INVESTIGATION OF WINDOWS AND SHIELDS FOR NEUTRON POINT SOURCES

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May 20, 1959

Printed for the U.S. Atomic Energy Commission
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ABSTRACT

An empirical approach for the evaluation of shielding materials for macrochemical manipulations of spontaneously fissioning heavy elements (curium and californium) has revealed interesting comparisons. High-density metal halide solutions were compared with lead glass and with rare earth glass for use as shielding windows. Laminated shields of lead-paraffin and uranium-paraffin were compared with water and with paraffin for shielding walls. Measurement of samples of the mixed shielding materials compared reasonably with published data, where available. On the basis of the figures obtained dose-attenuation factors were computed for various thicknesses of the materials tested.
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INTRODUCTION

The point-source problem has become more real with the production and isolation of increasing quantities of spontaneously fissioning isotopes. $^{1,2}$ At the Lawrence Radiation Laboratory at Berkeley $^{252}\text{Cf}$ has required special shielding considerations. $^{3,4}$ The problem of shielding a neutron point source with a finite medium was tested by F. R. Jones $^{5}$ for water and paraffin shields. National Bureau of Standards Handbook $^{6}$ and others $^{7,8}$ give methods for point-source calculation, but the results are dependent on the selection of a proper "B" (build-up) factor. $^{9}$ A more extensive treatment has been given to theory and confirming experiments on shielding of large-area sources by an "infinite" medium. $^{10-18}$ That is, the dose (or flux) is measured at a point within the shielding material, and boundary effects that arise when from the source of neutrons and the detector are in air at either side of the shield are not present to complicate matters. Some of the effects due to irregularities in the shield have been studied and corrections have been devised. $^{19}$

This study was made to discover what materials might be used for shields and shielding windows during macro- and microchemistry manipulations on several micrograms of $^{252}\text{Cf}$ associated with other heavy elements and fission products. Existing gamma shields in use were 2- and 6-inch lead walls with 4- and 12-inch high-density (6.2) lead glass windows. The neutron-attenuation characteristics of these shields, alone and in combination with other materials, were determined. Where gamma radiation was not a problem, an adequate shield or shielding window of minimum thickness to attenuate the neutrons to safe levels was desired.
SHIELDING RESULTS

Three classes of materials were tested for neutron-attenuation characteristics: aqueous solutions, compounds or elements (or their mixtures), and laminated combinations of these. Materials selection was limited to those readily available or already in use for other shielding at the laboratory. The results have been plotted for various shield thicknesses and are expressed as attenuation factors—the net count rate with a given source-to-detector distance in air divided by the count rate with the shielding material interposed without any change in the source-to-detector relationships.

Figure 1 shows these data as determined for water and several saturated aqueous solutions of high density. Figure 2 compares water with saturated solutions of materials commonly associated with high neutron cross sections. Figure 3 compares water with paraffin, lead, and lead glass of density 6.2 and 3.8 (Gd-Sm). Figure 4 shows water combined with lead glass—a combination to be used for direct-viewing windows. Figure 5 shows paraffin laminated with lead, and Fig. 6 paraffin with normal uranium.

One application of the data above is shown in Fig. 7. In the design of shielding windows consideration must be given to the source strength, the permissible exposure level, and the operating distance before the window thickness may be determined. Figure 7 permits the determination of one of these variables if the other three are known.

The data presented here have been used to design a minimum-weight shipping container for transporting several micrograms of Cs$^{252}$ and associated gamma emitters from the MTR (Materials Testing Reactor, Idaho Falls, Idaho) to Berkeley. The shield, consisting of 3.6 inches of uranium and 10 inches of paraffin, weighed less than 1200 pounds. Viewing windows for the manipulator boxes used with the separated californium consisted of 4 inches of lead glass and 10 inches of water.
Fig. 1. Neutron-attenuation factor, $I_0/I$, as a function of thickness of solution.
Fig. 2. Neutron attenuation in materials containing cadmium or boron.
Fig. 3. Neutron attenuation in water, paraffin, lead, and lead glass.
Fig. 4. Neutron attenuation in combinations of water and lead glass.
Fig. 5. Neutron attenuation in combinations of paraffin and lead.
Fig. 6. Neutron attenuation in combinations of paraffin and uranium.
Fig. 7. Window thickness $t$ of 50-50 6.2 glass-water needed to give required ratio of source level to dose rate at various spacings $S$ between window and neutron source.
DISCUSSION

Experimental Techniques

The data were taken with a fixed source-to-detector distance of about 70 cm. Shielding sections were built up so that the "cold" face was just touching the detector, as shown in Fig. 8. When available, shield sections 12 by 18 inches in cross section were used. These dimensions duplicate those of the standard window for our shielded manipulator boxes. The solutions were tested by filling 2-inch-diameter thin-wall brass tubes of assorted lengths, closed at each end. These were inserted into a paraffin collimator 10 inches square by 24 inches long. Cadmium sheets prevented the spill of slow neutrons from the collimator into the detector.

The measurements were taken with Nuclear Enterprises NE404 fast neutron scintillator, an adaptation of the Hornyak button,\textsuperscript{20} and an RCA 6655 photomultiplier tube. The tube was operated at 750 volts and fed an RIDL scaler model set to accept pulses of 2.5 millivolts or greater. A Po-Be source yielding about $10^6$ neutrons per second provided the neutron flux. Data were generally taken to maintain a standard error of 10%, but background counts, which varied from 15 to 50% of the lowest count rates in less than 1 hour, made the actual errors somewhat greater.

Gamma Sensitivity

The scintillator gave 50 counts per minute for each milliroentgen from radium gamma rays. Reasonable neutron count rates could not have been maintained at higher discrimination.

Neutron Sensitivity

Under the operating conditions used (750 volts, and 2.5 millivolts minimum pulse height) the absolute efficiency, on the assumption of inverse-square flux-density variation, was $4.7 \pm 0.9\%$, with a source strength of $2.34 \times 10^6$ neutrons per second. At 68 cm from this source the 2-inch-diameter scintillator gave $2300 \pm 100$ cpm, with an average low background of $20 \pm 2$ cpm.
Fig. 8. Experimental arrangement.
Neutron-Energy Sensitivity

Two factors concerning neutron spectral characteristics were worrisome. The first was that measurements of neutron attenuation were made with a source of neutrons (Po-Be) of different spectral characteristics from those required by the design specifications (fission neutrons). The second was the well-known phenomenon of spectral "hardening" as shield thickness is increased, and the question was how this would influence the counting efficiency of our plastic scintillators.

Although the spectrum of fission neutrons is reported with some precision and agreement, the californium spectrum has been compared to it as very similar, the Po-Be spectrum shows considerable variation in reported values. However, one calculation of the dose ratio between Po-Be and fission neutrons shows the Po-Be dose for a given flux density to be only 18% higher (ratio = 1.18) than the fission-neutron dose—very little difference, considering the difference in the spectral shape as shown in Fig. 9.

The problem of spectral hardening is both raised and ameliorated in the Handbook of Nuclear Reactor Shielding. On page 55 are examples of the shift in spectrum toward higher energies with increasing water-shield thicknesses. On this same page we are told, "It must be noted that this 'uncollided' spectrum is only of use in revealing the penetration characteristics of the neutrons; the actual spectrum will include the multiply scattered neutrons and will have the characteristic distribution similar to that of a fission spectrum..." Shields of the type required for exposure reduction will of necessity involve exposure to multiply scattered neutrons. Optimistically, then, we might assume that the Po-Be neutron spectrum suffers no appreciable distortion in its passage through a shield and therefore that the net response of the scintillation detector is proportional only to the flux density for a given type of neutron source.

The NE404 scintillator does not respond in proportion to dose from sources of differing spectrum because the ratio of absolute efficiencies at "zero" discrimination (extrapolated) is close to 2.3 (Fig. 10) instead of 1.18, as calculated for the dose ratio. However, the difference is not pertinent to the use of the attenuation factors reported here for calculation of fission-neutron shield thicknesses. In fact, fission-neutron attenuation factors should be somewhat higher because the effective energy is lower than that of Po-Be neutrons.
Fig. 9. Comparison of spectra from fission and from Po-Be.
Fig. 10. Absolute counting efficiencies of NE404 scintillator for Po-Be, Cf$^{252}$, and Ca$^{137}$. 
The attenuation-thickness curve for paraffin was checked within the statistical error by repeating the experiment with a Nuclear Enterprises NE 401 slow-neutron scintillator with 2.8 inches of paraffin as a moderator. These data are shown in Fig. 11. The curve of net counts per minute is shown as beginning at zero paraffin thickness to show the slow-neutron build-up. Attenuation factors were calculated from the point of maximum count rate (2.8 inches of paraffin) as zero shield thickness. The NE 401 is an enriched boron-loaded plastic scintillator utilizing an (n,a) reaction instead of the (n,p) attributed to the NE 404.

**Agreement with Published Values**

Most of the attenuation data, calculated or experimental, in the published literature show nearly straight lines on semilog plots. The data presented here show this relationship for heavy atoms in combination with hydrogeneous materials. But light materials exhibit a distressing tailing off, beginning at around 10 or 12 inches' thickness. Figure 12 compares water attenuation factors from four sources with the values derived in this investigation. The NBS Handbook 63 data are presented without the recommended "B" factor of 5 for shields 20 cm or more in thickness. Goldstein\(^2\) shows build-up factors (after NDA 15C-3a) up to 10 for 100 cm from a point isotropic source, but a factor of \(10^3\) would have to be applied to the Handbook 63 line to bring it close to the data derived in this investigation.

A reasonably apt comparison for shields of mixed composition is of Rockwell's 18% lead and 82% water, properly normalized, to this paper's interpolated data for a similar composition of lead and paraffin. If a "B" factor is calculated by dividing the Rockwell points by corresponding points found here, more reasonable values are found, although they are still higher than the Goldstein data. Figure 13 illustrates these relationships.
Fig. 11. Net counts per minute and attenuation factor as a function of paraffin thickness for NE401 detector.
Fig. 12. Comparative data for attenuation factors for water.
Fig. 13. Comparison of lead and hydrogen attenuation factors.
CONCLUSIONS

First approximations may be obtained for bulk shielding measurements in "bad-geometry" situations with some systematics. Certainly the values presented here will allow greater safety factors than earlier published data. The problems associated with shielding of increasing quantities of spontaneously fissioning materials of microscopic volumes will demand that efficient minimum-weight shields be designed, for reasons of economy and convenience.

ACKNOWLEDGMENTS

The authors wish to acknowledge the help of Rosemary J. Barrett in searching the literature and of Robert J. Walker in procuring and maintaining the necessary instrumentation.

This work was done under the auspices of the U. S. Atomic Energy Commission.
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9. Ibid., p. 596.

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