THE DESIGN AND CONSTRUCTION OF A PORTABLE IRRADIATION FACILITY USING CALIFORNIUM-252

A Thesis

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DEDICATED TO

MY FATHER AND MOTHER

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ABSTRACT

An irradiation facility using ²⁵²Cf has been designed and constructed to hold a source at least 100-µg. The facility met all its design specifications such as portability, minimum surface dose rates (100 mrem/hr) versus shield weight, ease of construction, minimum cost, significant biological shielding for protection of working personnel, good geometry for thermal neutron activation of Ca-48 in cement mixtures, and ease of use.

Experimentally measured dose rates on shield surfaces verified calculations for dose rates, source size, and shield dimensions. Calculated dose rates were larger than experimental dose rates by a factor of 1.25 or more due to the conservatism of ANISN (neutron transport code) and formulas used to calculate dose rates on the shield surfaces. The experimental dose rates proved a new shield could be designed with smaller dimensions than those calculated. Finally, the experimental dose rates gave evidence that a larger source could be used in the facility designed and constructed in this thesis.

CHAPTER I

INTRODUCTION

In the last few years at the Louisiana State University Nuclear Science Center, work has been conducted by F. A. Iddings and others in the area of the determination of percent cement in concrete by neutron activation.^{1,2,3} The process of neutron activation is accomplished by the use of a radioisotope called Californium-252 (²⁵²Cf) in a portable system so as to obtain results in the field.

Methods Studied other than ²⁵²Cf for Neutron Production

Methods other than the use of ²⁵²Cf have been studied for possible use in the activation of cement in concrete in a portable system. A neutron generator was the first method that was studied. This system though has several drawbacks, such as a high voltage D. C. power supply is needed; an unpredictable neutron flux; a very unstable system in regard to its ability to withstand such things as rough road conditions; a system requiring a lot of training and care in its use; and finally the cost of the system itself and its upkeep would be very high. One of the main advantages of a neutron generator for a portable system is in its ability to be turned off when not in use.

Since the concept of a neutron generator has been ruled out for use in a portable system, the only other alternative left is the use of a radioisotope for neutron production. The main requirements for a radioisotope to be used for neutron activation are a reasonably long

half-life, a sufficient neutron flux for activation of isotopes of interest in cement, an easily obtainable neutron source, and a low heat generation rate.

There are two methods of producing neutrons from radioisotopes. The two methods are composite sources such as PuBe, AmBe, etc., that produce neutrons by an (α, n) reaction, and sources such as 252 Cf that produce neutrons by spontaneous fission. Prior to this research, it was determined that a thermal neutron flux of at least 3.0×10^6 n cm⁻² sec⁻¹ was needed for activation of isotopes of interest in cement.⁴ Of the two methods of producing neutrons from radioisotopes, only 252 Cf can yield a sufficient neutron flux near that value as stated previously, and still meet all the requirements for a neutron source.

²⁵²Cf as a Neutron Source for Activation

Califorium-252 is a man-made radioisotope that decays by alpha, gamma, and spontaneous fission. Califorium-252 has a reasonably long half-life of 2.65 years, a large total neutron yield 2.4 x 10^{12} n - g⁻¹ sec⁻¹, a low heat generation rate of 39 watts per gram, and finally it is easily obtainable from the Energy Research and Development Administration's Califorium Demonstration Center at LSU.

Even though ²⁵²Cf has been chosen as the radioisotope to be used for activation of cement, several disadvantages of the use of this radioisotope should be mentioned here so as not to leave the impression that ²⁵²Cf is ideal. The major disadvantages in the use of ²⁵²Cf or any other isotopic source are as follows: a large and heavy biological shield is required to protect personnel from exposure

while using the source, an effective radiation safety program must be maintained, and finally the radioactive source can't be turned off.

Weighing the advantages and disadvantages against each other, the best choice for a method for neutron production for the problem in this research paper is by the use of the radioisotope ²⁵²Cf.

In all of the previous studies of cement determination in concrete by neutron activation using ²⁵²Cf, the portable irradiation facilities were either very difficult to build in regard to money and time spent in construction, ^{5,6} or the portable irradiation facility presented a potential radiation health hazard to someone not trained in its use.⁷

General Requirements of a Well Designed Portable Irradiation Facility

Design and construction of an optimum portable irradiation facility using ²⁵²Cf for determination of cement content of plastic concrete are the main themes behind this research paper. The general requirements for a well designed portable irradiation facility are as follows:

- Minimum weight-to-surface dose rate as per design specifications,
- Safe biological shielding for working personnel under all circumstances,
- 3. Reproducible irradiation geometry,
- Minimum training requirements in use and care of the facility,
- 5. Minimum care of facility required,

- 6. Minimum cost as related to design specifications,
- 7. Minimum construction requirements,
- Ability of facility to remain intact under normal and abnormal (accident) working conditions,
- 9. Easy operations of facility under design requirements, and
- 10. Safe source removal and replacement.

A discussion of how all the requirements for any well designed facility were met will be included in this research paper.

CHAPTER II

PRELIMINARY DESIGN CONSIDERATIONS

The shielding aspects of an irradiation facility must initially be considered before contemplating any possible design. In this chapter, the shielding aspects of the facility as related to radiation and federal law will be discussed.

Radiation Properties of ²⁵²Cf

Since 252 Cf has been chosen as the radioisotope for neutron activation of cement in concrete, a discussion of its radiation properties are important to the consideration of shielding. The radiation properties of 252 Cf are examined in terms of its neutron (n), alpha (α), beta (β), and gamma (γ) radiation.

Califorium-252 decays by alpha emission (97%) and by spontaneous fission (3%), as shown in Table 2-1 and Figure 2-1.⁸

The relative abundance and energies of the alpha particles emitted from ²⁵²Cf are given in Figure 2-1. The average alpha particle energy is 6.112 MeV.

No beta radiation has been reported in the decay process. The only sources of beta radiation are those which are associated with the formation of products caused by spontaneous fission, and with the decay of the fission products.

The gamma radiation from a ²⁵²Cf source consists of gamma rays following the alpha decay process, prompt spontaneous fission, and fission products continuously produced by spontaneous fission. Gamma

Decay Properties of ²⁵²Cf Alpha Decay Spontaneous Fission Total Specific activity, disintegrations or fissions per gram 1.92 x 10^{13} 6.14 x 10^{11} 1.98 x 10^{13} second Curies per gram^a 519.4 16.6 526.0 Half-life, years 2.731 25.5 2.676 Decay heat, watts/ 18.8 19.7 38.5 gram

^a A curie = 3.7 x 10¹⁰ disintegrations/second

^b Actually not a total, but rather an effective half-life given by

$$T_{eff} = \frac{T_{\alpha} T_{SF}}{(T_{\alpha} + T_{SF})}$$



Figure 2-1 Decay Scheme for ²⁵²Cf

radiation from each of these sources are described in Tables 2-2 and 2-3.

The energy and abundance of prompt gamma rays from spontaneous fission are listed in Table 2-3. The fission products formed from spontaneous fission approach equilibrium within a few hours after ²⁵²Cf separation. Equilibrium fission product gamma activities are listed in Table 2-3 also.

Fission fragment formation from spontaneous fission of ^{252}Cf is the main source for x-rays. Although relatively high in abundance, the x-rays produced are mostly less than 40 keV and are usually neglected in shielding calculations.

The neutron radiation from 252 Cf consists principally of neutrons from spontaneous fission. A very minor contribution arises from the (α ,n) reaction with high elements in the shield such as oxygen. The 252 Cf neutron energy spectrum can be represented by the Watt formula

$$N(E) = 0.373 e^{-0.88E} \sinh (2.0E)^{1/2}$$

in which N(E) is the fraction of neutrons per unit energy and E is the neutron energy in MeV.

Califorium-252 emits 2.4 x 10^{12} n g⁻¹ sec⁻¹, with 3.76 neutrons per spontaneous fission. The neutron distribution as a function of energy can be seen in Table 2-4. The average energy of the emitted neutrons in the neutron energy spectrum is 2.3 MeV, and the most probable energy is 0.8 MeV.

Gamma Rays from 252 Cf Alpha Decay Process

Energy, MeV	Abundance, photons/sec-gram		
0.043	2.8×10^{9}		
0.100	2.0×10^9		
0.156	4.0×10^8		

*

Gamma Rays from Spontaneous Fission of ²⁵²Cf

photons/(sec)(gram)

Gamma Rays from

Energy,	Prompt	Equilibrium Fission	
MeV	Gamma Rays	Products	Total
0 - 0.5	3.3×10^{12}	1.3×10^{12}	4.6 x 10^{12}
0.5 - 1.0	1.7×10^{12}	4.0×10^{12}	5.7 x 10^{12}
1.0 - 1.5	7.7 x 10^{11}	9.1 x 10 ¹¹	1.7×10^{12}
1.5 - 2.0	4.2×10^{11}	3.5×10^{11}	7.7 x 10^{11}
2.0 - 2.5	2.2×10^{11}		2.2×10^{11}
2.5 - 3.0	1.1×10^{11}		1.1×10^{11}
3.0 - 3.5	5.6 x 10 ¹⁰		5.6 x 10^{10}
3.5 - 4.0	3.0×10^{10}		3.0×10^{10}
4.0 - 4.5	1.7×10^{10}		1.7×10^{10}
4.5 - 5.0	8.2 x 19 ⁹		8.2 x 10 ⁹
5.0 - 5.5	4.9 x 19 ⁹		4.9 x 10 ⁹
5.5 - 6.0	1.8 x 10 ⁹		1.8 x 10 ⁹
6.0 - 6.5	1.0×10^{9}		1.0×10^9
Total	6.6 x 10 ¹²	6.6×10^{12}	1.3×10^{13}

Neutrons From Spontaneous

Fission Of ²⁵²Cf

Energy, MeV Neutrons/(sec)(gram)
$10.0 - 14.92$ 8.28×10^9
$6.70 - 10.00$ 6.42×10^{10}
$5.49 - 6.70$ 8.19×10^{10}
$4.49 - 5.49 \qquad 1.26 \times 10^{11}$
$3.68 - 4.49$ 1.68×10^{11}
$3.01 - 3.68$ 2.0×10^{11}
$2.02 - 3.01$ 4.36×10^{11}
$0.91 - 2.02$ 6.98×10^{11}
$0.41 - 0.91$ 3.36×10^{11}
$0.11 - 0.41$ 1.56 x 10^{11}
$0.015 - 0.11$ 2.74×10^{10}
0.0 - 0.0150
Total 2.3 x 10 ¹²

Selection of Shielding Material

With knowledge of the various types of radiation from the radioisotope (²⁵²Cf) to be used in the facility, selection of the optimum shielding material would seem to be the next logical step. Factors that must be considered when selecting the shielding material include cost, safety (as related to exposure to radiation), reliability, weight (as related to design specifications), and maintenance requirements. Properties to be considered include density, durability, and heat transfer characteristics (ignored for ²⁵²Cf due to low heat rate).

Safety as concerned with radiation exposure is the most important factor in shielding-material selection. To obtain maximum radiation safety (as per design specifications) for a 252 Cf shielding facility, knowledge of which of the types of radiation from 252 Cf are to be shielded must be known. The low penetrating power of α , β , and x-ray radiation from 252 Cf justifies neglecting the considerations of these radiations from shielding calculations. The design of shielding for 252 Cf must cope with fast neutrons and primary gammas from spontaneous fission, equilibrium fission product gamma rays, and finally capturegamma rays produced in the shield by a (n, γ) reaction.

Hydrogeneous materials such as water, polyethylene, and paraffin are used for shielding fast neutrons; heavy materials, such as lead and iron are used for shielding gamma rays. For ²⁵²Cf, shielding of thermal neutrons is not a significant problem due to the fact that the dose rate from thermal neutrons is less than 10% of the total dose rate on the surface of the shield. A comparison of different types of shielding materials that can be used for 252 Cf can be seen in Table 2-5. Water-extended polyester (WEP) was determined to be the best shielding material considering the various factors and properties that are required of a shielding material for 252 Cf in conjunction with Table 2-5. WEP was chosen from all the other possible shielding materials for the following reasons:

- Ease of fabrication WEP easier to fabricate than wood, paraffin, etc.;
- Fire resistant WEP more resistant to fire than paraffin, lucite, or polyethylene;
- Low cost WEP costs much less to make in large volume than lucite or polyethylene;
- Radiation shielding properties for WEP are excellent in regard to neutron radiation versus those properties of concrete;
- WEP does not leak or evaporate, as water might do;
- 6. Strength intermediate between wood and concrete;
- Low weight as compared to other shields as concrete;
- Prior experience at LSU Nuclear Science Center with its use;
- 9. Ability to add materials such as Li and B easily during construction so as to further reduce the dose rate on the shield surface; and

COMPARISON OF SHIELDING MATERIALS

	Density,	Number of H	
<u>Material</u>	grams/cm ³	atoms/cm ³	Comments
Ordinary Concretes:			
01	2.33	0.29×10^{22}	Inexpensive, fireproof, easily formed, structurally
02A	2.26	1.38×10^{22}	useful; but composition can vary widely: the water
03 ^a	2.37	1.20×10^{22}	content is strongly affected by type of aggregate
			used and by curing time and temperature.
"Lucite,"	1.18	5.7 x 10^{22}	Has structural strength; is easily formed into
(C ₅ H ₈ O ₂) _x			intricate shapes such as multiple source holders.
Water, H ₂ O	1.00	6.69×10^{22}	Cheapest (primary cost is that of containment
			vessel) and most available neutron shield; fireproof,
			but can leak and evaporate.

^a The concrete used at the Savannah River Plant has a density of 2.33 grams/cm³ and a hydrogen content of 1.26 x 10²² hydrogen atoms/cm³.

TABLE 2-5 (continued)

COMPARISON OF SHIELDING MATERIALS

	Density,	Number of H	
<u>Material</u>	grams/cm ³	atoms/cm ³	Comments
Water - extended	1.1	6.38 x 10^{22}	Solid containing up to 65% H ₂ O; easily prepared in
polyester, WEP-47 ⁵⁸			various shapes by curing aqueous emulsion of styrene
			monomer and polyester resin; radiation-resistent,
			fire-resistant; low cost (comparable to concrete);
			can contain added materials (suspended or dissolved)
			such as Li, B, Cd; can be machined, drilled; can be
			sealed by polyurethane paint to prevent dehydration;
			strength intermediate between wood and concrete.
Paraffin, C ₃₀ H ₆₂	0.92	8.18 x 10^{22}	A superior neutron shield due to its high hydrogen
			content; easily formed but must be used with care
			because of its low melting point and flash point;
			can be borated with B4C.
Iron, Fe	7.9	None	Heavy material for gamma shield.
Lead, Pb	11.3	None	Heavy material for gamma shield.

10. WEP is sturdy enough to hold dense materials

such as steel or lead at its core.

Description of Water-Extended Polyester (WEP)⁹

With WEP as the optimum choice of all the shielding materials for ²⁵²Cf, a general description of WEP (both physical and chemical) is presented below.

Unsaturated polyester resins readily emulsify with water. When properly catalyzed, the emulsion cures by an exothermic reaction into a hard material similar to a fine-grained plaster. WEP consisting of equal parts of styrene monomer and polyester resin can be combined with up to 65% water (by weight). The cured emulsion has an average atomic number of 3.50 and an average density of 1.1 g/cc.

The catalyst (H_2O_2) thermosets the WEP emulsion at room temperature in a matter of minutes. The rate of hardening of the WEP emulsion depends upon many factors, such as temperature of water, room temperature, ratio between water and WEP, completeness of mixing of water and WEP, and finally, volume of WEP and water.

The catalyst is added to the WEP-water mixture while the mixture is being stirred with an electric drill. The catalyst must be completely mixed with the WEP and water to insure uniform hardening. Care must be taken during mixing to prevent trapping of air bubbles. Large batches of WEP emulsion can be prepared without the final catalyst and stored for days with no loss of curability or other properties.

When molded, WEP shrinks about 0.5% - 1.5%, depending on water content, while it cures. The best mold material for smooth surfaces is polyethylene, but practically any mold would do if not interested in a smooth surface. As mentioned previously, the cured WEP has a physical strength between that of concrete and wood; its compressive strength is less than concrete, but four times that of wood. At greater than 65% water content, cell-to-cell diffusion of the water droplets occurs; water entrapped on exposed surfaces evaporates quickly after the curing process is completed. Polyurethane paints will seal the outer surface to prevent more than superficial dehydrating.

From results of work done by Oliver and Moore,⁹ samples of cured WEP submitted to a temperature of 400°C did not burn. At such temperatures, dehydration of the WEP occurs with loss of shape. Upon cooling, the sample was harder and more brittle than the original. Radiation effects tests showed that a 10⁷ Roentgen exposure (Cs-137) created no structural or decomposition problems.

WEP Shielding for Neutrons and Gamma Rays

Neutrons are best shielded by a material that has a high hydrogen content (greatest energy loss per collision of all elements). With WEP having a high hydrogen content near that of water (6.38 x 10^{22} atoms/cc versus 6.69 x 10^{22} atoms/cc), it is obvious that WEP would have good neutron shielding properties.

In systems composed of light nuclei, the moderation of neutrons results almost entirely from elastic scattering. As the average energy of the neutrons from ²⁵²Cf is 2.3 MeV, inelastic scattering is predominant until the neutron energy is decreased to less than 1 MeV where elastic scattering increases with decreasing energy.¹²

The most compact, effective shielding material for gamma rays are materials like iron, lead, and uranium. Materials like lead, uranium, etc., have a high cross section (probability) for gamma ray interaction due to their composition and density.

Primary gamma ray shielding by lead (gamma shield chosen in this paper) from the ²⁵²Cf source itself is most effectively accomplished by placing the lead shield directly next to or very near the source. The thickness of this shield will be determined in Chapter III under shielding calculations.

In regard to the production of capture gamma rays, 252 Cf shielding is similar to nuclear reactor shielding; the spontaneously emitted neutrons react with the shield to produce secondary radiation (capture gamma rays), which may in some cases be the dominant contributor to the penetrating dose rate. Thus the 1 H(n, γ)²H reaction; which has a thermal neutron cross section of 0.33 barns, produces a 2.23 MeV capture gamma in a hydrogenous shield. Production of this energetic gamma may be partially suppressed by adding boron to the hydrogenous shield; the competing 10 B(n, γ)⁷Li reaction, which is favored by a high thermal neutron cross section (3.84 x 10² barns), yields a more easily attenuated 0.48 MeV gamma. As an example of the

reduction in capture gamma dose rate see Figure 2-2. The use of Figure 2-2 will become much more apparent later on in this paper in the shielding calculations chapter.

The capture-gamma dose rate from the ${}^{1}H(n,\gamma){}^{2}H$ reaction can also be reduced by adding natural lithium (7.65% ${}^{6}Li$) to the hydrogenous shield. The competing ${}^{6}Li(n,\alpha){}^{3}H$ reaction, which has a thermal neutron cross section of 945 barns, produce no gamma radiation. It can be seen in Figure 2-3 how much the total (neutron + gamma) dose rate can be reduced by adding natural lithium to the hydrogenous shield.

Typically, the addition of boron to a hydrogenous shield also reduces the dose rate from the thermal neutrons to approximately 0.1 - 1.0% of the dose rate from neutrons of all energies. But since the thermal neutron dose rate constitutes no more than about 10% of the total dose rate even in the absence of boron, adding boron to the hydrogenous shield does not significantly reduce the total neutron dose rate to personnel outside the shield.

Boron was used in the design of the shield in this paper for several reasons. It is apparent from Figures 2-2 and 2-3, that the slope of the boron curve is much greater than the slope of the lithium curve. This greater slope means that the boron effects the total dose rate more than the lithium. Similarly, the relative concentrations of boron and lithium in the figures lead to the conclusion that a smaller amount of boron is needed than lithium to obtain the same results. Therefore to summarize, a smaller radius



shield could be built with boron than lithium, thus making the shield lighter and cheaper.

Thought was given to a lead shield completely around the facility which would shield both the primary gamma and capture gamma rays. The idea was immediately disregarded because the great weight of the lead would be in direct conflict with one of the main objectives of this paper which is minimum weight.

DOT Regulations Concerning ²⁵²Cf Facility

The application is the most important consideration in determining just what are the Department of Transportation's (DOT) regulations concerning the 252 Cf facility in this paper. The main application of the 252 Cf facility in this paper is that of a portable neutron irradiation facility, not a shipping facility.

Even though the facility in this paper is not classified as a shipping facility, the permissible dose rates for shipment according to DOT were adhered to. According to DOT the radiation dose rate must never exceed:

- (a) 200 mrem/hr at any point on the external surface of the cask,
- (b) 10 mrem/hr at 3 feet from any accessible external surface of the cask.

The permissible dose rates for storage according to DOT were also complied to. The dose rates for storage are given in Table 2-6.¹⁰

	Permissible Dose per	Permissible Dose per
Part of Body Irradiated	Quarter Year, rem	Year, rem
Whole Body; head and		
trunk; active blood-		
forming organs; lens of		
eye; gonads	1.25	5.0
Skin of whole body	7.5	30.0
Hands and forearms;		
feet and ankles	18.75	75.0

CHAPTER III

DETERMINATION OF THE OPTIMUM DESIGN OF

THE ²⁵²Cf IRRADIATION FACILITY

Knowing what the source is, the shielding materials to be used, and the federal laws governing the dose rates on the shield, it is now appropriate to begin the discussion concerning the shape, size and final dimensions of the irradiation facility.

Physical Shape of Shield

Consideration of the shape of the shield is extremely important in that shape is related closely to weight. In determining the final shape of the shield, several factors besides weight must be considered, such as portability, cost of construction as related to materials, and ease of construction.

The possible shapes of the shield in terms of volume are given below for a cube, cylinder, and a sphere. The volume formulas are:

Cube $V = s^3$ [s = length of side] Cylinder $V = \pi r^2 h$ [r = h/2 for minimum weight] = 0.785 s^3 Sphere $V = 4/3 \pi r^3$ [r = h/2] = 0.523 s^3

Obviously from the above formulas, a sphere would give the minimum weight. Due to the fact that a sphere doesn't meet two of the three requirements (ease of construction and cost of construction) in choosing the final shape of the shield as stated previously, a shield in some form of a cylinder was chosen.

A cylinder, as with a sphere, is extremely difficult to build due to its curved sides; therefore, an octagonal approximation to a circular cross section for a right cylinder was used to give minimum weight and ease of construction as seen in Figure 3-1. The shield in Figure 3-1 will be referred to as an octagonal or octagonshaped shield throughout the rest of this paper.

The cross section of a right cylinder can figuratively be represented by an octagon, as seen in Figure 3-1. Determination of the mathematical relationship between a cylindrical and a octagonal shield is as follows:

Volume of octagonal	shield = area of base x height
Area of base	= area of 16 right triangles
	$= [0.5] [H/2] \times r \times 16$
in which:	H/2 = base of right triangle [Figure 3-1]
	r = height of right triangle
	Height = 2r [minimum weight for shield]

Therefore,

Volume of octagonal shield = $4Hr \times 2r$ = $8Hr^2$ Since tan 22.5° = [H/2]/r.8284r = H

Volume of octagonal shield = $6.6272r_1^3$ [3-1]



Volume of cylindrical shield = $\pi r^2 h$ [h = 2r] = 6.2832r³.

With the volume given above for both that of a cylindrical shield (V_c) and that of an octagonal shield (V_o) , $V_c/V_o = 1.0548$. Therefore, an octagonal shield represents a cylindrical shield (for all practical purposes) both figuratively and mathematically within 5.48% by volume or weight.

With its rectangular sides and a weight nearly that of a cylindrical shield, an octagonal shield is the ideal choice for ease of construction, weight, and portability if a convenient cylinder is not available.

Mathematical Shielding Formulas

The final design limitations should be noted before beginning any shielding calculations. The limitations are as follows:

- 1. Shielding material is WEP
- 2. Primary gamma shield is lead
- 3. Borated WEP [BWEP] will be used in shield
- 4. Octagonal shield will be used
- 5. Boration¹¹: a $0.6^{10}B$
- 6. Dose rate on shield surface less than or equal to 100 mrem/hr for a maximum source size of 100 µg of ²⁵²Cf
- 7. Shield weight less than 1200 pounds [axle rating of most small trailers]

The above design limitations are those that are directly concerned with the shielding calculations. The first formula to be discussed concerns the total weight of the shield, which is a function of the weight of the WEP, the primary gamma shield around the source, and the weight of the WEP displaced by the primary gamma shield. The total weight of the shield in pounds equals A - B + C in which A, B, C are as follows:

> A = total weight of WEP if primary gamma shield not considered

 $A[1bs.] = [\rho] [V] \times 1/453.5$

in which:

 ρ = density of WEP = 1.1 g/cc V = volume of shield (cc) = 6.6272r³

Therefore,

$$A[1bs.] = \frac{[1.1] [6.6272r^{3}]}{453.5}$$
$$= 0.016072r_{1}^{3}$$

B[lbs.] = total weight of WEP displaced by primary

gamma shield

 $B[1bs.] = [\rho] [V] \ge 1/453.5$

in which:

 ρ = density of WEP = 1.1 g/cc V = volume of displaced WEP = $\pi r^2 h$ [h = 2r, cylinder] 1/453.5 = conversion factor grams to pounds
Therefore,

$$B[lbs.] = \frac{[1.1] [6.2832r_2^3]}{453.5}$$
$$= 0.015240r_2^3$$

 $C[1bs.] = [\rho] [V] \ge 1/453.5$

in which:

 ρ = density of lead = 11.35 g/cc

 $V = volume of cylinder [\pi r^2 h, h = 2r]$

Therefore,

$$C[lbs.] = \frac{[11.35] [6.2832r_2^3]}{453.5}$$

 $= 0.15722r_2^3$

Finally, the total weight of shield = A - B + C

$$= 0.016072r_1^3 - 0.015240r_2^3 + 0.15722r_2^3$$
$$= 0.016072r_1^3 + 0.14197r_2^3 \qquad [3-2]$$

in which:

r1 = total radius of shield in cm
r2 = cylindrical radius of primary gamma
shield in cm

There is no direct given formula that can be used to determine the total dose rate (neutron and gamma) on an octagonal shield surface. However, formulas for a spherical shield are available, and can be used to give slightly conservative results (a higher dose rate than expected). The total dose rate at the surface of an octagonal shield may be determined by the following formula which is:

 $D_{T}[mrem/hr] = [D_{nT} + D_{\gamma T}] \times 1/4\pi r_{1}^{2} \times 2.4 \times 10^{12} \times Z$ [3-3] in which:

> D_{nT} = total fast and thermal neutron dose rate in [mrem - hr⁻¹/neutrons - sec⁻¹] cm² $D_{\gamma T}$ = total primary and capture gamma dose rates 2.4 x 10¹² neutrons/g - sec = total neutron yield from spontaneous fission of ²⁵²Cf Z = source strength in grams = 100 x 10⁻⁶ grams r = total radius of shield (WEP radius and

primary gamma shield radius)

In using the formula given above for total dose rate at shield surface in conjunction with the graphs to be given¹², it is convenient to put the values obtained from the graph in a tabular form. D_{nT} (fast and thermal) and $D_{\gamma T}$ (primary and capture) dose rates without any correction factors can be obtained from Figure 3-2. Correction factors such as boration reduction and attenuation of gammas by lead can be obtained from Figures 3-3 and 3-4. An example of one shielding calculation is shown below:

Given: Radius of WEP = 35 cm

Source Size = $100 \ \mu g$

Radius of primary gamma shield = 2 cmTotal Radius (r) = 35 + 2 = 37 cmBoration = 0.6 mg/cc













Dose Constituents	$4\pi r^2 D(r)^a$	Correction	Total Dose Rate
Neutron			
Fast	2.0×10^{-3}		2.0×10^{-3}
Thermal	3.2×10^{-4}		3.2×10^{-4}

Gamma

Primary	2.9×10^{-3}	$2.9 \times 10^{-1}^{b}$	8.41 x 10^{-4}
Capture	2.0×10^{-3}	$3.4 \times 10^{-1}^{c}$	6.8×10^{-4}
			3.84×10^{-3}

^a these values obtained from Figure 3-2 for r = 35 cm in [mrem - $hr^{-1}/neutrons - sec^{-1}$] cm².

^b this value obtained from Figure 3-3 for r = 2 cm.

^c this value obtained from Figure 3-4 for $^{10}B = 0.6 \text{ mg/cc}$.

Using now Formula 3-3

$$D_{T}(mrem/hr) = \frac{(3.84 \times 10^{-3}) (2.4 \times 10^{12}) (100 \times 10^{-6})}{4\pi (37)^{2}}$$

 $D_{T} = 53.5 \text{ mrem/hr}$

The total weight of shield = $0.016072(37)^3 + 0.14197(2)^3$

= 815.2 lbs.

The above example was given to illustrate the basic calculations used to determine dose rate and shield weight. Changes in the primary gamma shield and WEP radius can easily be accommodated by Formulas 3-2 and 3-3.

Initial Determination of Optimum Shield Dimensions

Using Formula 3-2 and 3-3 and considering the limitations of 1200 pounds and 100 mrem/hr, the maximum and minimum values were chosen to be 40 to 25 cm for the amount of borated WEP (BWEP) and 0 to 10 cm for the primary gamma shield radius.

Determination of when a primary gamma shield is needed can be done both graphically and mathematically. Keeping in mind the maximum and minimum values of the amount of BWEP (BWEP radius slightly smaller than non-borated WEP, called just WEP, as will be seen later in this paper), the graph in Figure 3-1 shows that from about 20 to 37 cm of WEP the principal source of radiation is fast neutrons, whereas, from 37 cm and above, the principal source of radiation is due mainly to both primary and capture gammas. Therefore, from Figure 3-1, it can be concluded graphically that for a shield 37 cm or more in radius a primary gamma shield is needed.

Two important conclusions can be drawn from Table 3-1: Determination of whether a primary gamma shield is needed, and a facility with a primary gamma shield weighs about the same as one without (with the design specifications in this paper).

Looking at 35 cm of WEP and at 0 cm primary gamma shield radius, it is seen that the dose rate is 92 mrem/hr (close to 100 mrem/hr as per design specifications). This proves that it is possible to have a 252 Cf shield with no lead shield having a weight of about 690 lbs.

		Lead Shield Radius (cm)										
		Shield Weight (10 ² lbs.)					Dose Rate (10 ² mrem/hr)					
Amount of WEP(cm)	0.0	2.0	4.0	5.0	7.5	10	0.0	2.0	4.0	5.0	7.5	10
40	10.3	12.0	13.8	14.9	17.8	21.5	0.590	0.336	0.256	0.250	0.216	0 .179
35	6.89	8.15	9.62	10.5	12.9	16.1	0.920	0.537	0.411	0.384	0.325	0.278
30	4.34	5.28	6.41	7.07	9.07	11.7	1.97	1.26	0.996	0.924	0.781	0.681
25	2.51	3.18	4.01	4.52	6.11	8.30	4.79	3.28	2.64	2.43	2.02	1.74

* The amount of WEP is not the same as the radius of the main shield due to the addition of the lead shield

Table 3-1 - Dose Rates (at the external surface of the shield) and <u>Shield Weights</u> for various combinations of Amounts of WEF and Primary Gamma Shield Radii

ω S If a lead shield is used, the best choice would be a 4 cm radius shield as seen in Table 3-1. With a 4 cm lead shield and design specifications in mind, it can be seen from Table 3-1 that about 30 cm or more of WEP (shield radius 34 cm or more) will give a dose rate near 100 mrem/hr and a shield weight of about 640 lbs.

From the conclusions drawn from Table 3-1, a choice must be made as to whether a primary gamma shield is desirable. The choice between using or not using a gamma shield will depend not only on weight, but on ease of construction, and the effect of the lead on the neutron flux. Since 690 lbs. (no lead shield) is near that of 640 lbs. (4 cm lead shield) and that the values in Table 3-1 were calculated assuming the lead completely surrounded the source, a final source-lead-WEP arrangement must be decided upon before determining the final weight of the shield with a primary gamma lead shield so as to compare it to one without. The final source-lead-WEP arrangement must also be determined to see what, if any, the effect the lead near the source has upon the neutron flux.

Determination of Final Shielding Dimensions With and Without a Primary Gamma Shield

Much work has been done prior to this research concerning the optimum source-lead-WEP arrangement in regard to the required neutron flux for activation of ⁴⁸Ca (main isotope of interest) in cement in concrete. The research was conducted by Miller and

Iddings^{13,14}. From their results, a source-lead-WEP arrangement will be shown that gave an adequate neutron flux for activation, was easy to construct, and was of minimum possible weight.

The main problems that were investigated by the experiments of Miller and Iddings are as follows:

- Does lead right next to or completely around the source affect the flux and energy spectrum of the neutrons hitting the sample?
- 2. If lead near the source affects the neutrons, what is the optimum distance from the source to the lead shield with WEP in between to give an adequate neutron flux and energy spectrum for activation?
- 3. Does borated WEP (BWEP) near the lead shield effect the neutron flux and energy spectrum?
- 4. If BWEP effects the neutrons, what is the closest distance the BWEP can be placed near the source without much effect upon the neutrons?
- 5. What is the optimum thickness of a moderator to give maximum neutron activation of ⁴⁸Ca in concrete?

Questions 1 and 2 above are concerned directly with a primary gamma shield, whereas, questions 3, 4, and 5 are concerned with any shield whether it has a lead shield or not. The answers to the above questions will be presented in the order that they were given with some discussion about each.

It was determined, as expected, that both lead and BWEP near the source definitely did effect the neutron flux and energy spectrum. There are several possible reasons for this:

- Impurities in the lead, such as cadium, may have absorbed neutrons.
- 2. The scattering angle of lead is small (compared to hydrogen), therefore, there is little possibility of a neutron from scattering from any large angle into the direction of the sample.
- 3. With lead completely around the source, this prevents any hydrogenous material from being placed near the source to help moderate the neutrons.

The main reason that the BWEP affected the neutron flux was that the ^{10}B has a high thermal neutron cross section for a (n,γ) reaction.

In Figure 3-5 is a drawing of the source-lead-WEP arrangement that was determined to be optimum in terms of an adequate neutron flux and energy spectrum for activation, ease of construction,



Figure 3-5 <u>Source-Lead-WEP Arrangement</u> for a 4 cm Primary Gamma Lead Shield(cm)

and minimum weight. Note in Figure 3-5, the distance from the source to the lead shield. The distance from the outside of the lead shield to the BWEP was determined to be 12 cm. The thickness of the moderator on top of the source was determined to be 3.2 cm (1.25 inches) of plexiglass to give maximum activation of ⁴⁸Ca. With the final source-lead-WEP arrangement determined, thought must now be given to determining whether a primary gamma shield is needed to meet the design dose rates and shield weights.

The values for the dose rates and shield weights for the various amounts of WEP or shield radii given in Table 3-1 was only a rough estimate for BWEP. Since there is both BWEP and WEP in the types of shields considered in this thesis, provision must be made for determining the true dose rate on the shield surface. Beginning with Table 3-1 and using Formulas 3-2 and 3-3, in conjunction with the design limitations (100 mrem/hr and 1200 lbs.), various combinations of BWEP and WEP with and without a gamma shield was used to determine the optimum shield radius and weight as seen in Table 3-2.

The data from Table 3-2, is plotted in Figures 3-6 and 3-7. Note in Figure 3-6 that the total radius of the shield is plotted for the 4 cm primary gamma shield (not the amount of WEP) versus the dose rate. From Figure 3-6, considering the EWEP and WEP curves, it is concluded that a shield radius of about 34.5 cm is the optimum value with a 4 cm lead shield (amount of WEP = 31.75 cm), whereas, a shield radius of about 35.25 cm is the optimum value

1	4 cm Pri	mary Gamm	a Lead Sh	No Lead Shield			
Total Radius of Shield (cm)	Amount of WEP (cm)	Dose Rate B-WEP mrem/hr	Dose Rate WEP mrem/hr	Shield Weight (lbs.)	Dose Rate B-WEP mrem/hr	Dose Rate WEP mrem/hr	Shield Weight (lbs.)
29	25	263	301	401	227	261	392
30	26	200	234	443	197	229	431
31	27	168	201	488	163	190	478
32	28	140	169	536	142	167	527
33	29	119	147	586	123	146	578
34	30	99.6	125	641	10 9	129	632
35	31	78.4	99.6	698	92.0	112	689
36	32	69.3	89.3	759	<u>.</u>		
37	33	58.6	76.8	823			
38	34	50.8	67.5	891			
39	35	40.9	57.5	962			

Table 3-2 Dose Rates and Shield Weights for both a BWEP and WEP Shield with and without a Primary Gamma Lead Shield versus Total Shield Radii



Figure 3-6 Dose Rates for both a BWEP and WEP Shield with and without a Primary Gamma Lead Shield versus Total Shield Radii



Figure 3-7 <u>Dose Rates</u> versus <u>Shield Weights</u> for a BWEP Shield with and without a Primary Gamma Lead Shield

with no lead shield. From Figure 3-7, a facility with no lead shield would be approximately 30 lbs. heavier than one with. As seen in Figure 3-1 previously, no lead shield was needed for 37 cm or less of WEP. Therefore, for the reason stated above and knowing that a facility with no gamma shield would be easier to build and give a larger neutron flux than one with, a facility with no gamma shield was chosen.

With the final shield dimensions known (radius = 36 cm and height = 72 cm to give conservative dose rates on shield surface), the operational design of the shield as a portable irradiation facility to determine percent cement in concrete by neutron activation must now be considered.

Irradiation Facility Design and Dimensions

Several methods of irradiating concrete samples were thought of and disregarded due to flaws in their designs. Drawn in Figures 3-8, 3-9, and 3-10 are conceptual drawings of several methods of irradiating concrete samples that were disregarded. The movable shelf concept of irradiating samples (Figure 3-8) was disregarded due to the fact that as the shelf is pulled out there is a void of WEP above the source so as to give a high dose rate on the top of the shield. The movablecylinder concept of irradiating samples (Figure 3-9), where the sample was rotated over the source, was dismissed due to its great weight, difficulty in building, and the impracticality in its shape. Finally, the movable-top-half-of-shield concept of



Figure 3-8 Top and Side View of <u>Movable-Shelf Concept of</u> <u>Irradiation</u>



Figure 3-9 Movable-Cylinder Concept of Irradiation



Figure 3-10 Movable-Top-Half-of-Shield Concept of Irradiation

irradiating samples (Figure 3-10), where top-half of shield moved over source to irradiate the sample, was not considered (as in case of movable cylinder concept) because of its great weight and difficulty in construction.

Keeping in mind that the source must always be covered by WEP, a revised method using the movable-shelf concept was finally selected as the operational design of the irradiation facility as seen in Figure 3-11. The movable shelf extension in Figure 3-11 prevents any cavity from being formed above the source when installing or removing a sample.

With the physical dimensions of the shield (radius = 36 cm and height = 72 cm) having already been determined, the only dimensions left to be determined are those of the movable shelf. The dimensions of the movable shelf are determined partially by the dimensions of the sample. Determination of the ideal dimensions for a sample for maximum activation versus that of a commerically available plastic bucket was one of the major unforeseenable problems in this research paper.

Figures 3-12 and 3-13 were used to gain an insight into the approximate size of the sample needed to obtain maximum activation.¹⁵ From Figures 3-11 and 3-12, a search was begun to find a commerically available bucket with dimensions of at least 12.7 cm (5.0 inches) in height and a volume of 6.22×10^3 cc (380 in.³) to 6.56×10^3 cc (400 in.³).















Figure 3-13 Increase in cement content signal with sample size for concrete slabs 12.7 cm (thick)

The main ideas behind the search for a bucket was to find one that was plastic (metal would possibly get activated), easily obtainable, had minimum dimensions for maximum activation, and one that deformed little when filled with plastic concretee. After much searching for the ideal plastic bucket, it was found that there was none available.

At his point consideration was made as to whether the volume was the most important parameter for neutron activation. With this idea in mind, experiments were conducted using plastic concrete sample of different volumes and geometries (different diameters). One sample was about 6.22×10^3 cc (diameter = 23.5 cm) with 12.7 cm thickness of concrete, whereas, the other sample was about 4.59 x 10^3 cc (diameter = 20.3 cm) with 12.7 cm thickness of concrete. The larger diameter sample was formed in a plastic bucket that was cut to specified dimensions, whereas, the smaller sample was formed in a commerically available plastic paint bucket. The samples (2 of each the same kind) were irradiated using 20 µg of ²⁵²Cf (in a irradiation facility designed by Pepper¹⁶) for 5 minutes, allowed to decay 1 minute, and counted for 5 minutes using a single channel analyzer and a 5" x 5" NaI(TL) crystal. The results of the experiments are shown in Table 3-3.

The results from Table 3-3 indicate that pass a volume of about 4.59 x 10^3 cc (diameter = 20.3 cm) any more increase does not give any appreciable increase in total counts. The greatest

TABLE 3-3

Results of Experiment to Determine Importance of

Volume

Counting Sequence: 5 min. activation - 1 min. decay -

5 min. counting

Background/5 min. = 2695, 2516, 2682, 2715

	Diameter (20.3 cm)	Diameter (23.5 cm)
Wet Concrete:	15785	13168
	13993	12889
Dry Concrete:	14328	12920
	13997	12301

errors in counting of identical samples were from sample preparation. Therefore, the conclusion from the experiment is that the small-volume plastic bucket is optimum in terms of sample size, geometry presented to source, and availability (Figure 3-14).

Knowing now what the standard plastic bucket dimensions are, the final dimensions of the movable shelf can be determined.

The movable shelf must be one solid piece of WEP throughout. Knowing this, the final dimensions of the movable shelf are as follows. The width of the movable shelf is determined by considering the diameter of the sample bucket when filled with plastic concrete (20.3 cm) and by adding 2.54 cm to each side of the bucket to give strength to the shelf and to keep the shelf as one solid piece, this giving a width of 25.3 cm. Since 25.3 cm is close to 29.8 cm (width of edge of shield, see Figure 3-15), the width of the movable shelf was made 29.8 cm for ease of construction. The length of the movable shelf is determined by considering the distance the shelf must travel to remove the sample, keeping in mind the source must always be shielded. Wtih a 20.3 cm diameter sample, the length of the shelf is 72 cm (diameter of shield) plus 48 cm (distance shelf must be moved to remove sample), which equals 120 cm.

Before determining the height of the movable shelf, it should be noted that some sort of support plate is needed under the sample bucket to hold it in the shelf. Since a moderator



plate is needed on top of the source to obtain an optimum neutron flux and energy spectrum, a combination moderator-support plate (plexiglass) would be an ideal solution to the supportmoderator problem. A 1.25 cm thick moderator-support plate would be more than enough for strength, thus giving a total height of the movable shelf as 17.75 cm (16.5 cm for height of bucket plus 1.25 cm for moderator).

The final dimensions of the portable irradiation facility are shown in Figure 3-15 and Figure 3-18. The weight of the movable shelf was estimated at about 110 lbs. and the total weight of the shield about 750 lbs.











Figure 3-17 Side View of Both Top and Bottom of Shield(cm)





Figure 3-18 Top and Side View of Movable Shelf

CHAPTER IV

RESULTS

Dose Rates on Shield Surfaces

Presented in Figures 4-1, 4-2, and 4-3 are the gamma, neutron, and total (neutron + gamma) dose-equivalent rates (mrem/hr) on the shield surfaces with a $1000-\mu g^{252}Cf$ source installed. A $1000-\mu g$ source, instead of the designed source size of $100-\mu g$, was the only available high-intensity source with which the facility could be tested adequately.

From Figure 4-3, the largest total dose rate was 560 mrem/hr on top of the shield. Using Formula 3-3 and Figure 3-2 in Chapter III, the total dose rate for a 1000-µg source was calculated to be about 1000 mrem/hr for a shield constructed from either ordinary or borated WEP. The difference between 560 (experimental) and 1000 (calculated) mrem/hr is as expected for several reasons:

- The ANISN neutron-transport code was used to calcululate the values used in Figure 3-2.¹⁷ The ANISN code gives conservative values by a factor of 1.25 or more.
- 2. Formula 3-3 used to calculate the total dose rate is a formula for a spherical shield; therefore, the experimental values would be expected to be lower on an octagonal shield than the calculated.













Figure 4-2 External <u>Gamma Exposure Rates(mr/hr)</u> on Shield Surfaces for a 1000 microgram Source



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Figure 4-3 External <u>Total(Gamma + Neutron) Dose-Equivalent</u> Rates(mrem/hr) on Shield Surfaces for a 1000 microgram Source 3. The choice of a shield radius of 36 cm was determined considering both borated and nonborated WEP. The 36 cm radius was chosen to give the maximum biological shielding even if the shield was completely non-borated; thus, the calculated values were expected to be conservative.

As seen in Figures 4-1 and 4-2, the gamma dose rates are larger in most cases than the neutron dose rates, and the greatest dose rates are found near the cracks in the movable shelf, as expected.

Facility Operation

As may be seen from the data in Table 4-1 and the graph in Figure 4-4, the shield performed adequately as an irradiation facility for the activation of Ca-48 in cement in soil-cement mixtures. The data in Table 4-1 can not be compared directly to data from previously reported experiments because a smaller detector (3" x 3" vs. 5" x 5") was employed for the current work.

Shield Construction

Several comments should be noted about the actual construction of the shield. The form of the shield took approximately three sheets of plywood (4 ft. x 8 ft. x 5/8 in.) to construct. Silicone sealer was used to prevent leaking of liquid WEP from the form.

TABLE 4-1

Ca-48 Activation

Source Size: 1000-µg²⁵²Cf

Counting Apparatus: 400 Channel Multi-channel Analyzer,

3" x 3" NaI(T) Crystal

Counting Sequence: 6 Minutes Activation, 6 Minutes

Decay, 10 Minutes Counting

% Cement	Ca-49 Gross Activity (counts/10 min.)
0	2160
5	5590
7.5	8050
10	10160
12.5	11080
15	12820



in the switching

Aluminum foil and silicone lubricant were used around the plywood where the plywood was to be removed. It was found though that just plain bare plywood could be used instead of aluminum foil and silicone lubricant.

Several problems were encountered when mixing and pouring the WEP-water-catalyst mixture (called just WEP in this chapter). The major problem of mixing and pouring the WEP was the short time between the adding of the catalyst and the time the WEP gelled. The time before the WEP forms a gel after the catalyst is added to the emulsion is about 1.25 to 1.50 minutes. In less than 1.50 minutes, the catalyst must be completely mixed with the WEP and water (to insure uniform hardening) and poured into the form.

Several things can be done to insure uniform mixing of the catalyst and still give enough time to pour the WEP before it turns into a gel. Small amounts of WEP can be made so as to permit the mixing of the catalyst in the WEP in a short time. The maximum pour was determined to be about 18 to 20 gallons in about 90°F weather. Ice water can be used to slow the reaction time down by several minutes. Reduction of the catalyst concentration by a factor of 3 or 4 did not slow down the reaction time of the WEP.

The stopping blocks on the movable shelf were determined to be too fragile; therefore, either the blocks must be made larger or some sort of reinforcement must be put into the blocks when

pouring the WEP. The movable shelf was found to move easily through the shield with just a thin layer of silicone lubricant on the bottom. The plexiglass moderator-support plate and the cavity for the plastic sample bucket were easily moulded to the movable shelf by just pouring the WEP into a form containing them. The total materials' cost of the facility was less than \$200.

The completion of the shield required the placing of two locks and four strips of iron screwed to both the bottom and top of the shield to hold the two halves of the shield together.

CHAPTER V

CONCLUSIONS

The results in the preceeding chapter satisfy the main objectives of this thesis:

- 1. An irradiation facility has been designed and constructed to activate Ca-48 in cement mixtures. The facility can also be used to activate isotopes other than Ca-48 for students experiments with little or no modifications of the facility.
- 2. The facility will provide satisfactory biological shielding for working personnel when a 100- μ g ²⁵²Cf source is used. The facility actually provides at least 1.25 times the required shielding.
- 3. The facility was relatively easy to construct.
- The total cost of the construction materials was very low (less than \$200).
- 5. The time required to install and remove the source was minimum (less than 45 seconds).
- The facility is portable for all practical purposes.

The conservative dose rates on the shield surfaces allow two possible design changes in the irradiation facility. A facility could be built with a smaller radius than the one constructed for this thesis and still meet the design specifications. The facility constructed could hold more than the designed source size of $100-\mu g$.

Based upon experiences with the operation and construction of the shield, several recommendations can be made about improvements in future shields. The stopping blocks should be made larger or reinforced with some material while the WEP is still fluid. Instead of using WEP stopping blocks, lucite stopping blocks could be used that are screwed into the movable shelf.

Keeping the distance from the source to the top of the shield the same, the height of the shield can be reduced by at least 4 cm or more. The reduction in thickness results from: calculated versus experimental dose rates differ by a factor of 1.25 or more and dose rates can be higher below the shield than any other part of shield.

The final recommendation for a future shield is that the movable shelf could be made to fit into a notch in the bottom section, thereby, reducing the dose rates at the cracks between the bottom and top halves of the shield and at the movable shelf.

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 Engle, W. W. Jr., "ANISN Users Manual: A One Dimensional Discrete Ordinates Transport Code with Anisotropic Scattering." USAEC Report K-1693, Oak Ridge Diffusion. Glenn A. Keller was born on February 2, 1951 in New Orleans, Louisiana. He attended Chalmette High School in Chalmette, Louisiana and graduated in 1969.

In 1969 he entered Louisiana State University and graduated in May, 1973 with a B.S. in Electrical Engineering. Immediately after graduation, he started to work for Ebasco Services, Inc. as an electrical engineer till August, 1974 when he entered Graduate School in Nuclear Engineering at LSU. At that time, he received a graduate assistanship in the Nuclear Science Center. He worked in the area of student laboratory instruction with various professors.

He is presently a candidate for the degree of Master of Science in the Department of Nuclear Engineering.

VITA