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A METHOD FOR NEUTRON DOSIMETRY IN ULTRAHIGH FLUX ENVIRONMENTS

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ABSTRACT

A method for neutron dosimetry in ultrahigh flux environments is developed, and devices embodying it are proposed and simulated using a Monte Carlo code. The new approach no longer assumes a linear relationship between the fluence and the activity of the nuclides formed by irradiation. It accounts for depletion of the original "foil" material and for decay and depletion of the formed nuclides. In facilities where very high fluences are possible, the fluences inferred by activity measurements may be ambiguous. A method for resolving these ambiguities is also proposed and simulated. The new method and proposed devices should make possible the use of materials not traditionally considered desirable for neutron activation dosimetry.

1. Introduction

The motivation to devise a neutron activation dosimetry method for very high flux (or fluence) situations was initially provided in a talk by C. D. West about dosimetry in the (then proposed) Advanced Neutron Source reactor.[1] Among the problems that may arise in using traditional dosimetry foils in ultrahigh flux environments are the burnout or depletion of the foil material, the saturation of counting equipment by high decay rates, significant personnel doses from irradiated foils, and non-linearity of foil response to fluence.

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One approach used in the past to perform neutron dosimetry in some of the high flux facilities of the Advanced Test Reactors (ATR) includes as a crucial step the chemical dissolution of the activated foils and the careful assaying of a portion of the solution prior to counting. The method works but is cumbersome. Other steps involved computations to correct for depletion of the foil materials in long irradiations.

The method and devices proposed in this paper are a drastic generalization of these steps prior to irradiation rather than following irradiation, and purport to resolve some of the difficulties associated with dosimetry in ultrahigh flux or fluence environments.

2. Doped Wafers Concept

A material that is relatively neutronically "inert" or "transparent" is selected as a bulk material into which activation materials are dispersed uniformly. In the remainder of this paper, the background material is referred to as the bulk material, and the dispersed activation materials are referred to as dopants. By a judicious choice of the dopant or dopants included in a wafer, and their respective concentrations, a wafer can be produced that is optimized for the flux conditions under consideration and for the available counting equipment. In this manner counting rates that fall within the optimal ranges of the detectors can be produced, resolving the counting equipment saturation problem. An advantage of a multiply-doped wafer is the possibility to extract from a single wafer information that previously required a set of foils. Such a device would allow the measurement of the spectrum at a precise location in a facility where the flux or the spectrum have steep spatial gradients. Useful dopant concentrations that produce no significant flux depressions, and hence for which no self-shielding correction computations are necessary, can also be devised. Each of these features are modeled and studied in the following sections.

The difficulties arising from depletion of the original material and the depletion and decay of the activation materials are resolved by departing from the usual adherence to work within the linearity zone of the activation versus fluence relationship. Instead, depletion of the initial nuclides and decay as well as depletion of the activation product are explicitly taken into account. The resulting equations are used to produce a more rigorous activation versus fluence curve (or computer algorithm) from which counting results can be translated into fluence and hence reaction rates for the original nuclides. One of the potential difficulties of using such an approach, namely the possibility of ambiguous fluence readings from the activity curve, is resolved by a novel approach presented below.

3. Activation Equations

Traditional neutron dosimetry by neutron activation uses the linear portion of the activity versus fluence relationship. This practice inherently assumes that the fraction of

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target nuclei destroyed is negligible, that the radioisotope j produced has a neutron absorption cross section such that its radioactive decay constant λ_j is much greater than its rate of destruction by neutrons ($\lambda_j \gg \sigma_j \phi$, where σ_j is the microscopic absorption cross section for the j nuclide, and ϕ is the neutron flux), and that the irradiation times are short compared to the half-life of the activation nuclide (i.e., $\lambda_j t \ll 1$). Under these conditions, the activity is given by[2]

$$A_j(t) \approx \sigma_i N_i(0) \phi \lambda_j t \tag{1}$$

If the last condition is relaxed, the equation for activity displays a saturating behavior,

$$A_j(t) \approx \sigma_i N_i(0) \phi \left[1 - \exp\left(-\lambda_j t\right) \right] \tag{2}$$

In the present work all three assumptions are relaxed and in addition, it is assumed that the flux ϕ can be time-dependent. Under these conditions, the differential equations that govern the activity of the product nuclide cannot be fully integrated analytically. The expression for the activity $A(t, \phi(t))$ at time t caused by a time-dependent flux $\phi(t)$ is given by

$$A_j\left(t, \phi(t)\right) = \lambda_j \sigma_i^{n,\gamma} N_i(0) e^{-\lambda_j t} e^{-\sigma_j^a \Phi(t)} \int_0^t e^{\int_{t'}^{t}\left[\lambda_j + \left(\sigma_j^a - \sigma_i^a\right)\phi(t'')\right]dt''} \phi(t')dt' \tag{3}$$

where $\Phi(t)$ is the accumulated fluence at time t since the beginning of irradiation. Equation (3) assumed that initially no nuclides of species j were present.

If the total irradiation time of interest is denoted T, and if the power shape assumed to be a known function of time, $\phi(t)$, can be represented in the [0,T] interval by

$$\phi(t) = \Phi\, p(t), \tag{4}$$

where $p(t)$, which integrates to unity, is the power shape. With these definitions Φ is the final fluence, and equation (3) can be specialized to the case of the final fluence Φ for irradiation time T,

$$a_N = \frac{N_j(\Phi;T)}{N_i(0)} = \sigma_i^{n,\gamma} e^{-\lambda_j T} e^{-\sigma_j^a \Phi} E(\Phi, T). \tag{5}$$

Equation (5) is the equation for the normalized activity a_N, defined as the activity divided by the decay constant λ_j and by the initial atomic density of the target material. An expression for the term $E(\Phi;T)$ appearing in this equation is

$$E(\Phi;T) = \Phi \int_0^T e^{\int_0^t \left[\lambda_j + (\sigma_j^a - \sigma_i^a)\Phi p(t')\right]dt'} \, dt.$$

(6)

This expression can be evaluated numerically for any explicitly (numerically or analytically) prescribed normalized power history $p(t)$, maximum fluence Φ at end of irradiation, and irradiation time T. From a series of such evaluations, curves showing normalized activity a_N can be plotted. In the particular case where the power can be assumed to be constant (e.g. because the irradiation time at a constant power is much larger than the ramp up and down times), the expression for the normalized activity can be integrated. The result,

$$a_N(\Phi;T) = \frac{\sigma_i^{n,\gamma}\Phi}{\lambda_j T + (\sigma_j^a - \sigma_i^a)\Phi}\left\{e^{-\sigma_i^a\Phi} - e^{-\lambda_j T}e^{-\sigma_j^a\Phi}\right\},$$

(7)

is plotted in Figure 1 for various values of the irradiation time T and over a range of fluences that are plausible in the ANS reactor design. From curves such as these, or equivalently via a numerical procedure, the value or values of the fluence Φ that give a certain measured activity for a known irradiation time and power history can be determined. For example, in the case of a situation that can be represented by one of the curves in Figure 1, this determination has a unique solution for most cases of practical interest. In the instances where the activity can be caused by more than one plausible fluence, as shown for the normalized activity value of 0.03 in Figure 2 (17 days irradiation), the resulting ambiguity needs to be resolved. This resolution may be obvious from prior knowledge about the facility and the knowledge of the irradiation time. If this is not the case, then a method that assays the remaining atomic density of the target nuclides is applied. A possible approach is to re-irradiate the wafer immediately after counting in either the same field or an auxiliary known field. The consequence of this re-irradiation is a change of the activity of the wafer that is always an increase for the lower value of the candidate fluences and either an increase at lower rate or a decrease for the higher fluence, depending on how much time elapses between the end of the initial irradiation and the beginning of the second one. In Figure 2, two fluence levels are shown to correspond to the normalized activity of 0.03. Both of these fluences are plausible for this facility. It follows that assaying is necessary to resolve the ambiguity. For the ideal situation in which re-irradiation starts *immediately* after initial irradiation ends, the normalized activity during re-irradiation would behave just as a continuation from the two intersection points of Figure 2. In actual applications the initial activity at the beginning of the re-irradiation would have to be corrected for decay. Cases may arise for which the ambiguity is a continuum of values (plateau effect). Those cases could also be resolved via an assaying approach, but are beyond the scope of this paper.

4. Materials Characterization and Selection

Bulk materials were initially selected based on peak thermal, resonance, and fast neutron cross sections. Those selected as potential candidates had the lowest total neutron cross section. Further reduction of the substrate list was achieved by examining melting temperatures and by calculating neutron irradiation-induced activity using the ORIGEN2 code.[3] Materials were rated according to the types of activation products that resulted from irradiation. Those with interfering products, i.e., activation products which interfered with detection of dosimetry materials, were eliminated from consideration, as were those that produced long-lived activation products. The final selection for the bulk material was fused silica (SiO_2), because of its availability in very pure form and other overall desirable qualities, including chemical inertia and resistance to radiation damage over a wide range of fluences, and also because of widespread use of this material in nuclear applications.

A variety of dopant (dosimetry) materials, including the full spectrum of conventional choices, were considered for use in the new dosimeters. As the initial investigation concentrated on application to two research reactors, the Advanced Neutron Source (ANS) design and the Advanced Test Reactor (ATR), some potential candidates were rejected based on the specific criteria imposed by the operating cycles of the reactors. Candidate materials with short half-lives were initially eliminated. However, applications can be envisioned which would be well-suited to these candidates, and the actual final design of a set of dosimeters for a specific facility will necessarily include a very wide range of potential dosimetry materials.

5. Tailoring for Elimination of Self-Shielding

The design of the doped wafer devices allows their tailoring to match the counting equipment efficiency, by diluting the dopant to optimal levels. In the same way, dilution of the dopants can be used to produce devices for which the self-shielding, prevalent in foils, becomes negligibly small. When this is achieved, the often difficult self-shielding correction calculations become unnecessary, resulting in significant savings of time and effort in the dosimetry process. Dopant concentrations for dosimetry materials in the wafers that result in the elimination of the need for the self-shielding correction can be easily determined in the single dopant cases. In this work, this determination is made via simple geometry Monte Carlo transport calculations with varying concentrations of the dopant. In these simulations, the dopant concentration is increased until a region of non-linear response is encountered, or, if it occurs first, until the material is 100% dopant or an ordinary foil. Dopant concentrations below this level require no self-shielding correction. An example, the determination of self-shielding occurrence for the $^6Li(n,\alpha)^3H$ reaction is shown in Figure 3. The non-linearity is clearly seen for dopant concentrations above about 10^3 to 10^4 ppm. For dosimetry reactions with smaller cross sections, such as the $^{23}Na(n,\gamma)^{24}Na$ reaction, also shown in Figure 3, the non-linearity occurs at higher

concentrations and is less marked. Gold exhibits an intermediate behavior. From the figure it can also be seen that the self-shielding is absent for all three reactions below concentrations that correspond to achievable, and useful, dilution levels.

6. Conclusions

The doped wafer devices proposed and simulated in this work were initially intended for neutron dosimetry in ultrahigh flux or fluences situations. The simulations showed that the difficulties associated with ultrahigh flux situations, and enumerated in the introduction, are solved by the devices, using achievable and practical dopant concentrations. The examples shown in the paper relied on simplifying assumptions that made analytical solutions of the relevant differential equations possible. The use of a numerical approach for actual applications has been outlined. Besides the solution of the neutron dosimetry problem of ultrahigh flux and fluence situations, the use of complete activation and depletion equations, though a departure from conventional practice, may make possible the future use of materials not traditionally viewed as practical dosimetry targets.

7. Acknowledgments

7. References

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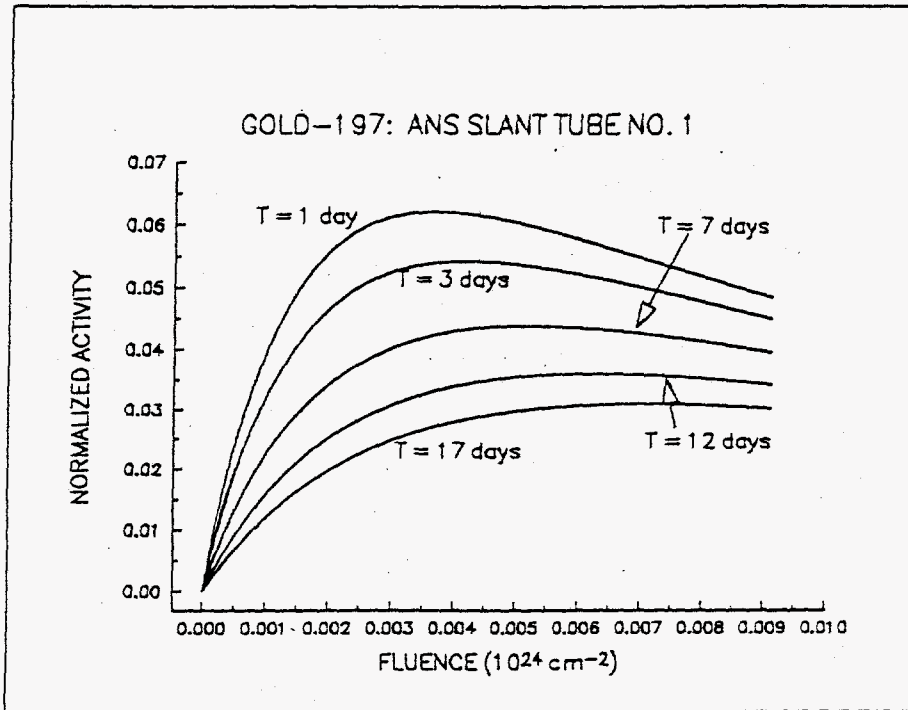
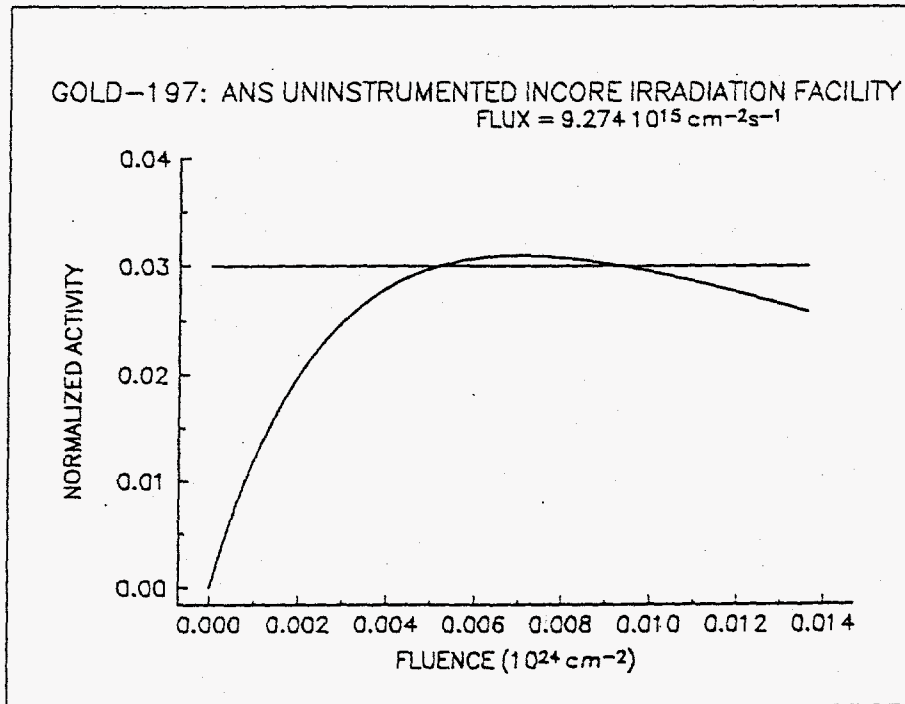Figure 1. Normalized Activity for Different Irradiation Times.

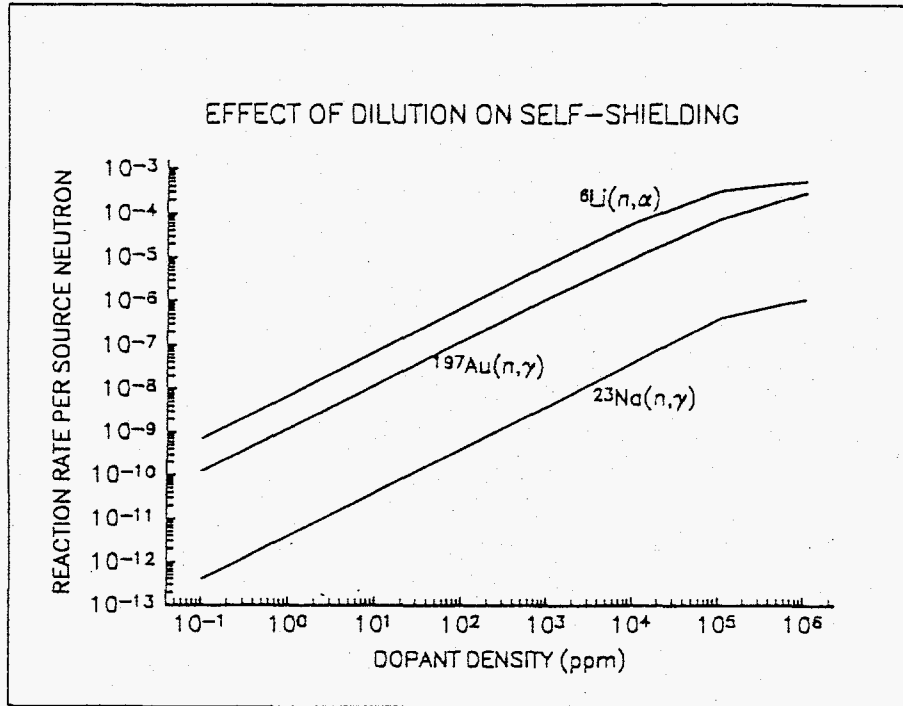Figure 2. Fluence Ambiguity in Au-197 Irradiation.

EFFECT OF DILUTION ON SELF-SHIELDING

Figure 3. Self-Shielding Effect on Reaction Rate at Various Dopant Dilution Levels.