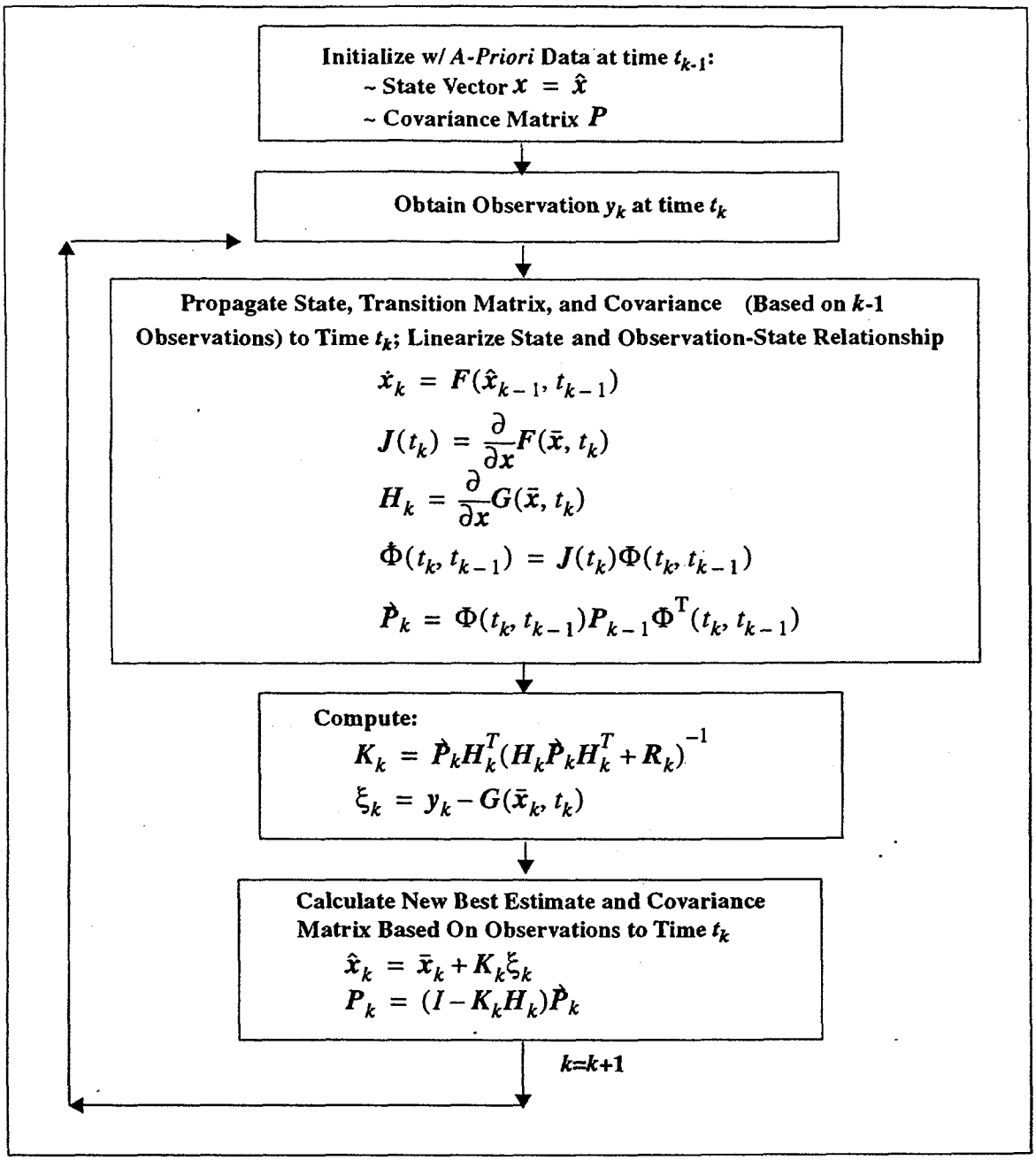

Figure 1. Kalman Filter Algorithm

When assaying samples containing both fissile and fertile isotopes, it is useful to perform two separate assays to obtain addition fissile/fertile isotope information. One irradiation is performed using an incident neutron flux of average energy e_1 and one using an incident neutron flux of average energy e_2 where e_1 is typically greater than 1.5 MeV and e_2 is less than 1 MeV. Thus, we assume two separate delayed neutron measurements are available, c_1 and c_2 corresponding to irradiation spectra e_1 and e_2, respectively. The system state x is then augmented with the constants of interest (4 isotopic masses) and the additional measurement. The resulting state vector is taken to be $z = [c_1, c_2 | m_1, m_2, m_3, m_4]$ with the corresponding dynamical system model defined as:

$$F(z,t) \equiv \begin{bmatrix} \dot{c}_1(t) \\ \dot{c}_2(t) \\ \dot{m}_1(t) \\ \dot{m}_2(t) \\ \dot{m}_3(t) \\ \dot{m}_4(t) \end{bmatrix} = \begin{bmatrix} \displaystyle\sum_{j=1}^{4} m_j \sigma_j \sum_{i=1}^{6} -A_{ji}\beta_{ji}(1-\exp(-\lambda_{ji}\tau))(1-\exp(-\lambda_{ji}\delta)) \\ \cdot \exp(-\lambda_{ji}t) \displaystyle\sum_{n=1}^{N}\sum_{l=1}^{n} \exp(-\lambda_{ji}[n-l]\theta) \sum_{m=1}^{M} \exp(-\lambda_{ji}[M-m]\tau) \\ \displaystyle\sum_{j=1}^{4} m_j \sigma^*_j \sum_{i=1}^{6} -A^*_{ji}\beta^*_{ji}(1-\exp(-\lambda^*_{ji}\tau))(1-\exp(-\lambda^*_{ji}\delta)) \\ \cdot \exp(-\lambda^*_{ji}t) \displaystyle\sum_{n=1}^{N}\sum_{l=1}^{n} \exp(-\lambda^*_{ji}[n-l]\theta) \sum_{m=1}^{M} \exp(-\lambda^*_{ji}[M-m]\tau) \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix} \tag{3}$$

where $\sigma, A, \beta, \lambda$ and $\sigma^*, A^*, \beta^*, \lambda^*$ are model constants associated with the e_1 and e_2 irradiations, respectively.

With this system description, the objective is the following: Expose an assay sample containing fissionable material to a neutron flux. Remove the neutron source and measure the cumulative delayed neutron response $y(t)$. Use Eq. (3), an observation-state relationship $H = \begin{bmatrix} 1 & 1 & 0 & 0 & 0 & 0 \end{bmatrix}$, and the Kalman filter algorithm of Figure 1 to estimate the mass of the individual fissionable isotopes contained in the sample using the cumulative temporal delayed neutron response.

III. Measurement Description

An assessment of the performance of Kalman filters in DN-NDA data processing, using simulated DN measurements, was presented in Reference 4. To test the capability of the algorithm and the approach using actual measurement data, delayed neutron nondestructive assays were taken of uranium and plutonium foils at the ANL-W DN-NDA facility. The DN-NDA apparatus consists of a 14 MeV neutron generator surrounded by a ~85 cm 4π shield of borated polyethylene. Neutrons exit the shield through a cylindrical opening with a diameter of 5 cm and enter a 71 cm x 71 cm x 66 cm block of lead which contains the assay container in its center. The purpose of the lead is to shield the ^{3}He neutron detectors against high gamma radiation associated with waste samples. The minimum thickness of the lead between the assay can and the detectors is 20 cm. The three sides of the lead shield not facing the neutron source and the bottom have 71 cm x 64 cm x 10 cm thick polyethylene blocks attached which serve as neutron moderators for 40 ^{3}He counters. The irradiating source is monitored using 4 fission chambers distributed within the assay room.

The assay sample is contained in a process scrap container (PSC), a cylindrical steel can with a diameter of 20.3 cm and a height of 22.2 cm. On the top of the PSC is a lead shield plug of 20 cm height. The PSC and the shield plug are in an outer can which is then placed into the counter through a vertical shaft from the top. The can is surrounded by 0.32 cm of cadmium in order to eliminate thermal neutrons. The assay sample is continuously rotated during the irradiation and measurements in order to average over inhomogeneous angular material distribution and detector efficiency variations. A retractable tantilum and polyethylene spectrum moderator is attached to a rail located between the neutron generator and the lead blocks.

Uranium and plutonium foils were distributed in a matrix of stainless steel cladding hull sections in the PSC. Both individual isotope and mixed isotope (aggregate) samples were assayed. Assays were first taken of individual samples of ^{238}U (111.95 g), ^{235}U (34.79 g), and ^{239}Pu (31.23 g) with the spectrum moderator inserted and retracted, resulting in individual calibration measurements c_1 and c_2 for each of the 3 isotopes. The mass of ^{240}Pu in the samples was negligible.

The samples were irradiated for 15 sec and the irradiating source turned off. Delayed neutrons were then counted for 30 sec following a 1.14 sec delay. The irradiation/DN measurement procedure was repeated (cycled) 200 times for the ^{238}U and ^{239}Pu samples and 40 times for the ^{235}U sample. Measurement data were integrated with a 0.5 sec dwell time. The counts from the individual cycles were summed to achieve desired statistics. The measurement data were then processed using a Kalman filter to determine assay-specific delayed neutron emission model parameters (delayed neutron constants, irradiating flux levels) for both the spectrum moderator inserted and retracted irradiations. This is accomplished by modifying Eq. (3) as:

$$F_j(z,t) \equiv \begin{bmatrix} \dot{c}(t) \\ \dot{\lambda}_1 \\ \vdots \\ \dot{\lambda}_6 \\ \dot{B}_1 \\ \vdots \\ \dot{B}_6 \\ \dot{\sigma} \end{bmatrix} = \begin{bmatrix} m\sigma \sum_{i=1}^{6} -B_i(1-\exp(-\lambda_i\tau))(1-\exp(-\lambda_i\delta))\exp(-\lambda_i t) \\ \cdot \sum_{n=1}^{N}\sum_{l=1}^{n} \exp(-\lambda_i[n-l]\theta) \sum_{m=1}^{M} \exp(-\lambda_i[M-m]\tau) \\ 0 \\ \vdots \\ 0 \end{bmatrix} \quad (4)$$

where the isotopic mass m is a known. This process was carried out for each irradiation (e_1 and e_2) and for each isotope. The constants appearing in the state of Eq. (4) for each individual isotope are then used in the process model of Eq. (3) when processing DN data from aggregate assay samples.

Three separate mixes of U/Pu were then assayed including 16.161 g ^{235}U and 109.597 g ^{238}U (Mix #1), 34.792 g ^{235}U, 2.45 g ^{238}U and 47.139 g ^{239}Pu (Mix #2), and 10.069 g ^{235}U, 0.709 g ^{238}U, and 6.209 g ^{239}Pu (Mix #3). These aggregate assay measurements, with the model constants determined from the single isotope calibration measurements, were then used to test the isotope discrimination capabilities of the Kalman filter algorithms.

IV. Results

The robustness of the calibrated models were first evaluated by processing the individual isotope calibration measurements using a Kalman filter with a priori isotope mass significantly in error from the correct mass. Both measurements c_1 and c_2 were processed simultaneously (see Reference 4). The results are presented in Table 2. The results show that the model developed from the calibration data provides for an accurate prediction of ^{235}U and ^{239}Pu mass and a marginal prediction of ^{238}U mass. The poorer performance of the ^{238}U model is due to the low DN count rate obtained from this fertile isotope. One would expect the ^{235}U and ^{239}Pu models to be representative of the DN measurement of similar assay samples and thus provide the necessary model structure for the Kalman filter. However, the poorer representation provided by the ^{238}U model constants will limit the accuracy of isotope estimates for samples containing a significant quantity of this isotope.

The constants (models) corresponding to the individual isotope measurements were then combined to create an aggregate isotope model descriptive of the mixed-isotope measurements. The three mixed-isotope measurement files were then processed using a Kalman filter to test the isotope discrimination capabilities of the Kalman filter. Results are presented in Table 1 and show accuracies better than 7.5% for small total fissile loading (Mix #3) and better than 4.5% for larger loadings (Mix #2). Estimates of total fissile mass were better than 2.5% in all cases. All estimates were obtained by running 100 iterations of the Kalman filter.

It was found that filter performance was not sensitive to the choice of a priori mass or covariance but was sensitive to the ratio of measurement errors for the spectral filter removed (c_1) and spectral filter inserted (c_2) data. Actual total error (statistical + systematic) is ~4%. By performing a parametric study using measurement data from Mix #3, optimal values of measurement error were found to be 3.6% for the

Table II: Model Performance

	^{235}U	^{238}U	^{239}Pu
Actual Mass (g)	34.79	111.95	31.23
A Priori Estimate (g)	100.0	100.0	10.0
Estimated Mass (g)	35.22 (+1.23%)	122.88 (+9.8%)	31.67 (+1.4%)

spectrum moderator out data and 3.0% for the spectrum moderator in data. The same values were also found to be optimal for the Mix #2 data set. However, for Mix #1 this choice of measurement errors resulted in a ^{235}U mass estimate error of 1.9% (comparable to that presented in Table 1) and 15% for ^{238}U (much larger than that presented in Table 1). The results for Mix #1 of Table 1 were obtained using measurement error estimates of 20% for spectral filter out data and 3% for spectral filter in data. This difference in performance is most likely due to the large ^{238}U loading in Mix #1, which is poorly represented by the models used in this analysis.

V. Summary and Conclusions

Reference 4 outlines the development of the Kalman filter algorithm for use in DN-NDA data analyses and presents an assessment of those algorithms using simulated data. We have shown in this work that it is possible to obtain accurate estimates of the mass contained in U or U/Pu mixed-isotope assay samples assayed at the ANL-W DN-NDA facility. This is accomplished by performing both individual isotope calibrations to develop suitable delayed neutron emission models and mixed isotope calibrations, similar in isotopic content to the assay sample of interest, to obtain optimal measurement error estimates. The results presented here demonstrate the capability of the Kalman filter in unfolding individual isotopic contributions to an aggregate delayed neutron measurement and determining total fissile mass with relatively good accuracy.

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Table III: U/Pu Assay Results

	Mix #1	Mix #2[a]	Mix #3[a]
^{235}U:			
Actual Mass (g)	16.161	34.792	10.069
A Priori Estimate (g)	50.0	20.0	15.0
Estimated Mass[b] (g)	16.06 (-0.6%)	35.07 (+0.8%)	9.32 (-7.4%)
^{238}U:			
Actual Mass (g)	109.597	2.450	0.709
A Priori Estimate (g)	150.0	2.450[c]	0.709[c]
Estimated Mass[b] (g)	111.28 (+1.5%)	N/A	N/A
^{239}Pu:			
Actual Mass (g)	0	47.139	6.209
A Priori Estimate (g)	N/A	60.0	3.20
Estimated Mass[b] (g)	N/A	45.23 (-4.1%)	6.58 (+6.0%)
Total Fissile Estimate (g)	16.06 (-0.6%)	80.3 (-2.0%)	15.9 (-2.4%)

a. Small quantities of ^{240}Pu neglected.
b. Percent error in parentheses.
c. Correct a priori mass with small covariance—no adjustment made.

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