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ARF 1214-TR 3
(Quarterly Technical Report No. 3)

A NUCLEAR-PHOTON
ENERGY CONVERSION STUDY

Aeronautical Systems Division
Wright-Patterson Air Force Base, Ohio
ATTN: ASRMFP-2

Contract No. AF33(657)-8527

Project 8173, Task 817301-17
BPSN: 2(3-3145)-60959-17

ARMOUR RESEARCH FOUNDATION

of

Illinois Institute of Technology
Technology Center
Chicago 16, Illinois

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(Covering the Period from 1 October 1962 to 31 December 1962)

January 15, 1963

NOTICE

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ARF 1214-TR3

(Quarterly Technical Report No. 3)

A NUCLEAR-PHOTON ENERGY CONVERSION STUDY

ABSTRACT

The temperature dependence and phosphor thickness dependence on the luminescence output was studied for several phosphors which appear promising for use in a nuclear-photon energy conversion system. In this system, beta particles from a radioisotope source are absorbed by the phosphor which emits a multiplicity of low energy photons. These photons are then converted to electrical energy by a photovoltaic device. The most promising phosphor to date, a ZnCdS:Cu, had about a 15 percent decrease in luminescence intensity as the temperature was increased from room temperature to 120°C; and was found to be relatively transparent to its own light with an optimum thickness for Sr^{90} beta excitation of 160 mg/cm^2. Strontium-90 beta particle radiation damage studies on this phosphor indicate an onset of damage at a total radiation dose of 10^8 ergs/cm^2. These data, in conjunction with the previously reported data on silicon cells, indicate that Sr^{90} can not be used and that the maximum operating temperature is about 120°C. Calculations were made on the temperature restrictions on the double-conversion power source geometry. These calculations indicate that, with a maximum temperature of 120°C allowed in the device, a maximum power input density of 200 mw/cm^2 is permitted. Experiments are in progress on the source-phosphor geometry of an intimate powder mix of promethium-147 and ZnCdS:Cu. Also, experiments are being conducted on the fabrication of a single crystal phosphor - photovoltaic device.

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A NUCLEAR-PHOTON ENERGY CONVERSION STUDY

I. INTRODUCTION AND SUMMARY

The overall objective of this research project is to study and design a nuclear radiation powered electrical energy source based on the following energy conversion scheme:

1. A primary source of nuclear radiation from a radioisotope.
2. A material which absorbs the nuclear particles and re-emits a multiplicity of low energy photons, i.e., a luminescent material.
3. A converter of low energy photons to electrical energy, i.e., a photovoltaic material.

The energy source is to have a ten watt output at 28 volts or less.

Early in this project we chose one or two candidate materials for each of the three components (the photovoltaic converter, the luminescent material, and the radioisotope) of the energy conversion scheme. These choices were based on a literature and information search and on materials which exist today as essentially "off-the-shelf" items; and these choices are, if not the best, at least representative of the necessary components. The photovoltaic converter choice was the p-n or n-p junction silicon photovoltaic cell; a transparent single crystal of silver or copper doped cadmium sulfide was the luminescent material choice; and the high specific activity radioisotopes of promethium-147 and strontium-90 are being considered.

During the second quarterly period a set of silicon p-n junction photovoltaic cells was experimentally studied to determine the effects of illumination intensity, temperature, and gamma radiation on the converter efficiency. The efficiency degradation observed with cobalt-60 gamma radiation, which simulates the bremsstrahlung from the high energy beta particles of Sr-90, becomes significant above a total dose of 10^5 rads. An n on p silicon cell should withstand about an order of magnitude more radiation than a p on n cell for identical damage. The bremsstrahlung radiation damage to the photovoltaic converter may require the use of a lower energy beta particle source.

The observed decrease in efficiency of the silicon cell at low illumination intensities (0.1 to 1.0 mw/cm^2) is of concern because the expected brightness from nuclear light sources is low. This fact suggests the consideration of silicon cells or other types of photovoltaic materials with wider junction transition regions where it is expected that there would be less efficiency at high light levels, but a less rapid decrease in efficiency at low light levels. In any case the optical coupling between phosphor and photovoltaic material need be carefully designed to get the maximum amount of light into the silicon.

Silicon cell efficiency was found to decrease nearly linearly with increased temperature in the range of 25°C to 180°C. The efficiency at 180°C

is twelve percent of that at 25°C.

Also in the second quarterly period we scanned seventeen commercial phosphor samples for their relative luminescence outputs as detected by a silicon cell. All of these phosphors were of the powder type. A ZnCdS: Cu and a ZnS: Cu phosphor gave the best response of the samples studied.

During this third quarterly period the experimental work has been primarily on phosphors and radioisotope source - phosphor geometry. The temperature dependence of phosphor light output was determined in the temperature range of 20°C to 200°C for several of the better response phosphors. Also the effect of strontium-90 beta radiation dose on the light output of the ZnCdS:Cu phosphor has been experimentally studied. Several newly received phosphors have been examined for use with a silicon cell, but none has given a better response than the ZnCdS:Cu.

Back surface strontium-90 beta excitation has been used to determine the light output versus phosphor thickness for this source-phosphor geometry. Experiments on the source-phosphor geometry of an intimate powder mix of promethium-147 oxide and ZnCdS:Cu are in progress.

In addition to the above, a novel single crystal phosphor-photovoltaic converter combination is discussed and initial experiments to form a photo-barrier at the surface of a CdS crystal are described. Also, calculations were performed to find the operational temperature reached in a double conversion power source of this type for various geometries.

II. PHOTOVOLTAIC CONVERTER

A. Silicon Photovoltaic Cells

No additional study on silicon cells has been performed since the last quarter when the effects of low illumination intensity, temperature, and nuclear radiation damage on the conversion efficiency of p on n silicon cells were determined. Concerning the radiation damage, however, it is known that an n on p silicon cell will withstand an order of magnitude more radiation than a p on n cell for the same degradation effect (ref. 1).

We have obtained a set of silicon cells which are one inch diameter size. These are being used for the detection of the phosphor light outputs. We have checked these cells against an Eppley thermopile with a tungsten light which is filtered with both a sharp cut red filter, passing radiation above 590 millimicrons, and a water filter, eliminating the long infrared radiation.

B. Single Crystal Phosphor – Photovoltaic Converter

One problem in fully utilizing the luminescence from a phosphor is the internal light trapping and Fresnel reflection losses at the boundary of a high refractive index material. This is especially true for large single crystals of cadmium sulfide which have a refractive index of 2.5. There are no suitable optical coupling materials in this high index range. A novel form of combined phosphor and photovoltaic converter in a single crystal would increase the optical coupling appreciably. Such a converter had been suggested in our proposal (ARF Proposal No. 62-221A, p. 18, November 3, 1961), for example, a single crystal of CdS:Ag with a rectifying barrier on one surface for the photovoltaic material. This type of photoconverter may also have advantageous properties at low illumination levels.

In the use of such a single crystal phosphor – converter the geometry of the exciting beta particles would be chosen so that all of the beta particle energy is expended in the phosphor type material. There should be no direct beta particle irradiation of the photobarrier region.

Experiments are in progress for making an integral phosphor–photovoltaic converter unit from a single crystal of luminescent CdS. To date, however, the required photovoltaic behavior has not been observed, apparently due to the lack of adequate forming of the barrier electrode. The basis for this remark and specific details of the experiments are discussed in the following paragraphs.

The starting materials for these experiments were single crystal pieces of CdS of two types: 1) "pure" (that is, not purposely doped) and 2) lightly doped with copper. The aim of these experiments was to convert one side of the single crystal into a photovoltaic cell. The CdS was sectioned and lapped. Sections were placed in an evaporator and a layer of indium was deposited approximately 130 Angstroms thick, based upon transmission measure-

ments through a microscope slide also placed in the evaporator.

The samples with the evaporated indium were placed in a pyrex tube along with some powder CdS. The powder was not touching the samples, however. The tube was evacuated using a mechanical and diffusion pump system overnight. The pyrex tube was then filled with helium to approximately one atmosphere, and sealed off. It was placed in a furnace which had equilibrated at 610°C over a length longer than the sealed off pyrex tube. The idea, here, was to make sure that the end of the tube without CdS was a temperature at least equal to that of the sample, to minimize sublimation. The tube was left in the furnace for 24 hours.

The choice of temperature and time was based upon work done with CdTe. It appears that indium diffuses into CdS and CdTe comparatively easily, and this determined the choice of temperature. The reason for the long diffusion time was that the desired end product of this part of the experiment was a nearly uniform concentration of indium in a thin layer. The result would be an n-type layer to which one could make electrical contact. The long time factor can be appreciated by considering a solution of the diffusion equation which is valid for a thin evaporated layer (ref. 2):

$$N = \frac{N_o \, \Delta \, e^{-x^2/4Dt}}{(\pi Dt)^{1/2}}$$

where

N_o = initial concentration of indium

Δ = thickness of evaporated layer

x = distance normal to evaporated surface

D = diffusion constant

t = time

Note that for long times, the exponential term approaches unity and that the concentration value N is nearly independent of depth, x. Obviously, the experiment is only an approximation to this condition, but it does offer one way to put a thin conductive layer in the crystal. Indium was soldered to the side to make contact with the n-type layer, and a resistance value of about 100 ohms was found. Based upon before and after measurements, the indium did diffuse in, and generate carriers. (It is known that indium is a donor for CdS.)

The surface on which the indium had been evaporated was now partly masked off, and the rest of the crystal was masked off. Copper was then deposited electrochemically on the unmasked portion of the originally indium evaporated surface. Following this, the mask material was removed, and a

solder contact was made to the copper. The "devices" were checked at this time for photovoltaic behavior with negative results. However, part of the prescription for making a good barrier or junction electrode involves a post-heating of the electrochemically deposited copper, based upon published work (ref. 3). The cells made in this laboratory were, therefore, incomplete. Specifically, the forming of the copper electrode involves heating at 460°C for about one minute. In addition, the surface on which the copper is deposited is to be roughened. These experiments are in progress.

III.   PHOSPHORS

A few additional commercial phosphors have been checked. None, to date, has a better usable luminescence output than the ZnCdS:Cu (NBS 1024) which was reported previously. The last of the phosphor materials on order have just been received, but no measurements are available for this report. Of special interest for examination are some new large single crystal CdS phosphors which under ultraviolet excitation visually appear to have a good red luminescence. These samples are CdS:Cu (25 ppm), CdS:Cu (75 ppm), and CdS:In (20 ppm) Ag (40 ppm), and they were obtained from Harshaw Chemical Company.

Also of interest is a set of six copper doped mixed ZnS, CdS powder phosphors. These phosphors have ratios of ZnS to CdS which vary in the following way:

ZnS:Cu

(0. 8) Zn (0. 2) CdS:Cu

(0. 6) Zn (0. 4) CdS:Cu

(0. 4) Zn (0. 6) CdS:Cu

(0. 2) Zn (0. 8) CdS:Cu

CdS:Cu

The peak of the emission band shifts to the red as the cadmium concentration increases. With this set of phosphors we will determine which one of this type matches the photovoltaic converter response.

A.   Temperature Dependence

The temperature dependence of the luminescence intensity under constant ultra violet excitation has been experimentally determined for the ZnCdS:Cu (NBS 1024), the ZnS:Cu (NBS 1022), the ZnS:Ag (NBS 1020), and the U. S. Radium 4890 powder phosphors. The ZnCdS:Cu and ZnS:Cu phosphors were examined since they have produced the best response with silicon cells; the ZnS:Ag was examined to check the difference between a copper, and silver activated phosphor; and the US Radium 4890 was examined since it has a greater proportion of emission in the red end of the spectrum than any other phosphor of significant emission except for the ZnCdS:Cu. (See Table 1 of ARF 1214-TR2, p. 16, October 9, 1962).

A modified laboratory hot plate was used to heat the phosphor con- tainers, whose temperature was measured with a thermocouple. Each tem- perature point was allowed to come to equilibrium. Luminescence was measured with a silicon cell both with and without an optical filter which passes light of wavelength greater than 0. 59 microns. A water filter was used in front of the silicon detector to reduce the heat radiation to the detector. The detector was removed between readings to prevent it from heating up. A complete temperature

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run was made with a "dummy" sample, during which time the detector was checked with constant illumination and the detector sensitivity remained constant.

Temperature data are shown in Figures 1 and 2. Phosphor light output is given relative to a normalized value of 1.0 at room temperature for all cases. The ZnS:Cu and ZnCdS:Cu phosphors retain a useable luminescence output up to temperatures of about 150°C. A shift of the emission to the red or longer wavelengths is indicated by the red transmitting filter data curves which retain a higher relative output than the total intensity curves. This is especially true for the ZnS:Cu phosphor.

The ZnS:Ag and US Radium 4690 phosphors show an appreciable decrease in light output with increased temperatures. This temperature behavior for a silver activated phosphor versus a copper activated phosphor is also reported in Leverenz (ref. 4) for several of the ZnS-CdS type phosphors. Thus for this program the copper activated phosphors are the most useable.

B. Nuclear Radiation Effect on Phosphor Light Output

A 160 mg/cm^2 thickness of ZnCdS:Cu phosphor powder was irradiated by a sealed strontium-90 beta source of 0.3 curie intensity. The beta particle energy deposition rate or dose rate in the phosphor was 3.2 x 10^5 ergs/cm^2 hr, which was determined by cobalt glass dosimetry. The luminescence output for this beta excitation was measured with a silicon cell at various times during the irradiation. In this experiment the silicon cell is inserted only at the time of measurement so that there is no radiation effect upon the detector. The strontium-90 source holder and sample and detector positioning are described in Appendix A.

The relative light output as a function of beta particle dose is shown in Figure 3. After 800 hours of Sr-90 irradiation the luminescence had decreased to 80 percent of that at zero time. Since this particular sealed Sr-90 beta source is a very low intensity source (0.01 mw/cm^2) compared to the estimated maximum intensity Sr-90 source (10 mw/cm^2), it appears that the Sr-90 beta damage to the phosphor would be much too severe for use.

This irradiation is being continued for another couple of weeks. At that time the phosphor will be heated to about 200°C to determine if some of the luminescence output may be restored by temperature annealing. Cobalt-60 gamma irradiations (at a much higher dose rate) will also be given to this and other phosphors to compare the radiation effects. A low energy beta irradiation is being given at this time to a ZnCdS:Cu phosphor which is mixed with a promethium-147 source. This is described in the following section.
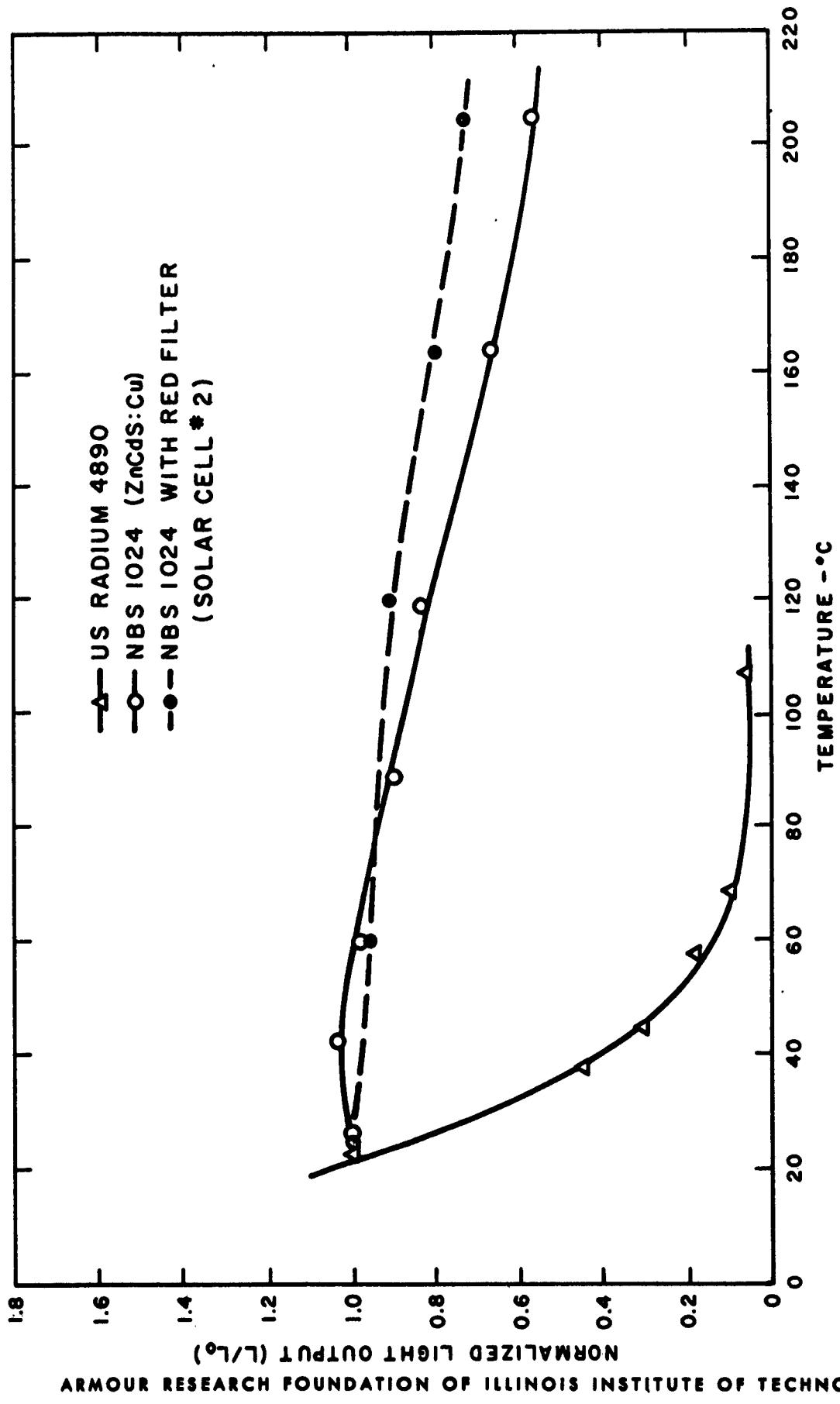
FIG. 1. EFFECT OF TEMPERATURE INCREASE ON THE LIGHT OUTPUT
OF VARIOUS PHOSPHORS WITH ULTRAVIOLET EXCITATION

Legend:
- △— US RADIUM 4890
- ○— NBS 1024 (ZnCdS:Cu)
- ●— NBS 1024 WITH RED FILTER (SOLAR CELL #2)

X-axis: TEMPERATURE – °C
Y-axis: NORMALIZED LIGHT OUTPUT (L/L₀)

FIG. 2   EFFECT OF TEMPERATURE INCREASE ON THE LIGHT OUTPUT
OF VARIOUS PHOSPHORS WITH ULTRAVIOLET EXCITATION

NBS 1020 (ZnS:Ag)
NBS 1022 (ZnS:Cu)
NBS 1022 WITH RED FILTER
           (SOLAR CELL #2)

TEMPERATURE – °C

NORMALIZED LIGHT OUTPUT (L/L₀)

FIG. 3 EFFECT OF STRONTIUM-90 BETA RADIATION ON THE
LIGHT OUTPUT OF A PHOSPHOR

PHOSPHOR NBS 1024 (Zn Cd S : Cu)
160 mg/cm^2
BETA PARTICLE DOSE RATE = 3.2 x 10^5 ergs/cm^2 hr

BETA PARTICLE DOSE, ergs/cm^2

RELATIVE LIGHT OUTPUT OF PHOSPHOR (L/L$_o$)
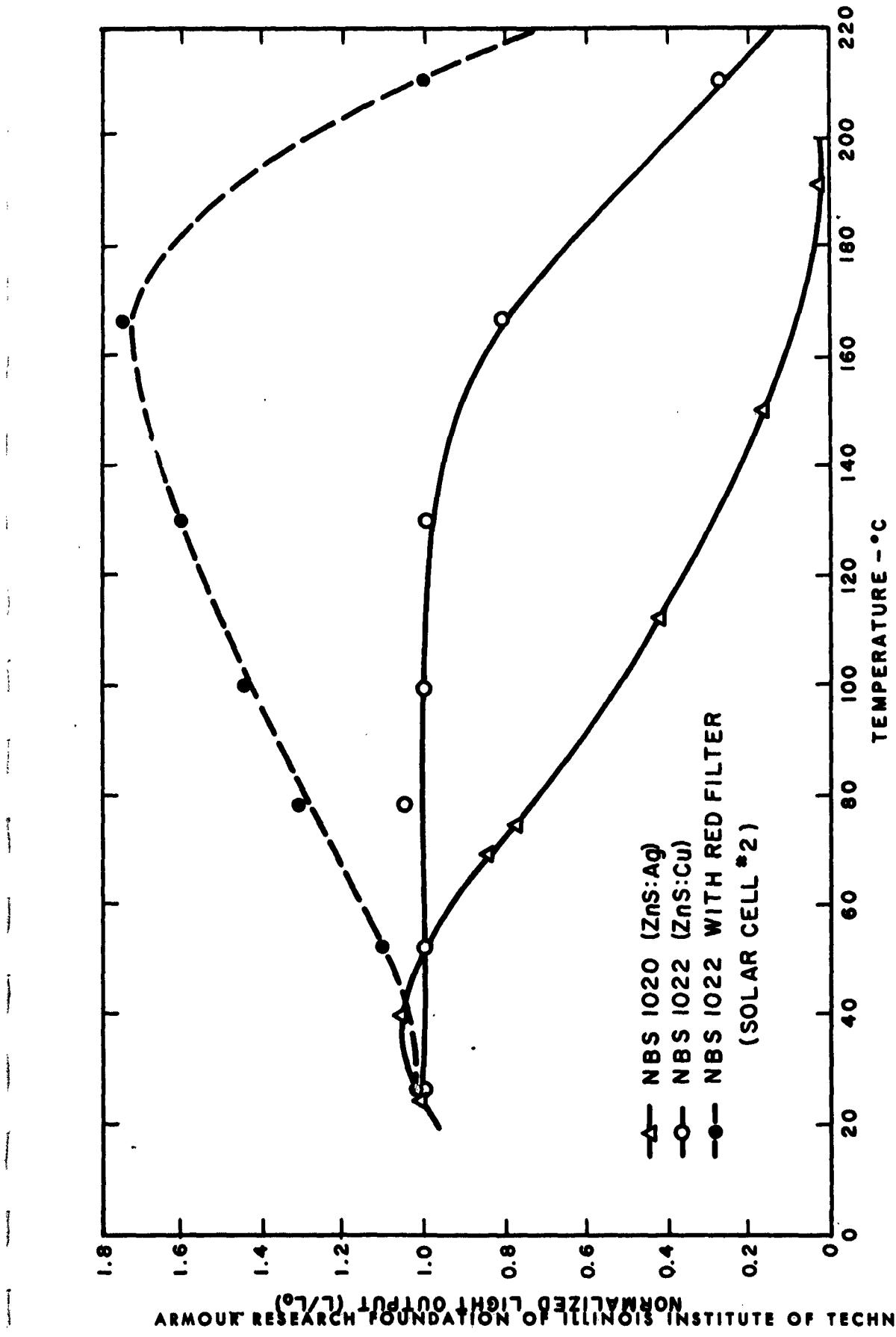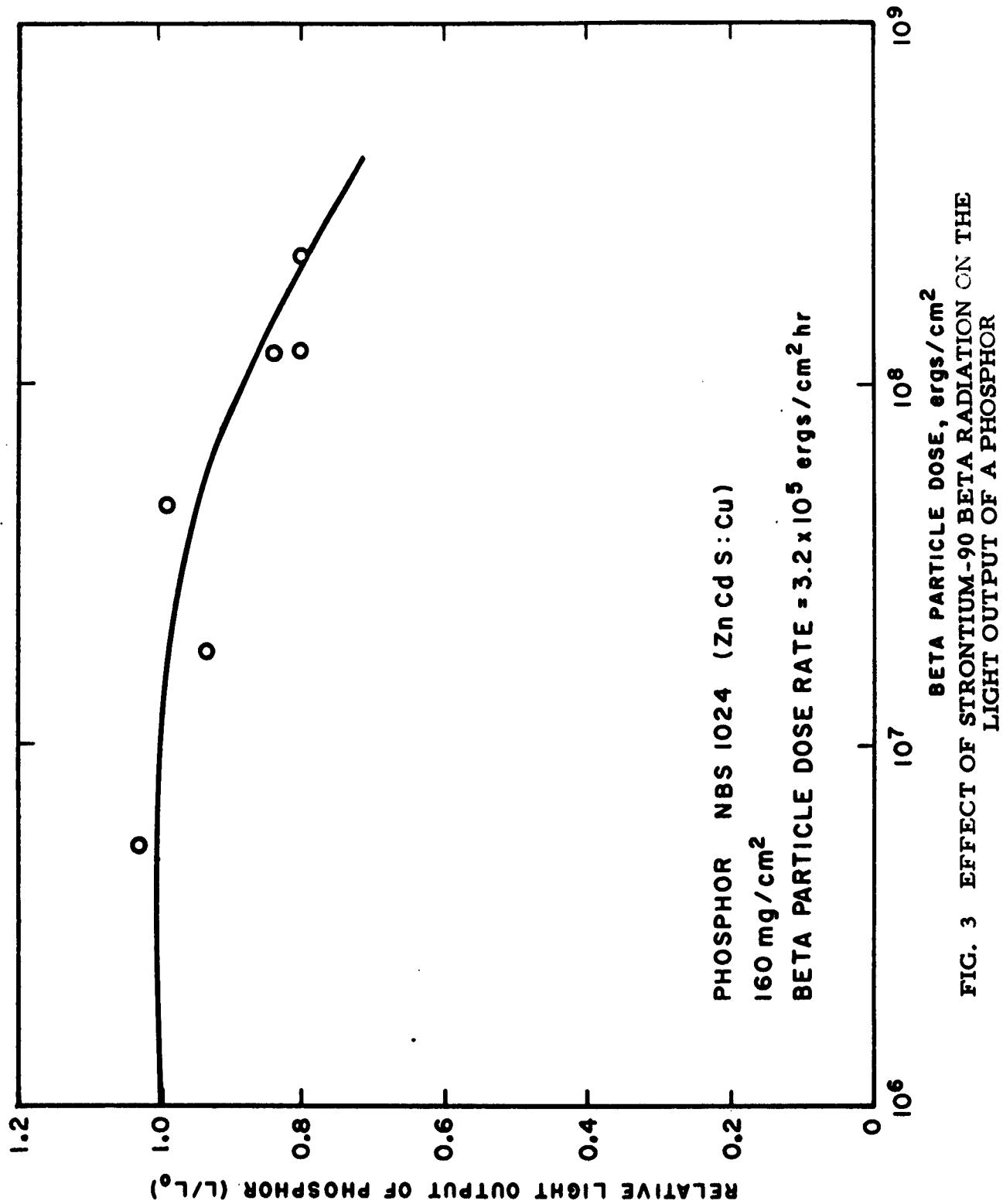
IV. SOURCE-PHOSPHOR GEOMETRY

A. Light Output versus Phosphor Thickness for Back Surface Sr-90 Beta Excitation

Three different powdered phosphors have been examined to determine the light output versus thickness for back surface Sr-90 beta excitation. The Sr-90 source holder and the phosphor and silicon cell detector positioning are shown in Appendix A. The data curves of relative light output as detected by a silicon cell versus phosphor thickness for the ZnCdS:Cu, ZnS:Cu, and ZnS:Ag phosphors are shown in Figure 4.

Maximum light outputs are observed at about 160 mg/cm^2 for ZnCdS:Cu, 100 mg/cm^2 for ZnS:Cu, and 20 mg/cm^2 for ZnS:Ag. These curves are influenced by both the beta particle excitation density as a function of distance in the phosphor and the optical transparency of the phosphor, as discussed in the first Quarterly Report (ARF 1214-TR1, pp. 16-19). Fortunately, the more promising powder phosphors for this application, the ZnCdS:Cu and the ZnS:Cu have greater optical transparencies to their own emission than that used in the calculations in the first Quarterly Report. For example, the optical half-thickness for the ZnCdS:Cu powder was measured for broad band "yellow-red" light and was found to be 64 mg/cm^2 compared to the 20 mg/cm^2 used in the earlier calculations.

B. Source-Phosphor Powder Mix

A second type of source-phosphor geometry is that where a radioisotope source and a powder phosphor are intimately mixed together. The parameters of interest in this geometry are the phosphor mass per curie of source and the total source-phosphor thickness for maximum light output. This geometry is most applicable for a low energy beta particle emitting isotope.

We are using the ZnCdS:Cu powder phosphor and a processed high purity promethium-147 radioisotope. The radioisotope was obtained from Oak Ridge National Laboratory in the form of a promethium-147 oxide powder. The specific activity, based on the purity quoted by ORNL, is calculated to be about 680 curies per gram of oxide material on December, 1962. Samarium and neodymium oxides are the major impurities.

Enclosed containers, shown in Figure 5, are being used for the source-phosphor mix study. These experimental containers are copper tubes which have been reamed out to have a one square centimeter inner cross section area. A radiation resistant fused silica (Corning 7940) window, also of one square centimeter area, is sealed at one end of each tube. A cylindrical steel piece is inserted inside the tube for the purpose of tamping the mixture and to serve as a back reflector. This tamper piece is controlled by a ring magnet which slips over the tube and allows one to raise and lower the tamper. The phosphor is added by a funnel through the side fill tube. When making additions

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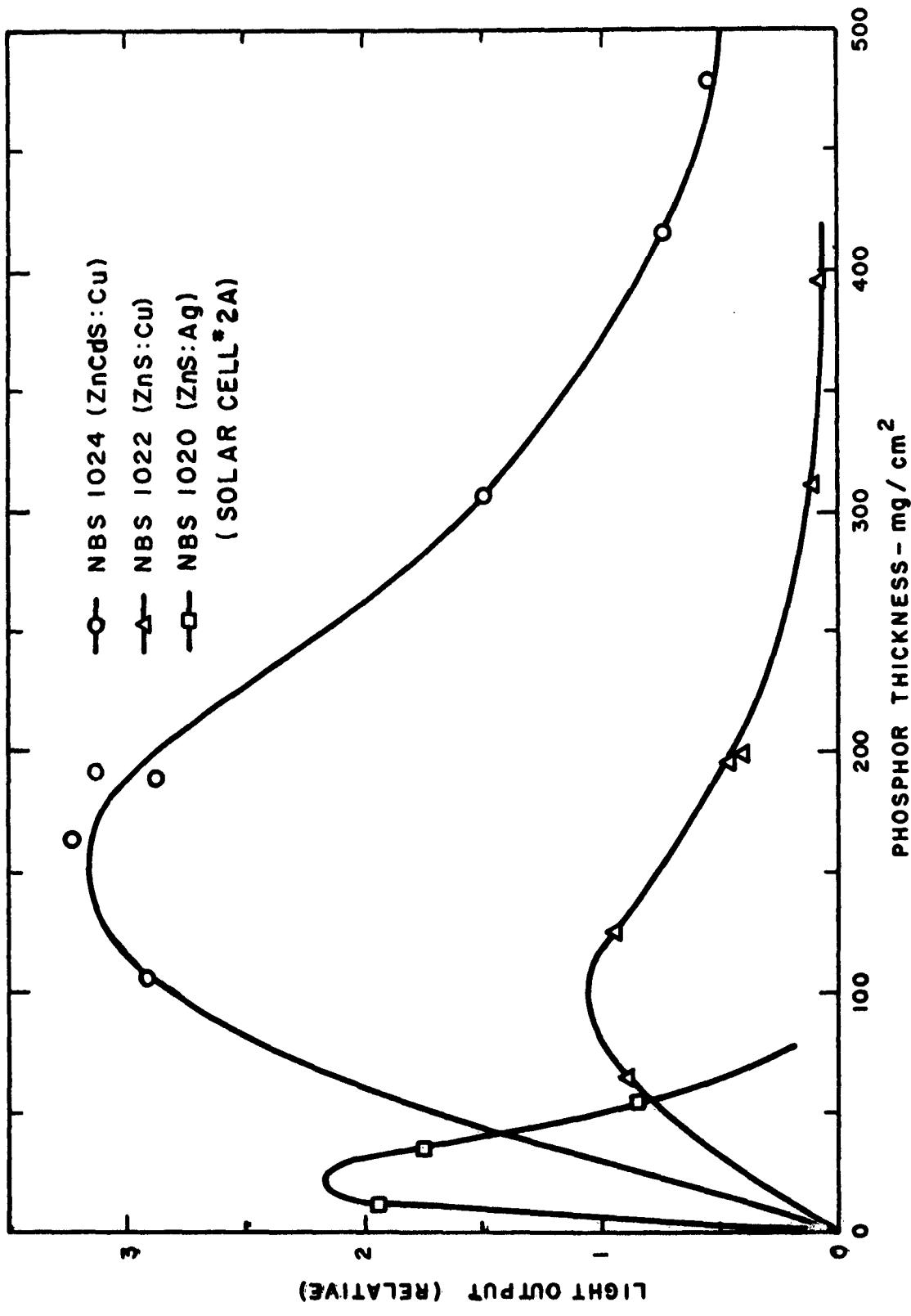
FIG. 4 MEASURED LIGHT OUTPUT OF VARIOUS PHOSPHORS WITH BACK SIDE Sr^{90}
EXCITATION AS A FUNCTION OF PHOSPHOR THICKNESS

Legend (on plot):

○ NBS 1024 (ZnCdS:Cu)
△ NBS 1022 (ZnS:Cu)
□ NBS 1020 (ZnS:Ag)
    (SOLAR CELL *2A)

X-axis: PHOSPHOR THICKNESS – mg / cm²
Y-axis: LIGHT OUTPUT (RELATIVE)

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12

Magnetic
Tamper

Fill Tube

Fused Silica Window
(Corning 7940)
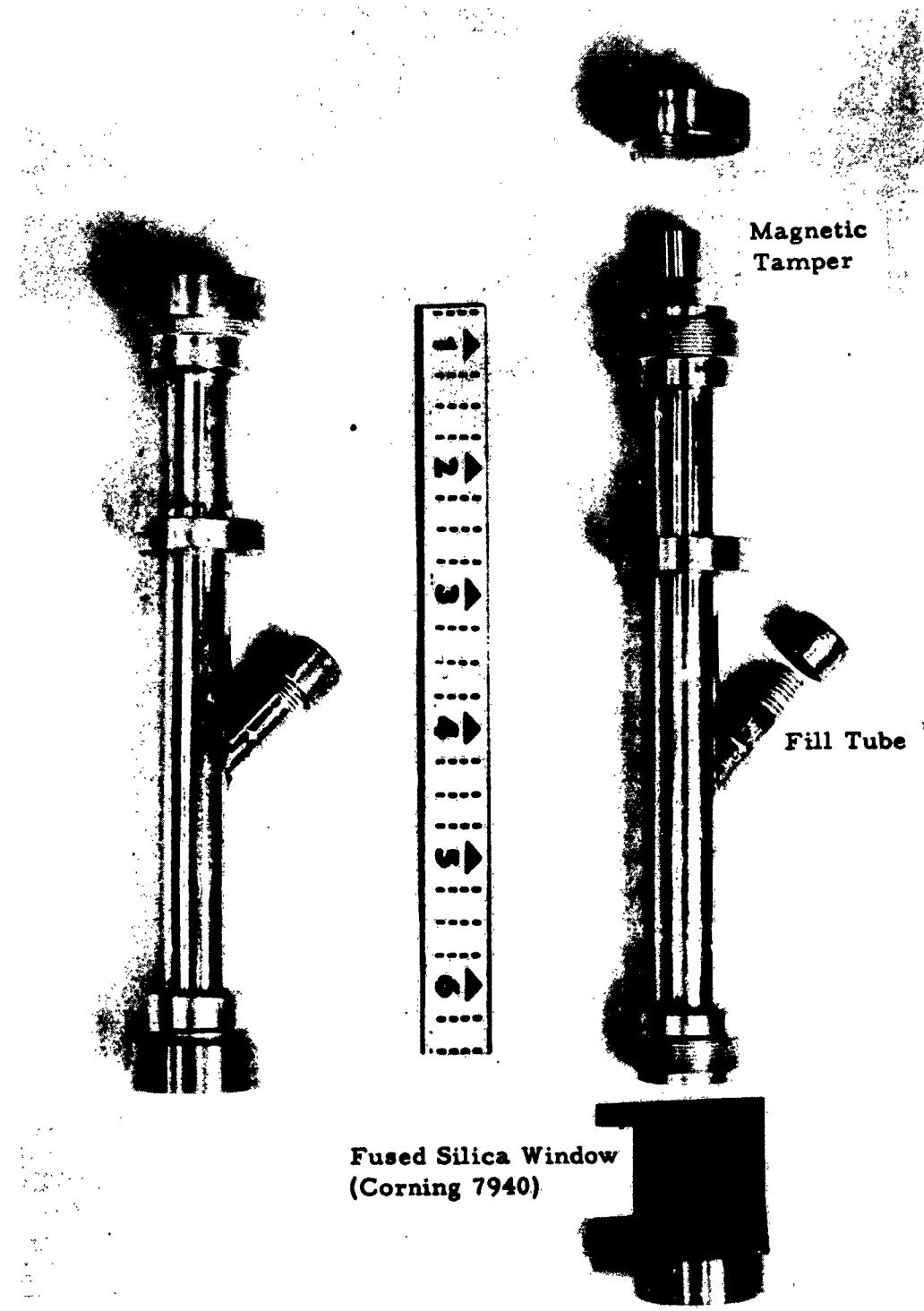
FIG. 5   APPARATUS FOR CONTAINMENT AND EVALUATION
OF PHOSPHOR-SOURCE MIXTURES

the tamper is raised above the fill tube. The cylindrical boss above the fill tube is used for positioning the sample tubes.

Three experimental containers have been loaded with 5.0 curies, 9.85 curies, and 16.7 curies of promethium oxide respectively. Pre-weighed increments of ZnCdS:Cu phosphor powder are then added to the containers and the light output is detected with a silicon cell. The results of this experiment form a three dimensional plot on which for each data point there is a light output value at a specific phosphor-source ratio (mg/curie) and a specific thickness (mg/cm^2). By using three containers with different source loadings there should be a sufficient range of parameters to determine at least the approximate optimum values. We have additional promethium oxide which can then be used to make an optimum mix.

To date, we have completed measurements on the 5.0 curie container with five different phosphor mass increments, and we have measurements on the other two containers for the initial phosphor additions. The remaining experiments with the three containers should be completed within the first half of January.

V.   TEMPERATURE RESTRICTIONS ON POWER SOURCE GEOMETRY

Since each unit of radioisotope-phosphor-photovoltaic cell is a heat source as well as a power source, there is a limit on the number of these units which may be stacked up. If too many are stacked, the innermost units will be operating at such a high temperature that the efficiency of power generation will be very low. An upper limit to the temperature, considering the effect of temperature on both the phosphor and the silicon photovoltaic device, has been set at 120°C. The maximum number of units which may be stacked without reaching a temperature greater than 120°C has been calculated for two types of power units and considering two methods of heat transfer. The first type of power units consists of a slab of strontium titanate ($SrTiO_3$) with a thickness of 600 mg/cm^2 ($\sim$ 20 curies) on a slab of single crystal cadmium sulfide (CdS) of thickness 1100 mg/cm^2, followed by a silicon cell with a thickness of 130 mg/cm^2. The second power unit considered contains a mixture of powdered promethium oxide (Pm_2O_3) and powdered phosphor with a total thickness of 105 mg/cm^2 on a silicon cell of thickness 130 mg/cm^2. The activity contained in this unit is about 26 curies.

The two methods of heat transfer considered are radiation to black space from the ends of a stack of units and convective cooling of the ends. The details of the calculation are given in Appendix B.

For radiative cooling to black space, the maximum permissible number of units which may be stacked without exceeding a temperature of 120°C were calculated as 2 units in the single crystal device and 22 units in the mixed source phosphor device. For convective cooling at an ambient temperature of 20°C, the maximum number of stacked units was calculated as 2 units in the single crystal case and 18 units in the mixed source-phosphor case.

The results of these calculations indicate that, regardless of the type of cooling employed, there is a maximum thickness and thus a maximum input area power density. For the single crystal-Sr^{90} device, the input power produced by the 40 curies of isotope is about 215 mw/cm^2. Likewise, the mixture of powdered Pm_2O_3 and phosphor, where the maximum permissible number of cells is about 20, gives a power input of about 200 mw/cm^2. Assuming an over-all conversion efficiency of one percent this would be an output power of 2 mw/cm^2 of stacked cells.

VI. CONCLUSIONS AND FUTURE WORK

The phosphor damage observed for the high energy Sr-90 beta radiation and the previously observed silicon cell damage from high energy gamma rays (simulating the bremsstrahlung of Sr-90 betas) tend to eliminate the use of Sr-90 in a ten watt output power device.

The maximum internal operating temperature of a power cell consisting of a copper activated ZnS, CdS type phosphor and silicon photovoltaic device is in the range of $120^{\circ}C$ to $150^{\circ}C$. Silver activated phosphors would require a lower operating temperature. Stacking of individual cells would thus be temperature limited to a specific input area power density of about 200 mw/cm^2, or about twenty cells using 26 curies of promethium-147 per cell.

Experiments are continuing on the source-phosphor powder mix geometry. These will be completed shortly and the results will give us a practical overall conversion efficiency value for the double energy conversion system of this type.

Several phosphors, both powder type and single crystals, remain to be examined. Also, experiments will continue on the fabrication of a single crystal phosphor-photovoltaic converter combination.

VII. LOGBOOKS AND CONTRIBUTING PERSONNEL

All data presented in this report are contained in ARF Logbooks numbers C12335, C12401, C12760, C12774, C12815, C12932 and C13124. The principal personnel contributing to this work are M. D . Oestreich, R. J. Robinson, and H. V. Watts. The temperature calculations were performed by Ludwig Wolf, Jr.

Respectfully submitted,

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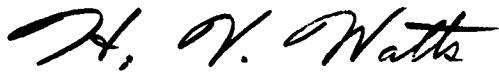
H. V. Watts, Research Physicist

S. Nudelman, Manager
Solid State Physics Research

APPROVED BY:

J. W. Buttrey, Assistant Director
of Physics Research

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APPENDIX A

STRONTIUM-90 SOURCE HOLDER

APPENDIX A

STRONTIUM-90 SOURCE HOLDER

In order to use an existing strontium-90 source for phosphor excitation and radiation damage studies, it was necessary to design a container which would permit maximum utilization of the beta particles from the source while minimizing personnel radiation exposure from both the beta particles and bremsstrahlung. The Sr-90 itself is a thin disc about 3/8 inch in diameter held in a brass disc about 1/2 inch thick and with a 1 inch diameter. The holder for the source, shown in Figure A-1, is a steel cylinder six inches in diameter and 3-1/2 inches thick with a hole in it allowing the source to be placed inside. A brass cover plate is then placed over the source to hold it in position. This cover plate is equipped with spring loaded tracks so that a drawer containing the materials to be examined may be inserted. The drawer is equipped to carry the brass phosphor container, described earlier, and a silicon solar cell. Electrical connections for the solar cell are made by a pair of insulated copper leaf spring terminals on the drawer and two insulated copper blocks on the brass cover plate. From the connections on the cover plate, leads are attached to a standard coaxial connector on the top of the steel cylinder.

The entire assembly is suspended from a top hook attached to a winch allowing the holder to be moved up and down in a concrete storage tube.

The distance from the surface of the beta source to the phosphor is about 1/2 inch and from the phosphor to the solar cell is about 1/8 inch. The total beta particle dose rate at the phosphor position, measured using cobalt glass dosimetry, is 6.0×10^5 ergs/cm² hr. Bremsstrahlung dose at the sample position is 10 r/hr and at the surface of the steel shielding cylinder about 10 mr/hr.

1. Source Position
2. Sample Position
3. Electrical Contacts
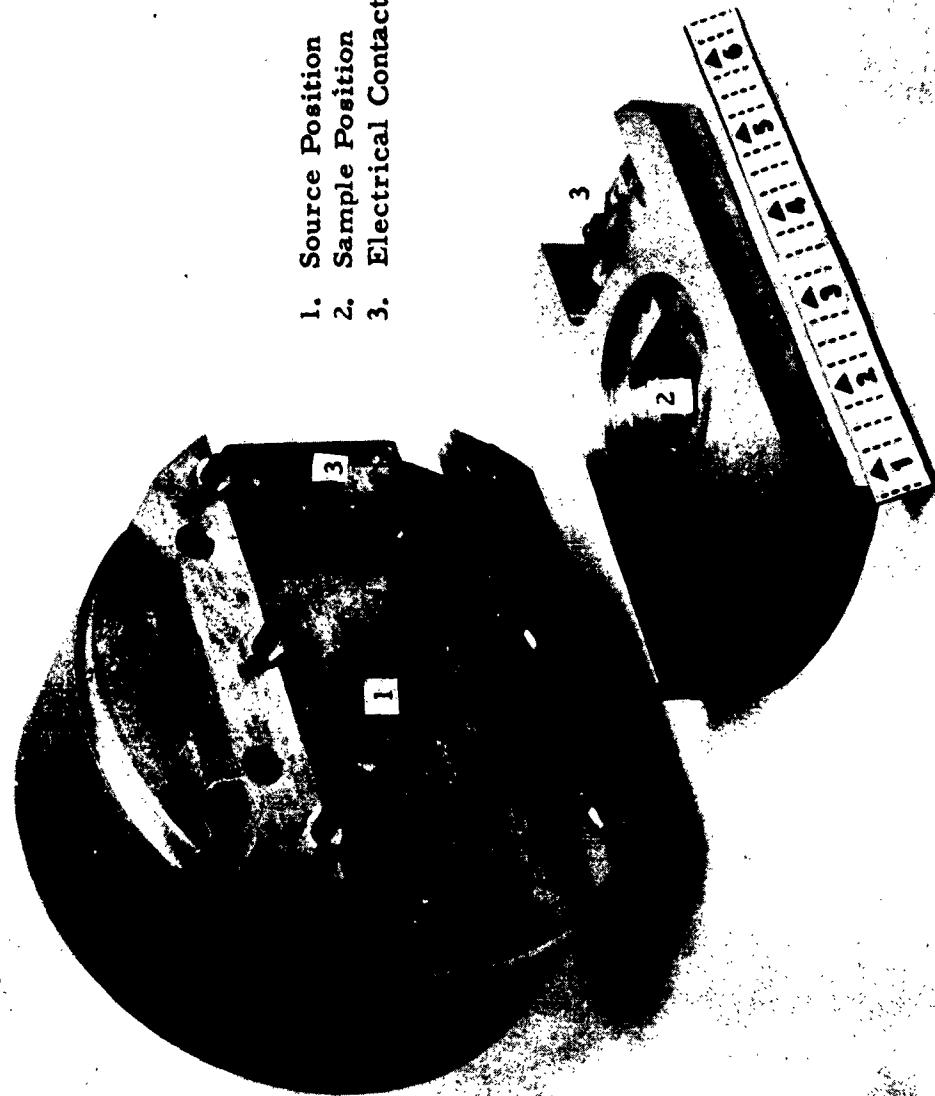
FIG. A-1 STRONTIUM-90 BETA SOURCE HOLDER AND SAMPLE DRAWER

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APPENDIX B

DETERMINATION OF THE TEMPERATURE
OF STACKED POWER CELLS

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I. INTRODUCTION

In order to develop an efficient power source using the principle of double energy conversion, space considerations require that individual radio-isotope-phosphor-photovoltaic cell units be stacked in some manner. Since each of these units is a heat source as well as an electrical power source, there is a limit on the number of these units which may be present in a single stack while maintaining the maximum power output of each unit. This stack thickness limit is determined by consideration of the possible mechanisms of heat transfer from the stack and by the maximum temperature permitted for best efficiency (120°C).

Two methods of heat transfer are considered: Radiation to black space from the ends of the stack and convective cooling of the ends. Two different compositions of the individual cells are to be considered. Case I consists of a slab of strontium titanate ($SrTiO_3$), a slab of single crystal cadmium sulfide (CdS), and a slab of silicon (Si). Case II consists of a slab of a mixture of powdered promethium oxide (Pm_2O_3) mixed with powdered cadmium sulfide on a slab of silicon.

II. THEORETICAL CONSIDERATIONS

For purposes of the derivation, each unit can be assumed to be a uniform slab with constant thermal conductivity and uniform internal heat generation. Since the heat relief occurs at the ends of the composite of individual units, a one-dimensional, steady heat flow analysis can be considered. Figure 1 shows the heat conduction - generation model.
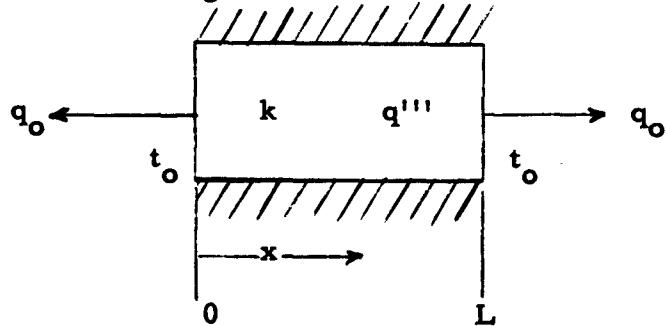

Fig. 1 One-dimensional, steady heat flow model

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The temperature distribution within the composite (t) can be found as (ref. 1),

$$t = t_o + \frac{q''' (L - x) x}{2k} \tag{1}$$

where k is the thermal conductivity, L is the composite thickness, q''' is the internal heat generation rate per unit volume, and t_o is the surface temperature of the ends. The maximum temperature in the composite occurs at its center (x = L/2) and is given as (ref. 1),

$$t_m = t_o + \frac{q''' L^2}{8k} \tag{2}$$

The heat transfer rate at the ends (q_o) is given as (ref. 1),

$$q_o = \frac{q''' AL}{2} \tag{3}$$

where A is the constant cross-sectional area.

In a stack of the individual power cells, the thermal conductivity is equal to the equivalent thermal conductivity (k) of each cell. The thermal conductivity of a heterogeneous cell, shown in Figure 2, is given as (ref. 1)

$$k = \frac{L_{13}}{\frac{L_1}{k_1} + \frac{L_2}{k_2} + \frac{L_3}{k_3}} \tag{4}$$
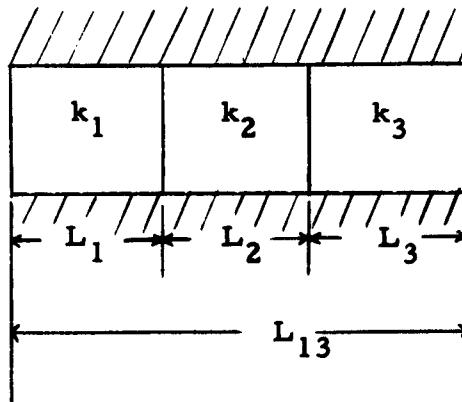
Fig. 2 Heterogeneous Cell

For convective heat relief (ref. 1),

$$t_o = \frac{q_o}{hA} + t_a \qquad (5)$$

where h is the convective heat transfer coefficient and t_a is the ambient temperature of the surroundings. Substituting (3) and (5) into (2),

$$t_m = \frac{q''' L}{2h} + \frac{q''' L^2}{8k} + t_a \qquad (6)$$

For radiative cooling to black space of the ends (ref. 1),

$$t_o = \left(\frac{q_o}{\sigma A \epsilon}\right)^{1/4} \qquad (7)$$

Thus, substituting in (2),

$$t_m = \left(\frac{q_o}{\sigma A \epsilon}\right)^{1/4} + \frac{q''' L^2}{8k} \qquad (8)$$

Where ϵ is the emissivity of the ends and σ is the Stefan-Boltzmann natural constant.

The assumption that the composite of individual cells can be represented by a uniform slab with uniform thermal conductivity and uniform internal heat generation is not completely true for small numbers of individual cells. The equivalent thermal conductivity of each cell no longer serves to represent the actual conductivity of each individual slab of the cell when only a few are stacked. For this situation a more rigorous analysis is necessary. The assumption of uniform internal heat generation may not be true for small stacks of individual cells since the heat generation within the cell may not be uniform.

III. PROPERTIES OF MATERIALS

The thermal conductivity and density of the materials under consideration were found as the following:

Strontium titanate (ref. 2)

$$\rho = 3.70 \ g/cm^3$$

$$k = \begin{cases} 0.0132 \ cal/cm \ sec \ K^o \ at \ 20^oC \\ 0.0144 \ cal/cm \ sec \ K^o \ at \ 120^oC \end{cases}$$

Mixed powder of promethium oxide and cadmium sulfide

$$\rho = 2 \ g/cm^3 \quad (assumed)$$

$$k = 0.024 \ cal/cm \ sec \ K^o \quad (assumed)$$

Cadmium sulfide (ref. 3)

$$\rho = 4.58 \ g/cm^3$$

$$k = 0.024 \ cal/cm \ sec \ K^o \ at \ 14^oC$$

Silicon (ref. 2)

$$\rho = 2.33 \ g/cm^3$$

$$k = \begin{cases} 0.308 \ cal/cm \ sec \ K^o \ at \ 20^oC \\ 0.207 \ cal/cm \ sec \ K^o \ at \ 120^oC \end{cases}$$

Although the density and thermal conductivity of the mixture of powdered promethium oxide and powdered cadmium sulfide could not be obtained, an assumption was made. Since the mixture was considered to consist of about 90 percent cadmium sulfide and 10 percent promethium oxide, the density of the powder mixture was taken as approxjmately half that of the cadmium sul- fide. This assumption may not be accurate, but it will not seriously affect the validity of the results since it was observed that the equivalent thermal conductivity had a secondary role in the determination of the maximum thick- ness.

When the thermal conductivity of a material was found as a function of

temperature, the average conductivity between 20°C and 120°C was used in the calculations. The convective heat transfer coefficient, h, assumed for the calculations corresponds to an average coefficient for free convection to air. Since the geometry of the system was unknown, this was the only reasonable choice that could be made.

Since the composition and heat generation rate of each cell are dependent upon the mass of each material per unit of cross-sectional area and the heat generation rate per unit of cross-sectional area, respectively, the following values were assumed.

For Case I:

$$M/A \text{ for strontium titanate} = 0.600 \text{ g/cm}^2$$

$$M/A \text{ for cadmium sulfide} = 1.100 \text{ g/cm}^2$$

$$M/A \text{ for silicon} = 0.130 \text{ g/cm}^2$$

$$q''' L_{13} = 0.108 \text{ watt/cm}^2 \quad (\sim 20 \text{ curies/cm}^2)$$

For Case II:

$$M/A \text{ for mixed powder of promethium oxide and}$$
$$\text{cadmium sulfide} = 0.105 \text{ g/cm}^2$$

$$M/A \text{ for silicon} = 0.130 \text{ g/cm}^2$$

$$q''' L_{12} = 0.010 \text{ watt/cm}^2 \quad (\sim 26 \text{ curies/cm}^2)$$

From these values and the proceeding properties of the materials, the length of each slab, the total cell length, and the equivalent thermal conductivity of the cells can be calculated. The results of these calcuations are:

For Case I:

$$L_1 = 0.162 \text{ cm}$$

$$L_2 = 0.242 \text{ cm}$$

$$L_3 = 0.056 \text{ cm}$$

$$L_{13} = 0.460 \text{ cm}$$

$$q''' = 5.61 \times 10^{-2} \text{ cal/cm}^3 \text{ sec}$$

$$k = 0.0208 \text{ cal/cm sec K}^\circ$$

For Case II:

$$L_1 = 0.0525 \text{ cm}$$

$$L_2 = 0.0558 \text{ cm}$$

$$L_{12} = 0.1083 \text{ cm}$$

$$q''' = 2.2 \times 10^{-2} \text{ cal/cm}^3 \text{ sec}$$

$$k = 0.0450 \text{ cal/cm sec K}^\circ.$$

IV. MAXIMUM PERMISSIBLE STACK

Combination of the above cell properties with the relationships for the composite temperatures yields the maximum tolerable thickness of the stack of individual cells for a maximum allowable internal temperature of 120°C.

For radiant cooling to black space at the ends, the assumption was made that the emissivity was unity $(\epsilon = 1)$. The Stefan-Boltzmann constant (σ) used was $1.372 \times 10^{-2} \text{ cal/cm}^2 \text{ sec K}^\circ$. Then, from equation (8), the maximum permissible stack thickness, L_m, is found to be 0.949 cm in Case I and 2.44 cm in Case II. This corresponds to a stack of 2 units and 22 units respectively. For this mode of heat transfer, it was found that the equivalent thermal conductivity of the material played a secondary role in the determination of L_m. The maximum thickness was almost entirely determined by the radiation rate.

For convective cooling, the ambient temperature t_a, was taken as 20°C, and the convective heat transfer coefficient h, was assumed to be 2.71×10^{-4} cal/cm^2 sec $^\circ$K. Using equation (6), the maximum permissible thickness, L_m, was found to be 1.1 cm in Case I, and 2.0 cm in Case II. This corresponds to a stack of 2 cells and 18 cells, respectively. In this mode of heat transfer, the equivalent thermal conductivity was a significant factor.

V. DISCUSSION

The results of this analysis indicate that, regardless of the type of cooling employed, there is a maximum thickness and thus a maximum power output density per unit area. For Case I, the single crystal phosphor with Sr^{90} excitation where the maximum permissible thickness is about two cells, the power in produced by the radioisotope is about 215 mw/cm^2. Assuming an overall efficiency of the conversion device to be 5 percent, yields a power output of 10 mw/cm^2.

For Case II, the mixture of powdered Pm_2O_3 and phosphor, the maximum permissible thickness is about 20 cells. Here, the power in is about 200 mw/cm^2 and again about 10 mw/cm^2 out.

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