NUCLEAR SCIENCE AND ENGINEERING: 20, 359-364 (1964)

Letters to the Editors

A Note on Delayed-Neutron Effects in Pulsed-Neutron Measurements on Multiplying Assemblies*

In pulsed-neutron measurements on subcritical multiplying systems[1,2], the delayed neutrons appear as a long 'tail' superimposed on the prompt-neutron decay following the source burst. If, as is frequently the case, the time of observation is short compared with the mean lifetime of the shortest-lived delayed-neutron precursor, the delayed-neutron component can be regarded as substantially constant and treated as a background to be subtracted from the observed decay to give the decay of the prompt neutrons alone. If higher harmonics are absent, this decay will be represented by a single exponential function, $\exp(-\alpha t)$, where α is the decay constant of the fundamental mode. In hydrogen-moderated systems where the prompt-neutron lifetime is on the order of 10^{-4} sec, the decay constant α will be on the order of 100 sec^{-1} in a near-critical assembly fueled with U^{235}, so that observation times of a few time $1/\alpha$ will be on the order of 0.01-0.02 sec, short compared with the emitter mean lifetimes. In well moderated systems using graphite or D_2O as moderator, however, the prompt-neutron lifetime may be on the order of 10^{-3} sec, α (near critical) on the order of 10 sec^{-1}, and observation times on the order of a few tenths of a second, comparable to the lifetimes of the shortest-lived emitters in U^{235}. It is therefore necessary in such systems to take the time dependence of the delayed neutrons explicitly into account in the analysis of the experiments. Fultz[2] has treated this problem in an approximate manner by lumping the various delayed-neutron groups into a single group. Analytic solutions for the delayed components have been given by Garelis and Russell[3] in connection with the '$k\beta/\ell$' method, but their solutions are not readily applied to measurements of the type under discussion here. More recently Küchle[4] has applied a method due to Fraude[5] to the analysis of pulsed-neutron measurements in a D_2O-moderated reactor. At BNL we have encountered this problem in the course of pulsed-neutron experiments on U^{235} graphite assemblies during the past year and a half. The method of correcting for delayed-neutron effects, presented below, differs somewhat from those described in Refs. 2 and 4, however, and is especially suited for calculation by a fast digital computer.

It is assumed in this method that a $t=0$ there is an infinitely short burst of thermal neutrons in the subcritical assembly from an external source, and that the essential features of the subsequent behavior of the neutron population are sufficiently well described by a single-energy group, m-delayed-neutron group model. It is also assumed that at all times the neutrons are distributed in the fundamental mode, so that $\nabla^2 n(\vec{r}, t) = -B^2 n(\vec{r}, t)$, where $n(\vec{r}, t)$ is the neutron density at the point $\vec{r}$ and the time t, and B^2 is the buckling. Under these circumstances the functions $n(\vec{r}, t)$ and $C_i(\vec{r}, t)$, where the latter is the concentration of the i-th delayed-neutron precursor, are separable in space and time and, since we are concerned only with the time-dependent parts, the spatial argument will be omitted in what follows. It is further assumed that the neutron density can be written as a sum of two terms,

$$n(t) = n_P(t) + n_D(t), \qquad (1)$$

where $n_P(t)$ is a function that decays exponentially with decay constant α; i.e.,

*Work performed under the auspices of the USAEC.

[1] B. E. SIMMONS and J. S. KING, "A Pulsed Neutron Technique for Reactivity Determination," *Nucl. Sci. Eng.* **3**, 595 (1957).

[2] S. C. FULTZ, "The Time-Dependent Thermal Neutron Flux from a Pulsed Subcritical Assembly," *Nucl. Sci. Eng.* **6**, 313 (1959).

[3] E. GARELIS and J. L. RUSSELL, Jr., "Theory of Pulsed Neutron Source Measurements," *Nucl. Sci. Eng.* **16**, 263 (1963).

[4] M. KÜCHLE, "Pulsed Neutron Measurements on a Heavy Water Reactor at Zero Energy," Preprint SM-42/6, IAEA Symposium on Exponential and Critical Experiments, Amsterdam, (Sept. 1963).

[5] A. von FRAUDE, "Theoretische Untersuchungen Zur Reaktivitätsmessung an einem Reaktor Mit Reflektor mit Hilfe einer gepulsten Neutronenquelle," *Nukleonik* **4**, 84 (1962).

$$\frac{\partial n_P(t)}{\partial t} = -\alpha n_P(t). \tag{2}$$

The initial conditions on $n_P(t)$ and $n_D(t)$ are that $n_P(0) = n(0) = n_0$ (the initial density of source neutrons), and $n_D(0) = 0$ (that is, at $t = 0$ there are no delayed neutrons). With the foregoing assumptions the following equations can be derived from the reactor kinetic equations:

$$\frac{\partial n_D}{\partial t} = -\alpha n_D + \frac{k_e \beta}{\ell} \sum_{i=1}^{m} \lambda_i \chi_i \tag{3}$$

and

$$\frac{\partial \chi_i}{\partial t} = -\lambda_i \chi_i + (\beta_i/\beta) n \quad i = 1,2, \ldots, m \tag{4}$$

with

$$\alpha = -\frac{(1-\rho/\beta)}{1-\rho} (\beta/\ell) . \tag{5}$$

In these expressions

λ_i and β_i are the decay constant and delayed-neutron fraction of the i-th precursor type,

β is the total delayed-neutron fraction,

ℓ the finite-medium thermal-neutron lifetime,

k_e the effective multiplication factor of the assembly, and

ρ its reactivity.

The quantity $\chi_i(t) = [(\ell_p N_f)/(k_e \beta)] C_i(t)$, where p is the resonance escape probability and N_f the fast non-leakage probability, is a modified precursor concentration and obeys the initial condition $\chi_i(0) = 0$.

The solution of Eq. 4 for $\chi_i(t)$ can be obtained by numerical integration of the observed decay, $n(t)$, and depends only on the delayed-neutron parameters λ_i and β_i/β. In our experiments the reactivity is known and the object is to measure ℓ/β. Initial estimates of α and ℓ/β are made from $n(t)$ (graphically, for instance), and these together with χ_i are inserted into Eq. 3. Numerical integration of the latter then gives $n_D(t)$, which is subtracted from $n(t)$ to give a first approximation to the prompt-neutron component, $n_P(t)$. This, in turn, is used to get a better estimate of α, through a least-square fit to an exponential, and hence of ℓ/β. The process is iterated as often as necessary to obtain a stationary value for the error of the fit.

The foregoing applies only to the decay following a single burst. If the reactor is pulsed repetitively but at a slow enough rate that all but the longest-lived delayed-neutron precursors have died away in the interval from one pulse to the next, then all that is necessary to apply the above method is to subtract from the observed count rates the pre-burst count rate that is usually recorded in the first channel of multi-channel time analyzers. Otherwise the assumed initial condition $C_i(0) = \chi_i(0) = n_D(0) = 0$ will not apply.

The one-energy-group model used here assumes that the source neutrons are thermal and distributed spatially in the fundamental mode. Neither assumption is likely to be true, of course. In graphite, however, the slowing-down time is on the order of 10^{-4} sec so that in 10^{-3} sec, say, the initially fast neutrons have been well thermalized. The relative error made in $n_D(t)$ by using for n_0 the value obtained by extrapolation of $n(t)$ back to t=0 in an exponential fashion will be proportional to the relative difference between the integral of $n(t)$ (extrapolated exponentially to t=0) and that of the true thermal neutron distribution; this, however, in the case cited, will be quite small. On the other hand if, as is frequently the case, an appreciable delay is introduced between the source burst and the first post-burst channel of the analyzer, it is important that it be taken into account in the integrations of Eqs. 3 and 4. In the example cited below, the delay was equivalent to two channel widths or about 5000 μsec, and neglecting it introduces an error in $n_D(t)$ of 4%.

The neglect of the higher harmonics in the initial portion of the decay is justifiable if the reactor is very close to critical. In the case discussed below, the most slowly decaying higher harmonic had a computed decay constant nearly 50 times that of the fundamental; its neglect is therefore comparable, in terms of the error introduced into $n_D(t)$, to the neglect of the thermalization of the source neutrons.

Pulsed measurements on a bare U^{235} graphite critical assembly at Brookhaven provided experimental unification of the inadequacy of the "constant delayed-neutron method" of analysis in those cases where the decay times are long. This assembly was a rectangular block measuring 55 × 66 × 72 inches; the carbon-to-U^{235} atom ratio was 1.718×10^4. The pulsed-neutron source, a Kaman Model NT-60-8A Pulsatron unit[a], was located at the center of one face of the assembly, and a $B^{10}F_3$ neutron proportional counter at the center of a face making an angle of 90° with the source face. The signals from the counter were amplified and fed into a 256-channel time analyzer[b] which also triggered the source. The width per channel of the analyzer was 2560 μsec (with a 10 μsec delay between adjacent channels) and the delay between the source burst and the first post-burst channel was

[a] Kaman Nuclear Division of the Kaman Aircraft Corporation, Colorado Springs, Colorado.

[b] Technical Measurements Corporation, North Haven, Connecticut.

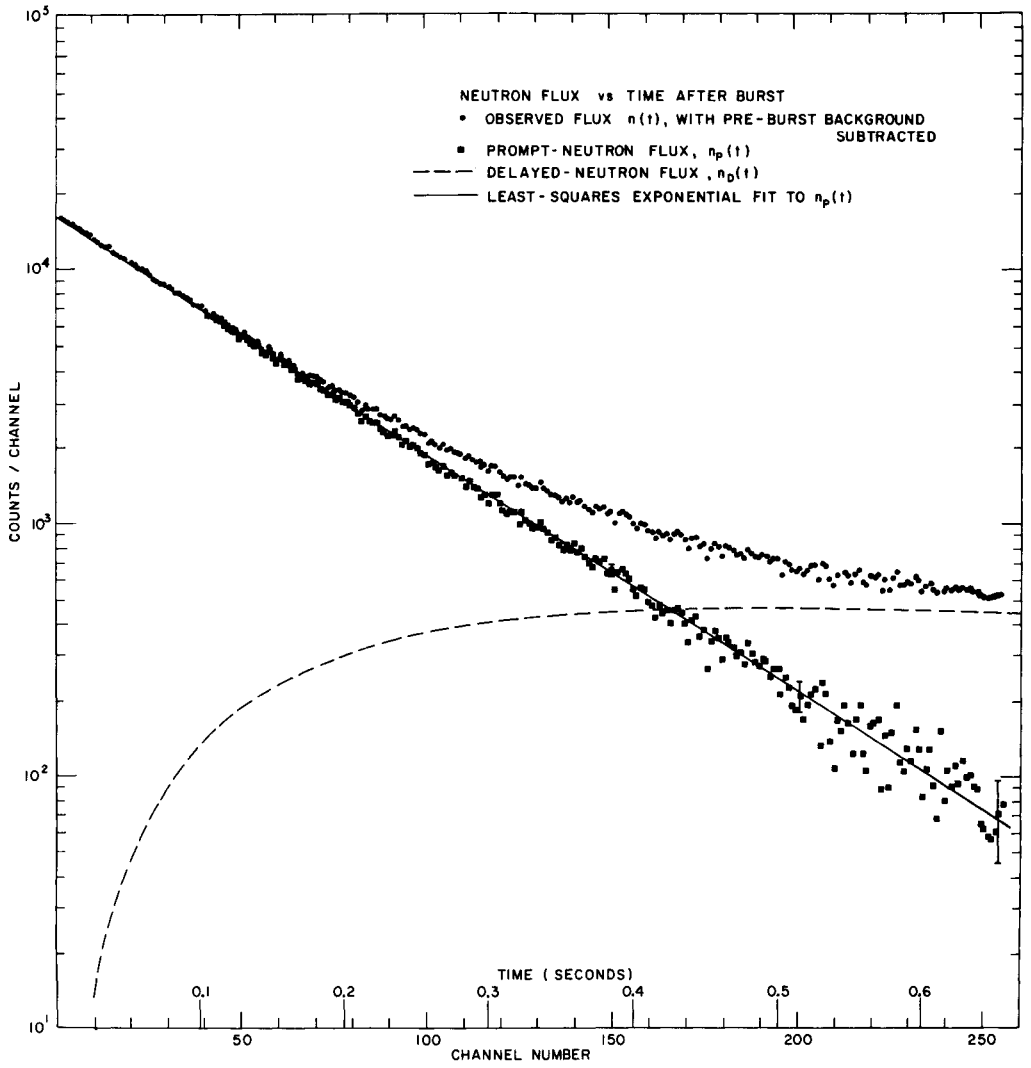

Fig. 1. Neutron decay following source burst in U^{235} -graphite assembly. Reactivity is -20 cents; channel width of time analyzer is 2560 μ sec.

set at two channel widths. The assembly was held at a reactivity of -20 cents (measured with a reactivity meter of the analog-computer type[6]) while being subjected to a total of about 100 source bursts separated in time by 3 minutes each. The observed channel count rates were entered as input data in a Fortran program that integrates Eqs. 3 and 4 numerically. Also submitted as input information were the reactivity, the channel width, the pre-burst background and an initial estimate of ℓ/β. The program computes $n_D(t)$ and $n_P(t)$ by iteration, prints out the final values of these, and also performs a least-squares fit of $n_P(t)$ over any selected range of channels to the function $\exp(-\alpha t)$ to determine α and therefore ℓ/β.

The results are given in Table I and displayed graphically in Fig. 1. Also given in the table are the results obtained by the constant-background method. In both methods, the data in the channels preceding the 31st were omitted in the least-squares analysis for α because of the large dead-time losses (>10%). It will be noted from Table I

TABLE I

Comparison of Experimental Results Obtained by Two Different Methods of Analysis. Errors quoted are statistical errors only.

Fitted Region (Channel #1)	Time-Dependent Method ℓ/B (sec)	Constant-Background Method ℓ/B (sec)
31-106 (I)	0.140 ± 0.001	0.139 ± 0.001
107-182 (II)	0.142 ± 0.002	0.147 ± 0.002

[6] C. SASTRE, "The Measurement of Reactivity," *Nucl. Sci. Eng.* 8, 443 (1960).

that the constant-background method gives results for ℓ/β in regions I and II that differ by almost 6%, whereas those obtained by the time-dependent method agree within the experimental error. For channel numbers greater than 182, the delayed-neutron correction ranges from 60 to 90% of the total signal so that the results are very sensitive to errors in $n_D(t)$; this region is therefore excluded from the analysis, as would normally be the case.

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Received March 20, 1964
Revised July 6, 1964

ACKNOWLEDGMENTS

We are grateful to Walter Kley, who assisted us in the taking of the data and the interpretation of the results, and to Joan Felberbaum, who wrote the Fortran program used in the analysis of the data.

Synergism in the Solvent Extraction of Alkali Metals*

Solvent extraction studies of the removal of fission products from nuclear fuel reprocessing wastes are under intensive study in many laboratories. Removal of strontium-90, cerium-144, promethium-147, and cesium-137 from process waste is of particular interest because of the long half-lives, intense heat generating rates and the biological hazards of these isotopes. Interest in recovery of these materials is also prompted by their potential uses as radiation and heat sources. Waste solutions from which separation of the above isotopes is normally to be effected are usually acidic and contain high concentrations of sodium, iron, and other non-radioactive isotopes. It has therefore been imperative to find an extraction technique which will remove these fission products from acid solutions with large separation factors.

Processes which extract strontium and the rare earths, but not cesium, from acidic solutions with di(2-ethylhexyl)phosphoric acid (D2EHPA) in kerosene have been developed at the Hanford Laboratories[1] and at the Oak Ridge National Laboratory[2].

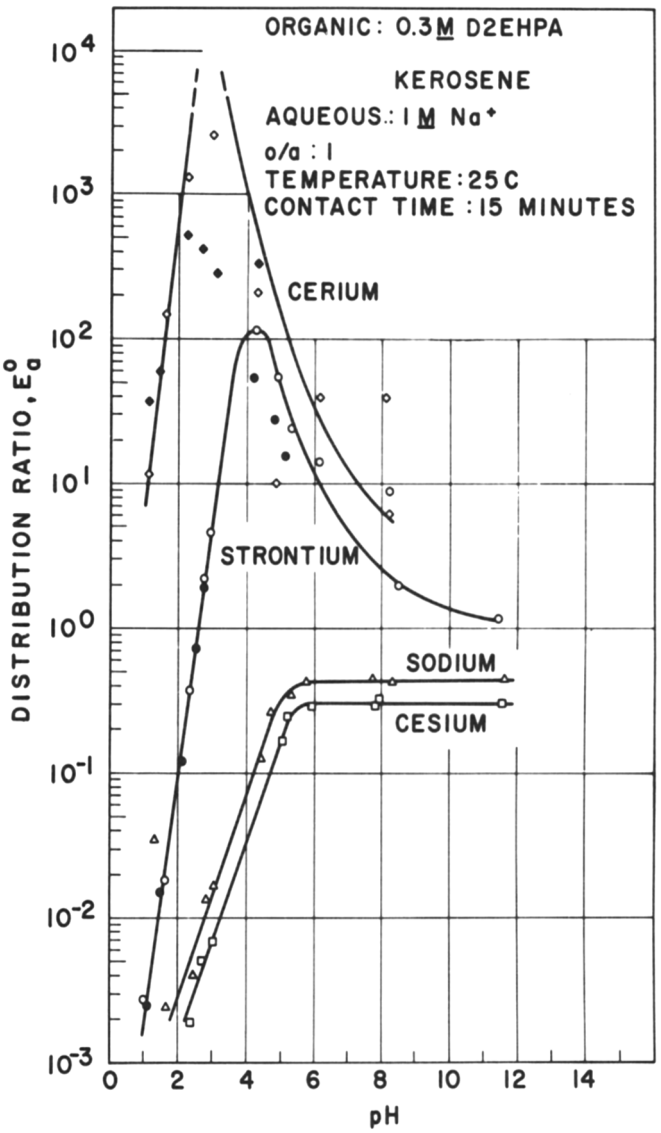

Fig. 1. D2EHPA extraction as a function of pH (open points indicate the organic contained 0.15M TBP to prevent third phase formation).

Cesium extraction has been a more difficult problem. Only moderate success was achieved in earlier work in which cesium was extracted from near neutral or alkaline solutions into high dielectric constant solutions as the dipicrylaminate, polybromide, or the polyiodide[1,3,4]. Recently,

*Work performed under Contract No. AT(45-1)-1350 for the USAEC.

[1]R. E. BURNS, W. W. SCHULZ and L. A. BRAY, "Recent Solvent Extraction Studies at Hanford Laboratories," *Nucl. Sci. Eng.* **17**, 566-575, (December, 1963).

[2]D. E. HORNER, D. J. CROUSE, K. B. BROWN and B. WEAVER, "Fission Product Recovery from Waste Solutions by Solvent Extraction," *Nucl. Sci. Eng.* **17**, 234-246, (October, 1963).

[3]LOUIS M. SLATER, "Extraction of Sodium and Cesium Polyiodides into Nitrobenzene," *Nucl. Sci. Eng.* **17**, 576-585, (December, 1963).

[4]F. C. W. PUMMERY and J. H. MOSS, "The Solvent Extraction of Cesium by the Poly-Bromide Method from Mock Windscale Highly Active Wastes," AERE-R-3596, Harwell, (December, 1960).