Determination of Buildup Factors in Titanium and Depleted Uranium*

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INTRODUCTION

Approximately 13% by volume of the U. S. Department of Energy (DOE) current backlog of radioactive waste is characterized as high-level waste. Transportation of these wastes requires that the waste package have adequate shielding against gamma radiation. This project investigates the radiation shielding performance of titanium and depleted uranium, which have been proposed for use as gamma-shielding materials in DOE transportation packages, by experimentally determining their buildup factors.

Buildup factors are important in shield heating and radiation damage calculations. A point-isotropic-source type of buildup factor is the most useful for application in the point-kernel approach utilized in many simple shielding codes. The point-kernel method provides reasonable results for cases in which the shield is made of one solid material and the source can be approximated as one homogeneous material. The point-kernel method has been incorporated into a large number of shielding codes treating three-dimensional geometry using buildup factor data in some form. Buildup factors vary with a number of parameters such as the distance of penetration through the attenuating medium; the geometric configuration of the attenuating medium, source, and detector position; the composition of the medium; the detector response function; and the energy and direction of emission of the source photons, ideally taken to be monoenergetic and isotropic.

The primary focus of this project is to measure buildup factors for titanium and to compare these values with published point isotropic buildup factors for elemental iron. Since measured data for titanium is not in the literature, buildup factors for iron have been used in the past to characterize the behavior of titanium. These experimental values are then compared with those predicted using the MCNP computer code (Briesmeister

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* This work, performed at the University of New Mexico in Albuquerque, NM, was supported under Contract AJ-9008 by Sandia National Laboratories for the U. S. Department of Energy under Contract No. DE-AC04-94AL85000.

1993). The experimentally measured buildup factors for depleted uranium are compared with the point isotropic buildup factors for uranium. Finally, empirical formulas are developed to give an accurate representation of the buildup factors as a function of distance.

EXPERIMENTAL DESCRIPTION

The authors utilized information about the fission product and actinide content of irradiated PWR fuel (Benedict et al. 1981) to determine which gamma source would be most appropriate to represent spent nuclear fuel. The average gamma energy from spent nuclear fuel is approximately 0.660 MeV. After approximately 5 years, the gamma spectrum is dominated by the ^{137}Cs gamma, and after about 10 years, over 90% of the spent fuel activity is due to ^{137}Cs. Based on this, the authors decided to use a single ^{137}Cs source to represent the gamma activity of the spent nuclear fuel. Additional analyses were performed to determine the source strength required to characterize buildup factors through multiple thicknesses of shielding materials. These analyses indicated that a 20 Curie ^{137}Cs source would be optimum; however, discussions with the manufacturers of ^{137}Cs calibrated sources revealed that the largest available calibrated ^{137}Cs source strength was 10 Curies. Based on this information, a nominal 10 Curie, National Institute of Standards and Technologies (NIST) calibrated ^{137}Cs source was purchased from Amersham Corporation. The source was integrated into a shielded holder with a 15-degree collimator. The calibrated activity as of September 30, 1994 was 9.59 Curies, and the source is housed in a Model HBC5.1 Irradiator. The HBC5.1 Irradiator contains a lead and tungsten shielded source with a key actuated electronic remote control which automatically moves the source into the exposed position within the collimator.

Titanium samples that were 98.8% pure were purchased from Atomergic Chemetals Corporation. Six titanium samples were in the form of 20.3 cm x 20.3 cm x 2.54 cm thick (8 inches x 8 inches x 1 inch) plates, and four titanium samples were in the form of 20.3 cm x 20.3 cm x 0.635 cm thick (8 inches x 8 inches x 0.25 inches) plates. Twenty depleted uranium samples that were 99.98% pure with an enrichment of 0.2 weight percent ^{235}U were obtained from Babcock & Wilcox Idaho. The samples were in the form of 20.3 cm x 20.3 cm x 0.83 cm thick (8 inches x 8 inches x 0.325 inches) plates. After receipt, the depleted uranium plates were found to be coated with an oxide layer that posed a contamination risk. This was minimized by covering each plate with a thin plastic adhesive. The presence of the plastic film should have a negligible effect on the buildup behavior.

Since the ^{137}Cs source is collimated to a 15-degree cone, there was no need for additional collimation or shielding beyond that found in the irradiator. The distance from source to detector was determined by the space required for about 20.3 cm (8 inches) of sample material added to the outside of the shielding around the source. The distance from the source to the outside of its shielding material is 10.95 cm (4.31 inches). The detector position was then fixed at 36.9 cm (14.5 inches) from the source to allow adequate room for samples but not encounter too much beam dispersion or $1/R^2$ losses. At this distance, the beam has a radius of 4.8 cm (1.9 inches). Figure 1 illustrates the experimental setup.

A boron-lined ionization chamber designed for both neutron and gamma detection was initially utilized in the buildup factor measurements; however, due to problems in obtaining data for any substantial thickness of the depleted uranium, the authors decided to use Panasonic model 802 thermoluminescent dosimeters (TLDs). Readings were taken from the two LiBO elements. For each thickness, five TLDs were positioned on the backside of the sample plate: one in the center of the beam, and one at the top, left, bottom, and right of the beam centerline. The TLDs are capable of handling exposures in the range of 10 mR to 100 R. In this experimental configuration, the maximum exposure with no sample material was about 20 R.

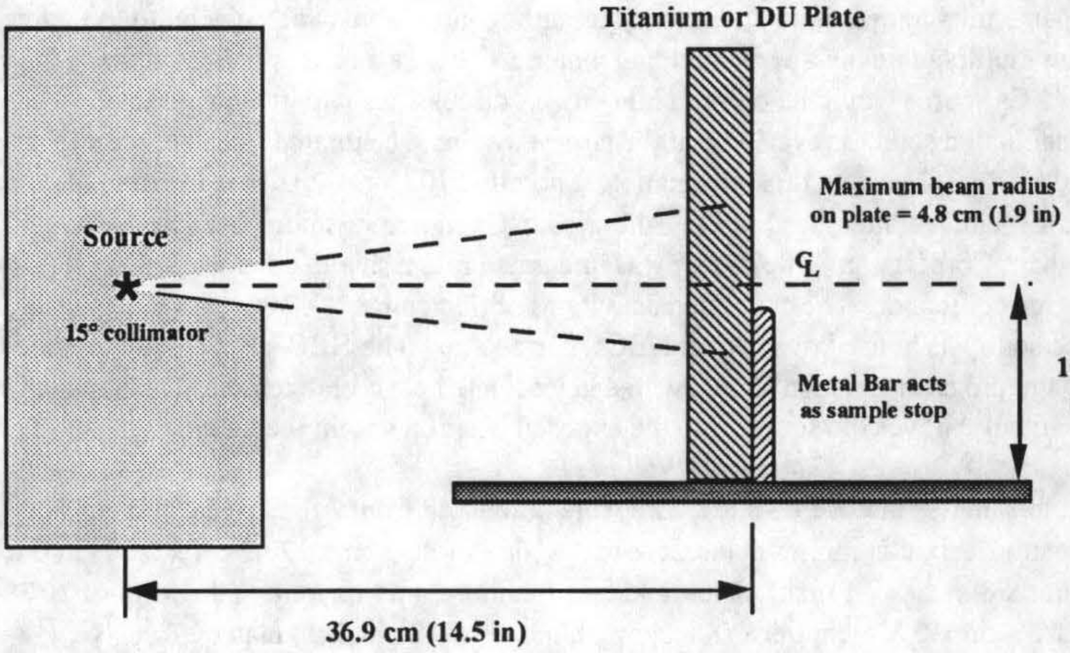

Figure 1. **Experimental Setup for Buildup Factor Measurements.**

TITANIUM RESULTS

Using the experimental setup illustrated in Figure 1, transmission measurements for various thicknesses of titanium were made using the Panasonic 802 TLDs. The average element values were compared against uncollided, attenuated values to determine the buildup. The value at zero thickness was used for the uncollided flux and an attenuation coefficient of 0.32535 cm^{-1} was used to calculate the attenuated flux without buildup. The values with the titanium plates were then ratioed to the uncollided values to determine the buildup.

As shown in Figure 2, the published point isotropic buildup factors for iron (ANS 1993) are greater than the experimentally determined values. However, the experimental geometry was significantly different than that for a point isotropic buildup measurement. A point isotropic measurement is made with a source surrounded by material. This means that radiation can backscatter and then be detected at a point on the outside of a shield. For our slab buildup experiment, only scattering within a 15-degree cone which then is detected at a point on the outside of the shield contributes to the buildup factor. This suggests that the experimentally determined values of buildup for a slab-shield geometry should be less than those determined for a point isotropic buildup geometry. MCNP analyses were performed on the actual experimental configuration to check the effect of detector location on buildup.

Figure 2 also shows a plot of the MCNP calculated buildup factors for titanium. Note that the calculated values are much less than the point isotropic values, but are still higher than the experimental values. The experimental values lie within two standard deviations of the MCNP calculations, but are always below the calculated values. This indicates that using published point isotropic buildup factors for iron for the appropriate geometry is acceptable. In fact, using point isotropic buildup factors for iron to estimate exposures at the surface of titanium shields is quite conservative for a point-source, slab-shield geometry similar to this experiment.

A polynomial fit to the experimental slab-shield geometry buildup data for titanium indicates the following relationship:

$$\mathit{buildup} = 0.0001 * \mathit{thickness}(\mathit{cm})^2 + 0.1614 * \mathit{thickness}(\mathit{cm}) + 1.0000$$

with a correlation coefficient of 0.9737. A similar polynomial fit to the MCNP calculated slab-shield geometry buildup data indicates the following relationship:

$$\mathit{buildup} = -0.0009 * \mathit{thickness}(\mathit{cm})^2 + 0.2579 * \mathit{thickness}(\mathit{cm}) + 1.0413$$

with a correlation coefficient of 0.9988. Given a source shield configuration similar to that of the experiment, either of these two equations could be used with a simple shielding code such as QAD or MicroShield to provide quick estimates of exposure levels on the outside of titanium shields.

DEPLETED URANIUM RESULTS

Using the experimental setup illustrated in Figure 1, transmission measurements for various thicknesses of depleted uranium were made using the Panasonic 802 TLDs. The average element values were compared against uncollided, attenuated values to determine the buildup. The value at zero thickness was used for the uncollided flux and an

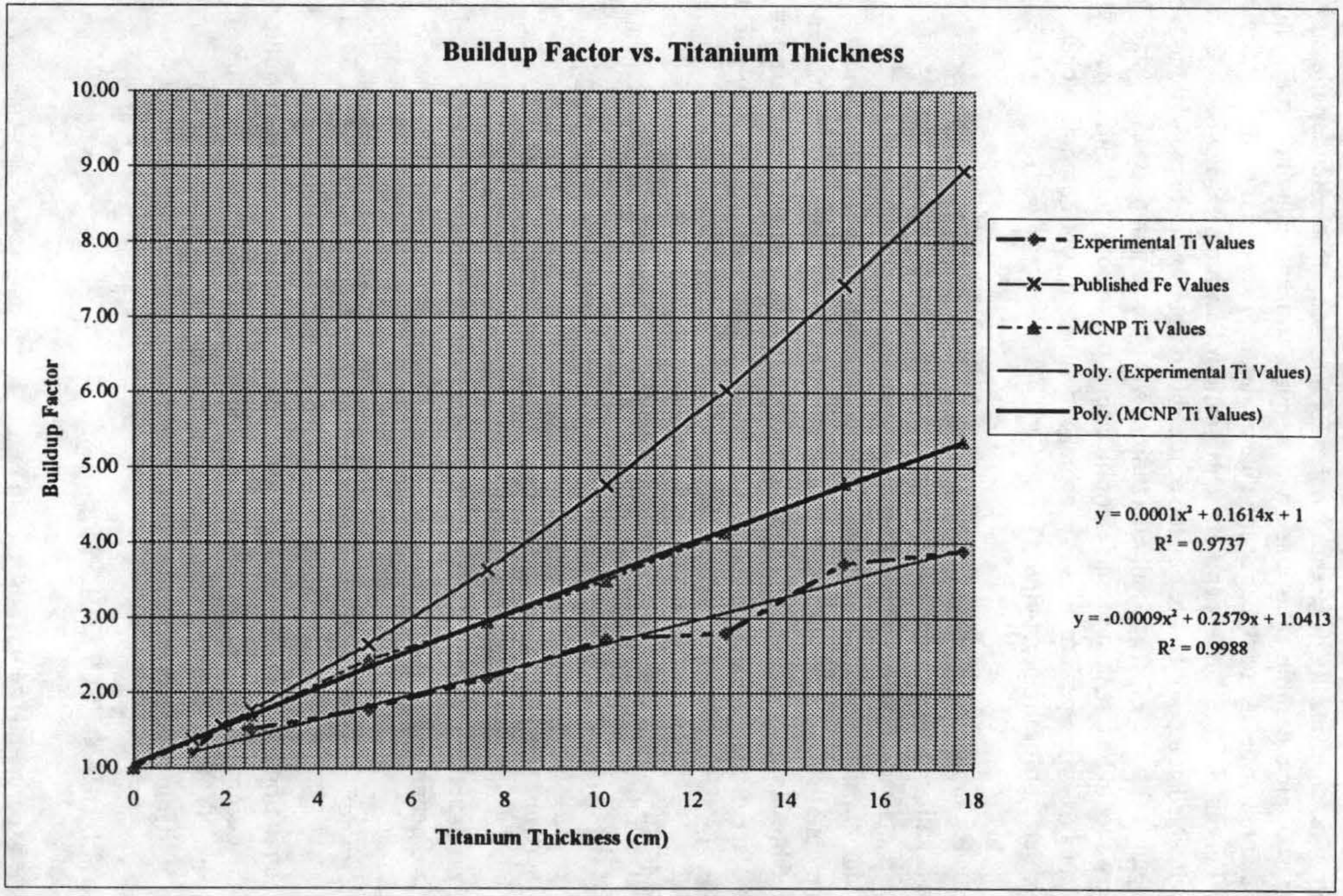

Figure 2. Experimentally Measured and Calculated Buildup Factors for Titanium.

attenuation coefficient of 2.46078 cm^{-1} was used to calculate the attenuated flux without buildup. The values with the depleted uranium plates were then ratioed to the uncollided values to determine the buildup.

Experimental results for buildup in depleted uranium had a much wider statistical variation. Since depleted uranium is quite dense with a mean free path of about 0.41 cm, it only took about 4 cm of depleted uranium to reduce the exposure by four orders of magnitude. Therefore, determination of the buildup factor for more than a few cm of depleted uranium was difficult due to the low signal-to-noise ratio. In addition, beta and gamma radiation associated with the decay of the uranium daughter products and spontaneous fission products significantly increased the background radiation level and reduced the signal-to-noise ratio. As a result, the experimentally determined buildup factors for depleted uranium had very large standard deviations.

As shown in Figure 3, comparison of the experimentally determined values with published point isotropic buildup factors for uranium (ANS 1993) shows that the published values are everywhere lower than the measured values. This is unexpected since the point values include isotropic scatter while the experiment only measured scattering from a small angle. It is surmised that the increase in the experimental values is due to the creation of bremsstrahlung in the high-Z depleted uranium. Bremsstrahlung production is accounted for in the point isotropic buildup factors, but may dramatically change with our experimental geometry. Thus, conservatism in the design of depleted uranium shields would suggest using the experimental data which accounts for bremsstrahlung production in the appropriate geometry. Figure 3 also shows the experimentally determined buildup factors increased by sigma (one standard deviation of the measurement) and decreased by sigma. There is a 66% probability that the measured buildup value lies between the two curves given in Figure 3.

A polynomial fit to the experimental buildup factor for depleted uranium indicates the following relationship:

$$buildup = \exp(0.4049 * thickness(cm))$$

with a correlation coefficient of 0.9838. However, as noted the values are nonconservative as compared to the published data, and the curve shapes are significantly different.

Attempts to calculate depleted uranium buildup factors with MCNP for the experimental configuration were not successful. The problem lies with achieving statistically significant results due to the density and small mean free path of depleted uranium. Even with variance reduction methods, MCNP runs are requiring a few days to obtain enough particles for a 66% confidence interval. Presently, the confidence intervals obtained are so large that the calculational results are essentially meaningless.

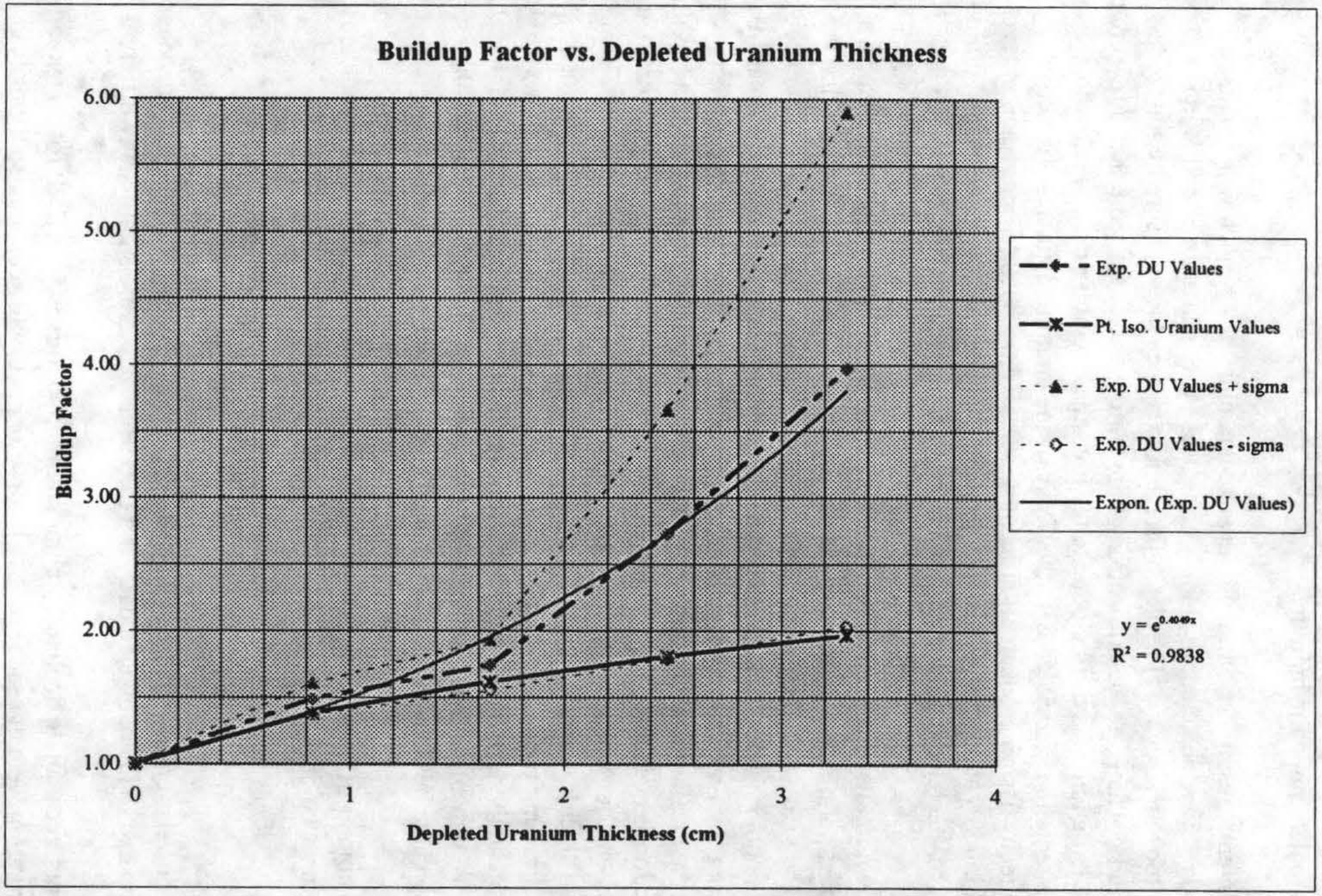

Figure 3. Experimentally Measured Buildup Factors for Depleted Uranium.

CONCLUSIONS

Since the experimental data for titanium are within two standard deviations of, but always below, the MCNP calculated buildup values, this implies significant conservatism in the calculated values. Due to the sensitivity of the buildup factor to detector geometry, it is strongly suggested that MCNP analyses or equivalent be done on each configuration to determine the buildup factor and resulting exposure/dose which is appropriate to the situation. If concern is centered on point exposures at the outside of the shield, then the results of this experiment should be appropriate. However, if concern is centered on personnel exposures at a distance from the shield, then neither published point isotropic data nor these experimental data would be appropriate. The wide variation in geometries and difficulty in obtaining good experimental statistics from many thicknesses of titanium make the experimental data useful for initial characterization of the exposure, but with limited accuracy for dose or exposure calculations.

For titanium the use of published point isotropic buildup factors for iron is suitable for preliminary design work, although the calculated doses will be larger than the actual doses. Moreover, the use of Monte Carlo methods for refined design will provide results which are closer to the actual doses, although they too will be somewhat larger than the actual doses.

For depleted uranium the use of published point isotropic buildup factors for uranium in preliminary design work provides a good initial estimate; however, the calculated doses may be less than the actual doses. Furthermore, the use of Monte Carlo models for refined design is more difficult due to poor statistics and adequate consideration of radiation from spontaneous fission products. Finally, for handling purposes, the depleted uranium should be clad with some material to avoid contamination. Because of the bremsstrahlung and decay radiation, this cladding material should be low-Z to reduce radiation levels at the outside of the shield.

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