The Monte Carlo Method: Versatility Unbounded In A Dynamic Computing World
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STORAGE SAFE SHIELDING ASSESSMENT FOR A HDR CALIFORNIUM-252 BRACHYTHERAPY SOURCE

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ABSTRACT

In pursuit of implementing HDR ^{252}Cf brachytherapy, a suitable external storage safe must be designed and constructed to store the medical source when not in use. The shielding capabilities of polyethylene, RICORAD™, and Pb were examined for both neutron and photon radiations using Monte Carlo methods. RICORAD™ is a heat-resistant plastic, with 2.00%-mass boron in a polyethylene-based matrix having a mass density of 0.945 g cm^{-3}. Due to the high-hydrogen content, the fission-energy ^{252}Cf neutrons are readily attenuated by RICORAD™ and polyethylene, and the boron loading in RICORAD™ further helps prevent creation of 511 keV gamma-rays from neutron capture by hydrogen. Pb readily attenuates ^{252}Cf photons. Results of shielding calculations using MCNP were incorporated into the design of a spherical RICORAD™/Pb safe for storage of 1 mg of ^{252}Cf that would reduce the external radiation fields to levels below regulatory concerns. Practical considerations governing safe design included exposure levels, material availability, and total safe weight and size. Results are presented per milligram of ^{252}Cf and can be scaled as needed for other source strengths.

Key Words: Californium-252, neutron shielding, photon shielding, Monte Carlo

1 INTRODUCTION

Californium brachytherapy was first proposed nearly forty years ago as the promise of radiotherapy with neutrons first became apparent. Since that time, a handful of clinical centers around the world have employed low dose rate (LDR) intra-cavity or interstitial californium therapy to treat tumors from head-to-toe. Thorough reviews of the clinical results and radiobiological advantages have been published by Maruyama.[1,2]

Miniaturized high dose rate (HDR) ^{252}Cf sources (~1 mg) recently became available through advances in radiochemistry.[3] As such, HDR ^{252}Cf brachytherapy sources compatible with modern remote afterloader technology are feasible. For standard-of-care brachytherapy employing a HDR ^{192}Ir source, a tungsten-alloy storage safe has been integrated into the remote afterloading unit. However, it is not feasible to construct a compact storage safe to attenuate the mixed-field radiation emissions from ^{252}Cf due to the increased path length of neutrons and photons in suitably hydrogenous materials in comparison to the shorter path length of ^{192}Ir

photons in the W-alloy. Therefore, an external storage safe must be employed. Additionally, to minimize the size of the storage safe, a multiple component shield is needed to address the different requirements of the mixed neutron/photon radiation fields.

In this study, the shielding properties of polyethylene, Pb, and RICORAD™ – a borated polyethylene product manufactured by Reactor Experiments, Inc. (San Carlos, CA)* – were investigated using Monte Carlo methods. In addition, preliminary models of a multi-component safe, employing Pb and RICORAD™ to maximize photon and neutron attenuation, respectively, were investigated.

2 MATERIALS AND METHODS

2.1 ^{252}Cf Radiation Spectrum

Californium-252 is one of the few radionuclides that decay via spontaneous fission. One milligram of ^{252}Cf emits 2.314 x 10^9 neutrons per second through this decay mode, although spontaneous fission accounts for only 3.092% of disintegrations.[3] Historically, ^{252}Cf neutron emission was modeled using either a Maxwellian or Watt fission model [4] and the appropriate built-in MCNP source probability function, *e.g.*, f = -2 or –3 on the SP card, respectively. In this work, neutron emission data published by Mannhart and integrated into the Evaluated Nuclear Data File (ENDF) were used.[5] **Figure 1** compares the three neutron spectra normalized to unity at the most probable energy: 0.7, 0.9, and 1.2 MeV for the Mannhart, Watt, and Maxwellian models, respectively.

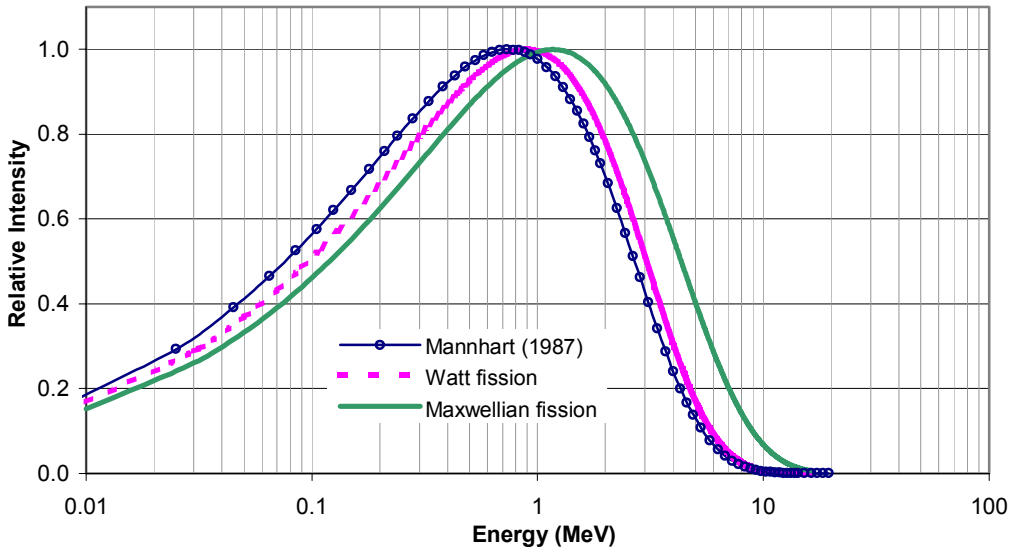

Figure 1. Comparison of normalized Mannhart ^{252}Cf neutron emission spectrum, Watt fission spectrum, and Maxwellian fission spectrum. In this study, the Mannhart neutron spectrum was utilized.

Prompt ^{252}Cf gamma rays are emitted stochastically through spontaneous fission, but a representative spectrum was not available in the ENDF. Verbinski *et al.* measured the prompt

* The company has been acquired by Thermo Electron Corporation, Yokohama City, Japan.

gamma ray emission from ^{252}Cf using 13 energy bins between 0.14 and 10 MeV.[6] Skarsvåg similarly measured the prompt gamma ray spectrum for photons above 0.114 MeV using 18 energy bins, but binned photons above 2.54 MeV.[7] Above 2.54 MeV, the number of gammas per fission was 0.43 and 0.426 and the percent of total emission was 4.42% and 5.46% for Skarsvåg and Verbinski *et al.*, respectively. Thus, the data of Verbinski *et al.*, which had finer energy resolution, was used for prompt gamma ray energies above 2.54 MeV.

The photon emission rate from 1 mg of ^{252}Cf was taken to be 1.3 x 10^{10} s^{-1}, which was employed in shielding calculations performed by Hootman and Stoddard (1971) and by da Silva and Crispim (2001).[8,9] Dividing the neutron emission rate by the average number of neutrons per fission (3.7676) and multiplying by the average number of prompt photons per fission (9.7), as measured by Skarsvåg, yielded a prompt gamma emission rate of 5.96 x 10^{9} s^{-1}. There is a factor of 2.2 between the two values because delayed gamma rays from the decay of fission product progeny were not included by Skarsvåg. Hootman and Stoddard published photon spectral data for fission product gamma rays, but did not cite the source of their spectral data. Additionally, only four energy bins between 0 and 2 MeV were used. **Figure 2** displays the relative intensities of prompt and delayed gamma rays published by Skarsvåg and by Hootman and Stoddard.

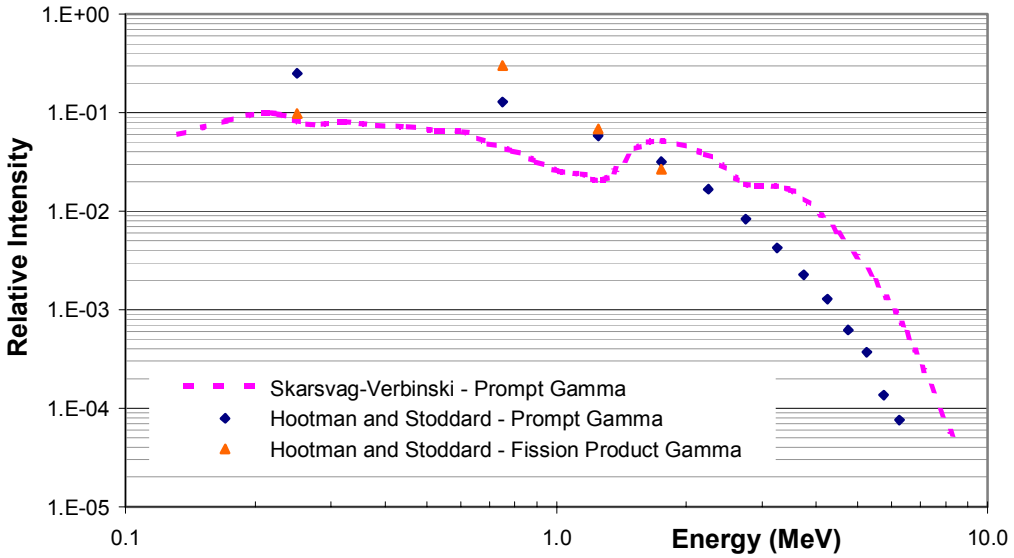

Figure 2. Prompt and delayed photon emission spectrum from spontaneous fission of ^{252}Cf and decay of fission daughter products, respectively. The prompt gamma ray spectrum measured by Skarsvåg and modified using data by Verbinski *et al.* was used in this study.

Although the prompt photon spectrum of Skarsvåg underestimates the total number of low-energy photons (*e.g.*, 0.25 and 0.75 MeV) presented by Hootman and Stoddard, similar trends are exhibited between the two datasets. Since modern measurements or calculations of the delayed photon spectrum are lacking, the prompt photon emission data of Skarsvåg was employed with the total photon emission rate of 1.3 x 10^{10} s^{-1} mg^{-1}.

2.2 Radiation Transport Calculations

2.2.1 Monte Carlo tally types and simulation defaults

Radiation transport calculations were conducted using version 5 of the Monte Carlo N-Particle Transport Code System (MCNP5), which employs the DLC-220 cross section library published in 2004.[10] Initial calculations were performed on a 1.4 GHz computer running the Windows 2000 (Microsoft Inc.; Redmond, WA) system, and final calculations were undertaken at Oak Ridge National Laboratory's C-Pile.[11,12] For each shield thickness, two separate calculations were performed. One simulated the penetration of ^{252}Cf neutrons through the shield in N P mode to include the generation and transport of secondary photons. The second simulated the attenuation of ^{252}Cf photons through the shield in mode P.

For neutron transport, the F4 tally estimator was used in conjunction with a tally modifier (*i.e.*, DE and DF) to determine the equivalent dose to International Commission on Radiation Units and Measurements (ICRU) four-component tissue in a vacuum.[13] The neutron tally modifier included two components: (1) kerma coefficients to convert neutron fluence (cm^{-2}) to absorbed dose [Gy] and (2) an energy-specific radiation weighting factor (w_R) to convert absorbed dose to equivalent dose. The ICRU published neutron kerma conversion coefficients in Report 63 for twelve elements.[14] To determine the kerma coefficient for a compound containing any of these elements, the individual coefficients are weighted by their respective mass-percent and summed. Energy-specific neutron w_R factors were taken from Publication 60 of the International Commission on Radiological Protection (ICRP).[15] In addition, thermal neutron scattering functions, *i.e.*, $S(\alpha,\beta)$, was utilized when appropriate to provide accurate modeling of neutron thermalization and incoherent scattering.

For primary photon transport, the *F4 tally estimator was used in conjunction with a tally modifier (*i.e.*, DE and DF) to determine the absorbed dose to ICRU four-component tissue in a vacuum. Energy absorption coefficients published by Hubbell and Seltzer [16] transformed energy-flux per neutron [MeV cm^{-2}] to absorbed dose [J kg^{-1}]. Additional unit conversion was included to determine the absorbed dose [Gy], which is equal to the equivalent dose as w_R is unity for photons. Due to the small population of high-energy photons, photonuclear physics (PHYS:P) was not incorporated.

A sufficient number of photon histories were used to ensure statistical uncertainties remained less than 2%. For most cases, preliminary calculations using 10^6 histories were sufficient to meet this requirement. Final calculations performed at C-Pile computed between $1x10^7$ and $2x10^9$ starting particles, and variance reduction techniques were not utilized.

Three shielding materials, polyethylene, RICORAD™, and Pb, were evaluated towards the construction of a HDR ^{252}Cf storage safe. Elemental compositions of the shielding materials used in this study are given in **Table I**. The composition of ICRU four-component tissue is included, which was used to determine the neutron kerma coefficients and photon energy absorption coefficients.

Table I. Percent composition by mass of materials included in shielding analyses.

Element	Material			
	ICRU Tissue	POLYETHYLENE	LEAD	RICORAD™
H	0.101174	0.143716	-	0.1206
B	-	-	-	0.0200
C	0.111000	0.856284	-	0.8493
N	0.026000	-	-	-
O	0.761826	-	-	0.0046
Al	-	-	-	0.0015
Si	-	-	-	0.0020
Fe	-	-	-	0.0020
Pb	-	-	1.000	-
ρ [g cm^{-3}]	1.00	0.93	11.35	0.945
S(α,β)	-	POLY.60t	-	POLY.60t

For elements that did not have DLC-220 neutron cross sections for their natural isotopic abundances, relative isotopic abundances by atom were assigned considering the available neutron libraries. Hydrogen was modeled using 99.985 % ^{1}H and 0.015% ^{2}H; boron was modeled using 19.9% ^{10}B and 20.1% ^{11}B; oxygen was modeled as 100% ^{16}O; and, Pb was modeled as 24.4% ^{206}Pb, 22.4% ^{207}Pb, and 53.2% ^{208}Pb.

2.2.2 Simulation geometry

A spherical geometry was used that was similar in design and composition to that employed by both Hootman and Stoddard and by da Silva and Crispim. An unencapsulated point source was positioned at the center of a solid aluminum sphere with radius 0.6204 cm and a volume of 1 cm^3. While da Silva and Crispim employed a 0.02 cm-thick aluminum shell with inner radius 0.62 cm, a solid sphere more accurately represented the geometry and density of a physical source without requiring a vacuum, which would remove the effects of self-shielding. Shells of varying thickness were added outside of the source for the three shielding materials evaluated, as shown in **Figure 3**. F4 and *F4 tallies for neutrons and photons were determined in a vacuum for a shell of thickness 0.1 μm surrounding the outer boundary of the shield.

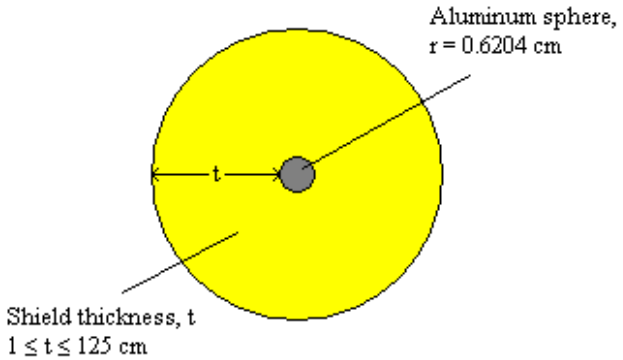

Figure 3. Schematic of the spherical shielding geometry employed in these Monte Carlo calculations. A shield of varying thickness, t, was placed around an isotropic source at the center of an aluminum sphere.

A two-component Pb:RICORAD™ shield was evaluated by fixing the total shield thickness and varying the relative amount of the two materials for a 1 mg ^{252}Cf source. To minimize total weight, Pb was employed as the internal layer and RICORAD™ surrounded it externally. Pb thickness of 2, 4, 6, 8, and 10 cm were calculated for a shield of total thickness 50 cm. For a larger shield, 100 cm thick, Pb thickness of 5, 10, 15, and 20 cm were calculated.

3 RESULTS AND DISCUSSION

3.1 Single Component Shield Analysis

Figures 4, 5, and 6 show the product of the equivalent dose rate and the distance squared [mSv h^{-1} x d^2] from 1 mg of ^{252}Cf for neutrons, secondary photons, and primary photons, respectively. The square of the distance was incorporated to remove the influence of divergence. The total equivalent dose for each shielding material is shown in **Figure 7**.

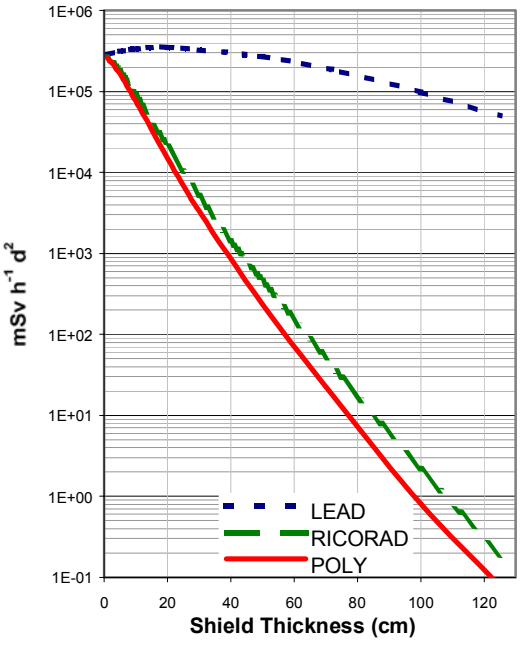

Figure 4. Equivalent dose to ICRU four-component tissue from ^{252}Cf neutrons for the three shielding materials.

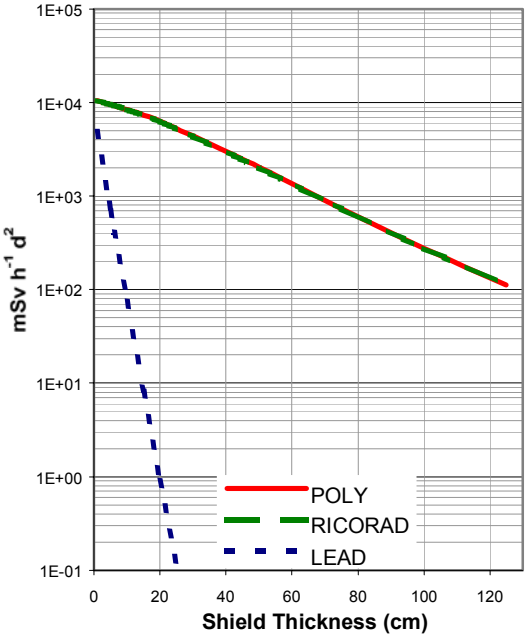

Figure 5. Equivalent dose to ICRU four-component tissue from primary gamma rays for the three shielding materials.

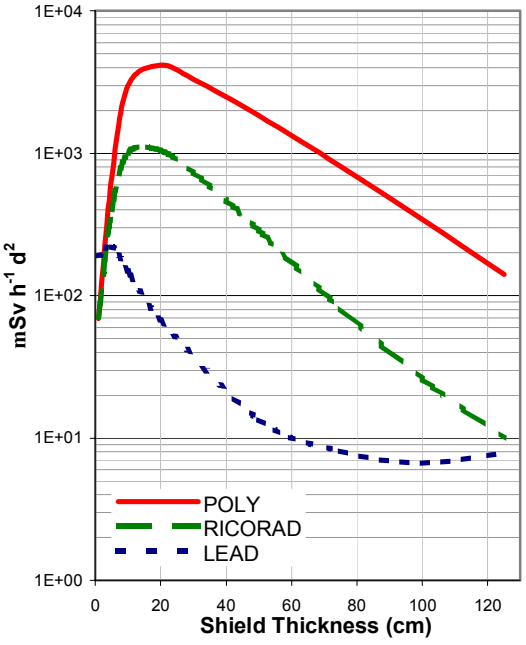

Figure 6. Equivalent dose to ICRU four-component tissue from secondary gamma rays (neutron capture photons) for the three shielding materials.

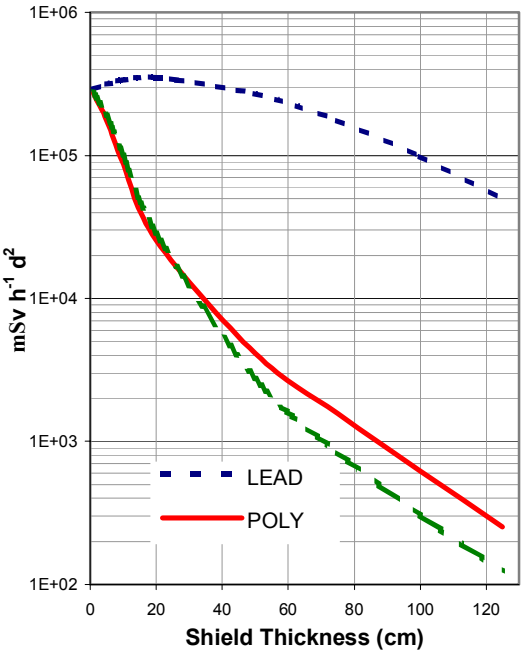

Figure 7. Total equivalent dose to ICRU four-component tissue after attenuation through Pb, polyethylene, and RICORAD™.

Pb provides significant attenuation of the primary photons, but does not provide neutron protection. RICORAD™ and polyethylene provide similar primary photon and neutron attenuation. Fewer secondary photons escape from the RICORAD™ shield because thermal neutrons are preferentially captured by boron, which minimizes capture gamma ray emission after capture by hydrogen.

3.2 Multiple Component Shield Analysis

Table II displays the neutron and photon equivalent doses at the exit of the 50 cm-thick Pb:RICORAD™ shield.

Table II. Comparison of neutron and photon equivalent dose (H) at the exit of a 50 cm spherical Pb:RICORAD™ safe for a 1 mg ^{252}Cf source.

Pb Thickness [cm]	Equivalent Dose [μSv h^{-1}]			
	H_n	$H_{n,\gamma}$	H_γ	H_{total}
2	173.0	121.6	270.2	564.8
4	181.8	121.1	112.9	415.7
6	185.8	119.6	48.5	353.9
8	190.4	118.7	21.0	330.1
10	199.6	119.4	9.2	328.1

ICRP 60 recommends limiting the annual effective dose to a radiation worker below 50 mSv. Therefore, a Pb:RICORAD™ shield designed to maintain a radius of 50 cm would allow

only 89 to 149 hours of work per year adjacent to the safe for the 2 and 10 cm of Pb designs, respectively.

For a Pb:RICORAD™ shield designed to maintain a radius of 100 cm, the annual workload would increase to over 7,800 hours. Equivalent dose values for a multi-component safe with a total shield thickness of 100 cm diameter are shown in **Table III**. Assuming 2,000 work-hours per year, a radiation worker positioned at the exterior of the safe would receive 12.8 mSv per annum, one-quarter of the ICRP 60 allowance. Thus, a safe employing one of the designs in **Table III** would provide adequate protection for a source containing almost 4 mg of ^{252}Cf.

Table III. Comparison of neutron and photon equivalent dose (H) at the exit of a 100 cm spherical Pb:RICORAD™ safe.

Pb Thickness [cm]	Equivalent Dose [μSv h^{-1}]			
	H_n	$H_{n,\gamma}$	H_γ	H_{total}
5	0.25	2.86	3.35	6.46
10	0.20	2.53	0.43	3.16
15	0.23	2.42	0.06	2.71
20	0.20	2.53	0.01	2.74

Although storage safes are typically placed in restricted areas and require minimal maintenance, a robust shield would be desirable to assure minimal radiation exposure to personnel and the public. One disadvantage with the 100 cm safe design is that it would occupy a volume of 4.2 m^3. The total shield weight would be 4,040, 4,085, 4,199, or 4,415 kg for safe models with 5, 10, 15, or 20 cm of Pb, respectively. Potential installation sites would require thorough engineering analyses prior to construction.

3.3 Comparison to Other Studies

Hootman and Stoddard modeled the shielding properties of 32 different materials in their 1971 report, but only one of those materials is in common with those studied here - polyethylene. Using a spherical shield and a point source, Hootman and Stoddard calculated the equivalent dose to tissue. **Figure 8** displays the ratio of the results presented here to those of Hootman and Stoddard.

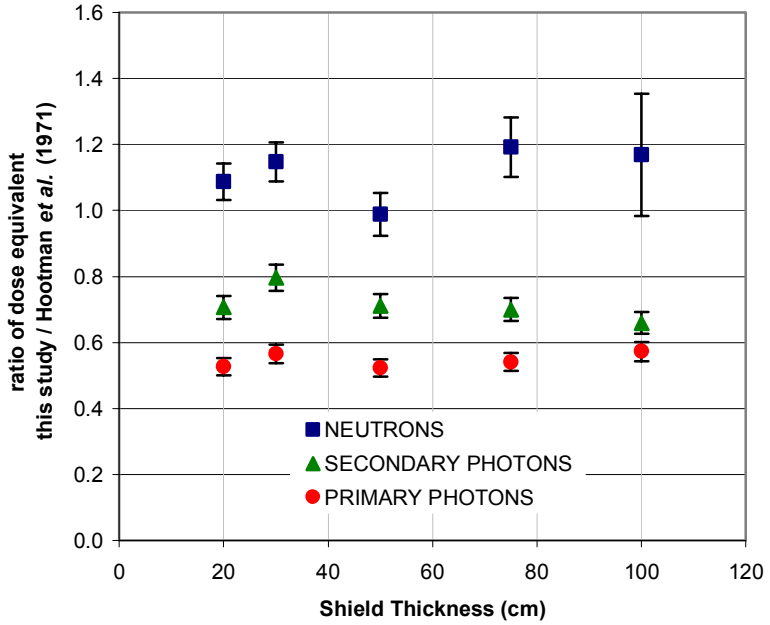

Figure 8. Ratio of Monte Carlo shielding calculations for polyethylene from this study to Hootman and Stoddard (1971).

Our neutron equivalent dose data in polyethylene was within 20% agreement of those presented by Hootman and Stoddard. The secondary photons and primary photons exhibit a near constant ratio, although they differ from the current Monte Carlo results by approximately 71% and 55%, respectively.

The models employed by da Silva and Crispim were designed for the construction of a neutron irradiator and included a 30 cm sphere of high-density polyethylene around the source. Three shielding materials were studied, but none were similar to those investigated here. In addition, da Silva and Crispim included a thin (0.1 cm) shell of cadmium at the shield outer surface to absorb thermal neutrons. Because of these differences, it is difficult to directly compare the results obtained herein to those obtained by da Silva and Crispim. However, direct comparisons can be made at the exterior of a 30 cm radius sphere of polyethylene that is included to moderate the neutron spectrum. **Table IV** shows this comparison.

Table IV. Comparison of equivalent doses (H) at the exterior of a sphere of polyethylene containing 1 mg of ^{252}Cf.

	Equivalent Dose [μSv h^{-1}]		
Investigator	**H_n**	**$H_{n,\gamma}$**	**H_γ**
da Silva and Crispim	2790	3230	5950
This Study	3510	3550	4940
Ratio	1.26	1.10	0.83

As shown in **Figure 8** and **Table IV**, neutron equivalent dose values in this study were typically higher than those reported by Hootman and Stoddard or by da Silva and Crispim. While this difference is partially due to employing different neutron spectra, differences in

dosimetric conversion factors likely caused the discrepancies, *e.g.*, differences between the radiation weighting factors and neutron kerma coefficients. For photons, the energy spectra of Hootman and Stoddard included a larger component of low-energy photons that resulted in increased equivalent dose at the shield outer surface. In addition, the results of this study included the most up-to-date photon cross section libraries. The results of Hootman and Stoddard and of da Silva and Crispim are subject to incorrect photon cross sections. Additional research is required to determine the magnitude of the effect of the different neutron and photon spectra, as well as, the impact of using older (*e.g.*, MCLIB02) photon cross sections.

4 CONCLUSIONS

Monte Carlo methods have been used to determine the shielding characteristics of three materials: polyethylene, RICORAD™, and Pb. Each has desirable shielding characteristics for either neutrons or photons. A storage safe with spherical geometry which combines Pb and RICORAD™ has been evaluated. Using a total shield thickness of 50 cm, a 1 mg ^{252}Cf source can be safely stored according to ICRP 60 standards, allowing over 100 hours of work directly adjacent to the shield. Fabrication of this or a similar design is needed for confirmation with experimental measurements to compare with calculated exposure rates preceding installation of the storage safe and implementation of HDR ^{252}Cf brachytherapy treatments.

5 ACKNOWLEDGMENTS

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6 REFERENCES

1. Y. Maruyama, "Neutron brachytherapy for the treatment of malignant neoplasia," *International Journal of Radiation Oncology Biology Physics*, **15**, pp. 1415-1429 (1988).
2. Y. Maruyama, J. G. Wierzbicki, B. M. Vtyurin, and K. Kaneta, "Californium-252 neutron brachytherapy," *Principles and practice of brachytherapy*, S. Nag (ed), Futura Publishing Co., Inc., Armonk, NY USA (1997).
3. R. C. Martin, R. R. Laxson, J. H. Miller, J. G. Wierzbicki, M. J. Rivard, and D. L. Marsh, "Development of high-activity ^{252}Cf sources for neutron brachytherapy," *Applied Radiation Isotopes*, **48**, pp. 1567-1570 (1997).
4. L. L. Anderson, "Status of dosimetry for ^{252}Cf medical neutron sources," *Physics in Medicine and Biology*, **18**, pp. 779-799 (1973).
5. W. Mannhart, "Evaluation of the Cf-252 fission neutron spectrum between 0 MeV and 20 MeV," *IAEA-TECDOC-410*, pp. 158-171 (1987).
6. V. V. Verbinski, H. Weber, and R. E. Sund, "Prompt gamma rays from ^{235}U(*n,f*), ^{239}Pu(*n,f*), and spontaneous fission of ^{252}Cf," *Physical Review C*, **7**, pp. 1173-1185 (1973).
7. K. Skarsvåg, "Differential angular distribution of prompt gamma rays from spontaneous fission of ^{252}Cf," *Physical Review C*, **22**, pp. 638-650 (1980).
8. H. E. Hootman and D. H. Stoddard, "^{252}Cf shielding guide," Savannah River Laboratory,

Aiken, SC (1971).
9. A. X. da Silva and V. R. Crispim, "Shielding design studies for a neutron irradiator system based on a ^{252}Cf source," *Radiation Protection Dosimetry*, **95**, pp. 333-338 (2001).
10. X-5 Monte Carlo Team. MCNP – A general Monte Carlo N-Particle transport code, Version 5, Los Alamos National Laboratory, Los Alamos, NM USA (2003).
11. "Computational Pile – LINUX Cluster," Nuclear Science and Technology Division, Oak Ridge National Laboratory, http://cpile.ornl.gov/index.html (2004).
12. M. J. Rivard, K. E. Evans, L. C. Leal, and B. L. Kirk, "Selective perturbation of in vivo linear energy transfer using high-Z vaginal applicators for Cf-252 brachytherapy," *Nuclear Instruments and Methods in Physics Research B*, **213**, pp. 621-625 (2004).
13. ICRU, "Tissue substitutes in radiation dosimetry and measurement," *International Commission on Radiation Units and Measurements Report 44*, Bethesda, MD USA (1989).
14. ICRU, "Nuclear data for neutron and proton radiotherapy and for radiation protection," *International Commission on Radiation Units and Measurements Report 63*, Bethesda, MD USA (2000).
15. ICRP, "1990 Recommendations of the International Commission on Radiological Protection," *International Commission on Radiation Units and Measurements Report 60*, Pergamon Press, Oxford, United Kingdom (1990).
16. J. H. Hubbell and S. M. Seltzer, "Tables of x-ray mass attenuation coefficients and mass energy-absorption coefficients (version 1.4)," National Institute of Standards and Technology, Gaithersburg, MD USA, http://physics.nist.gov/xaamdi (2005).