RESPONSE OF THERMOLUMINESCENT DOSIMETERS

TO CALIFORNIUM-252 NEUTRONS

A Thesis

Submitted to the Graduate Faculty of the
Louisiana State University and
Agricultural and Mechanical College
in partial fulfillment of the
requirements for the degree of
Master of Science

in

The Department of Nuclear Engineering

by
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B.S., Louisiana State University, 1971
August, 1975

Dedicated to

My Wife, Maida

ACKNOWLEDGMENT

The author would like to express his sincere appreciation to his major professor, Dr. John C. Courtney, for his assistance, encouragement, and advice during the course of experimentation and preparation of this paper. Dr. Courtney exercised meritorious patience during the long period of time when this work was only an idea.

The author also wishes to give a special thanks to Dr. Frank Iddings for his valuable assistance and advice.

The author is thankful to Dr. Edward Lambremont, Dr. William Curry, and Dr. Robert Miles for their helpful advice.

Mr. John Landry and Mr. Russell Timothy Kracht provided invaluable encouragement. Finally, without the support and encouragement of the author's wife and family, this work would not have been possible.

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ABSTRACT

The objective of this work is to determine the response of thermoluminescent dosimeters to radiations from californium-252. Two basic experimental configurations are used. The first experiment is performed with both the source and dosimeters suspended in air without any moderators present. In the second experiment, the dosimeters are suspended in air, near the source, which is located at the boundary of a "semi-infinite" homogeneous moderating medium. Gamma ray, thermal neutron, and fast neutron dose rate determinations are made by dosimetry methods other than thermoluminescence, thus defining the components of the mixed radiation field. Lithium fluoride thermoluminescent dosimeters of type TLD-100, TLD-600, and TLD-700 are then exposed to the californium-252 source. The thermoluminescent dosimeters are exposed bare, cadmium covered, and lithium-6 covered. The use of thermal neutron absorbers and lithium fluoride dosimeters of different lithium-6 content allows a degree of separation of the responses of the thermoluminescent dosimeters into the components caused by the gamma ray, thermal neutron, and fast neutron fluxes. A correlation is thus achieved between the components of the thermoluminescent response and the components of the dose due to the mixed neutron and gamma radiation field associated with californium-252 sources.

CHAPTER I
Introduction

The use of thermoluminescence in the dosimetry of radiation
fields that are composed of both neutrons and gamma rays, such as
associated with californium-252, has increased considerably in the
past few years.[1,2] Such combinations of particle and photon
radiations are termed "mixed fields". Recent developments have
demonstrated that, in some applications, thermoluminescence is the
only method acceptable for dosimetry of californium-252.[3]
Judicious application of thermoluminescence to californium-252
dosimetry should increase the effectiveness and use of californium-252.
This intense neutron source is now being used in oil well logging,
neutron radiography, radiotherapy, and numerous other applications.
The main advantage of californium-252 is the high rate of neutron
emission, 2.34 x 10^{12} neutrons per second per gram. Other isotopic
neutron sources require beryllium. They are orders of magnitude
larger in size and have high heat generation rates[4] for a neutron
emission rate comparable to that of californium-252.

On the average, 97 percent of the decays of californium-252 are
by alpha emission and only 3 percent are by spontaneous fission.
Each fission yields an average of 3.76 neutrons in a fission
spectrum of energies which can be approximately represented by the
Watt spectrum[5]. The equation which describes the Watt spectrum is

$$N(E) = 0.373 \exp(-0.88E) \sinh(2.0E)^{\frac{1}{2}} \qquad \text{(Equation 1-1)}$$

where E is the energy of the neutron in million electron volts (Mev) and $N(E)$ is the fraction of neutrons per unit energy range[6]. In the fission spectrum of californium-252, the most probable energy is approximately 0.8 Mev and the average energy is approximately 2.35 Mev. In addition, californium-252 emits 1.3×10^{13} gamma photons per second per gram due to prompt fission, fission products, and alpha decay. The effective half life is 2.646 years which is long enough to make its use feasible and yet short enough to render californium-252 a high specific activity [7].

The Nuclear Science Center at Louisiana State University has been designated as a Californium Demonstration Center and charged with the development of applications for this isotope. Approximately 100 mg of californium-252 have been provided by the U.S. Atomic Energy Commission.* The sources are available for loans at no cost to the user. The facilities of the Nuclear Science Center are available to investigators who prefer to use these sources at Louisiana State University. By providing prospective customers with the opportunity to use these sources at a minimum cost, it is hoped that many applications will be developed.

Experimentation with californium-252 is carried out at the Nuclear Science Center in various research fields. An accurate determination of the dose or dose equivalent delivered to the target

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* In January 1975, the U.S. Energy Research and Development Administration was formed and took over the responsibility for this program.

during a californium-252 irradiation is usually an essential part of any experimental procedure. The ^{252}Cf Shielding Guide[8] has been prepared by the U.S.A.E.C. to provide estimates of neutron and gamma levels from encapsulated sources of californium-252. This guide considers point sources in one-dimensional spherical shields. The actual experimental configurations are much more complex. Contributions from secondary radiation and scattering from other materials outside the basic shield system are not considered. Also, perturbations of the radiation field by the target may be severe. Dosimetry is quite complex and cannot be handled accurately by generalized calculations. Therefore, dosimetry must be performed for each experimental arrangement used. The staff of the Nuclear Science Center assists experimenters in this regard. As the dosimetry capabilities of the Nuclear Science Center improve, the Center becomes more valuable to its clients.

Mixed field dosimetry is difficult by any method known. Each component of the radiation field interferes with the dosimetry of the other. Regardless of where the irradiation is carried out, matter will be present. The interaction of fission neutrons with matter complicates the radiation field by the creation of capture gammas and inelastic scattering gammas. Scattered gammas and neutrons from the surroundings impinge on the detector, adding to the measurement.

Dosimetry of californium-252 at the Nuclear Science Center previously consisted of such techniques as metal foil activation, ionization chambers, and ethyl disulfide activation[9]. All of

these require much time, effort, and skill. The possible advantages

of thermoluminescent dosimetry over these other methods prompted this

research. This thesis reports the first results on thermoluminescent

dosimetry monitoring of neutron radiation at the Nuclear Science

Center. It is hoped that it will form the basis for further research

and give direction toward future improvements in the thermoluminescent

dosimetry capabilities at the Nuclear Science Center for mixed

radiation fields.

A major interest of the Nuclear Science Center is the effects

of neutron radiation on biological systems. The more notable work

has been done by Greene with the wax moth[10] and Sgrillo with the

sugar cane borer (Appendix I). In such experiments, the targets and

the corresponding irradiation chambers are very small. The ability

to mix the dosimeters among the target insects or to implant the

dosimeter into the specimen is an advantage of a small dosimeter

such as the thermoluminescent dosimeter. These techniques allow

more accurate dosimetry which is important when dealing with biological

systems. Accurate dosimetry is also important in order to compare

the effects of neutron radiation with those due only to gamma

radiation.

To utilize thermoluminiscent dosimetry, it is advantageous to have

some understanding of the mechanism by which it operates. Ionizing

radiation impinging upon matter produces ionization and excitation.

Some of the absorbed energy is used to break chemical bonds and a

small fraction of the energy is stored in metastable states in

materials which exhibit thermoluminescence. Electrons are raised to

metastable or higher energy levels and, upon heating, return to the ground state. The stored energy is released as visible light. TL is the phenomenon of light emission upon heating[11]. The amount of light emission is linearly proportional to the radiation dose received by the thermoluminescent dosimeter. It is this fact that makes thermoluminescent dosimetry possible.

For thermoluminescent dosimetry to be an effective dosimetry method in mixed radiation fields, the responses of the dosimeter to gamma and neutron radiation must be separable. A correlation should be made between the components of the thermoluminescent response and the components of the dose due to the mixed neutron and gamma radiation field associated with californium-252. In order to achieve this, gamma ray, thermal neutron, and fast neutron dose rate determinations are made by dosimetry methods other than thermoluminescent dosimetry. Thus, the components of the mixed radiation field are known. Thermoluminescent dosimeters are then exposed to the same mixed radiation field. By the use of thermal neutron absorbers and lithium fluoride thermoluminescent dosimeters of different lithium-6 content, the responses of the thermoluminescent dosimeters can be separated into the components caused by the gamma ray, thermal neutron, and fast neutron fluxes.

The thermoluminescent dosimeter that makes the separation of response possible is lithium fluoride. It is available in three different isotopic compositions of lithium as shown in Table 1-1. The $^6Li(n,\alpha)^3H$ reaction has a high cross section (950 barns) at thermal energies. Since 4.8 Mev of localized energy is deposited

TABLE 1-1

Isotopic Composition of Various Lithium Fluoride Dosimeters

Isotope	^{N}LiF (TLD-100)	^{6}LiF (TLD-600)	^{7}LiF (TLD-700)
^{6}Li	7.5%	95.6%	0.01%
^{7}Li	92.5%	4.4%	99.99%

in the dosimeter by this reaction, the thermal neutron sensitivity of ^{6}LiF is very high. In the range of 20 kev to 15 Mev, the average cross section for the ^{6}Li(n,α)^{3}H reaction is one barn (Figure 1-1). The total cross section of lithium-6 is approximately 950 barns at thermal energies with a "1/v" variation into the epithermal energy region.

FIGURE 1-1

Variation of Cross Section with Energy for the ^{6}Li(n,)^{3}H Reaction
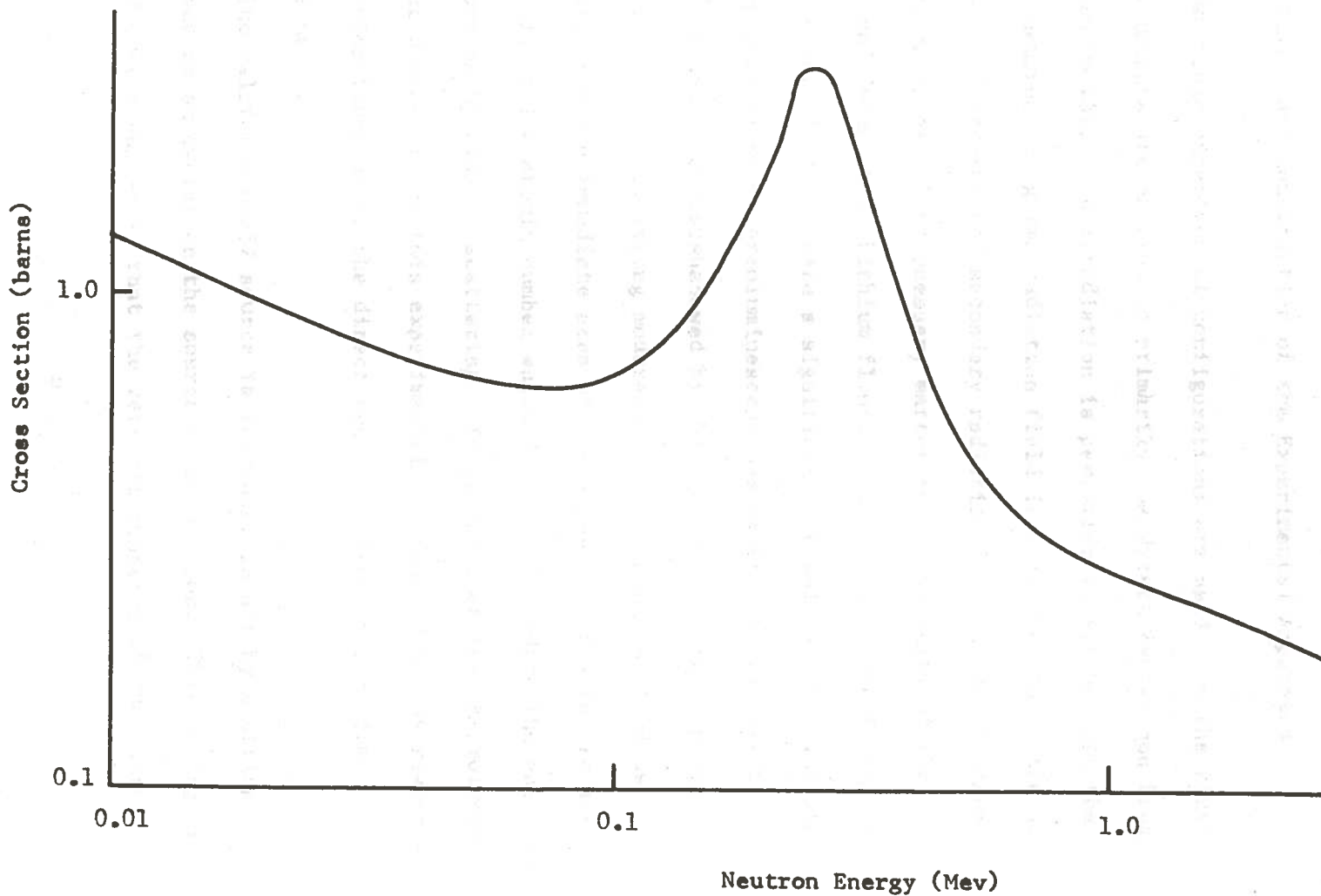
CHAPTER II

Design and Construction of the Experimental Apparatus

Two basic experimental configurations are used. In the first, the dosimeters are exposed to primarily the direct radiations from californium-252. The irradiation is performed in air so that the direct neutron and gamma radiation field is perturbed as little as possible. Scattered and secondary radiation is kept to a minimum by the exclusion of all unnecessary matter in the vicinity of the source and detectors. Lithium fluoride has a high thermal neutron sensitivity. If there were a significant thermal neutron flux present, the response of the thermoluminescent dosimeters to the direct radiation would be overshadowed by the thermal neutron response. For this reason, all strong moderators are excluded as much as possible from the immediate area of the exposure. Moderators are materials of low atomic number which drastically reduce the energies of neutrons by elastic scattering. By exposure of the thermoluminescent dosimeters to this experimental configuration, the response of lithium fluoride to the direct radiation from californium-252 can be measured.

The californium-252 source is suspended in air by a string attached to an eyelet on the source encapsulation. This string is draped over a pulley so that the vertical position of the source

can be controlled from a remote location. The source normally
occupies a safe, shielded position in a tank of water below the
location for this experiment.

The dosimeters are arranged in an array around the position
that the exposed source would occupy as shown in Figures 2-1 and 2-2.
The source to detector distance is the same for each dosimeter so
that each is subjected to the same radiation field. Each dosimeter
is suspended by a string.

The primary criterion for this experimental configuration is
the exclusion of as much matter as possible near the source and
dosimeters. Materials of low atomic number are good moderators
and cause thermalization of the neutron flux. Materials of high
atomic number have a high inelastic scattering cross section and cause
an increase in the gamma dose. Any type of material in a radiation
field will interact with or perturb that field in some way. There-
fore, the primary consideration for this configuration was to use
the least amount possible of any type of matter. The method of
using strings to suspend the source and dosimeters provided the
least possible perturbation to the radiation field.

There is some perturbation to the radiation field by the
building and the tank of water used to store the source. The
exposed source position is 8 feet above the surface of the water.
At this position, there still remains 6 feet from the source to the
ceiling. The nearest wall is 10 feet away from the source. The
scattered and secondary radiation from the building, at these
distances, has a very small effect on the dosimetry.

FIGURE 2-1

Side View of Geometry for Experiment Conducted

in Air without Moderation in Vicinity of Source

20 cm   20 cm

Ethyl disulfide

R-meter probe

Californium-252
source

Thermoluminescent
dosimeters
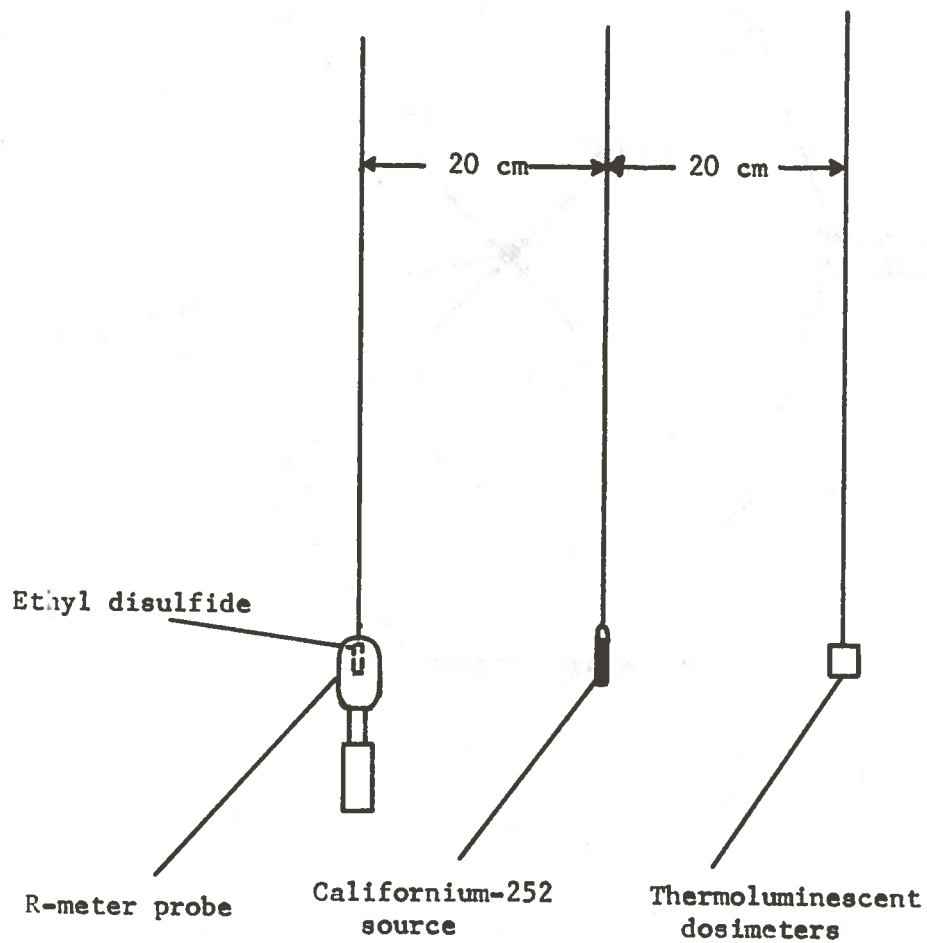
FIGURE 2-2

Top View of Geometry for Experiment Conducted

in Air without Moderation in Vicinity of Source

Ethyl disulfide

20 cm

20 cm

Thermoluminescent
dosimeters

Californium-252
source

20 cm

20 cm

R-meter probe
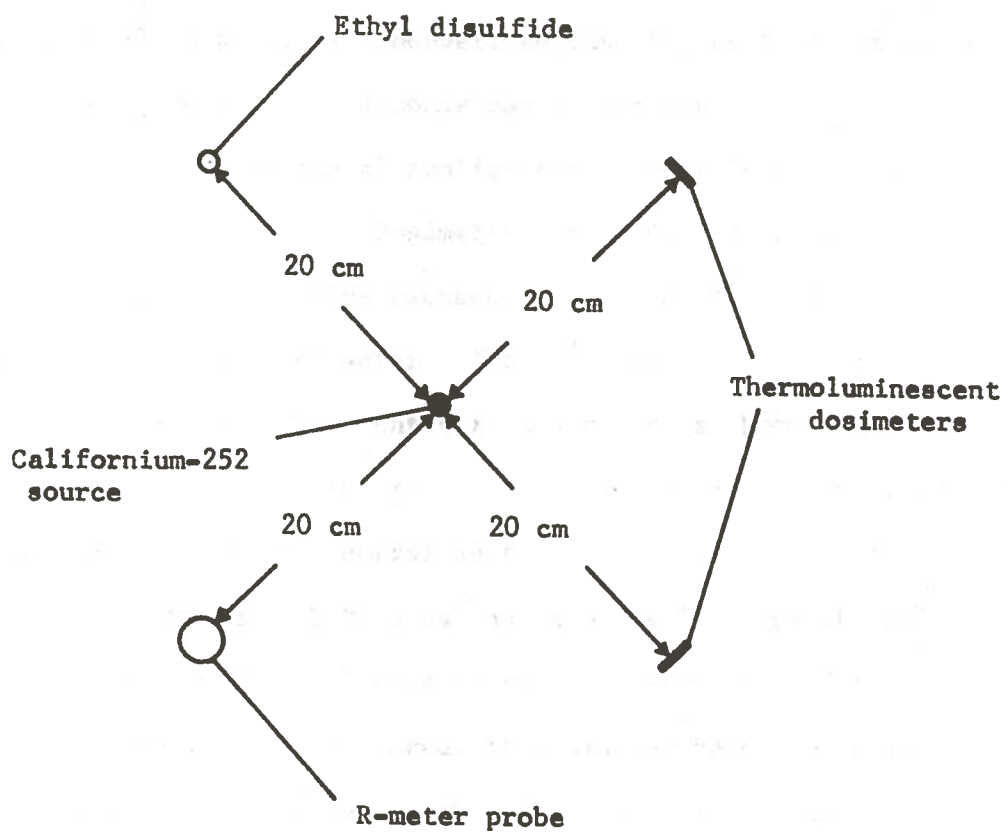
Although undesirable, a small increase in gamma dose from scattered or secondary gamma radiation is tolerable. The primary objective of this experiment is to correlate the components of the thermoluminescent response to the components of the radiations from californium-252. As long as the gamma dose delivered to the thermoluminescent dosimeter is known, the gamma dose can be accounted for in the thermoluminescent response. The gamma dose should be kept as small as possible, however, so that the neutron component of the thermoluminescent response can be discerned.

The second experimental configuration was a "semi-infinite" homogeneous water medium. Dosimetry was conducted in the air adjacent to the water. The exposed source position was at the interface of the air and water. This arrangement is shown in Figure 2-3 and 2-4. This configuration yields a thermal neutron flux at the point of dosimetry to allow determination of the thermo-luminescent response to thermal neutrons. However, the thermal flux is not large enough to cause an excessively large thermal neutron component in the thermoluminescent response. If the thermal neutron component were too large, it would dominate the total thermoluminescent response and the responses to the other components of the radiation would be indiscernible.

This configuration was achieved by utilizing the unshielded side of the source storage tank for the Californium Demonstration Center. Three sides of the tank are shielded by concrete blocks but one side is left exposed for experimentation. With the source

FIGURE 2-3

Side View of Geometry for Experiment Conducted

near a "Semi-Infinite" Homogeneous Water Medium
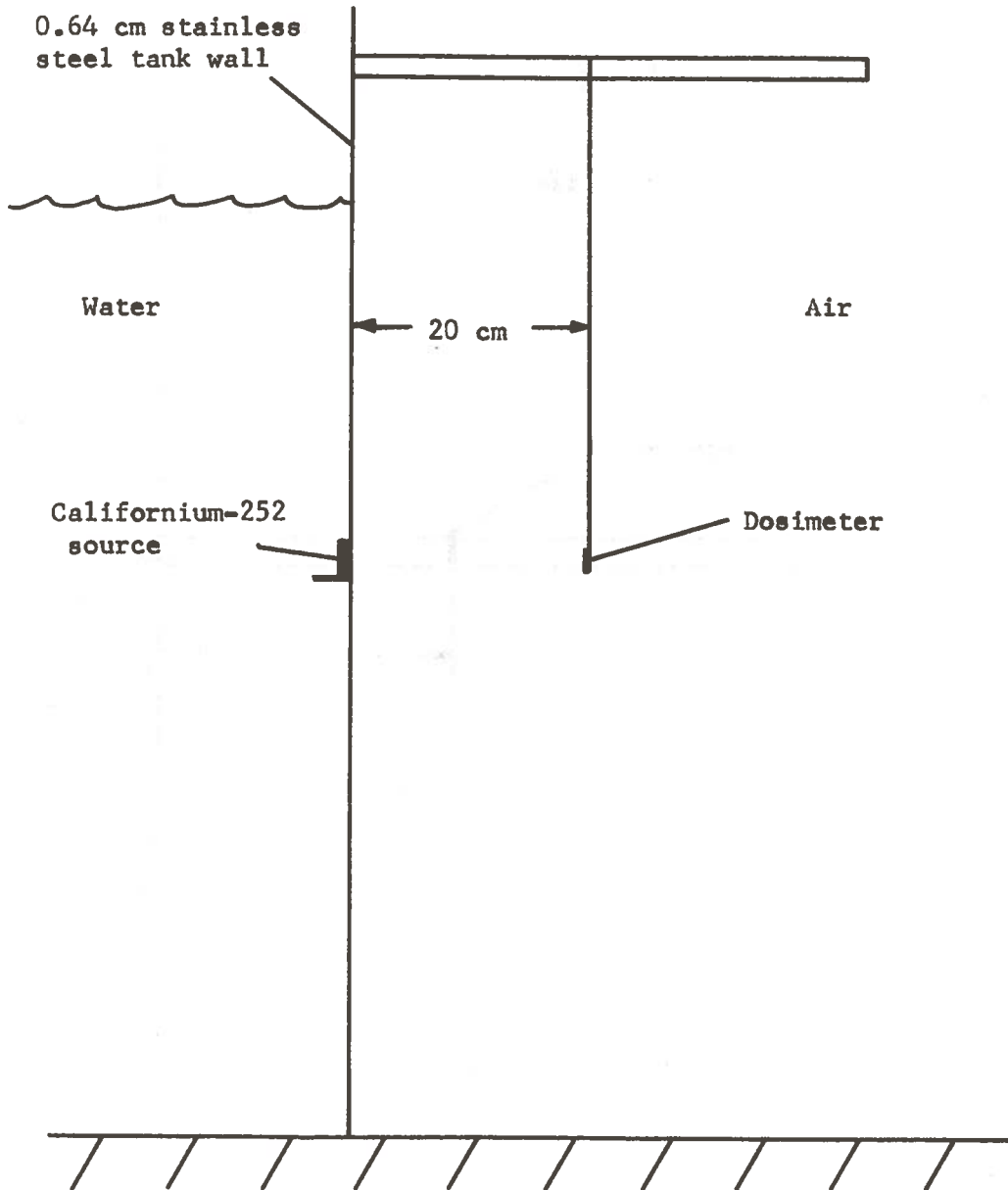
FIGURE 2-4

Top View of Geometry for Experiment Conducted

near a "Semi-Infinite" Homogeneous Water Medium
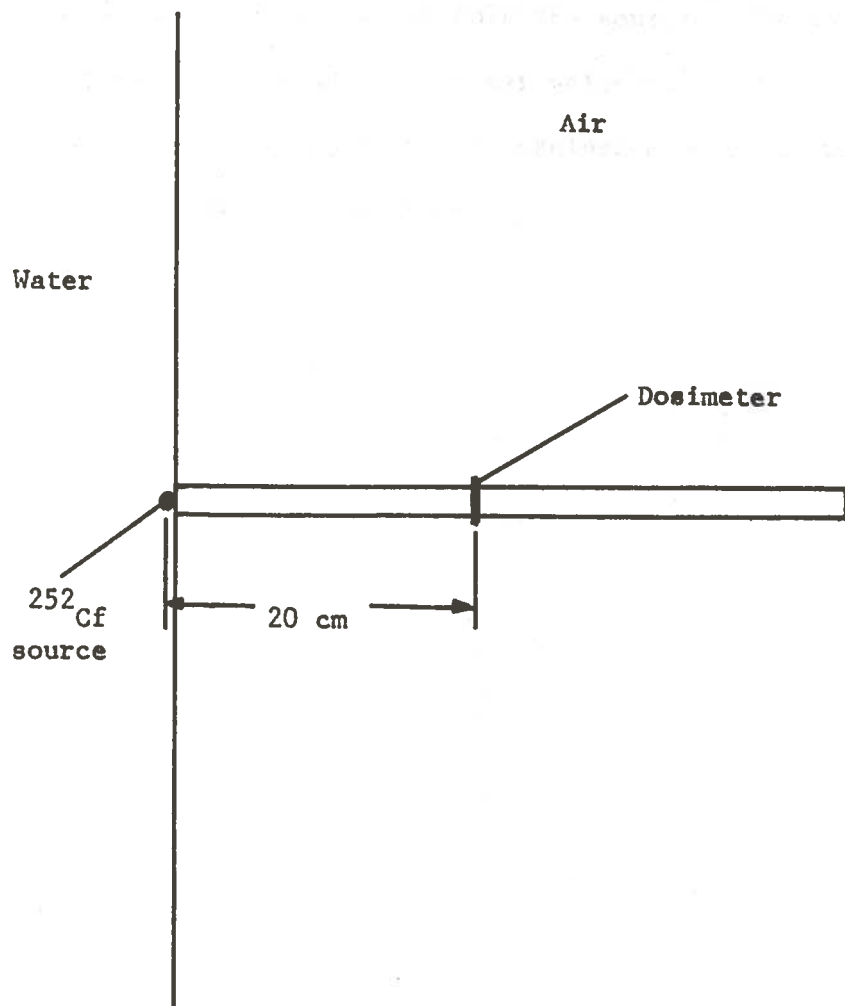
positioned against the inner face of the tank, the only perturbation to the direct radiation is the 0.64 cm thick, stainless steel wall of the tank.

The source holder had been constructed for previous experimentation and was still in place. It consisted of a steel block placed on a ledge on the tank wall below the water level. In the steel block, a notch was cut to hold the source. The dosimeters were suspended by strings in the air adjacent to the source. In this experimental arrangement, the exclusion of scattered and secondary radiation was not important.

CHAPTER III

Dosimetry

Trial calculations were performed to select a source of suitable strength. Sources available ranged from one to five micrograms (μg) and from four to six milligrams (mg). Since the neutron emission rate from a californium-252 source is 2.34×10^9 neutrons per sec per mg, the expected fast flux and dose rate at any distance from the source can be determined from the ^{252}Cf Shielding Guide[12]. The sources in the microgram range are unsuitable since they would require a very small source to dosimeter distance and excessively long irradiation times. Calculations determined that the source containing 5.25 mg of californium-252 (May 25, 1975) would provide the experimental conditions to yield reliable results.

Once the source size is determined, other factors such as radiation safety could be considered. The ^{252}Cf Shielding Guide is used to make estimates of the dose equivalent rates to operating personnel. During experimentation the exposure rate to personnel from gamma radiation is monitored by a Victoreen Model 440 ionization chamber survey meter. In addition, the neutron flux level at operating positions is monitored by an Eberline Model PNC-4 neutron counter.

It should be emphasized that a thermoluminescent dosimeter is not an absolute radiation detection device. That is, there are no

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mathematical means to correlate the thermoluminescent response and the energy deposited in the dosimeter. The radiation field must be defined first by dosimetry methods other than thermoluminescence. Then, the thermoluminescent dosimeters can be exposed to the known radiation field and a relationship between dose and response obtained. The radiation field in this experiment consists of neutron and gamma radiations.

In both experimental configurations, determinations are made of the gamma dose rate and the fast neutron dose rate. In the experiment with the "semi-infinite", homogeneous water medium, a large thermal neutron component was expected. Therefore, a measurement of the dose rate due to thermal neutrons is required for this geometry. A negligible thermal neutron flux is expected in the experiment in air because of the absence of moderator material in the vicinity of the source.

The source to detector distance used is important to achieve accurate results. Also, the irradiation time is a function of the distance. If the dosimeters were exposed very close (less than 10 cm) to the source, geometry effects could cause significant error. The smaller the source to detector distance becomes, the greater the deviation from the point source and point detector concept. At large distances, excessively long irradiation times are required. Therefore, a compromise is made by the choice of 20 cm. Trial calculations were performed which indicated that useful results can be obtained within several hours with a source to detector distance of 20 cm.

Gamma dose rate determinations are made by a Victoreen Instrument Company R-meter, Model #70, and an associated 25 roentgen ionization chamber probe. The center of the sensitive volume is suspended 20 cm from the axis of the source in each experiment. A proper irradiation time of 30 minutes had been predetermined to give results within the range of the probe. Division of the results of the R-meter reading by the time of the irradiation gives the gamma exposure rate in roentgen per unit time. The dose rate in rad per unit time is then determined by multiplication of the exposure rate by a factor of 0.867[13].

The fast neutron flux was determined by use of the ethyl disulfide activation procedure which employs the $^{32}S(n,p)^{32}P$ reaction with a threshold of about three Mev. There is at least one variation of the basic method. In the experiment with the "semi-infinite" medium, the method as described by Greene[14] was employed. In the experiment in air, a variation described by Morel[15] was used. In both, the activation and counting procedures are the same. Six milliliters of ethyl disulfide are exposed to the californium-252 source at a distance of 20 cm for six hours and then allowed to decay for at least 24 hours. The variation in methods involves a difference only in the preparation of the counting solutions. In the method described by Greene, the same amount of activated ethyl disulfide is added to each of ten liquid scintillation vials. Then a different amount of unactivated ethyl disulfide is added to each vial. The ethyl disulfide is a chemical quencher to

liquid scintillation counting. Thus, each vial has the same amount

of phosphorous-32 activity, but a different amount of quencher.

In the variation described by Morel, no unactivated ethyl disulfide

is used. Different amounts of activated ethyl disulfide are added

to ten liquid scintillation vials. Thus, each vial has a different

amount of phosphorous-32 activity and a different amount of the

quenching material. These solutions are counted for phosphorous-32

activity in a Beckman liquid scintillation counter, Model LS-250.

From this radioassay, the induced activity per gram of ethyl

disulfide can be calculated. By use of the standard activation

equation, the fast neutron flux over three Mev is determined for

each experimental arrangement. The activation equation is given

by

$$A = \sigma \phi_f N \ (\ 1 - e^{-\lambda t_a}) \ e^{-\lambda t_d} \qquad \text{[Equation 3-1]}$$

where A is the activity at the time of counting in units of

disintegrations per second and ϕ_f is the fast neutron flux over

three Mev. In this equation, N is the number of sulfur atoms per

gram of ethyl disulfide, λ is the decay constant and t_a is the

activation time in units comparable to that of the decay constant.

Further, t_d is the decay time in units comparable to that of the

decay constant. Since the fast neutron flux over three Mev is

28.17 percent of the total uncollided fission spectrum from

californium-252, the total fast neutron flux can be determined.

The dose rate due to the fast neutron flux can be found by

multiplication of the factor 1.375×10^{-5} rad per hour per unit flux by the total fast flux as recommended by the American Nuclear Society Committee on Shielding Standards[16].

The thermal neutron flux is determined in the "semi-infinite" homogeneous water medium experiment by the indium foil activation technique[17]. Bare and cadmium covered indium foils are exposed to the californium-252 source for one hour each at a distance of 20 cm. The irradiation time at this distance was predetermined to yield a suitably high counting rate. The radiation detector used for the indium-116m activity is a 3 in by 3 in NaI(Tl) scintillation crystal manufactured by Harshaw (Type 12-S-12) and a Hewlett-Packard Model 6515-A high voltage power supply. A technical Measurements Corporation multi-channel analyzer, Model 401D, is used to record the activity. The detector system is calibrated absolutely using a National Bureau of Standards cobalt-60 source. The indium foils are counted by the NaI(Tl) crystal with the foils held 1.5 cm from the crystal by a plastic beta shield inbetween. With the indium-116m induced activity determined by this count, the thermal neutron flux can then be calculated since the thermal neutron activation cross section for the reaction is known. The dose rate due to thermal neutrons is determined by multiplication of the factor 1.838×10^{-6} rad/hr per n/sec cm^2 by the thermal neutron flux. This thermal neutron flux to dose conversion factor is recommended by the American Nuclear Society Committee on Shielding Standards[18].

Another experiment is carried out to determine the variation in fast neutron flux with distance from the source for the "semi-infinite" medium experiment. A Reuter-Stokes Model P5-0805-201 fast neutron detector using helium-4 as the sensitive gas was used. The associated electronics are a Canberra Model 3015 high voltage power supply, a Canberra PAD Model 814 amplifier, an ORTEC Model 484 scaler, and an ORTEC Model 719 timer. Measurements were made from 10 to 100 cm in increments of 10 cm. The helium-4 fast neutron detector is not calibrated absolutely, so it merely gives relative values of the fast flux as a function of distance from the source.

With the radiation field defined by the previously mentioned dosimeters, thermoluminescent dosimetry is performed. The types of lithium fluoride dosimeters used are TLD-100, TLD-600, and TLD-700 hot pressed chips purchased from Victoreen Instrument Company. The nominal dimensions of these thermoluminescent dosimeters are 0.125 in by 0.125 in by 0.030 in. The dosimeters are prepared for exposure by individually subjecting them to the "LiF anneal" cycle of a Victoreen Model 2800 thermoluminescent dosimeter reader. In this cycle, the dosimeters are preheated to 115 C to release thermoluminescence from any low temperature, unstable traps. They are heated to 255 C to integrate the light output from the main thermoluminescent glow peak. Further heating to 340 C anneals the dosimeters and restores their proper sensitivity. After removal from the reader, the dosimeters are packed in thin (0.005 in) acetate sheet to keep them clean and facilitate handling.

Dosimeters of all three types are exposed with only the plastic covering at a distance of 20 cm for one hour. All three types of lithium fluoride thermoluminescent dosimeters are also sandwiched between two sheets of cadmium (0.060 in thick) for another exposure with all other parameters the same. A third exposure is made with the three types of lithium fluoride dosimeters sandwiched between two sheets of ^{6}LiF. Ten individual dosimeters of each type of lithium fluoride dosimeter are exposed with each type of shielding: bare, cadmium covered, and lithium-6 covered. The entire procedure is then repeated to test the reproducibility and lend confidence to the previous results. The preceding series of exposures are performed in air and with the "semi-infinite" medium at the side of the tank. After exposure, all dosimeters are read individually in the "LiF anneal" cycle of the thermoluminescent dosimeter reader. This cycle automatically reads, anneals and resets the sensitivity of the dosimeters in preparation for another exposure.

To assure that all pertinent parameters remained the same from exposure to exposure within the same geometry conditions, control thermoluminescent dosimeters are used. One dosimeter of each lithium fluoride dosimeter type is exposed along with the primary dosimeters for each exposure. The control dosimeters were positioned apart from the primary dosimeters in an accurately reproduceable geometry.

CHAPTER IV

Experimental Results

The results of the experimentation described in Chapter III are listed in Tables 4-1 through 4-4. All values are normalized to those which would be encountered by exposure to a 1 mg source of californium-252. The actual source content, however, was approximately 5.5 mg of californium-252 during most of the experimentation. The source to detector distance, 20 cm, is the same for all measured and calculated values.

Tables 4-1 and 4-2 give the results of experimentation conducted to determine the gamma, fast neutron, and thermal neutron dose rates in the two experimental configurations considered. The first column lists the values determined by the experimental procedures and calculations of the thesis. The second column gives the predicted values according to the ^{252}Cf Shielding Guide. The results of the experiments conducted in air to determine the radiation field which the thermoluminescent dosimeters would be subjected to are listed in Table 4-1. In this experimental configuation, there are no moderators in the vicinity of the source. The results are listed in Table 4-2 for the determination of the radiation field adjacent to a "semi-infinite" homogeneous water medium. The source was positioned at the interface of the water and the air.

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TABLE 4-1

Results of Dosimetry to Determine the Radiation Field 20 cm

from a 5.5 mg Californium-252 Source Suspended in Air

All Values Normalized to 1 mg of Californium-252

Gamma Dose Rate in rad/hr per mg

Measured	^{252}Cf Shielding Guide
5.25	3.99

Fast Neutron Flux in $n\text{-}cm^{-2}sec^{-1}$ per mg

Measured	^{252}Cf Shielding Guide
4.15×10^5	4.66×10^5

Fast Neutron Dose Rate in rad/hr per mg

Measured	^{252}Cf Shielding Guide
5.71	6.66

TABLE 4-2

Results of Dosimetry Performed in Air to Determine the

Radiation Field 20 cm from a "Semi-Infinite" Homogeneous

Water Medium with a 5.5 mg Californium-252 Source at the

Interface of the Air and Water

All Values Normalized to 1 mg of Californium-252

Gamma Dose Rate in rad/hr per mg

Measured	^{252}Cf Shielding Guide
5.50	3.99

Fast Neutron Flux in $n\text{-}cm^{-2}sec^{-1}$ per mg

Measured	^{252}Cf Shielding Guide
3.75×10^5	4.66×10^5

Fast Neutron Dose Rate in rad/hr per mg

Measured	^{252}Cf Shielding Guide
5.16	6.66

Thermal Neutron Flux in $n\text{-}cm^{-2}sec^{-1}$ per mg

Measured

9.91×10^4

Thermal Neutron Dose Rate in rad/hr per mg

Measured

0.18

TABLE 4-3

Results of Thermoluminescent Dosimetry Performed 20 cm

from a 5.5 mg Californium-252 Source Suspended in Air

All Values Normalized to 1 mg of Californium-252

Thermoluminescent Response in equivalent roentgen/hr per mg

	Bare	Cadmium	Lithium-6
TLD-100	2.63	2.74	2.80
TLD-600	3.70	3.73	3.53
TLD-700	2.74	2.93	2.64

TABLE 4-4

Results of Thermoluminescent Dosimetry Performed in Air

20 cm from a "Semi-Infinite" Homogeneous Water Medium

with a 5.5 mg Californium-252 Source at the

Interface of the Air and Water

All Values Normalized to 1 mg of Californium-252

Thermoluminescent Response in equivalent roentgen/hr per mg

	Bare	Cadmium	Lithium-6
TLD-100	13.70	3.55	3.60
TLD-600	51.08	10.69	11.18
TLD-700	3.15	2.99	2.85

In Table 4-3, the results of the exposure of thermoluminescent dosimeters to the radiation field defined in Table 4-1 are given. The results are listed in Table 4-4 for the exposure of thermoluminescent dosimeters to the radiation field defined in Table 4-2. Each value listed in Tables 4-3 and 4-4 is the average of the readings of ten dosimeters. The units of the values listed in Tables 4-3 and 4-4 are equivalent roentgen per hour per milligram of californium-252. An equivalent roentgen is a thermoluminescent dosimeter response to neutron radiation[19]. That same thermoluminescent dosimeter response is produced by exposure to one roentgen of cobalt-60 gamma radiation. The number of equivalent roentgen per rad of neutron radiation deposited in the phosphor is a function of energy and the amount of lithium-6 in the lithium fluoride phosphor.

Figure 4-1 shows the result of the experiment to determine the variation in fast flux with distance from the source. A diagram of the electronics used in this experiment is shown in Figure 4-2.

Tables 4-1 and 4-2 show a good correspondence between the experimentally determined values and the values estimated according to the ^{252}Cf Shielding Guide. The experimental values of gamma dose rate for both configurations are about 36 percent higher than the estimated values. The higher value could be due to secondary gamma radiation which the ^{252}Cf Shielding Guide does not consider. One type of secondary gamma radiation possible for the experimental configurations used is capture gammas due to the (n,γ) reaction in materials near the experiment. Capture gammas could be caused by hydrogen in the air, water, and concrete. The iron, nickel, and

FIGURE 4-1

Relative Fast Neutron Flux at Various

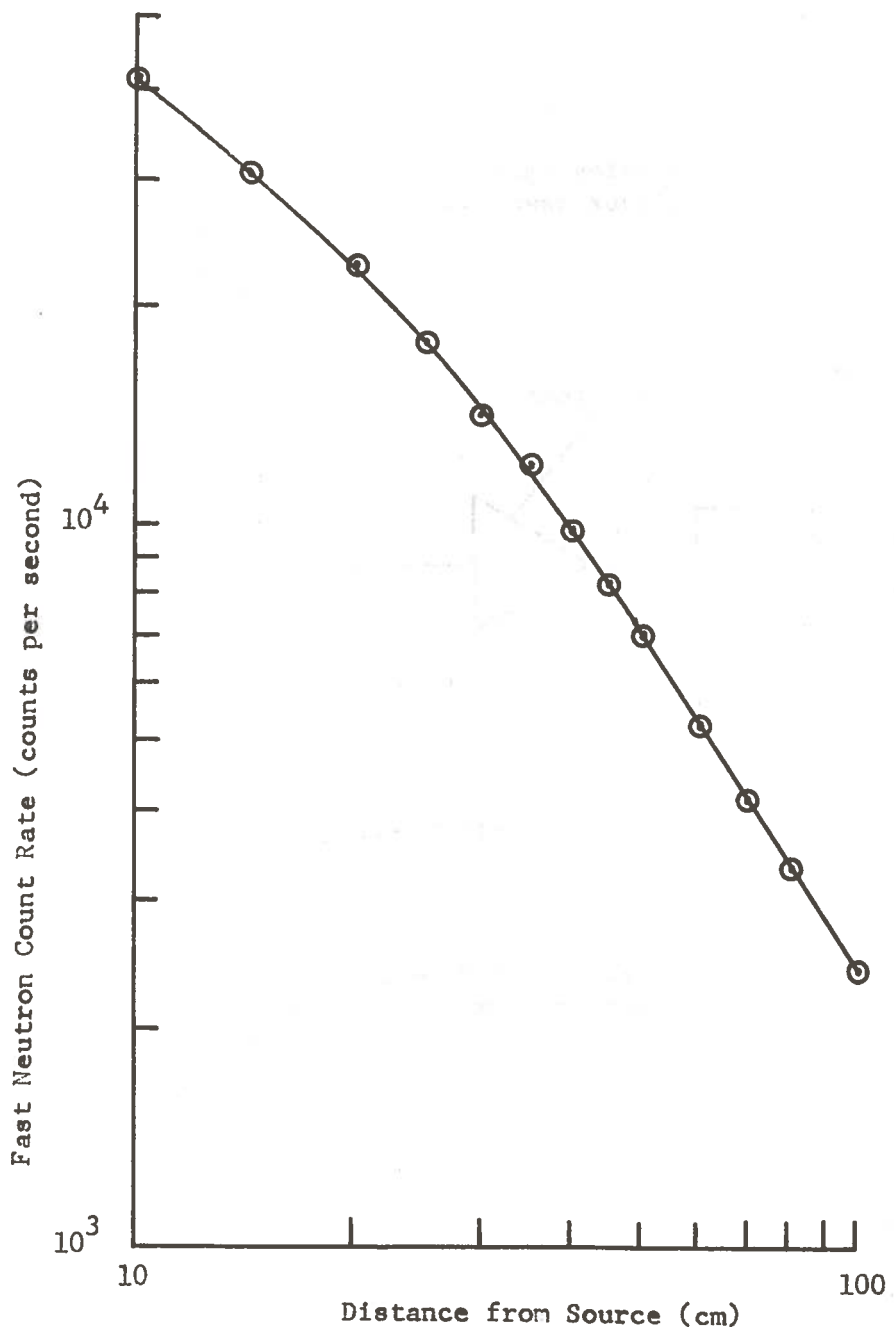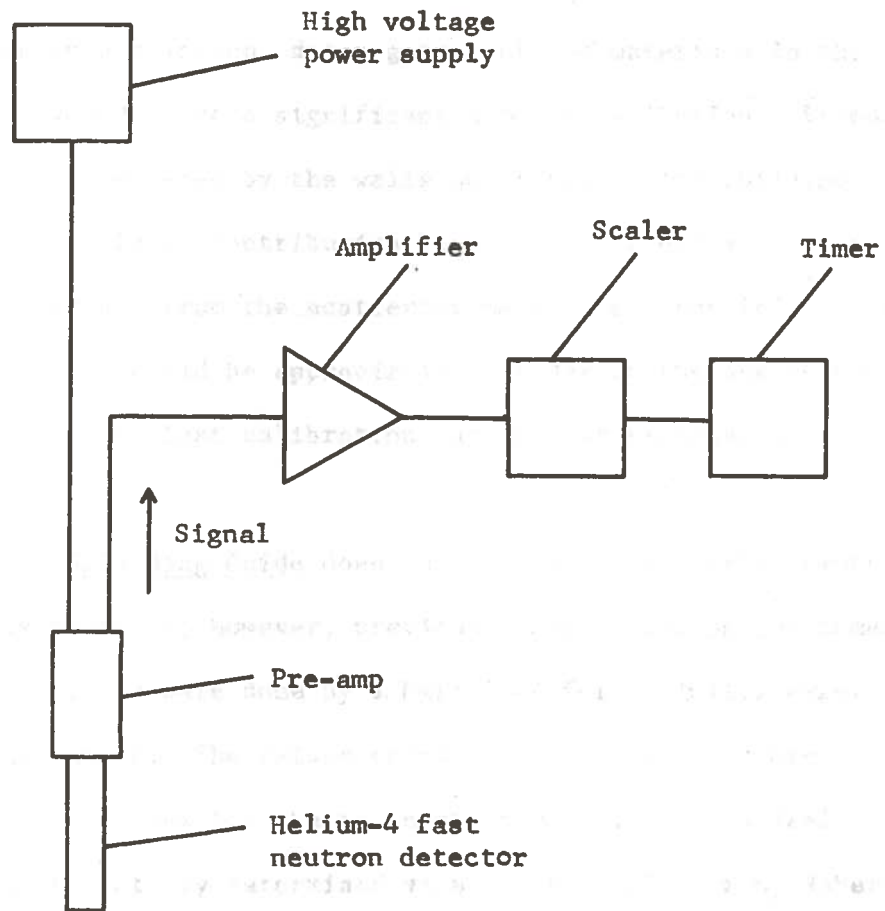Distances from Source

FIGURE 4-2

Schematic of Detection System for Determination

of Relative Fast Neutron Flux at Various Distances from Source

High voltage
power supply

Amplifier

Scaler

Timer

Signal

Pre-amp

Helium-4 fast
neutron detector

chromium of the stainless steel tank could also be a source of
capture gammas. Another type of secondary gamma radiation which
could cause a significant contribution to the gamma field is inelastic
scattering gammas. Inelastic scattering gammas occur mostly in
materials of high atomic number, as in the steel of the storage
tank and the materials of the source encapsulation. Due to activation
of the source encapsulation, decay gammas of the materials in the
encapsulation could cause a significant gamma contribution. Primary
gamma radiation scattered by the walls and floor of the building
should not make a large contribution because of the distance of the
source and detectors from the scattering materials. Possible error
in the measurement could be appreciable considering the age of the
detector, time since last calibration, and energy response of the
detector.

The ^{252}Cf Shielding Guide does not estimate the thermal neutron
flux for this geometry; however, previous experimentation and numerical
methods calculations were done by Robert [20] for a similar experi-
mental configuration. The values calculated by Robert compare
favorably to the values for thermal neutron flux listed in Table
4-2. The experimentally determined value of thermal flux by Robert
is a factor of two less than the value obtained by this thesis. The
difference could be accounted for by the slight difference in
geometry. There are many errors possible in the determination of the
thermal neutron flux. An estimate is given by Robert of $\pm$ 15
percent accuracy due to combined errors in the activation cross
section for indium, counting conditions and flux perturbations for
thermal neutron flux determination by indium foil activation.

The results of the fast neutron flux determinations compare favorably to the values from the ^{252}Cf Shielding Guide. The experimental determinations are less than 20 percent lower than the actual values of fast flux. Robert [21] estimates a possible error of $\pm$ 25 percent due to combined errors in the average sulfur cross section and counting statistics. The cross section of sulfur-32 for the (n,p) reaction does not have a clearly defined threshold at 3 Mev, but instead, rises slowly over the range from 2-5 Mev. The effective reaction threshold of 3 Mev has been computed by numerical techniques, but could represent considerable error for the neutron distribution used here. The accurate fast flux determination hinges on the fact that the flux over 3 Mev has been found and is 28.17 percent of the total uncollided fast flux. Another possible source of error is the 0.060 inch thick cadmium shield used to prevent competing reactions. The cross section of cadmium is not zero at energies above 0.4 ev. It is very small at higher energies but could still decrease the fast flux available to the detector. Price [22] recommends a cadmium correction factor of 1.22 for 0.060 inch of cadmium shielding on indium foil to account for the absorbtion of epi-cadmium neutrons by the shield. This factor only applies to indium foils but is indicative of the degree of reduction in fast flux by cadmium shields. Scatter is not a strong source of error in the fast neutron determination, since most neutrons with energies above the threshold of sulfur are uncollided neutrons. That is, it is unlikely that scattered neutrons from the californium-252 source would be above three Mev. Therefore,

it is assumed that neutrons which would activate sulfur come directly from the fission spectrum of the source.

From Figure 4-1, it can be seen that the variation in fast flux over 35 cm from the source is inversely proportional to the square of the source to detector distance. A "$1/r^2$" variation in count rate is expected because of the angular divergence of radiation from a point source. An explanation for part of the deviation at distances close to the source may be due to counting losses. Close to the source, the count rate approaches 10,000 counts per second which is high enough to cause significant count losses. These losses are due to dead time in the electronics. Assuming the resolving time of the detection system is approximately 1 microsecond, ten percent of the deviation would be accounted for. A major source of error could be geometry effects of the large volume detector (300 cm^3) at positions close to the source. The smaller the source to detector distance becomes, the greater the deviation from the point detector concept. The straight portion past 35 cm show that the detector is sensitive to fast neutrons only, because the californium-252 source is the only origin of fast neutrons. If the detector had more than negligible sensitivity to other than fast neutrons, it would respond to the scattered neutrons from the walls and tank. This would be shown by other than straight line behavior in Figure 4-1 at distances greater than 35 cm.

Thermoluminescent dosimeters of the same lithium-6 content give reproducible results from dosimeter to dosimeter irradiated under the same conditions. Table 4-5 demonstrates this by listing the thermoluminescent responses of 10 TLD-100 dosimeters irradiated under the

TABLE 4-5

Responses of Ten Thermoluminescent Dosimeters of

Type TLD-100 to a 5.5 mg Californium-252 Source

Thermoluminescent Response in Equivalent Roentgen

72	77
76	78
73	78
73	76
78	76

same conditions. The reproducibility of the same dosimeter from irradiation to irradiation is also excellent. All dosimeters responded within the $\pm$ 5 percent accuracy (as guaranteed by Victoreen). The values tabulated in Tables 4-3 and 4-4 are the averages of the actual responses of the dosimeters.

In the first experimental configuration, in air, the different types of lithium fluoride dosimeters have various responses to the different shielding conditions. The most notable difference in response is between TLD-600 and TLD-700. TLD-600, bare, has a 38 percent higher response to the same radiation field. The gamma response of all three types of lithium fluoride dosimeters should be the same. Future experimentation may show that the gamma responses are not the same. At the current state of development, however, there is no evidence that the gamma response of different lithium fluoride dosimeter types should be different[23]. Therefore, the increased response of TLD-600 must be due to neutrons. In this experiment, there is a negligible thermal flux due to the building and tank of water below. In addition, the thermal neutron flux dose contribution to the fission spectrum is less than 0.1 percent as determined by Stone [24]. If there were a significant thermal flux, there would be a decrease in TLD-600 response with cadmium or lithium-6 covers which effectively screen out most thermal neutrons. A sheet of 0.1 mm of ^{6}LiF absorbs 50 percent of incident thermal neutrons[25]. In this experiment, a sheet of ^{6}LiF approximately 0.8 mm thick was used. Therefore, the increased response of TLD-600 over TLD-700 must

be due to fast neutrons and is approximately 1.0 equivalent roentgen per mg of californium-252 for this experiment. The relative neutron response (k) can be defined as[26]

$$k = \frac{\text{TLD response produced by a neutron field which will deposit one rad in tissue}}{\text{TLD response produced by an exposure of one roentgen of cobalt-60 gamma radiation}}$$

The relative neutron response of TLD-600 to fast neutrons (k_{F-600}) is therefore equal to 0.17 equivalent roentgen/rad. Recent work in the area of TLD response to fast neutrons by McGinley[27] had shown that TLD-700 has a response to fast neutrons also. His work shows that the relative neutron response of TLD-700 to fast neutrons (k_{F-600}) from californium-252 is 0.033. The accuracy and methods available in this thesis do not allow the resolution of this response. It is of interest to note in Table 4-3 that the gamma response of all lithium fluoride dosimeters is significantly less than the gamma exposure they were subjected to. Experimentation by ionization chamber methods indicates an exposure of approximately 6.0 roentgen/mg whereas the thermoluminescent dosimeters have a response of approximately 2.7 roentgen/mg. This difference could possibly be due to an error in calibration of the lithium fluoride dosimeters or a difference in energy response.

Analysis of the results in Tables 4-2 and 4-4 shows a marked response of TLD-600. It is evident that this response is due to the thermal neutron flux present. The neutron radiation field is basically the same as the neutron field described in Table 4-1 for the bare

source experiment, except for a large thermal neutron component superimposed on the fast neutron flux. Since the fast neutron flux in each experimental configuration is the same, the thermoluminescent response to the fast neutron flux should be the same also. Thus, in Table 4-4, approximately 1.0 equivalent roentgen of the total response of TLD-600 is possibly due to fast neutrons. Again the TLD-700 response to fast neutrons would be indiscernible.

The gamma dose rate for the experiment carried out adjacent to the "semi-infinite" water medium should be slightly higher than the gamma dose rate for the bare source experiment. The increase in dose rate should be primarily due to capture gammas from the elements in the stainless steel of the tank and hydrogen in the water. In Table 4-4, TLD-700, bare, shows a 15 percent greater response than TLD-700 in Table 4-3 under the same shielding conditions. The difference could be due to thermal neutrons or the increased gamma dose. Completely pure ^{7}LiF is not available, and trace amounts of lithium-6 can cause a noticeable response by thermal neutrons. This 15 percent greater response is significantly greater than statistical variations. The values for TLD-700 in Table 4-4 are slightly lower for the shielded dosimeters. It is believed that the cadmium and lithium screens absorbed some of the thermal neutrons. It is not possible with these results, however, to conclusively state whether the increased response by TLD-700 in Table 4-4 is due to thermal neutrons or the increased gamma dose. The increased response of TLD-600, bare, in Table 4-4 over the comparable value in Table 4-3

is due largely to thermal neutrons. Assuming approximately the same response to gammas and fast neutrons as in Table 4-3, the thermal neutron component is 47.38 equivalent roentgen per mg. Since the thermal neutron dose delivered to produce this response was 0.18 rad per mg, the relative neutron response by TLD-600 due to thermal neutrons (k_{th-600}) is 263 equivalent roentgen/rad. Further analysis of the data in Table 4-4 shows that TLD-600, cadmium covered and lithium-6 covered, still show a response to thermal neutrons. Cadmium and lithium-6 do not screen out all thermal neutrons. The very small fraction of the thermal flux which passes through the cadmium or lithium-6 is enough to cause an appreciable response in TLD-600.

CHAPTER V

Conclusions

Lithium fluoride thermoluminescent dosimeters show a response
to the gamma and neutron radiations from californium-252. The
resolution and determination of these responses is not straight-
forward because of the complex radiation field caused by the inter-
action of the radiations with matter. Some degree of resolution of
the components of the thermoluminescent response is possible under
special irradiation conditions. In particular, this investigation
uses measurements made in radiation fields characterized by techniques
other than thermoluminescent dosimetry. Experimental configuration
that vary the thermal neutron component, neutron shields, and
measurements with lithium fluoride dosimeters of different
lithium-6 content are used to quantify the response of the thermo-
luminescent dosimeters. Some of these responses cannot be resolved
in the total thermoluminescent response because of statistical
variation in the response of the thermoluminescent dosimeters.
If significant thermalization of the neutron flux occurs, the thermal
neutron response in TLD-600 will dominate due to the high cross
section for the $^6Li(n, \alpha)^3H$ reaction.

Control thermoluminescent dosimeters are an effective means to
monitor for possible changes in thermoluminescent dosimeters or
irradiation conditions. Control thermoluminescent dosimeters can

give an indication of radiation damage or change in sensitivity of the dosimeter, and changes in source geometry or activity.

It has been found that a high degree of accuracy in the reproduction of the thermoluminescent response can be obtained. The level of accuracy is determined and guaranteed by the manufacturer. Thermoluminescent dosimeters with even higher levels of accuracy are available from the manufacturer at increased cost. Such thermoluminescent dosimeters would be useful in the determination of the thermoluminescent responses to components of radiation fields which otherwise could not be resolved.

It is felt that a basis has been formed for further research and development of the thermoluminescent dosimetry capabilities at the Nuclear Science Center. Some recommendations for further research are:

1. Control thermoluminescent dosimeters should, whenever possible, be incorporated into experimentation with thermoluminescence.

2. Gamma dosimetry by a method other than thermoluminescence should be improved. Recently calibrated, highly accurate survey meters could be used with a closed circuit television system to accurately obtain the gamma dose from californium-252. The thermoluminescent dosimeters used in the experimentation of this thesis should be exposed to an accurately known amount of cobalt-60 to check the accuracy of the calibration factor determined by the manufacturer of the thermoluminescent dosimeters. With data from

experimentation such as this, the discrepancy in the gamma
response of calibrated thermoluminescent dosimeters can
be investigated. Such research should reveal whether the
discrepancy in the gamma response of the thermoluminescent
dosimeters is due to inaccurately calibrated thermoluminescent
dosimeters, errors in dosimetry by methods other than
thermoluminescence, or a difference in energy response.

3. Research should be done to improve methods of screening out
    thermal neutrons. Thermoluminescent dosimetry could be
    greatly improved by the ability to determine the fast
    neutron and gamma response in a radiation field with a large
    thermal neutron component.

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APPENDIX A

Application of Thermoluminescent Dosimetry to

Biological Research with Californium-252

Research is being conducted to determine the suitability of californium-252 to induce genetic damage in <u>Diatraea sacharalis</u> (L.), the sugar cane borer. Pupal stages of the sugar cane borer are being irradiated by neutron and gamma radiation from a 6.5 mg californium-252 source. Gamma dosimetry in the irradiation chamber (shown in Figure A1-1) is being performed by thermoluminescent dosimetry. Thermoluminescent dosimeters of types ^{7}LiF and CaF_2, both of which are relatively insensitive to neutron radiation, are used to determine the gamma dose at various positions in the chamber. Because of the dimensions of the chamber and the large gradient in the radiation field, most methods of gamma dosimetry are not suitable in this application. Thermoluminescent dosimeters (nominal dimensions 0.125 in by 0.125 in by 0.030 in) were able to estimate the gradient in the radiation field by determination of the gamma dose at small volumes within the irradiation chamber. The positions the thermoluminescent dosimeters occupy are shown by astericks on Figure A1-1.
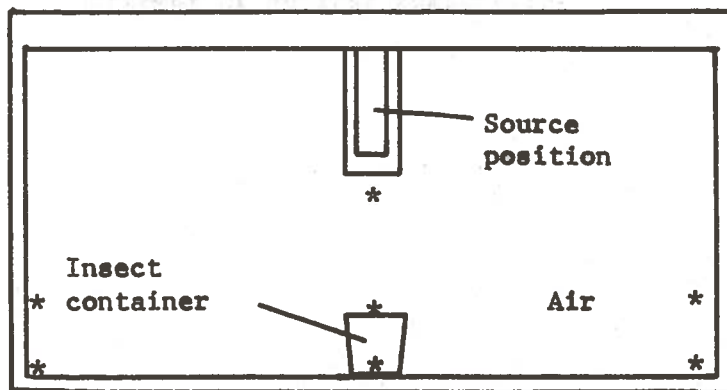
FIGURE A1-1

Chamber for Irradiation of the Sugar Cane Borer

Locations of Thermoluminescent Dosimeters Indicated by Asterisks

Water

Air

Plastic
chamber

Top View

Source
position

Insect
container

Air
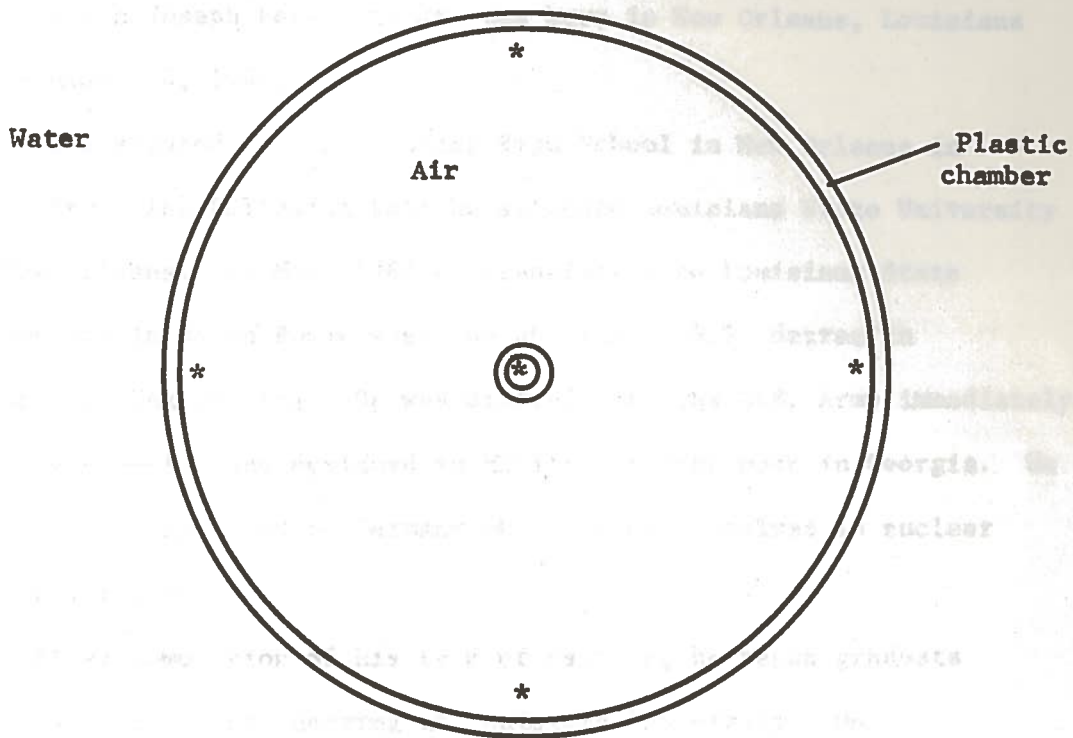
Side View

VITA

Calvin Joseph Bergeron, Jr. was born in New Orleans, Louisiana on December 14, 1947.

He graduated from Holy Cross High School in New Orleans in May, 1965. The following fall he attended Louisiana State University in New Orleans. In May, 1968 he transferred to Louisiana State University in Baton Rouge where he obtained a B.S. degree in Mechanical Engineering. He was drafted into the U.S. Army immediately after graduation and assigned to Military Police work in Georgia. He was later transferred to Germany where he was involved in nuclear weapon security.

After completion of his term of service, he began graduate study in Nuclear Engineering at Louisiana University. On December 21, 1974, he married the former Maida Elizabeth Owens of Baton Rouge. He is presently a candidate for the degree of Master of Science in the Department of Nuclear Engineering.

THE GRADUATE SCHOOL—LOUISIANA STATE UNIVERSITY

Report of _____Oral_____ Examination for the degree of __M.S.__

We, the undersigned, as a Committee, have on __July 11, 1975__
(Date)

examined __Calvin J. Bergeron__ , a candidate for the degree

of __Master of Science__
upon work done by

__him__ in the field of __Nuclear Engineering__ and report the
(her, him)

following results: ___Passed___

Committee:

John Courtney (signature)   John C. Courtney
(Chairman)

(signature)   Frank A. Iddings

Edward N. Lambremont (signature)   Edward N. Lambremont

Date __July 14, 1975__   Approved: _James P. Traynham_ (signature)
(Dean of the Graduate School)

To be returned in duplicate to the Dean of the Graduate School