CESIUM-137 POWER PROGRAM

QUARTERLY REPORT III

PREPARED FOR THE DIVISION OF ISOTOPE DEVELOPMENT
U. S. ATOMIC ENERGY COMMISSION

ROYAL RESEARCH CORPORATION
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THE CESIUM-137 POWER PROGRAM

This work performed for
The Division of Isotopes Development
United States Atomic Energy Commission
Contract No. AT(04-3)-366

3rd QUARTERLY REPORT

by

ROYAL RESEARCH CORPORATION
a subsidiary of ROYAL INDUSTRIES, INC.
Hayward, California

Hampden O. Banks, Jr.
Project Director

October 31, 1961
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FOREWORD

This report has been prepared by Royal Research Corporation, Hayward, California, as required under U. S. Atomic Energy Commission Contract AT(04-3)-366. It is the third quarterly report for the Cesium-137 Power Program and covers the period June 22 to September 22, 1961.
SUMMARY

The third quarter of the Cesium-137 Power Program was devoted to completion of a definite fuel form and the investigation of those parameters which regulate a reproducible product material. Such qualities as color of the polyglass were analyzed insofar as it identified contaminants, incomplete reaction and material density. The presence of minute surface bubbles were found to be attributable to a trace of water in the starting constituents. It was discovered that preheating minimized this phenomenon. Final removal of all remaining bubbles could then be accomplished by mechanical means.

Fuel quality is predicated upon safety. Toward this end, a series of long-term solubility measurements was continued. Aqueous media, including natural sea water and distilled water, were used, with all physical and chemical parameters maintained at a constant. Initial dissolution did occur because some surface cesium was found to be in an unreacted state. Following this dissolution, the rate decreased markedly. After 100 days immersion, the total dissolved cesium was in the low parts-per-million region, although this is purely a function of testing vessel volume. Weekly increase is in the parts-per-billion range. For this work, tracer cesium was added to stable polyglass since the solubility is well below conventional measurement levels. Fuel quality, as a function of radiation dosage, was investigated and found to have a stabilizing effect that retards dissolution. This may be due to co-polymerization of the complex borosilicates.

The amount of Cesium-137 required to produce 120 thermal watts was recalculated, employing the more recently accepted half-life for Cesium-137, of 26.7 years. Individual energy contributions from primary beta decay was redefined, based upon a more rigorous treatment of the method of calculation. As a result of these revisions in available energy, a total of 25,500 curies will be needed.

A second and more accurate shielding experiment was performed, using a 10-curie Cesium-137 source containing 1.5 per cent of Cesium-134. This sample was prepared by the Oak Ridge National Laboratory, and was made representative of the 25,500-curie batch accumulated for fueling the 5-watt generator. Half-value layers for lead agreed with those values obtained by ORNL researchers. Both half and tenth-value layers were determined for tungsten, and showed that over 3 in. of that material would be needed to reduce the radiation to a level of 100 mr/hr at 4 ft. from the center of the generator. Since this would result in an unacceptable over-all

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generator size, depleted uranium was selected. Uranium has twice the attenuation qualities as lead, therefore, not more than 2 in. would be needed. Precise shielding experiments are in progress.

The Westinghouse effort on power conversion has yielded a package which is approximately half-scale, with an output of 2.75 watts at 0.99 volts. Thermocouple cladding was made by means of a poured insulator; this has resulted in a high heat loss through the package and a required thermal wattage of 140 watts to provide the required 5 watts electrical.

Royal Research is continuing its backup program and has instituted life tests on a typical Royal Research configuration. This includes printed circuitry, brazed "n" and "p" elements and a suitable ceramic cladding. Elements are then insulated with felted fiberglass. The outer container will be a right circular bellows type of wall with end plates of a highly (thermal) conductive alloy. Coupler resistance has been reduced to 0.027 ohms.

The fuel will be doubly encapsulated in two separate containers, each holding three elements. This was adopted to permit use of the ORNL remote welding apparatus. Final dimensioning of the inner fuel can and cladding have been completed. Two full-scale 7075-T6 aluminum generator shells have been ordered. These will be used for pressure testing and air dropping to determine integrity.

Reduced scale models of the generator shell were tested at the Naval Electronics Laboratory for resistance to deep-sea pressure. Excellent results were obtained, indicating that as little as 1.40-in. wall thickness would afford ample protection against a 6,000-fathom pressure. At this same installation, underwater exposure tests were initiated to determine a suitable coating agent to protect the aluminum from sea water attack. Four samples, each of different cladding, were immersed in running sea water, to be left for 90 days. Interim inspection will be made by means of closed circuit television provided at the offshore site.

Two acceptable dc-dc converters have been defined and are being assembled by Electrosystems Co. One is a solid state device of greater than 85 per cent efficiency but with a longevity assured for approximately 10,000 hours. This device is not position sensitive. The second will operate at from an estimated 85 to 92 per cent efficiency, has a component longevity well in excess of 30,000 hours and must be operated within $\pm$ 10 deg. of vertical. Since both devices represent a breakthrough in longevity, no decision will be made at this time as to which will be used in the generator package.

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The basic information which has evolved from the various tasks of this program has made possible the dimensioning of the main components comprising the 5-watt generator. From this information, a full-scale model was designed and fabricated. Although inoperable, this model was sufficiently detailed to permit inspection of all major parts, as well as indicate their relative size and position in relation to their function. The model consisted of three sections: the generator body, the dc-dc converter cavity and the lid. The model was delivered to the customer in a carrying case, together with color slides and descriptive literature.

At the convenience of the customer, the site selected for fuel fabrication and cladding has been changed from the Vallecitos Atomic Laboratory, California, to Oak Ridge National Laboratory. Necessary hot cell equipment required to fabricate the fuel elements is under design by the contractor and will be delivered, subject to approval by the fabricating facility.
I. SUBTASK 1.2 - CESIUM COMPOUND CONVERSION

Research into the methodology of converting starting cesium chloride (in a dry state) to aqueous cesium carbonate was completed during the third quarter of the program. Evaluation of contaminants in a typical sample of starting Cesium-137 fuel material has shown that no appreciable dissimilar elements are present as radioisotopes. For this reason the preliminary scrubbing step, which had been assumed necessary, may be omitted from the process.

Oak Ridge National Laboratory has provided the project with a 10-curie sample, representative of the 25,500-curie batch being reserved for fueling the operational generator. A small aliquot of this material was extracted, diluted and counted, on both the end window Geiger-Mueller counter and the liquid-well counter. No short-lived constituents were identified other than the Cesium-134 component. A sample was then assayed with the 256-channel pulse-height analyzer. No gamma peaks attributable to radioactive contaminants were seen above the photoelectric region. This sample contained a reported 1.5 per cent Cesium-134.

Because of the relative purity of the above sample, the anticipated use of ammonium phospho-molybdate (in a column) for decontaminating the fuel material, has been obviated. In addition to reducing the number of steps required for the overall cesium polyglass fabrication, the processing time also has been reduced. Instead of the fuel entering the compound conversion column diluted to 1.68 liters and then coming off as the carbonate with a volume of 13.4 liters, only 200 ml. of chloride are introduced, and 570 ml. are collected as the carbonate. This will result in smaller vessels and a tremendous saving of time, when evaporating the carbonate solution down to the desired 200 ml. prior to mixing it with the stable borosilicates. Perpass time through the compound conversion column has been reduced, as well. However, information gained as a result of considering the presence of a contaminant is useful for future fuel processing, particularly if the charge of a future generator is found to be radiocontaminated.

Changes in the physical dimensioning of the generator fuel array, as well as a reduction of Cesium-137 needed to provide the necessary 120 thermal watts, has resulted in a change in cesium processing volumes, equipment and running time. Because of restrictive dimensions encountered in the cladding welder equipment, a single, doubly-encapsulated Hastelloy C configuration cannot be used. Instead, two smaller doubly-encapsulated cans must be employed. In so doing, an even number of equally-filled platinum cups must be fabricated, instead of seven, as originally planned. For this reason, the number of cups has been reduced to six, and this makes mandatory a greater Cesium-137 content per cup. This, in turn, has required a reduction in the
number of incremental process columns, although resulting in increased total volume. Therefore, each compound conversion column will contain 1100 ml. of ion exchange resin. The column diameter will be 4.84 cm. and contain a resin height of 59.6 cm. This material represents a 10 per cent excess amount of resin (as a safety factor) and is capable of converting 169 g. of cesium chloride. Tests have shown that such a column has an efficiency of from 97 to 99 per cent, with a column retention of <0.5 per cent after washing. To process six fuel increments, six such columns would be required. This approach is superior to repeated recharging of a single ion exchange column in that it saves time, reduces the possibility of channeling and guards against the pass-through of unreacted cesium chloride. The columns will be medium-walled pyrex with a coarse frit forming the bottom of each. A 3-in.-long, tapered stopcock will be incorporated in the column, positioned below the frit. Interior bore on the discharge end will be 4 mm. in diameter.

Dowex 1X8 anion-exchange resin has been replaced by the more uniform and more efficient type -- AG 1X8. The latter is simply a more refined form of Dowex 1 which has an increased exchange capacity of 15 per cent. The AG 1X8 has a reported value of 1.4 meq/ml. of moist resin. This material is provided, as the carbonate, by the BioRad Laboratories, Richmond, California.

The procedure which has been found most suitable for fuel processing is as follows. The entire charge for one increment (169 g. cesium chloride) is dissolved in 200 ml. of water. Occasionally, a gentle warming has been found necessary. This then, is fed into the conversion column from the dissolver. Because of dimensional characteristics of the column described earlier in this report, the volume would result in an 11 cm. head above the resin. Wash water collected in the dissolver is not added at this time. Instead, the original solution is passed through the column at a rate of 8.5 ml. per minute, until the column head has been reduced to 1 cm. above the resin. The wash water is then (introduced at the top of the column) added to maintain a 1-5 cm. head. Fig. 1 shows a typical elution curve obtained from a full scale run. Tracer Cesium-137 was employed as the elution indicator. A total of 570 ml. of solution was passed through -- 200 ml. of original cesium chloride solution and 370 ml. of wash water. After elution, the resin was analyzed for residual cesium. Sensitive counting showed that only 0.01 per cent of the cesium was retained. This would amount to less than 15 mg. of radiocesium lost during a hot run, a negligible number.
169 gr. CsCl in 200 ml. solution passed through column - \( \text{Cs}_2\text{CO}_3 \) eluted

**Total Elapsed Elution Time in Minutes**

**Fig. 1 - Compound Conversion with AG 1X8 Resin**
As indicated by the curve in Fig. 1, the break-point is clean and well defined. The rise time is as narrow as could be expected from any resin. It is flattened through the use of a concentrated cesium chloride solution. Flattening is necessary so that the steep drop-off can be more readily anticipated and better control exercised. While a more concentrated starting solution would provide a smaller product volume (classical data suggests a much higher solubility in water), it has been our experience that such is not possible.

Control of the endpoint during hot cell operations may be achieved by means of a shielded counter tube having a high saturation point. This could be positioned perpendicular to the column and would identify the cut-off in radiocesium passage through the resin.

The carbonate form of AG 1X8, as supplied by the BioRad Laboratory, will be made available to Oak Ridge National Laboratory for use in their processing of the cesium fuel.
II. SUBTASK 1.3 - CESIUM FUEL FORM INVESTIGATION

This subtask is concerned not only with cesium polyglass as a product material of certain chemical and physical characteristics, but with heat producing capabilities of the final product as well. A suitable hot cell scale-up of laboratory techniques involved in the manufacture of cesium polyglass, has also been included. Evolution of specific pieces of apparatus which become identified for fabrication are discussed in Task IV.

Originally, 28,000 curies were identified as being needed to provide the 120 thermal watts of energy for the cesium generator. As described in the first quarterly progress report, this was predicated upon calculations which assumed the useful beta energy as 1/3 the total energy. A more rigorous examination has recently been made of beta energies available from Cesium-137 decay chain, resulting in certain revisions. It was shown that the 514 Kev. beta has a useful energy of 183 Kev. and the 1,176 Kev. beta has a useful energy of 283 Kev., based upon relative abundance of 92 and 8 per cent, respectively. These values are derived in Appendix A. As before, no contribution to thermal energy has been derived for the Cesium-134 addition to the calculated 137-curie strength. In resolving the generator fuel requirement, the more recent value of 26.7 yrs. for Cesium-137 half-life was used. As a consequence, the customer has been advised that 25,500 curies of radioisotope will be sufficient to provide the required 120 thermal watts.

The reduction in fuel quantity has resulted in a dimensional revision of the fuel core. Further changes were dictated by the Oak Ridge National Laboratory requirement that the double encapsulants fit in the existing hot cell welding enclosure. Therefore, the decision was made to fabricate the 25,500 curies in the form of six increments, instead of the previously reported seven. While the dimensions of each platinum fuel cup will be reduced in height, the 2.50-in.-dia. has been retained. This is being done partly to conform to external design commitments already made, to optimize self-absorption and thereby reduce external shielding, and lastly, to keep the linear heat path through the fuel cans to a minimum. If a narrower fuel can and shield were employed, the previously determined thermal insulability would be invalidated, it being more difficult to force heat through a long, narrow cylinder. By reducing the fuel load and retaining the original diameter, essentially we have foreshortened the fuel-configuration height and the shield height, thus resulting in improvement of thermal economy.

The following characteristics, based upon final revision of fuel increment dimensions, are descriptive of the fuel package:

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TABLE I

Physical Characteristics of the Revised Fuel Configuration

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<th>Fired Product</th>
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<tr>
<td>Weight (grams)</td>
<td>1,687</td>
</tr>
<tr>
<td>1,874</td>
<td>1,687</td>
</tr>
<tr>
<td>Density (grams per cc.)</td>
<td>3,00 ± .02</td>
</tr>
<tr>
<td>1,53 ± .02</td>
<td>3,00 ± .02</td>
</tr>
<tr>
<td>Volume (cubic centimeters)</td>
<td>563</td>
</tr>
<tr>
<td>1,230</td>
<td>563</td>
</tr>
<tr>
<td>Activity (curies)</td>
<td>25,500</td>
</tr>
<tr>
<td>26,100</td>
<td>25,500</td>
</tr>
<tr>
<td>Diameter (centimeters)</td>
<td>6.35</td>
</tr>
<tr>
<td>Increments</td>
<td>6</td>
</tr>
<tr>
<td>Increment Height (centimeters)</td>
<td>3.44</td>
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</tbody>
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These changes were brought about, in part, by a revision upward (by Oak Ridge National Laboratory) in specific activity of the starting fuel material. Originally, they had estimated a specific activity of 25 curies per gram. However, the strength of the 10-curie representative source, as provided for the shielding experiment, showed an increase of 5.62 per cent, or 26.7 curies/g. Further consideration of specific activity of the fuel is unnecessary; it will be assumed that a mechanical mixing-drying device will not affect product density, or specific activity. Physical loss of fuel will be kept to a 2 per cent maximum.

Repeated use of the false fuel-cup wall, as provided by the dental porcelain, has demonstrated its reliability. It is strong enough to insure against accidental breakage from handling, yet brittle enough to permit later removal with suitable tools. Its intrinsic non-wetting qualities insure against fuel-material creep upward on the cup wall. Samples of a fired false wall, starting powder and complete fabricating instructions have been provided ORNL representatives for their evaluation. The alternative to this method would be a full-height platinum cup. However, if the latter method is used, several problems must be overcome. First of all, molten cesium polyglass has a tendency to "wet" the inner wall and then creep up it. Furthermore, removal of the excess platinum can be achieved only through the use of a lateral saw, if rim irregularities are to be avoided.
The presence of tiny air bubbles in the upper 1/8 in. of fuel increment surface has been investigated, in an effort to explain their presence and seek a method of removing them, if practical. Bubbling, in glass, has been a major problem associated with glass manufacture. These bubbles are caused by (1) water vapor, (2) decomposition of impurities, and (3) internal volatilization. There has been no convenient method found, to date, for the rapid removal of such bubbles, or mixing glass without this side effect occurring. In the case of cesium polyglass, the second cause can be omitted since pure constituents are used. However, the extremely high hygroscopicity of cesium carbonate makes complete water removal virtually impossible. When this was attempted, through evacuated heating furnaces, some cesium carbonate decomposed to form cesium oxide, which readily volatilizes unless it can be made to react with a borosilicate. Internal volatilization of cesium oxide does occur but because of its high reactivity with the borosilicates and an excess of silica present in the charge, this would be of a momentary nature.

Extreme care exercised in the post-drying stage (400 deg. F.) does evaporate almost all of the moisture. When fusion occurs, moisture bubbles form and then migrate upward through the viscous matrix. Surface tension inhibits total escape of the lowermost ones. Therefore, after the proper fusion period, a 1/8-in.-deep layer usually contains tiny bubbles.

Surface volatilization of cesium oxide has been minimized to where it accounts for a less than 2 per cent loss. However, it can not be omitted completely. Earlier researchers, who endeavored to entrain cesium in a vitreous matrix, were unsuccessful because they sought thin-film sources. No fused head was present to prevent the high loss of cesium, as is true with cesium polyglass increments.

As explained in preceding subtasks, bubbles have been present in samples which were subjected to long-term solubility determinations, the irradiated sample test and all fuel-product evaluation tests. Chemical and physical properties associated with cesium polyglass tolerate the presence of bubbles or vacuoles, which are only 1 mil in diameter and approximately 10 mils apart.
III. SUBTASK 1.4 - LONG TERM SOLUBILITY TESTING

A precise determination of the solubility of cesium polyglass in various aqueous media is perhaps the most important single test being conducted on the program. Thus, the destined subsea use of the five-watt generator, makes contact with sea water far more probable than any other credible incident. Corrosion of the outer metallic components could proceed independently of a physical accident which might rupture the generator. Therefore, in addition to the evolution of a structural array to protect the fuel core from impact, fire or vandalism, a chemical compound was sought which would be inert to natural aqueous media. Cesium polyglass exhibited acceptable resistance to both distilled and natural sea water during early testing. As the immersion time extended to months, this fact was substantially confirmed. Testing will continue throughout the duration of the contract, in order to make possible the most accurate extrapolation of ultimate dissolving time.

Although solubility is reported as parts per million of aqueous medium, this terminology does not reflect the unusually low solubility of cesium polyglass. Physical restrictions of laboratory space prevented controlled testing in larger vessels; and the use of granular samples prevented measurement of surface area exposed to the testing medium. This is the true parameter of measurement, since dissolution is a surface reaction. Dissolution has proceeded so slowly, that the 13 liter vessel used actually represented an infinite volume to the polyglass specimens. Although spiked with a relatively high specific activity of Cesium-137, the amount found in interim aliquots has represented the lowest limit of counting efficiency, and results are most probably an order of magnitude higher than they should be.

A distilled water experiment consisting of two independent 13-liter vessels, each containing specimens of cesium polyglass, was run for a period of 100 days. During this time, the volume was kept constant, temperature was maintained at 50 deg. C. and the water constantly agitated. Samples were drawn every 5 days and subjected, in duplicate, to end-window beta counting of mounted evaporated samples, and scintillation-crystal gamma counting of liquid samples. Results of this experiment are concluded in Table 2. Here, uniformity of increase obviates random-type results. During the last 50 days of testing an average increase of less than 2 ppm. was observed for both runs. A plot of these results, which includes earlier testing for comparison, may be seen in Fig. 2. Although physical testing has been terminated, an acceptable extrapolation is readily possible, suggesting a dissolution-rate of 0.095 g. of cesium polyglass per year. Naturally, it is understood that dissolution studies conducted in distilled water have only academic value.

RRC-0102
### TABLE 2

**Solubility Data Sheet**

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<tr>
<th>Sample</th>
<th>Weight</th>
<th>Elapsed Time, Days</th>
<th>Avg. ppm Cs in Solvent (13 L.)</th>
<th>Solvent</th>
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RRC-0102
### TABLE 2 (Continued)

**Solubility Data Sheet**

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<th>Weight</th>
<th>Elapsed Time, Days</th>
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Fig. 2 - Solubility of Cesium Polyglass in Aqueous Media

Days in Solution

Parts Per Million

Sample IV
Sample III
Sample II
Sample I
Extrapolated

Solvents
Distilled Water - I and II
Sea Water - III and IV

0 10 20 30 40 50 60 70 80 90 100 110 120

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At the conclusion of testing, the 13 liter contents of both fresh water experiments were evaporated down to 100 ml. with gentle warming. A 3 ml. aliquot from each was taken and evaporated to dryness for counting purposes. Results indicated that total cesium dissolved during the testing period was only 67 per cent of the amount reported at the end of 100 days. From this then, it may be assumed that all interim values reported were also in error by a like amount.

Of interest is the method of interim aliquoting employed on these and all other solubility measurement samples. The procedure is as follows: Every 5 days, a 1-liter portion of the original solution is drawn and evaporated down to 100 ml. From this, 2-ml. aliquots are taken for counting purposes. At the end of each analysis, all liquid is collected, re-diluted to 1 liter and returned to the solubility test vessel. In this manner, no accumulated cesium is lost from the testing medium.

The second series of solubility studies under evaluation is natural sea water testing. This is perhaps the most important test, since the generator's initial application requires deep sea immersion. Therefore, attack by sea water is a more probable hazard. The third quarter period saw a continuation of tests throughout the 120-day period. As in the case of the distilled water phase, interim samples were drawn every 5 days. A tabulation of these data may be seen in Table 2. Confirmation between the beta and gamma counting techniques is good, and the gradual increase indicated is uniform. The net increase in dissolved cesium for the past 50 days represents only 1 ppm. From the 15-g. sample, this is only 95 mg. per year of dissolved cesium polyglass. Since cesium represents only 50 per cent of it by weight, over three centuries would be required to dissolve the 15-g. specimens.

A graphic interpretation of dissolution, as a function of time, may be seen in Fig. 2. Weekly buildup in the 13 liter testing solution is of the order of parts-per-billion, as in the case of the distilled-water experiment. It is interesting to note that the solubility increase in a sea water medium is only 35 per cent greater than in distilled water.

Upon conclusion of distilled water testing, the equipment was thoroughly cleaned, wipe-tested for residual radioactivity and prepared for the fresh water tests. Although present considerations do not include fresh-water bodies as locations for isotopic powered generators, such tests still are in order since there is a possibility of an accidental drop in a lake or river. Such impact could rupture the external case under special circumstances, as when near-surface reefs are in the impact area. Therefore, running long term solubility determinations of cesium polyglass in this aqueous medium would be most useful in the hazards evaluation. Should future application extend to generator placement in fresh water, these studies would become ever more valuable.

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Potable water was obtained, with mineral content, pH and organic count approximately that of large fresh-water bodies, rather than that which is typical of any one local area. It was found during previous testing that when polyglass is fabricated, there is a thin surface film of unreacted cesium remaining, or deposited upon the product after firing. This cesium dissolves off the specimen quite rapidly, and in doing so, suggests a greater than actual dissolution rate. Examination of the distilled and sea water solubility runs illustrates this fact. Since a superficial cleaning action is feasible in the fuel fabricating cell, this step was introduced prior to immersion.

After 5 days of immersion, no activity was found in the aliquot samples drawn from the fresh-water testing vessels. This, is in comparison to the 3 ppm, reported at the end of a similar period for both previous tests. Since the rate of dissolution in fresh water should be between distilled and sea water, the absence of early dissolving action must be attributed to the added cleaning step. This should result in a curve whose general shape resembles that of the sea and distilled water tests, but which will level off below either of their maxima.

The foregoing solubility tests were conducted for the purpose of determining the absolute dissolution rate of cesium polyglass in various aqueous media. Radioactivity was introduced, in order to measure cesium in amounts not detectable by conventional methods. However, the six fuel increments will contain a total of 25,000 curies of radiocesium. The intense radiation field to which the fuel would be constantly subjected could conceivably change the rate of dissolution, after sufficient dosage had been received. In order to resolve this question, several samples of polyglass were prepared. They contained sufficient Cesium-137 to permit detection of even a trace of dissolution. These samples were then subjected to a preliminary immersion in distilled water (at room temperature) for a period of 15 days, as indicated in Fig. 3. Dissolution progressed to 4.5 and 3.7 ppm, respectively, for the two samples. The former weighed 2.1 g. and the latter, 1.8 g., size being kept to a minimum in order to accommodate the samples in the isotope cans provided for irradiation.

After establishing non-irradiated dissolution rate, the samples were subjected to a gamma flux of $1.7 \times 10^7$ r/hr for a period of 10 days. This amounted to a massive dose of $4 \times 10^9$ r received by each specimen. Post-irradiation examination showed perceptible darkening of the polyglass, as illustrated in Fig. 4. No powdering was observed on the surface, and no fracturing had taken place during irradiation. When subjected to sudden jarring, no disintegration occurred. In general, no physical change was detected, other than an optical one.
Fig. 3 - Solubility of Irradiated Cesium Polyglass

Rate of Dissolution
Before Irradiation

Rate of Dissolution
After Irradiation

Parts Per Million

Immersion Time (Days)
Fig. 4 - Polyglass Samples Before and After Irradiation
(Sample size was limited by small isotope can shown in photo above)
The fuel core will be effective only if it dumps internal heat promptly. The color transformation is not detrimental to this thermal transfer since the polyglass will transmit heat to the platinum cup by conduction. The heat referred to in this instance, is primarily beta-particle energy conversion heat, as gamma-ray energy undergoes conversion in the radiation shield.

When the irradiated specimens were immersed in containers of new distilled water, the dissolution rate was appreciably lower, as shown in Fig. 3. Since both samples had been given a preliminary rinsing before initial immersion studies began, the lower rate observed after irradiation must be attributed to this treatment. One possible explanation is that radiation-induced cross-polymerization occurred in the glass structure, further tying up the cesium present and linking it to more durable bonds. A second consideration is that a tempering action occurred, making the polyglass structurally harder, so that it better resisted the erosive action of the swirling water. This last effect would add to true dissolution by causing a temporary suspension of minute particles, whose high surface-to-volume would be more vulnerable to attack.

In order to more clearly define the chemical properties of the new cesium compound which has evolved as a result of the program, a series of spiked polyglass samples were subjected to the action of various common reagents. Table 3 shows the effect of 0.10N solutions of nitric acid, hydrochloric acid, sulfuric acid and sodium hydroxide. It is interesting to note that nitric acid proved to be the most corrosive, while sodium hydroxide had the least effect. This may, in part, explain the fact that sea water did not exhibit a particular affinity for cesium as might be expected, since sea water also has a slight alkalinity.

**TABLE 3**
In reviewing the relative action of distilled, fresh and natural sea water upon cesium complex borosilicate, it becomes increasingly obvious that the Debye-Huckel Theory of "uncommon ion effect" does not apply. If cesium loss by corrosive action of an aqueous medium were the only factor involved, then the loss would be greater by many orders of magnitude. The extremely low solubility of cesium polyglass in any aqueous medium (with or without salts dissolved in it) suggests that the silica and silicates may be the determining factor. Dissolution, then, would be a two-stage process. First the silica linkage must be severed, leaving cesium less protected than before. Next, the cesium borosilicate may be reduced to a simple peninsular shape. This, in turn may be fractured by the swirling water action, suspending tiny fragments of cesium-containing material in the testing medium. Orderly increases in cesium content preclude a gross random effect. Since so little is known of the nature of glasses, an immediate solution and explanation is not possible. However, reproducible results substantiate the claim that the action is predictable and the experimentally obtained dissolution rate is reliable.

These tests have all been conducted with samples of polyglass containing surface bubbles. These bubbles are of the order of magnitude of 1 mil, in diameter. They occur during processing of the polyglass and in no way influence solubility of the product. While they can be removed, either by mechanical stirring, or extended time in a molten state, the former involves a difficult manipulator step, while the latter will cause a slight increase in cesium loss. Since excellent insolubility has been demonstrated with samples containing the bubbles, and since they are too small and too localized to affect heat flow, they cannot be considered injurious to the fuel. No measurable density change has ever been detected between samples taken from the pellet surface (containing the bubbles) and those taken from the middle or base of the pellet. Therefore, it is strongly felt that they need not be removed.

A corrosion problem which more closely relates to generator longevity rather than fuel containment, is the resistivity of the outer shell. For the sake of compactness and saving of weight, 7075 T6 aluminum has been selected for the pressure resistant outer envelope. Since this material is subject to attack by sea water, a means was sought to provide a thin film barrier between the aluminum and the ocean. Several test blocks of alloy were coated with different materials. Each was subjected to a pressure depth of 3,600 fathoms (9,500 psi) in sea water by means of a compression chamber. Phenoline-300 failed, in that water seeped under the film, causing it to lift away from the aluminum. The three remaining test blocks which successfully withstood the pressure test, were then subjected to refluxing boiling sea water for a period of 24 hrs. Having withstood this, they were then taken down
to the Naval Electronics Laboratory, San Diego, California, and there put
into running sea water at a recommended depth of 15 ft. for 90 days. Interim
reports from the offshore station indicate, that after 60 days numerous col­
onies of barnacles and other marine life have collected on the surfaces of
each one. This prevents underwater television from examining the surfaces
for failure. At the end of the 90 day period they will be retrieved and examined.
IV. SUBTASK 2.1 - HEAT TRANSFER ANALYSIS

Heat transfer experiments performed earlier in the program and reported in the second quarterly report, have identified the optimal thermal insulants for a 5-watt device. Based upon the then defined fuel-can dimensions, a shield height of 9.5 in. was used. Results indicated that, in addition to the double reflector configuration, an additional 3/4 in. of compacted AA Felted Fiberglass would be necessary. This combination of insulators would waste only 21 thermal watts in maintaining an upper shield temperature of 500 deg. C. Such loss represents the passage of 0.108 thermal watts per square inch. However, it must be remembered that the experimental configuration consisted of an insulating envelope completely surrounding the shield. Therefore, the above number actually represents heat loss per unit area of insulation. Additional heat was also lost through the thermocouple shield and leads, as well as the electrical heater leads.

Subsequent increase in fuel-can dimensions, necessitated by restrictions in the ORNL remote welding fixture, have caused an increase in shield dimensions. The final radiation shield is a right cylinder, 7 in. in diameter by 13 in. in height. As in the experiment, complete thermal insulation must cover the sides and base. The upper section will be in intimate contact with the thermoelectric convertor packages. This will have an active area whose thermocouples will cover 30% of the exposed area. The remaining 70% must be thermally insulated. The combined exposed surfaces represent a sizable increase over the original consideration. If only 3/4 in. of insulation were employed, the heat loss would be 46.6 thermal watts. It must be remembered that the overall generator diameter cannot exceed 13 in.; therefore, unlimited insulation cannot be used. However, by increasing the fiberglass thickness to 1.25 in., it is possible to reduce loss to 25.6 watts, and still maintain an acceptable generator outside diameter. An additional 3.8 thermal watts are saved by subtracting the lead-wire loss present in the experimental apparatus. This reduces total loss to 21.8 watts, which is approximately the same value as reported earlier.

There are three principle avenues through which internally-produced heat may travel. The first, lateral heat loss, had been discussed in the above. The second is through the thermoelectric assembly. Here, heat loss is a function of couple efficiency and hot and cold junction temperatures. The last is through the metal envelope which surrounds the power conversion system.
Power, through thermoelectric conversion, can only become accepted on a widespread basis if cost is optimized. Since prototype models must be hand-fitted, the unit cost is understandably high. This practice must, however, be phased out of any volume production. One of the most time consuming phases of generator construction is assembly of the conversion system. In an attempt to anticipate production requirements, a pre-packaged conversion system has been pursued. Suitably high thermally conductive metals were found with little difficulty. Selection of the vertical wall material for the package presented a more complex problem. Such parameters as flexibility, nonporosity, low thermal conductivity and compatibility with metal bonding made most materials unacceptable. Stainless steel was selected, since a double, 5-mil wall best met all requirements. Stainless, with an ambient thermal conductivity of 15.65 Btu/hr/ft²/deg. F/ft., drops sharply with a rise in temperature. At operating conditions, it is actually more resistant to thermal flow than electrical and mechanical ceramics, whose ambient conductivity is 2 Btu/hr/ft²/deg. F/ft. This is true, partly because a 1/4-in. thick wall would be required to provide structural strength and prevent diffusion inward during vacuum pumpdown. A 1/4-in. thick ceramic wall would have no flexibility, would bond to metal only with extreme difficulty and waste more heat through its cross section than stainless steel.

An examination of Fig. 5 shows the cross section of the power conversion package. The bellows shape of the vertical outermost wall permits intimacy of contact under reduced pressure. This insures good heat flow by minimizing heat backup immediately in front of the hot junction plate and external to the cold junction shoe. Since heat loss through the insulation located between couples has already been considered, all that remains to be done is to identify the heat loss through the stainless-steel wall.

A calculation of heat loss through the stainless steel walls may be made in the following manner:

\[
Q = (T_h - T_c) \cdot (u) \cdot (K)
\]

Where:
- \(K\) (Cross section exposed to heat) = 0.208 in² or 0.0014 ft²
- \(T_h\) (hot junction) = 900 F.
- \(T_c\) (cold junction) = 145 F.
- \(\Delta T\) (temperature drop) = 755 F.
- \(u\) (thermal conductivity) = 15.65 Btu/hr/ft²/deg. F/ft.

The heat loss \(Q\) (heat) may then be determined:

\[
Q = (900 - 145) \cdot (15.65) \cdot (K)
\]

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Fig. 5 - Stainless-Steel Bellows Envelope for Thermocouple Package
\[ Q = 16.54 \text{ Btu/hr.} \]

(or) \[ Q = 4.8 \text{ thermal watts/linear foot} \]

The length of the pleated vertical wall, when unfolded, is 5 inches. Therefore, this length factored into the above value (4.8 thermal watts) increases it to:

\[ Q = 2.4 \times 4.8 = 11.5 \text{ thermal watts loss} \]

If we add this to the heat loss sustained through the insulation surrounding the shield and between couples, there will be a total conductive heat loss of 33.3 thermal watts experienced by the present 5-watt generator design.

It is strongly believed that the above loss is pessimistic, since several contributory factors were purposely omitted. Available heat was predicated upon only that which is derived from Cesium-137. Analysis of the Cesium-134 contribution indicates that an additional 4.7 thermal watts will be created by this isotope. While relatively shorter half-life (2.4 yrs.) suggests a rapidly decreasing contribution, there will be heat available, regardless of the time factor since the generator performance is predicated upon a 3 to 5 yr. operational requirement.

The experiment that was run to determine lateral heat loss, employed twin polished reflectors as well as a gold-plated shield surface. After termination of the experiment, the apparatus was dismantled and it was seen that the gold had suffered from exposure to air, resulting in loss of most of its reflective surface. This occurred during initial running when a leak developed and vacuum was lost overnight. For this reason, no change in heat loss was observed. Improved methods of sealing in assembly of the actual generator, will obviate such a possibility and make for improved insulability.

Tungsten was originally selected as the radiation shield, heat-sink material. This was predicated upon the feasibility of obtaining a massive piece, which would have a uniform density, and be of sufficient size for fabricating the shield. Subsequent testing of various samples of material indicated that 86% of theoretical density is the best that is presently available. Shielding experiments (discussed later in this report) indicated that over 3 in. of cross section would be required to attenuate the radiation down to an acceptably safe value. This, in turn, would increase the outermost generator dimensions beyond 13 in. dia. For this reason tungsten is totally unacceptable.
Depleted uranium is a high-density metal whose attenuation is almost twice that of lead and over 1.5 times that of tungsten. A rigorous investigation has shown that, with uranium, the original dimensions apply. However, in making the change, it was necessary to re-evaluate the heat transfer properties of a uranium shield. Sintered tungsten has a high (89 Btu/hr/ft²/deg. F/ft) thermal conductivity. At 49 deg. F., however, this value decreases to 75 at operational temperature as indicated in Fig. 6. Uranium, Fig. 7, exhibits the opposite effect. At ambient temperature, its conductivity is 15 Btu/hr/ft²/deg. F/ft whereas, at operational temperature, (900 F.) this value is 20. Since the operational temperature reflects a thermal conductivity lower than tungsten values, a rigorous treatment was given the problem of "hot-spot" formation throughout the uranium shield. As may be seen in Appendix B, solution of the problem is intricate. Certain conservative assumptions were made in this calculation. They tend to make the results reflect a temperature differential considerably higher than actual. Despite this, the greatest difference found was only 140 degrees F. between top and bottom of the shield. Because of these results employment of a depleted uranium shield (in cast form) is not only a dimensionally economical means of shielding but completely safe from the hazards aspect as well. It must be remembered that the present design criterion inverts the shield, so that the threaded cap is at the base, rather than at the top of the heat flow pattern. By so doing, the air gap between threads does not impede heat transfer.

Additional protection will be given the uranium by plating it with a heavy layer of nickel on all exposed surfaces. By so doing, the heat transfer is actually improved, while obviating the possibility of pyrophoric consumption in the event of an accident.
Fig. 6 - Thermal Conductivity of Tungsten (Sintered) As a Function of Temperature.*

Fig. 7 - Thermal Conductivity of Depleted Uranium (Theoretical Density) As a Function of Temperature.

*Data Supplied by Stauffer Metals Co.

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V. SUBTASK 2.2 - POWER CONVERSION ANALYSIS

As indicated in earlier reports, a dual effort on the power conversion system for the cesium generator is being expended by both Royal Research Corporation and the Westinghouse Electric Corporation. That system which most closely meets contractual requirements as to element cladding, couple cladding and dimensional compatibility, while optimizing the available heat, will be used. Data resulting from RRC and Westinghouse efforts are presented individually in this subtask, for the purpose of delineation and comparison.

*A. WESTINGHOUSE EFFORT

Westinghouse design and fabrication of the thermoelectric couples for the Cesium-137 Power Generator were described in the second Quarterly Report. Since then, thermoelectric couples have been assembled and enclosed in a module for endurance evaluation. It is our feeling that sufficient data points will be obtained by testing a miniaturized thermoelectric module. From these data, calculations will be made to determine the specifications necessary to produce 5 watts power output at 3 volts load.

The miniature thermoelectric module, with power terminals, is shown in Fig. 8. This module contains 15 thermoelectric couples connected electrically in series and thermally in parallel to form the thermoelectric ladder.

The thermoelectric couples are sandwiched between but electrically insulated from, the hot and cold sides. The electrical insulation used maintained its dielectric properties at the high, 482°C, hot side temperature while insuring reasonably good thermal conductance. A potting material having low thermal conductivity, was also incorporated in the module. This material will be described later.

The module hot and cold sides were machined from a stress-free 18-8, 300 series steel.

Special welding was employed to join a thin 18-8 tubular steel shell to the hot and cold sides, to insure a leak-free assembly.

The module temperature was raised to 100°C with an inert gas flowing through the module. This final precautionary step was used to minimize oxidation of the thermoelectric materials. After sufficient time, a positive pressure of the inert gas was sealed in the thermoelectric module.
Fig. 8 - Westinghouse Miniature Thermoelectric Module
Fig. 9 - Test Setup of Miniature Thermoelectric Module
Performance

The evaluation of the miniature thermoelectric module shown under test in Fig. 9, was to be conducted in two separate tests. The first test was conducted only on a temperature gradient criterion, i.e., the thermal losses were not considered.

At the maximum $\Delta T$ of 433°C, the thermoelectric module power output is 2.75 watts at 0.99 volts.

Tabulated results are listed in Table 4, Column 1.

The $\beta$ factor in Column 1 is a ratio of conductor resistance plus contact resistance to bulk thermoelectric material resistance. Special designing and assembly techniques would reduce $\beta$ further; but a range between 0 - 10% for $\beta$ is most acceptable.

Column 2 in Table 4 compares the measured data with values calculated from theoretical curves. The curves will be published by Westinghouse and are proprietary information.

Table 5 is a full-powered extrapolation, based on results obtained with the miniaturized module evaluation, for a 3-volt load voltage and 5 watt power output. The thermoelectric module will be 3-3/4 in. square and enclose 35 thermocouples (1/4 x 1/4 x 1-1/8 in.).

The second test has been designed to minimize thermal losses and requires a special testing apparatus. The mechanisms involved in heat transfer and the techniques for measurement are complex. It is planned to conduct the test in the near future.
### TABLE 4

**Reduced Scale Thermoelectric Module Output**

<table>
<thead>
<tr>
<th></th>
<th>Column 1</th>
<th>Column 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hot wall temperature ($T_h$ °C.)</td>
<td>482</td>
<td>482</td>
</tr>
<tr>
<td>Hot junction temperature (°C.)</td>
<td>472 (estimated)</td>
<td>472</td>
</tr>
<tr>
<td>Cold wall temperature ($T_c$ °C.)</td>
<td>49</td>
<td>49</td>
</tr>
<tr>
<td>Cold junction temperature (°C.)</td>
<td>73</td>
<td>73</td>
</tr>
<tr>
<td>Couple voltage (open circuit)</td>
<td>0.132 volt/couple</td>
<td>0.132 volt/couple</td>
</tr>
<tr>
<td>Module voltage (open circuit)</td>
<td>1.98 volt</td>
<td>1.98 volt</td>
</tr>
<tr>
<td>Module voltage (matched load)</td>
<td>0.99 volt</td>
<td>0.99 volt</td>
</tr>
<tr>
<td>Couple power output</td>
<td>0.183 watt/couple</td>
<td>0.183 watt/couple</td>
</tr>
<tr>
<td>Module power output</td>
<td>2.75 watt</td>
<td>2.75 watt</td>
</tr>
<tr>
<td>Couple resistance (27°C.)</td>
<td>0.0074 ohm/couple</td>
<td>0.0074 ohm/couple</td>
</tr>
<tr>
<td>Module resistance (482°C.)</td>
<td>0.361 ohm</td>
<td>0.336 ohm</td>
</tr>
<tr>
<td>$\beta$</td>
<td>0.0745</td>
<td>0</td>
</tr>
<tr>
<td>$\dot{Q}_c$ (Conducted through couple)</td>
<td>26.3 watt</td>
<td></td>
</tr>
<tr>
<td>$\dot{Q}_p$ (Heat absorbed at hot junction)</td>
<td>10.15 watt</td>
<td></td>
</tr>
<tr>
<td>$\dot{Q}_j$ (Joule heat returned to hot junction)</td>
<td>1.36 watt</td>
<td></td>
</tr>
<tr>
<td>Couple efficiency</td>
<td>7.75%</td>
<td></td>
</tr>
</tbody>
</table>

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### TABLE 5

**Anticipated Output of the 5-Watt Conversion System**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hot wall temperature ($T_{n}$ °C.)</td>
<td>482</td>
</tr>
<tr>
<td>Cold wall temperature ($T_{c}$ °C.)</td>
<td>49</td>
</tr>
<tr>
<td>Temperature difference ($\Delta T$) °C.</td>
<td>433</td>
</tr>
<tr>
<td>Module voltage (open circuit)</td>
<td>4.62 volts</td>
</tr>
<tr>
<td>Module voltage (1.74 A load)</td>
<td>3.00 volts</td>
</tr>
<tr>
<td>Module resistance (482 °C.)</td>
<td>0.936 ohms</td>
</tr>
<tr>
<td>Power output (1.74 A load)</td>
<td>5.18 watts</td>
</tr>
<tr>
<td>$\dot{Q}_c$ (Conducted through couples)</td>
<td>54.7 watts</td>
</tr>
<tr>
<td>$\dot{Q}_p$ (Absorbed at hot junction)</td>
<td>14.9 watts</td>
</tr>
<tr>
<td>$\dot{Q}_j$ (Joule heat returned to hot junction)</td>
<td>1.39 watts</td>
</tr>
<tr>
<td>Couple efficiency</td>
<td>7.6 %</td>
</tr>
</tbody>
</table>

**Potting Thermal Insulation**

Considerable effort was applied to the development of several potting thermal insulators by Westinghouse. These thermal insulations are characterized by four parameters.

1. The thermal conductivity of the thermal insulator has to be considerably less than that of the thermoelectric materials.

2. The thermal insulator has to support the thermoelectric pellets during shock and vibration loadings.

3. The thermal insulator must retain a high electrical resistance ($10^7$ ohm) at the hot junction temperature with negligible chemical reactivity.

4. It must have handling properties which permit its incorporation within the thermoelectric module simply and economically.
Satisfying the first requirement of the above was the most difficult. It was necessary to construct a thermal conductivity measuring apparatus for testing the insulating materials.

After conducting tests on a number of materials and mixtures, the final potting compound was formulated by mixing exploded silica and a smooth, working refractory material. This potting thermal insulator best fitted the outlined requirements.

**Thermal Conductivity**

The apparatus used for the thermal conductivity measurements was a modification of the 8-in. guarded hot plate method approved by the National Bureau of Standards (A.S.T.M. Designation: C177-45).

Fig. 10 shows such an apparatus, consisting essentially of a circular heater (2), guard heaters (1) and (6), a section of the insulating material to be tested (3) and the cooling plate (4). The heater (2), established heat flow through the insulating material. The guard heaters, (1) and (6) and heater (2), are kept at the same temperature by adjustments of the input to the guard heaters. This procedure prevents heat flow from heater (2) to heater (1) and (6), resulting in all the heat flow from heater (2) being perpendicular to its surface.

The space between the insulating material to be tested, heaters (2) and the guard heaters, is filled with alumina insulating grain.

As indicated in Fig. 10, temperatures are measured at various points in the insulating material and heater section by means of chrome-alumel thermocouples. The differential thermocouple method was employed between the electrical powered heater (2) and guard heaters (1) and (6). By then measuring energy input to the heater (2), insulating material thickness, and temperature gradient across the insulating material, the thermal conductivity may be determined by the standard equation:

\[ K = \frac{Q \times L}{A \times T} \text{ (Btu)} \frac{A \times T}{\text{hr, ft}^0 \text{ F}}. \]
Fig. 10 - Thermoelectric Module Evaluation Apparatus
B. ROYAL > RESEARCH CORPORATION EFFORT

Royal > Research Corporation's effort on the power conversion system, which is designed to produce the required electrical power through conversion of thermal energy of the isotopic fuel source, is continuing with favorable results. Design parameters have been defined and attention is now focused on the elements themselves. To assure maximum efficiency of individual thermocouples, problems such as heat flow through the couple, internal resistance, contact resistance and compactness must be solved. Research was initiated, by defining a thermocouple design completely different from any known to date. In our design, considerable emphasis was placed on couple resistivity to thermal and vibrational shock.

A comprehensive evaluation was next made of various electrical insulator materials. The criterion for acceptability was high electrical resistance with good thermal conductance. Many kinds of materials were tested by applying them to thermocouples which were then placed under test. Only those materials which survived extended periods of subjection to test conditions were selected for use in the thermoelectric package. As shown in Fig. 11, the cold end-cap design has made possible the assembly of couples having extremely close tolerances. Materials having superior bonding characteristics are employed in the cold junction assembly.

The need for a method of joining the completed thermocouples in a series array, which would be capable of reproducing the required output voltage, was resolved by means of the special printed circuit board shown in Fig. 14. Capable of withstanding temperatures over 300°F., the printed circuit board permits less than .002 ohms combined contact resistance, per thermocouple. Fabrication and testing of an array designed to accommodate 6 couples, Fig. 12, showed that the internal resistance per couple decreased, when an inert-atmosphere enclosure was used to fabricate and assemble our P and N elements, Fig. 13.

Early couples of Royal's design resulted in an average resistance of .027 ohms. At the present time extensive work is being undertaken to lower couple resistance to the alternate value of 0.0055 ohms, the material resistance of lead telluride. For a completed thermocouple to be efficient and durable, every problem associated with construction must be defined. Toward this end, each detail is under study, with the goal that of obtaining the highest efficiency for all of the thermocouples.
Fig. 11 - Enlargement of Royal Research Lead Telluride Thermocouple Design, With Operating Temperature Profile
Fig. 12 - Reduced-Scale Printed-Circuit Board Used in Life Testing of Royal Thermocouples

Fig. 13 - Thermocouple Assembly in Inert Atmosphere Enclosure
Fig. 14 - Royal Thermocouples Assembled on Printed Circuit Board
To insure that a completed element will be free of any oxidation during fabrication and assembly, all thermoelements are handled within an inert gas enclosure. Here, an argon atmosphere protects the materials until fabrication and assembly are complete. Actual testing is now under way to identify maximum operating temperatures of the hot and cold junctions and so determine the most efficient temperature differential. In the thermocouple tester (described in the 2nd Quarterly Report), temperature-indicating thermocouples were placed on the hot junction, iron shoe of the couple, copper cold junction cap and the cold junction plate. Individual temperatures were recorded, which later proved very valuable in determining the insulation best suited for both the hot junction iron shoe and cold junction copper cap. Anticipated operating conditions are simulated within the tester, in order to realistically analyze behavior of the thermocouples.

A study also has been initiated to identify those methods which aid in assembling the completed package. At this writing, the method decided upon is as follows: After successfully completing acceptance tests, each thermocouple will be attached to the printed circuit board. When all 46 thermocouples have been attached, the assembly will be tested to determine the power output from the system. After satisfying testing requirements, the system shall be enclosed in an hermetically sealed envelope. A stainless steel bellows comprising the walls of the envelope shall be bonded to the cold junction plate. Copper was chosen because it can be brazed to the bellows. This container is then inverted and an electrical insulator placed on the interior surface of the copper plate. Next, the printed circuit board, containing the thermocouples, will be inserted in the container so as to make physical contact with the cold junction. A pre-punched, circular piece of AA Fiberglas shall be inserted between the thermocouple pattern. At this time, an Armco iron plate, with location holes matching the position of each thermocouple, will be placed over the insulator. The electrical insulator will then be attached to individual iron hot shoes. Locating holes are threaded, to permit individual adjustment of the thermocouples so that intimate contact and pressure necessary for both the cold and hot junctions, can be maintained. After all thermocouples have been adjusted, a thin, stainless steel plate is placed over the hot junction plate. This disk is then welded to the stainless steel bellows, completing assembly of the thermoelectric package. A valve located on the cold junction copper plate will permit the package to be pumped down, under vacuum. After such time as is needed to outgas the system, a partial atmosphere of reducing gas and argon will be introduced into the package. Thermocouple performance is optimized by such an atmosphere. Finally, the package will be sealed off and evaluation initiated in preparation for its use in the Cesium-137 generator.
Royal Thermocouple Performance Under Test

A typical temperature profile encountered in our thermocouple tests may be seen in Fig. 11. These couples have an open-circuit voltage of 0.218 volts when the shoe temperature is 925° F. and the cold junction temperature is 175° F. Each thermocouple is designed to dissipate 2.07 watts of heat, with 1.97 watts of it going through the lead telluride and 0.10 watts through the ceramics.

Theoretical resistance of a typical couple at room temperature would be 5.53 milliohms, 33 milliohms at operating input temperature of the couple. It is anticipated that these resistances actually will be achieved in future fabrication research. To date, these couples have a room temperature resistance of 19 to 25 milliohms; resistance increases to about 100 milliohms per couple at operating temperature. At this time, it has been established that this increase in resistance is not due to junction resistance but rather to micro-fractures found in the lead telluride material. Work is progressing on developing an annealing process to eliminate excessive resistance.

A design objective of our thermoelectric program is to produce 0.128 watts per thermocouple, with an efficiency of 6.2 per cent at the designed operating temperature. Thus, the output of the thermocouple package, which contains 46 couples, will be 5.89 watts. This power then, will produce 5.0 watts of electricity, based on an 85 per cent efficient dc-dc converter. Open-circuit voltage of the thermocouple package will be 10 volts, d.c. To date, experimental data on these couples indicate a maximum power when the internal load is 1.25 times the external load. Therefore, maximum power will be produced when the thermoelectric power package is operating at 4.4 volts. This figure will increase when the power is optimized as a function of the dc-dc converter efficiency and the efficiency of the thermoelectric system, although this primary voltage to the dc-dc converter will remain within the range of 4.4 to 5.0 volts.
VI. SUBTASK 2.3 - GENERATOR DESIGN ANALYSIS

A basic overall design was presented and described in the Second Quarterly Report. This has been subject to several internal changes, due primarily, to development of more detailed information regarding fuel fabrication, arrival at a better understanding of shielding requirements and the results of experimental data on outer pressure shell integrity.

The fuel-increment configuration has been changed from a single, doubly-encapsulated stack of seven increments, to two stacks of three each. This was necessitated because of limitations of the Oak Ridge National Laboratory hot-cell welding enclosure. Because of this, the fuel configuration had to be shortened and increased in diameter, or fabricated as two separate units. The former alternative was undesirable, since it would have invalidated all previous work done on lateral dimensioning of the generator. Further, it would have resulted in a design whose outer diameter would have exceeded 13 in. Since the requirement for the device specifically defined the tolerable limits of diameter, the fuel increment diameters were held at 2.50 in. The only alternative was in going from a seven increment fuel load, to an even numbered one and dividing the increments into two batches. Six elements would reduce the fabrication time previously required for seven, and would not materially affect the fabrication technique or require a gross enlargement of process equipment. The final design of the fuel element array is shown in Fig. 15. All dimensional thicknesses and spacers are requirements identified by ORNL and transmitted to the contractor. The lateral walls will be ground to size, base plates welded in place and the resulting containers delivered for fuel loading. Additional hardware to be included, will be fitted caps and interior can spacers of varying thicknesses. The latter are for filling any air voids which may develop from increment stacking. Since the exact height of each increment is not known, vertical can dimensions can not be anticipated. In addition to two cans and lids of each size, one extra set of cans will be delivered, to permit orientation welding and familiarization prior to actual fuel enclosure.

Although not shown in the full scale model, the shield has been modified. Originally specified to be fabricated from sintered tungsten, it will now be made of cast, depleted uranium. Additionally, the screw cap originally shown at the top, will become the base. This enhances heat flow in the desired direction, upward, by removing the air gap which must exist between cap and body of the shield. Finally, both ends will be tapered to conserve on weight and improve insulability of the component. Since uranium is to be used, all inner
and outer surfaces will be heavily nickel plated to obviate combustion in the event of a disaster. The external, nickel surface will be polished and gold plated, to serve as a reflector and improve radiant-heat insulation.

The shield cap will be provided with three indented slots to permit easy assembly. A tapered pin slot and hole will be provided so that final closure may be made by means of a pin hammered home. It is interesting to note that the three holding slots may also be used to rigidly position the shield in the inverted position, preventing eventual sag into the felted fiberglass. Should this occur, intimacy of contact between shield and thermoelectric converter would be lost, causing a power drop.

No change has been made in the thermal insulation materials. AA Felted Fiberglas will be used to insulate against conductive heat loss. This material will be compacted to a density of 7 pounds/ft$^3$, which the manufacturer quotes as having a thermal conductivity of less than 0.01 Btu/hr/ft$^2$/ft in a vacuum of less than 100 microns. The dual, gold plated radiant-heat reflectors will be used as described in the Second Quarterly Report. Braided fiberglass spacers will be used as standoffs between shield and spacers.

One significant fact was brought to light after the last thermal insulation experiment was concluded. When the apparatus was disassembled, it was seen that the gold plating had all but disappeared. Improper determination of air tightness of the outer shell prior to testing, resulted in development of an air leak during evaluation. Since this occurred during the night, considerable oxygen was admitted to the interior before remedial measures could be taken. At the high operating temperature, to which the shield must be continually subjected, exclusion of air is imperative. Measures have been taken to prevent the admittance of air during generator pump-down. Although AA Felted Fiberglas will also be used to thermally shield the dc-dc converter (in the upper compartment), the higher efficiency made possible with a partial vacuum is not necessary here, and therefore no pump-down will be needed. The converter itself will be sealed within a potted package which will exclude oxygen.

Final dimensioning of the outermost 7075-T6 aluminum shell will be made after full scale testing to verify experimental results obtained. For these tests, reduced-scale containers were fabricated as shown in Fig. 16. Wall thickness was predicated on the Lamont pressure curves, which though limited to 2024 aluminum, were the only available data predicated upon actual testing rather than on calculated resistivity to pressure. Although the inside diameter of the test vessels was 3 in., the results were believed to be extrapolatable to anticipated dimensions of the generator.

RRC-0102
Fig. 15 - Double-Walled Hastelloy C Fuel Can
Fig. 16 - Reduced Scale Aluminum Test Vessels

Fig. 17 - Vessel Pressure Testing at Naval Electronics Laboratory, San Diego, California

Official U. S. Navy Photograph

RRC-0102
Fig. 18 - Imploded Test Vessel, Showing Cracked Wall, Above, and Fragmented Bottom, Below
These test vessels were rounded at the bases and provided with a single "O" ring seal at the lid. A raised area on the lid was machined, drilled and taped to provide a threaded hole for the lifting lug. In this way, the lid thickness was not weakened by perforation. In threading the lid to the vessel body, a gap of 0.030 inches was left, to determine if the two parts would pressure-weld under pressure.

Pressure testing was performed at the Battery Whistler Test Area, Naval Electronics Laboratory, San Diego, California. Here, a naval rifle has been foreshortened, the tube plugged and inner bore tapped for sea water admission and a two-stage pump connected to provide pressure. Because implosion could damage breech parts, the inner cavity of the first test vessel was filled with wood and then wrapped in burlap. Fig. 17 shows interim removal of the vessel after a typical run.

Runs were accomplished by alternately pressure testing and machining metal from the outside of bottom and side walls. After four such runs, the wall and base thickness had been reduced to a point considerably below Lamont recommendations. The tests were then temporarily concluded with no rupture occurring.

For the second set of runs, the reserve test vessel was used. Here, the starting wall and base thickness was equal to final dimensions on the first experiment. The purpose of this test series was to determine the rupture point in wall thickness and/or base thickness. This test vessel differed from the first, in that the inner base was rounded, so that no right angle was presented to the outside. Table 6 shows the results of the second series of tests. After each pressure test, first at 9,500 psi, and then at 15,900 psi, the vessel was removed and machined down. The foregoing pressures represent sea pressure at depths of 3,600 and 6,000 fathoms respectively.

During machining, an effort was made to maintain the original ratio of wall to base thickness, so that ultimate collapse could be attributed to a final configuration, rather than to a weak spot in the cross section thickness. However, despite all precautions, rupture did occur, as indicated by test No. 7. The appearance of the vessel and fragmented base may be seen in Fig. 18. Analysis of the rupture point indicates that a weak area developed circumferentially along a line opposite the interior round on the bottom. During machining, an outside radius was recut on the corner formed by the sides and base, to maintain a curved surface. However, in so doing a portion of the bottom roundness was reduced to a point that it did not exceed the vertical wall in thickness, as it should have. This may be seen by the concavity of the break surface where the base imploded and also by the symmetrical curvature on the outermost fragments as shown in the photo of the reassembled base.
TABLE 6  
Generator Shell Pressure Test - Reduced Scale  
(Pressure Testing, 5 Min. at Pressure)
7075-T6 Aluminum - 81,000 psi. max. tensile strength
72,000 psi. max. yield strength

<table>
<thead>
<tr>
<th>Test No.</th>
<th>Side Thickness</th>
<th>Bottom Thickness</th>
<th>Pressure 9,500 psi</th>
<th>Pressure 15,900 psi</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Wall 0.58</td>
<td>Bottom 0.875</td>
<td>OK</td>
<td>OK</td>
</tr>
<tr>
<td></td>
<td>O. D. 4.15</td>
<td>Length 4.28</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>Wall 0.559</td>
<td>Bottom 0.750</td>
<td>OK</td>
<td>OK</td>
</tr>
<tr>
<td></td>
<td>O. D. 4.088</td>
<td>Length 4.155</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>Wall 0.528</td>
<td>Bottom 0.625</td>
<td>OK</td>
<td>OK</td>
</tr>
<tr>
<td></td>
<td>O. D. 4.026</td>
<td>Length 4.03</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>Wall 0.497</td>
<td>Bottom 0.550</td>
<td>OK</td>
<td>OK</td>
</tr>
<tr>
<td></td>
<td>O. D. 3.964</td>
<td>Length 3.915</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>Wall 0.466</td>
<td>Bottom 0.550</td>
<td>OK</td>
<td>OK</td>
</tr>
<tr>
<td></td>
<td>O. D. 3.902</td>
<td>Length 3.915</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>Wall 0.388</td>
<td>Bottom 0.550</td>
<td>OK</td>
<td>OK</td>
</tr>
<tr>
<td></td>
<td>O. D. 3.777</td>
<td>Length 3.915</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>Wall 0.326</td>
<td>Bottom 0.460</td>
<td>OK</td>
<td></td>
</tr>
<tr>
<td></td>
<td>O. D. 3.652</td>
<td>Length 3.79</td>
<td>A t 15,850 psi, B ottom Collapsed</td>
<td></td>
</tr>
</tbody>
</table>

The extreme thinness of base and wall at the time of fracture verifies the value of the experiment. According to these data, a full scale pressure shell of approximately 1.4 in. side-wall thickness and 2.2 in. base thickness would withstand immersion to a full 6,000 fathoms. This is a considerable reduction over initial estimates of 2.5 and 3.5 in. thickness for sides and base, respectively.

Repeated cyclings of pressure, during the numerous tests performed on both test vessels produced no leaks through the single "O" ring. (This is highly desirable since researchers at Lamont have reported that placement at depth and subsequent retrieval of instrument packages, caused the con-
tainers to "breathe" e.g., the extremely high pressure caused slight contraction of the entire container. This, in turn, resulted in leakage.) When the first test vessel was examined, no pressure welding was found to have occurred, despite repeated compressions. A galling effect was identified, which made reopening extremely difficult. This phenomenon will be re-evaluated during full-scale pressure testing.

The success of reduced-scale testing provided wall thickness approximations which will be applied to two full-scale pressure shells. In anticipation of these tests, two forgings have been ordered from the Harvey Aluminum Company, Oakland, California. These will consist of bases and caps only, since the upper portion of the configuration will be structurally stronger by design. Starting as forgings, the pieces will be stress relieved and machined to approximate dimensions. After receiving them, final machining and thread cutting will be performed. Inside diameters will be predicated upon a 2-in.-thick radiation shield since this thickness appears adequate when using depleted uranium as shield material.
VII. SUBTASK 2.4 - GENERATOR SHIELDING ANALYSIS

Particular emphasis has been placed upon the shielding of gamma radiation in the proposed cesium generator. This is important from the standpoint of safety and efficiency. Concerning safety, this means that the nuclear core must have sufficient shielding to reduce to a tolerable level the dosage received by persons working in close proximity to it. As to efficiency, adequate shielding means effective utilization of gamma-ray conversion to heat. Cesium is a useful power source only if both beta and gamma decay radiation can be converted to heat. Since the shield is also the heat accumulator, all attenuated radiation is made available as heat to the thermoelectric converter.

While both foregoing situations suggest the generous use of shielding material, a third parameter influences the careful avoidance of excessive shielding. The cesium generator is predicated upon a subsea application. As such, its diameter may not exceed 13 in. (for the Lamont application) and its overall weight must be kept to a practical minimum. The latter feature is necessitated by ultimate plans for retrieval of the generator from great ocean depths.

Initial consideration of the radiation shield design suggested the use of pressed and sintered tungsten. This was partly due to its high structural strength, good thermal conductivity and high density. The most promising material available (several manufacturers of malleable tungsten have been contacted) has a quoted 92 per cent theoretical density. Preliminary shielding experiments using lead were made and later, a tungsten equivalent thickness calculated, based on a 100 per cent tungsten density. A one-curie source was used for this determination. However, it was felt that results reported earlier in the program, although carefully obtained, were subject to inaccuracies introduced by extrapolation from one curie to 28,000 curies. For this reason, a larger source was procured from Oak Ridge National Laboratory. To improve reliability, this source was aliquoted from the same massive batch of radiocesium held in reserve for fueling the 5-watt operational generator. This precaution was important since the amount of Cesium-134 present can determine the amount of shielding required if it exceeds 1 per cent of the fuel source. The assay on the 10-curie source indicated 1.5 per cent content of the shorter-lived isotope (2.4 yr.) of cesium. Therefore, this fraction was used in the shielding experiment.

In order to provide a safe working area, as well as to exclude scatter radiation from the meter, the 10-curie source was shielded within a 2-in.-thick lead cave, as shown in Fig. 19. The lead glass windows and castle manipulators permitted safe handling of the source and attenuator disks. A heavy-walled
Fig. 19 - Exterior and Interior Views of Lead Cave Used In Shielding Experiments
Fig. 20 - Attenuation of Cesium-137 (Incl. 1.5% Cs-134) by Tungsten and Lead
Fig. 21 - Attenuation of Cesium-137 (Containing 1.5% Cs-134) by Aluminum
inner cave was so designed as to permit only a circular beam of radiation to emerge from the configuration. The diameter of the beam was controlled by an iris large enough to expose the entire liquid contents of the source bottle, permitting an unshielded reading to be taken. Approximately 3 in. outward from this circular orifice was a second orifice 0.5 in. greater in diameter. Into this circular tunnel were placed the attenuator disks used to determine the half and tenth-value layers for the Cesium-137, Cesium-134 source. The enlarged diameter of the second, continuing aperture, provided a stepped radiation seal against scatter radiation. A calibrated ionization chamber meter was positioned at a distance of 4 ft. from source center of the experiment. In this way, the meter was protected from stray radiation which may have existed within the confines of the outermost shield. With the beam hole plugged inside, the meter registered no radiation field, verifying the containment geometry of the experiment.

The purpose of this experiment was threefold. We wished to experimentally determine the combined, unshielded gamma radiation from an aqueous source of radioesium, to find the half and tenth-value layer of lead and also, to obtain like values for sintered tungsten. The unshielded dose at 4 ft. was measured and found to be 2.00 r/hr. Various thicknesses, first of lead and then sintered tungsten, were then used to obtain attenuation curves shown in Fig. 20. From the lead curve, a half-value thickness of 0.24 in. and a tenth value of 0.79 in. were obtained. Similar thicknesses obtained from the tungsten curve were found to be 0.20 in. (half-thickness) and 0.67 in. (tenth-thickness). As indicated by the tungsten curve, the slope continued on course up to the point that random results overshadow the values obtained. This is unfortunate, since the curves did not reflect a resolution of Cesium-134 gammas, after attenuation of the softer Cesium-137 gammas. During the conduct of this experiment, duplicate values were obtained for all points during attenuator stacking and subsequent individual removal of attenuators.

Since it is difficult to accurately anticipate the shielding required to reduce the gamma ray energy from a high kilocurie source, several safety factors were introduced into our calculations. The contribution to attenuation made by the steel reflectors surrounding the shield was omitted as well as that of the Hastelloy C cans. In a similar manner, attenuation by the (outermost) thick aluminum pressure shell, will also be left out of the preparation of final shielding dimensions. Although these contributions to overall shielding are valid, they will not be included in shield requirements. However, it is important to know how valid they are in reducing radiation, in order to define them as safety factors. For this reason, a third shielding experiment was run. Permanent attenuators (tungsten and steel) were first positioned to intercept the beam. Then varying known thicknesses of aluminum were added. Results of this experiment are indicated by the curves shown in Fig. 21.
Here too, it can be noted that no break in the plot is seen, indicating that only the Cesium-137 fraction was evaluated. Values for the half and tenth thicknesses of lead obtained in the foregoing experiments were subsequently verified by similar work performed at Oak Ridge National Laboratory. For their work a 20-curie source was employed, obtained from the same kilocurie batch as the 10-curie source used in the experiments described above.

Temporarily overlooking the inaccuracy of experimental results, a shield thickness was determined for tungsten. For this calculation, commercially available tungsten of 86 per cent purity (tungsten content) and of 92 per cent theoretical density, was considered. After extrapolating the unshielded dose to 25,500 curies, it was found that at 4 ft. some 5,100 r/hr. must be attenuated down to 100 mr/hr.

Using the experimentally obtained half and tenth values for sintered tungsten, a thickness in excess of 3 in. would be required. Actually, the amount needed would be a net value obtained after factoring-in the Cesium-134 contribution minus self-absorption and source geometry. Never-the-less, 3 in. of thickness is completely untenable, since it would result in a generator of decreased thermal efficiency due to increased surface. Of greater consequence is the fact that the resulting 5-watt device would have a diameter of nearly 24 in. Original requirements have made mandatory a 13-in. maximum diameter.

Although the density of uranium is not appreciably different from tungsten (W = 19.3 g/cc; U = 18.48 g/cc), its shielding properties are superior. Moreover, uranium can be procured and machined in cast form of theoretical density; machinable tungsten is only 86 per cent pure since it has lower Z adulterants added to permit machining. Since tungsten must be pressed and sintered, theoretical density can never be reached, the best being about 92 per cent. Although uranium in finely divided form is pyrophoric, in a massive shape combustion is quite unlikely. Electroplating of nickel on all outer surfaces renders it noncombustible and provides a resistance to corrosion which is far superior to tungsten. Depleted uranium is plentiful and has been used before to provide high density ball swivels for shielded cave manipulators. Since it has no other use, fabrication of shields would provide an excellent outlet for the stockpiles which have accumulated over the past 15 yrs.

Before final consideration could be given the use of depleted uranium, it was necessary to determine the attenuation factors of it. Shielding experiments were performed at the Oak Ridge National Laboratory in a similar manner to those performed at the Royal Research Laboratory. The same 20-curie source was used here, as was employed for the lead shielding experiment described earlier. Although data has not been completely resolved,
the contract was provided with data showing half and tenth values to be 0.11 and 0.37 in., respectively. ORNL researchers reported that they were unable to resolve the shielding plot into a two-component curve. Because of omission of this important factor, common to all experimental work, an analysis was made of all contributing gamma rays from both radioisotopes of cesium. These data, shown in Appendix C, include a consideration of buildup factors, internal conversion coefficients, self-absorption and primary radiation. From this it is seen that 2.1 in. of depleted uranium (cast) would be adequate. The dose rate at 4 ft. from the center of the generator, in a plane perpendicular to, and bisecting the generator axis, would be 104 mr/hr. The effects of the 1.5 per cent Cesium-134, a finite volume of fuel material and dose buildup, are included.

While the uranium alone would reduce radiation to a safe level, additional contributions to shielding by the Hastelloy C cans, reflector cups and aluminum pressure shell will make a further contribution. They represent a margin of safety and are not included in shielding calculations.

A final consideration is Cesium-134 content. The 1.5 per cent used throughout the analysis was predicated upon an August use date, whereas the generator will not be delivered until next spring. Decay of this 2.3-yr. isotope will reduce its curie strength to 83 per cent of the August value. This will result in a reduction of shielding requirements.
The basic parameters of a 5-watt, cesium-fueled thermoelectric generator have already been defined, based upon a deep-sea application at an intended operational depth of 3,600 fathoms. Containment to 6,000 fathoms, compactness and extreme structural strength are emphasized. In addition to the operational requirements, complete containment of the nuclear fuel load has dictated certain departures from original design.

The fuel element configuration has been changed from seven increments to an array of six. This was necessitated by limitations of the ORNL remote welding enclosure, which cannot accommodate the Hastelloy C can described in the last report. Instead, two separate cans will be fabricated, each to contain three fuel increments of equal size. After closure by welding, each can will be sealed within a second one of the same design and construction, thus effecting double encapsulation of fuel increments. Fig. 15 shows this modification.

Although revision of the shielding requirements has resulted in a change from tungsten to depleted uranium material for its construction, the general configuration remains the same. This will consist of a hollow cylinder with threaded cap. After closure, the two parts will be locked by means of a taper pin. Top and bottom will be beveled to reduce weight and exposed surface.

One important design change involves repositioning of the shield within the shell, which will have a beneficial effect on thermal efficiency. This has been accomplished by reversing the shield ends, so as to locate the threaded cap at the bottom, instead of at the top. Thus transposed, the heat path will consist of a continuous wall of uranium, from heat source to power conversion system. The bottom location of the cap-thread interface air gap will not materially impede heat flow. Fuel cans will be loaded in such a manner as to be in upright position when the generator is completely assembled.

The change in fuel cladding, to conform to the Oak Ridge National Laboratory specifications of wall thickness and tolerances, has made necessary an increase in dimensions of all components external to it. The inside diameter of the shield has been increased to 2.88 in., necessitating an outside diameter of 7.07 in. Vertical dimensions of the shield have been increased to 8.81 in. cavity height, 13.01 in. for overall height. Initial thermal insulation requirements were predicated upon a smaller shield. However, by increasing it, additional felted fiberglass thickness is needed. This has been increased from 3/4 in. compacted, to 1.25 in., compacted. Similarly, the 7075-T6 aluminum pressure shell now must be increased to a 10-in. inside diameter, with a
1.5-in. wall. The overall generator diameter has been held to 13 in. as first promised the customer.

The thermoelectric conversion package has been reduced in size to optimize the thermocouple array and reduce heat loss through the stainless steel envelope. The dc-dc converter package is almost complete. Final closure will be made by joining the package to the top lid of the pressure shell.
IX. SUBTASK 2.6 - HAZARDS EVALUATION

Research relating to the hazards evaluation comprises two areas. The first consists of tests affecting the structural design and/or efficiency of the thermoelectric generator as an adequate means of containing the fuel load. The second area is restricted to tests of the type that identify the limits of the final design's endurance to natural and accidental situations to which a generator could be subjected. Since safety is of primary concern in the evolution of a portable device employing high kilocurie quantities of Cesium-137, a comprehensive recapitulation of all information pertaining to this subject will be prepared as a separate report. Interim work is described under the subtask most appropriate to it, or where results affect the continuing effort. For that reason data under this heading which is described elsewhere in the report, will not be reiterated here.

The original scope of work of the hazards evaluation was devoted to identification of a safe fuel compound, design of a structurally-sound package and attenuation of radiation through it. Physical testing, in relation to credible incidents, was to be restricted to thermal insulation by burial. Recently, an addition was made to the original scope of work. Experimentation was outlined to test under realistic conditions the following parameters of safety:

1. Biological Uptake Studies of Cesium Polyglass.
2. Radiation Effects on Cesium Polyglass.
3. Pressure Testing the Generator Shell.
5. Generator Shell Impact Evaluation.

1. Biological Uptake Studies.

The contractor will fabricate cesium polyglass sources in size, quantity and dose rate required by the investigating laboratory. This material will be used to determine the uptake of Cesium-137 in marine plants and animals. This work has not yet been initiated.

2. Radiation Effects Study.

Described in subtask 1.4, this experiment was concerned with the change in cesium polyglass, after irradiation by massive gamma ray dosage. Testing showed that while optical change occurred, other physical properties were
retained. Although solubility testing is incomplete, results indicate that (if anything) the dissolution rate was decreased by the radiation effect. Irradia-

tion was performed by the High Level Gamma Facility of Vallecitos Atomic Laboratory.


Since little was known of how the shape and thickness of a 7075-T6 aluminum vessel would affect its resistance to water pressure at 9,500 to 15,900 psi., specimens were subjected to tests which would determine this factor. Reduced scale hollow cylinders, whose lids were sealed with a single "O" ring, were employed for initial work. Pressure welding of the threaded surfaces was sought, as a means of permanent closure against tampering. A ratio of inside diameter to wall and base thickness was estab-

lished. While pressure welding did not occur, galling made lid removal ex-

remely difficult.

The successful completion of reduced-scale models led to the design and testing of a full scale generator shell. After being subjected to the water pressure anticipated at 6,000 fathoms (15,900 psi) for 3.5 hrs., the specimen showed no deformation of cracks. The single "O" ring seal prevented any water seepage, and the lid galled so tightly to the case that disassembly by a team of men was impossible. All testing was performed at the high pressure test facility, Naval Electronics Laboratory, San Diego. All testing and data gathering was performed by authorized naval personnel.

4. Generator Shell Surface Coating.

Several potentially acceptable materials were given preliminary tests, first refluxing in sea water for 24 hrs., and then subjection to 10,000 psi, water pressure. Of the coatings tested, three resisted attack satisfactorily: hard anodized surface with chromate cover, Kel-F and Bisonite. Aluminum slugs with surface coatings of these three materials were placed in shallow, running sea water for a period of 90 days. Interim reports have been received from the Oceanographic Tower, Naval Electronics Laboratory, San Diego, where the slugs are under test. These reports, which are based upon observations through a submarine TV scanner, indicate that all specimens are coated with colonies of marine plants and crustaceans but that corrosive attack on the coatings is not perceptible. At the conclusion of tests, the specimens will be retrieved and examined.
5. Generator Shell Impact Evaluation.

The present design of the 5-watt generator represents the safe minimum of structural strength capable of withstanding a submerged depth of 6,000 fathoms. This is in keeping with the basic objectives of producing a generator of minimal size and weight. Its diameter has been held to 13 inches, the allowable maximum, while the weight is well under 500 lbs. Internal shielding reduces the dose to approximately 100 mr/hr at 4 ft. It is obvious then, that a credible incident, such as an air accident involving free fall to terminal velocity, cannot be readily interpreted by calculations.

A second full-scale aluminum pressure shell identical to the one used for pressure testing is being fabricated. When completed, it will be air dropped, after which structural containment and the impact burial pattern will be evaluated.

Tests performed by original contractual agreement covered the possibility of a thermal excursion occurring as a result of accidental burial of a generator in sand, or envelopment by burlap or canvas. The former situation could obtain only as a result of impact after air drop, while the latter (highly probable) could occur during storage or transportation of the device. To be efficient, a thermoelectric generator must pass thermal energy rapidly through its conversion system. To do so, the generator must be in an environment of infinite heat capacity, whose ambient temperature is lower than that of the device. All conceivable operating conditions would fulfill this requirement. Carefully planned storage or transportation methods would take this into consideration and provide ventilation even in locations of high temperature. However, accidental covering by burlap or canvas would cause an insulation effect which might produce a critical temperature rise at the outer skin and then move inward to the fuel core. It was the purpose of these tests to determine -- by actual duplication of conditions -- the effect of accidental insulation upon the generator core.

For the burlap experiment, the same configuration was used as reported in the Second Quarterly Report (core and shield mockup containing a 700-watt cartridge heater with output control). This was protected by double gold-plated radiant heat reflectors and compacted, felted fiberglass. The assembly was housed in an evacuated aluminum cylinder. Thermocouple probes were positioned internally, to indicate temperatures throughout the configuration. This then, was wrapped in seven thicknesses of burlap and covered by a canvas tarpaulin.
Fig. 22 - Core Temperature As a Function of External Insulation
When the core heater was energized to an output of 120 thermal watts (anticipated output of the Cesium-137 fuel core), the core temperature rose steadily, reaching a maximum of 820 deg. C. after 42 hrs., as indicated in Fig. 22. Since the melting range of cesium polyglass is above 900 deg. C., a modest safety margin is provided. More important is the fact that the external surface of the test vessel was only 1/5 the anticipated surface of the 5-watt generator. Greater surface would dissipate heat more readily, even in an insulated environment. Naturally, prolonged insulation at the temperature indicated above would result in total destruction of the thermoelectric conversion system, but this is not a criterion of the hazards evaluation.

The second thermal insulation experiment employed the same device (described above) in a sand-burial situation, to determine core temperature as a function of time. To reproduce natural conditions, bags of sand were stacked around the apparatus, after placing it upon several moist sandbags. Loose sand was then packed between the bags, to fill existing air gaps. Core temperature again rose steadily, reaching a maximum of 625° C. after 90 hrs. of elapsed time. Bags of sand were employed instead of bulk sand, to provide an additional insulant in the burlap, itself. This would simulate any organic material which might be present under actual terrain conditions. Absence of the burlap would, of course, reduce the temperature peak and provide a greater margin of safety against fusion of the polyglass.

Earlier experiments have shown that, if exposed to air, a generator will not experience a hazardous temperature excursion, even when in a still, ambient atmosphere maintained at 90 deg. F.
X. TASK III - DESIGN AND CONSTRUCTION OF THE MODEL

When the conceptual design of the generator had been formulated, the design parameters were incorporated into a model. As per contractual agreement, this model is full scale and in sufficient detail to illustrate all component parts, although not necessarily to final dimension. The model, with suitable carrying case which can also be used as a portable pedestal, has been delivered to the customer.

Designed in three separate increments, the model may be assembled in such a manner as to give an exploded view, using clear plastic spacers to separate the increments. Thus, the original contractual requirements of construction of a thermoelectric generator could be illustrated, apart from the dc-dc power converter package. Threaded increments permitted integration of parts to illustrate present plans for a 5-watt device capable of a 12-volt output.

The basic generator, which includes the complete power generating system and lid with power-outlet connector, is shown in Fig. 23. Interior components are illustrated in relief by means of a pie-section cutaway. Six individual fuel increments, with cutaway platinum cups, are innermost. External to these are shown the two, double-walled Hastelloy C fuel cans. The radiation shield has been quarter-sectioned, with the threaded lid in full round to illustrate dimensional characteristics. A cutaway of the power conversion system (simplified) is positioned directly above the shield, with several conceptual thermocouples in full round and thermal insulation located between. The envelope surrounding the power converter, also is simplified and is shown as a right cylinder, instead of the bellows wall which actually will be used in the operational generator. The two gold-plated reflector cups which surround the radiation shield may be seen as lines in the vertical and as full diameter disks at the base. External to them is shown the conductive insulation. A thick pressure shell surrounds the components, which being pie-sectioned to 120 degrees exposes the generator interior. The threaded pressure shell lid is provided with two, concentric "O" rings, so that when joined to the shell proper, the full size of the device is seen.

The fully integrated generator, including the dc-dc converter section, is shown on page ii. While the latter section is not pie-sectioned, a view of the integrated system is provided, when the plastic spacers are in position. Cooling ribs may be seen on the circumference of the upper section. When in the exploded view, a thermal insulation layer also may be seen surrounding the dc-dc converter package. Since this converter is being purchased outright and not developed within the program, interior components have not been pie-sectioned.
Fig. 23 - Generator Model, Less DC-DC Converter Accessory

RRC-0102
A more basic reason for omitting a pie-sectioned view is because two applicable but mechanically different converters are available and final selection cannot be made until completion of prolonged life testing of both devices.

On the top of the lid may be seen two lifting rings. They are to be used for lowering the generator into position on the ocean bottom. The male section of a through-shell electrical outlet plug is positioned at the center of the lid.

Since the purpose of the model is to illustrate the basic principle of this device, components have been simplified to reduce cost, and exaggerated where necessary, to permit visual identification from a distance. Color has been used to delineate between different metals and alloys to be used in the actual device. The exterior surface of the generator was painted to suggest a protective coating over the aluminum shell. The nuclear fuel core is illuminated by means of a dry-cell battery system. The switch is hidden at the upper left corner of the generator cutaway.

In addition to the model and carrying case, the customer was also provided with descriptive literature to aid in making oral presentations. Color slides of the model were prepared in 35 mm and lantern-slide sizes.
XI. SUBTASK 4.1 - FABRICATION ANALYSIS

The conceptual design shown in the second quarterly progress report is currently under re-evaluation, based upon certain changes which have since been made. Machining details and assembly techniques are being formulated in accordance with fabrication requirements. Data, derived by testing and calculations in previous subtasks, is being reviewed. Various suppliers of raw materials and certain fabricated parts have been contacted in an effort to synchronize delivery of necessary items, which must be procured before going ahead with dependent parts.

An AEC permit has been requested, to allow possession by Royal Research of the 25,500-curie fuel load, as well as the heavy-walled depleted-uranium shield. Auxiliary equipment is being constructed, including a heavy-duty vacuum pumping system. This will be used to evacuate the fiberglass insulation and thereby enhance heat economy within the generator.

Coordination has been established with the Lamont Geological Observatory, so that the electrical leads and physical shape of the generator will meet the requirements of all associated seismographic equipment.

Initially, this subtask is concerned with identifying any necessary deviations to the conceptual design which may become necessary as the result of final testing, or changes required in certain generator components. During generator assembly, surveillance will be maintained to anticipate needed modifications and thereby save time by obviating rework effort.
A change in program scope has resulted in redefinition of the facility responsible for fuel processing. The Vallecitos Atomic Laboratory, General Electric Company, will not process the 25,500 curies of Cesium-137. Instead, this work will be performed at Oak Ridge National Laboratory. The project has been notified that Royal Research is to provide the following items for fuel processing and encapsulation:

1. Seven platinum fuel cups.
2. Three complete double-walled fuel cans and caps.
3. Sufficient ion exchange resin to process fuel.
4. A suitable fusion furnace, with blueprints.
5. A suitable slurry mixer, with blueprints.
6. Complete fuel processing data.

Oak Ridge representatives will reserve the right to utilize either a ceramic-walled, platinum fuel cup, or a full platinum cup. If the latter is employed, they will provide a cutting wheel to remove excess platinum wall after fusion. The cups will be spun and will have 0.004-in.-thick walls and 0.005-in.-thick bases. Although only six are required, one extra will be furnished as a precautionary measure.

Three sets of double-walled Hastelloy C fuel cans will be fabricated to the required dimensions, with the bases sealed by welding. All thicknesses and tolerances will be to specifications supplied by ORNL. In addition, Hastelloy C spacers of thicknesses varying from 0.032 to 0.125 in. thickness will be provided. Shaping will be achieved through the use of seamless tubing. If unavailable, rolled sheet may be substituted for tubing, provided it is accompanied by X-ray certification. Machine cutting will not be employed; instead, high speed grinding will be used to fabricate cup parts to dimension. All welds will be stress relieved. Final closure will be performed at ORNL, after fuel loading.

Royal Research has developed a high temperature furnace for fabricating the cesium polyglass fuel, as shown in Fig. 24. The lower section may be swung away from the rigid top and loaded with a fuel cup. The fuel charge can then be added as shown in photo A. Next, the lower section is returned to a position directly under the top, photo B, and elevated tight against it by
Fig. 24 - High Temperature Furnace Used for Cesium Polyglass Fabrication
Fig. 25 - Electrically Driven Cesium-Fuel Mixer
means of a remotely operated screw drive, as in photo C. A stainless steel
off-gas vent that projects from the furnace top, carries away moisture and
fusion gases. Furnace temperature is controlled by means of a rheostat
mounted external to the hot cell. Heat is provided by special nichrome heat­
ing elements wound around a ceramic cylinder. Thermal insulation is pro­
vided at the sides and base of the furnace. This type of device has been used
by Royal researchers to fabricate cesium polyglass test specimens and has
proven reliable and efficient.

A second piece of hardware designed by the contracting laboratory and
under development, is an electrically driven auger. Its purpose is to mix an
aqueous solution of Cesium-137 carbonate with stable complex borosilicates.
Water is removed by heating during the mixing process. As the slurry goes
first to a paste and then to a dry mix, the revolving screw action breaks up
lumps and finally reduces the contents to a dry, finely divided and intimately
mixed product. Since this is a complex mechanism, considerable development
time is needed to perfect it. Components which comprise the basic machine
are shown in Fig. 25. Power is derived from a variable speed, d. c. Bodine
motor. This propels a spiral auger member housed within a hopper. When
the hopper is inclined, the slurry is forced up the ramp in the interior of the
hopper, whereupon it returns to the rear by the pushing action of the auger on
the slurry. Heat is provided to the hopper walls by externally-mounted heating
elements. Considerable flexibility is provided, permitting the angle of eleva­
tion, or depression to be changed, the auger speed varied, and its direction
reversed. The hopper may be tilted for dumping and the entire apparatus
rotated on its base for convenient charging with the dry and liquid constituents.

Problem areas include the shape of the hopper, the position of the auger
and redundancy of motion. In addition, the search for a suitable bearing
material has been extremely difficult. Such parameters as resistance to radia­
tion, heat and the abrasive action of the borosilicates have damaged all bear­
ings. An offset chain drive proved to be impractical, when dry product caked
in the chain and caused breaks in the chain links.

Several highly-successful runs have been made with the experimental
auger-hopper yielding a finely divided, dry powder of uniform particle size,
after only 2 hours running time. The loss of product on the walls was less
than 3 per cent. This would not become an additive loss for each of the six
fuel-processing runs since subsequent addition of aqueous cesium carbonate
would redissolve any dry residue. This would be further aided by the abrasive
action of the borosilicate particles in the slurry.
Resin Dimensions

CsCl
Dissolver

169 g.
in
200 ml.
H₂O

Temp: 70° C.
Time: 30 min.

Volume in: 200 ml. CsCl
+265 ml. wash water

Volume out: 465 ml.
Elution time: 105 min.

Fig. 26 - Cesium Polyglass Flow Sheet
Because of the complexity of the auger-hopper approach to mixing and drying, alternative methods are also under evaluation. One particularly promising approach involves rotating knife blades within an externally heated hemispherical bowl. Constant rotation of the specially shaped blades continually blends the slurry, then breaks up the lumps and finally, reduces particle size by erosive action of the particles themselves. Water is driven off by a heater which surrounds the bowl. Although somewhat slower than the auger method, basic simplicity of the concept suggests easier reproducibility.

The revised fuel-process schedule appears in Fig. 26 and includes such improvements as the elimination of a cesium cleanup step and reduction of eluent from the carbonate conversion column. This, in turn, results in the elimination of a liquid-volume reduction vessel since the carbonate eluent may be fed directly to the borosilicate mixer. This is shown as a broken, lined box since the apparatus has not been selected as yet. Changing from a seven increment system to one containing six increments of fuel has caused an increase in processing reagents. Despite this, the overall process requires less time than that reported earlier in the program.
The number of curies of Cs\textsuperscript{137} required to produce 120 watts of thermal energy depends on the average energy release per disintegration and the percentage of this energy deposited within the radiation shield. Calculation of the latter presents no problem. All the betas will be easily stopped within the shield and since it is designed to provide many tenth-value layers for the 662 kev gamma rays, it is safe to assume that essentially all the gamma energy will be available thermally. It should be noted that although the 662 kev gammas are partially converted, the resultant electrons are monoenergetic, with no accompanying neutrino, as in true beta decay. Thus, assumption of complete availability of the gamma energy remains valid, regardless of the actual conversion coefficient.

Calculation of the average energy per disintegration is not straightforward because of the continuous spectrum characteristic of beta decay. Since the energy carried away by the neutrino in each disintegration is lost, an average beta energy must be determined. For this, the shape of the spectrum and the end-point (maximum) energy must be known. If there is more than one branch, as in the case of Cs\textsuperscript{137}, the branching ratios also are required. From a search of the literature, the best values for end-point energies *1 and branching ratios *2 and *3 were determined. These are shown on the decay scheme below.
Data on average beta energies found in the literature are sparse, with the following rule-of-thumb frequently quoted: \(^5\) \(E \approx \frac{1}{3} E_{max}\). That this is a rather risky assumption, is amply demonstrated by Marinelli et al., \(^6\) whose compilation shows a substantial scatter of values about the figure of 1/3 for the well-behaved "allowed" transitions.

Neglecting screening effects, \(^7\), we have for any beta spectrum:

\[ N(\eta) \, d\eta = F(Z, \eta) \, C(\eta, \eta_0, Z) \, \eta^2 \left( W_0 - W \right)^2 \, d\eta \]

where \(N(\eta) \, d\eta \) is the number of betas with momenta between \(\eta\) and \(\eta + \, d\eta\)

- \(\eta = \frac{2p}{mc}\) is relativistic momentum in units of \(mc\)
- \(W = \frac{E + mc^2}{mc^2}\) is total energy in units of \(mc^2\)
- \(E = \) kinetic energy
- \(W_0 = \frac{E_0 + mc^2}{mc^2}\) is total energy corresponding to \(E = E_0 = E_{max}\)

and \(W^2 = \eta^2 + 1\) is the relation between \(W\) and \(\eta\)

- \(F(Z, \eta)\) is relativistic nuclear Coulomb factor
- \(C(\eta, \eta_0, Z)\) is statistical weight factor
- \(C(\eta, \eta_0, Z)\) is shape correction factor depending on the type of transition

The average beta energy, \(\bar{E}\), is then defined as:

\[ \bar{E} = \frac{\int_0^{\infty} W F(Z, \eta) \, C(\eta, \eta_0, Z) \, \eta^2 \left( W_0 - W \right)^2 \, d\eta}{\int_0^{\infty} F(Z, \eta) \, C(\eta, \eta_0, Z) \, \eta^2 \left( W_0 - W \right)^2 \, d\eta} \]

The factor \(F(Z, \eta)\) is rather complicated and includes a \(f^2\) function of complex argument. Fortunately, this function has been tabulated \(^8\) in the form \(f(Z, \eta) \approx \frac{F(Z, \eta)}{\eta F(2, \eta)}\), for \(Z = 1\) to 100 and a sizeable range of \(\eta\). (The value of \(Z\) to be used in the tables is that of the daughter nucleus, \(Z = 56\) in this case.) The above integrals thus simplify somewhat to:

\[ \bar{E} = \frac{\int_0^{\infty} W f(Z, \eta) \, C(\eta, \eta_0, Z) \left( W_0 - W \right)^2 \, d\eta}{\int_0^{\infty} f(Z, \eta) \, C(\eta, \eta_0, Z) \left( W_0 - W \right)^2 \, d\eta} \]

and numerical integration can readily be carried out. For the case of \(F(Z, \eta) \approx 1\), which is a fair approximation for allowed transitions in low-\(Z\) nuclei, the integrations, though somewhat tedious, can be carried out directly.
Cesium-137 Energy Calculations

The shape factor, $C$, for allowed transitions, is unity. For the 514 Kev beta of Cs$^{137}$, a unique first-forbidden transition, $C = (W_b - W)^2 + (W^2 - 1)^2$, and for the 1176 Kev beta, a second-forbidden transition, $C = (W_b - W)^2 + 0.03(W^2 - 1)^2$ and 9. Referring to the shape factor for the 514 Kev beta, we note that it augments both the high and low energy ends of the spectrum relative to an allowed transition, while the shape factor for the 1176 Kev beta enhances mainly the low energy contribution of the spectrum.

Using NBS tables *8, and the above shape factors, the average beta energies for both branches were determined by numerical integration and the results tabulated below.

<table>
<thead>
<tr>
<th>$E_{max}$</th>
<th>%</th>
<th>$\bar{E}$</th>
<th>$\bar{E}/E_{max}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>514 Kev</td>
<td>92%</td>
<td>183 Kev</td>
<td>.356</td>
</tr>
<tr>
<td>1176 Kev</td>
<td>8%</td>
<td>283 Kev</td>
<td>.240</td>
</tr>
</tbody>
</table>

The only value for $\bar{E}/E_{max}$ found in the literature was a value of 0.367 (=192/523) for the 514 Kev branch that appears unreferenced in an ORNL compilation of fission product properties *10. As was suggested by the comment on the 1176 Kev shape factor, the calculated $\bar{E}/E_{max}$ evidences a substantial energy depression.

Summary

The results of the average beta energy calculations and the assumption of essentially complete deposition of the unconverted 662 Kev gammas within the radiation shield indicate a value of $(0.92)(183) + (0.08)(283) + (0.92)(662) = 800$ Kev for the average available energy per disintegration.

Since $P$ (watts) = $5.94 \times 10^{-3} \times$ Mev/disintegration $\times$ Curies, we find that 120 watts of thermal energy requires 25,300 curies of Cs$^{137}$. 

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*References*


APPENDIX B

Calculation of Temperature Distribution in Uranium Shield

Summary

The possibility of "hot spots" developing in the uranium shield of the Royal Research 5-watt Cesium-137 thermoelectric generator has been investigated as reported in this note.

The assumptions necessary to provide a mathematically tractable solution are discussed and justified. Sample calculations show a maximum possible temperature differential of 140° F. from the cooled top surface of the uranium shield to the hottest point in its interior.

Introduction

The uranium shield of the Royal Research 5-watt Cesium-137 generator is shown in the sketch below. The beta and gamma decay energy of the 8.8-in.,-high fuel cylinder containing 25,300 curies of Cesium-137, produces 120 watts of thermal power within this shield. The division of energy between the beta and gamma components is calculated as follows: Of the .800 Mev released, on the average, per Cesium-137 disintegration (see section on Cesium-137 energy calculations), \((0.92 \times 0.183) + (0.08 \times 0.283) = 0.191\) Mev is apportioned the betas arising from true beta decay. About 10% of the .662 Mev gammas are internally converted and emerge as monoenergetic conversion electrons, bringing the total average beta energy per disintegration to .252 Mev. The remainder, or .548 Mev per disintegration, is the gamma component. Since the range of the betas is small compared to the fuel cylinder diameter and the major portion of the gamma rays will escape the fuel cylinder, we will assume the following behavior for the spatial deposition of the nuclear radiation:

\[<2.88\text{'}><1.88\text{'}><8.0\text{'}><13\text{'}>\]
1. All the beta energy (32% of the total) is deposited within the fuel cylinder.

2. All the gamma energy (68% of the total) is deposited exponentially in a radial direction in the uranium shield.

We can now write the equation governing the temperature distribution within the uranium shield, which is simply the time-independent, non-homogeneous heat conduction equation:

\[ \nabla^2 T + \frac{S}{\kappa} = 0 \]

where \( S \) is the source term arising from the exponential deposition of gamma energy within the shield.

Assumptions

1. The nature of the source term and the solution of the partial differential equation are considerably simplified if we extend the cesium fuel cylinder the entire height of the uranium shield without changing the cesium loading per unit length or the fuel cylinder diameter. This most certainly produces a worse situation than encountered in the actual generator since we are now generating \((120) (13/8, 8) \approx 177 \) watts of power, instead of 120. This geometry is shown in the sketch below.

2. If we neglect axial conduction along the fuel cylinder, then the heat must flow through the annulus \( a < r < b \) at the top, a smaller area and therefore requiring a larger temperature gradient than in the actual case.

3. If we assume the bottom and sides of the uranium shield to be perfectly insulated, we require all the heat to flow through the annulus \( a < r < b \), which again requires a larger temperature gradient.

4. The top of the cylinder \( a < r < b \), \( z = h \), is assumed to be at a constant temperature. This is plausible since the resistance to heat flow across the interface at \( z = h \) will be greater than the
resistance to heat flow in the radial direction at \( z = h \). In any event, the temperature distribution in the area of greatest interest, near the bottom of the uranium shield, will be fairly insensitive to any departure from this assumed boundary condition.

Statement of Problem

The above assumptions allow us to completely define the problem mathematically. The differential equation is:

\[
\frac{\partial^2 T(r, z)}{\partial r^2} + \frac{l}{r} \frac{\partial T(r, z)}{\partial r} + \frac{\partial^2 T(r, z)}{\partial z^2} + \frac{\mu a}{k r} Q_r e^{-\mu (r-a)_+} = 0
\]

for \( a < r < b \)

\( 0 < z < h \) \hspace{1cm} (1)

and the boundary conditions are:

\[
\frac{\partial T(a, z)}{\partial r} = -\frac{Q_B}{k} \quad (2)
\]

\[
\frac{\partial T(b, z)}{\partial r} = 0 \quad (3)
\]

\[
\frac{\partial T(r, 0)}{\partial r} = 0 \quad (4)
\]

\[
T(r, h) = T_S \quad (a \text{ constant}) \quad (5)
\]

where

- \( Q_r \) = gamma energy flux (BTU/hr in²)
- \( Q_B \) = heat flux into uranium shield from betas deposited in fuel cylinder (BTU/hr in²)
- \( \mu \) = linear energy absorption coefficient for gamma radiation (in⁻¹)
- \( k \) = thermal conductivity (BTU/hr in °F.)
- \( h \) = height of cylinder (in.)
- \( a \) = inner radius of cylinder (in.)
- \( b \) = outer radius of cylinder (in.)

The above problem is amenable to solution by a separation-of-variables technique which proceeds, briefly, as follows:

(The main problem is first broken up into two sub-problems.)

RRC-0102
Sub-problem 1

Differential equation:
\[
\frac{\partial^2 T_1(n, z)}{\partial n^2} + \frac{1}{n} \frac{\partial T_1(n, z)}{\partial n} + \frac{\partial^2 T_1(n, z)}{\partial z^2} = 0
\]  
(6)

with boundary conditions:
\[
\frac{\partial T_1(n, 0)}{\partial n} = 0 \quad (7) \quad \frac{\partial T_1(n, h)}{\partial n} = 0 \quad (9)
\]
\[
\frac{\partial T_1(n, z)}{\partial n} = 0 \quad (8) \quad T_1(n, h) = T_0 \quad (10)
\]

Sub-problem 2

Differential equation:
\[
\frac{\partial^2 T_2(n, z)}{\partial n^2} + \frac{1}{n} \frac{\partial T_2(n, z)}{\partial n} + \frac{\partial^2 T_2(n, z)}{\partial z^2} + \frac{\mu a Q \eta}{R} e^{-\mu(n-\alpha)} = 0
\]
(11)

with boundary conditions:
\[
\frac{\partial T_2(n, 0)}{\partial n} = 0 \quad (12) \quad \frac{\partial T_2(n, h)}{\partial n} = 0 \quad (14)
\]
\[
\frac{\partial T_2(n, z)}{\partial n} = 0 \quad (13) \quad T_2(n, h) = 0 \quad (15)
\]

The sum of the solutions to (6) and (11) with their respective boundary conditions, is then the solution to (1) with boundary conditions (2-5). That is:

\[
T(n, z) = T_1(n, z) + T_2(n, z)
\]
(16)

Now the solution to (6) is obviously

\[
T_1(n, z) = T_0
\]
(17)

The solution to (11) is obtained by first applying the separation of variables technique to the homogeneous part of (11). The characteristic functions which represent the z dependence of the product solution and which satisfy homogeneous boundary conditions (14) and (15) are:

\[
\cos \frac{2\pi n + 1}{h} z \quad n = 0, 1, 2, \ldots
\]

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Since these functions form a complete orthogonal set, we are permitted to expand any piecewise differentiable function in terms of these orthogonal characteristic functions. It should be pointed out that the original problem could have been broken up in a different fashion, such that the characteristic functions representing the radial dependence with appropriate homogeneous boundary conditions were orthogonal, and then the expansions would be carried out using these characteristic functions instead. For the case at hand, we let

\[ \frac{\mu}{k \pi} Q_y \, e^{-\mu(n-a)} = \sum_{n=0}^{\infty} S_n(r) \cos \frac{2m+1}{2} \frac{\pi}{h} \, Z \]  

(18)

and

\[ T_2(r, Z) = \sum_{n=0}^{\infty} t_n(r) \cos \frac{2n+1}{2} \frac{\pi}{h} \, Z \]  

(19)

The \( S_n(r) \) are readily shown to have the form

\[ S_n(r) = \frac{4(-1)^n \mu}{(2n+1) \pi k \pi} Q_y \, e^{-\mu(n-a)} \]  

(20)

and the result of substituting (18) and (19) into (11) is

\[ \sum_{n=0}^{\infty} \left[ \frac{\partial^2 t_n(r)}{\partial r^2} + \frac{1}{r} \frac{\partial t_n(r)}{\partial r} - \left( \frac{2n+1}{2} \frac{\pi}{h} \right)^2 t_n(r) + S_n(r) \right] \cos \frac{2n+1}{2} \frac{\pi}{h} \, Z \, = 0 \]  

(21)

Now the functions \( t_n(r) \) appearing in (19) can be found by solving the second-order, non-homogeneous (ordinary) differential equation involving these functions, which appears within the brackets in (21). The solution is obtained by the variation of parameters technique which yields:

\[ t_n(r) = \left[ A_n + u_n(n) \right] K_0 \left( \frac{2m+1}{2} \frac{\pi}{h} \, n \right) + \left[ B_n + v_n(n) \right] I_0 \left( \frac{2m+1}{2} \frac{\pi}{h} \, n \right) \]  

(22)

where the \( A_n \) and \( B_n \) are constants which are determined by the two remaining boundary conditions (12) and (13), the \( I_0 \) and \( K_0 \) are zero-order modified Bessel functions of the first and second kind, respectively. The nature of the \( u_n(r) \) and the \( v_n(r) \) will be discussed shortly.
Thus the solution to our original problem, from (16), (17), (19), and (22) is:

$$T(r, z) = T_0 + \sum_{n=0}^{\infty} \left\{ A_n + V_n(n) \right\} K_0 \left( \frac{2n+1}{2} \frac{\pi}{L} r \right) + \left[ B_n + V_n(n) \right] I_0 \left( \frac{2n+1}{2} \frac{\pi}{L} r \right)$$

\[ \times \cos \frac{2n+1}{2} \frac{\pi}{L} z \]  \hspace{1cm} (23)

This solution would be amenable to straightforward calculation, using available tables and modified Bessel functions, were it not for the nature of the $u_n(r)$ and $v_n(r)$. The latter are of the form

$$u_n(n) = -E_n \int_C^n I_0 \left( \frac{2n+1}{2} \frac{\pi}{L} r' \right) e^{-\mu (r' - a)} dr'$$  \hspace{1cm} (24)$$

and

$$v_n(n) = E_m \int_C^n K_0 \left( \frac{2n+1}{2} \frac{\pi}{L} r' \right) e^{-\mu (r' - a)} dr'$$  \hspace{1cm} (25)$$

where $C$ and the $E_n$ are constants. It appears that (24) and (25) cannot be integrated directly and numerical procedures would be required. It turns out that, if the problem had been broken up such that the orthogonal functions in which we expanded were the $r$-dependent set mentioned earlier, there would again arise integrals involving the product of Bessel functions and exponentials.

**Simplified-Solution Technique**

All the foregoing assumptions and solution techniques are equally applicable if we unfold the cylinder into a slab of the same thickness and height. The fact that the cross-sectional area perpendicular to the cylinder axis is now reduced, because of the unfolding, makes the resistance to heat flow in both the $r$ and $z$ directions even greater and therefore requires an even larger temperature gradient in both directions. With this modification the problem becomes easily manageable since the solution now involves only hyperbolic and trigonometric functions and ordinary exponentials. Most important, however, is that each assumption and departure from the actual generator configuration has been shown to require a larger temperature gradient than will be met in reality and thus will make any so-called hot spots appear even hotter.
Equation (1) now becomes

\[
\frac{\partial^2 T(x,z)}{\partial x^2} + \frac{\partial^2 T(x,z)}{\partial z^2} + \frac{\mu Q(x) e^{-\mu x}}{k} = 0
\]  

(26)

with the boundary conditions corresponding to (2)-(5)

\[
\frac{\partial T(0,z)}{\partial x} = -\frac{Q(x)}{k} \quad (27) \quad \frac{\partial T(x,0)}{\partial z} = 0 \quad (29)
\]

\[
\frac{\partial T(L,z)}{\partial x} = 0 \quad (28) \quad T(x, h) = T_5 \quad (30)
\]

The nomenclature is the same as before, with the exception of the independent variable \( r \), which is now replaced by \( x \); and the interval \( a \leq r \leq b \) now corresponds to \( 0 \leq x \leq L \). The solution procedure follows identically that which was outlined previously. The main problem is again split up into two sub-problems.

Sub-problem 1

Differential equation

\[
\frac{\partial^2 T_1(x,z)}{\partial x^2} + \frac{\partial^2 T_1(x,z)}{\partial z^2} = 0
\]

(31)

with boundary conditions

\[
\frac{\partial T_1(0,z)}{\partial x} = 0 \quad (32) \quad \frac{\partial T_1(x,0)}{\partial z} = 0 \quad (34)
\]

\[
\frac{\partial T_1(L,z)}{\partial x} = 0 \quad (33) \quad T_1(x, h) = T_5 \quad (35)
\]

Sub-problem 2

Differential equation

\[
\frac{\partial^2 T_2(x,z)}{\partial x^2} + \frac{\partial^2 T_2(x,z)}{\partial z^2} + \frac{\mu Q(x) e^{-\mu x}}{k} = 0
\]

(36)
with boundary conditions

\[
\frac{\partial T_2}{\partial x} (0, Z) = -\frac{Q_0}{R} (37) \quad \frac{\partial T_2 (x, 0)}{\partial z} = 0 \quad (39)
\]

\[
\frac{\partial T_2 (L, Z)}{\partial x} = 0 \quad (38) \quad T_2 (x, h) = 0 \quad (40)
\]

Where \( T(x, Z) = T_1 (x, Z) + T_2 (x, Z) \) (41)

Again we find that \( T_1 (x, Z) = T_S \) (42)

For the solution of (36), we use the same device of expanding in terms of the orthogonal characteristic functions

\[
\cos \frac{2n+1}{2} \frac{\pi}{h} Z \quad n = 0, 1, 2, \ldots
\]

obtained from a separation-of-variables solution of the homogeneous part of (36) and satisfying boundary conditions (39) and (40).

Thus we let

\[
T_2 (x, Z) = \sum_{n=0}^{\infty} t_n (x) \cos \frac{2n+1}{2} \frac{\pi}{h} Z \quad (43)
\]

and

\[
\frac{\mu}{R} Q_0 e^{-\mu x} = \sum_{n=0}^{\infty} S_n (x) \cos \frac{2n+1}{2} \frac{\pi}{h} Z \quad (44)
\]

The \( S_n (x) \) are readily shown to be

\[
S_n (x) = \frac{4Q_0 \mu (-1)^n e^{-\mu x}}{R (2n+1) \pi} \quad (45)
\]

We now substitute (43), (44), and (45) into (36), with the result:

\[
\sum_{n=0}^{\infty} \left[ \frac{\partial^2 t_n (x)}{\partial x^2} - \left( \frac{2n+1}{2} \frac{\pi}{h} \right)^2 t_n (x) + \frac{4Q_0 \mu (-1)^n e^{-\mu x}}{R (2n+1) \pi} \right] \cos \frac{2n+1}{2} \frac{\pi}{h} Z = 0 \quad (46)
\]
The $t_n(x)$ of (43) are then found by solving the second-order, non-homogeneous (ordinary) differential equation bracketed in (46), with the result:

$$t_n(x) = A_n \cos k \frac{2n+1 \pi (L-x)}{2} + B_n \sin k \frac{2n+1 \pi (L-x)}{2} + \alpha_n e^{-\mu x}$$  \hspace{1cm} (47)

where

$$\alpha_n = \frac{2 Q x^\mu (-1)^n}{k h \left(\frac{2n+1 \pi}{2} \right)^3 - \mu^2 \frac{2n+1 \pi}{2}}$$  \hspace{1cm} (48)

and the $A_n$ and $B_n$ are constants determined by substituting (47) back into (43) and satisfying the two remaining boundary conditions (37) and (38).

From (41), (42), (43), and (47) we obtain the final solution:

$$T(x, z) = T_s + \sum_{n=0}^{\infty} \left[ A_n \cos k \frac{2n+1 \pi (L-x)}{2} + B_n \sin k \frac{2n+1 \pi (L-x)}{2} + \alpha_n e^{-\mu x} \right]$$  \hspace{1cm} (49)

where

$$\alpha_n = \frac{2 Q x^\mu (-1)^n}{k h \left(\frac{2n+1 \pi}{2} \right)^3 - \mu^2 \frac{2n+1 \pi}{2}}$$  \hspace{1cm} (48)

$$B_n = \frac{-\mu \alpha_n e^{-\mu L}}{2 n+1 \pi}$$  \hspace{1cm} (50)

$$A_n = \frac{4 Q x^\mu (-1)^n}{k (2n+1 \pi)} - \frac{2n+1 \pi}{2} B_n \cos k \frac{2n+1 \pi}{2} + \frac{2n+1 \pi}{2} L e^{-\mu x} \alpha_n$$

$$+ \frac{2n+1 \pi}{2} \sin k \frac{2n+1 \pi}{2} + \frac{2n+1 \pi}{2} L$$  \hspace{1cm} (51)
Calculations

Values of the required parameters are as follows:

\[ h = 13 \text{ in.} \]
\[ L = 1.88 \text{ in.} \]
\[ k = 1.66 \text{ BTU/hr. in. } \text{oF. (for } T = 900 \text{ oF.)} \]
\[ \mu = 6.2 \text{ in.}^{-1} \]
\[ Q_y = 3.47 \text{ BTU/hr. in.}^2 \]
\[ Q_{\beta} = 1.65 \text{ BTU/hr. in.}^2 \]

The first five of each of the coefficients appearing in (49), i.e., (48), (50), and (51) have been calculated and are listed below.

\[
\begin{align*}
A_0 &= +143.0 \\
B_0 &= +19.2 \times 10^{-5} \\
\varpi_0 &= -0.431 \\
A_1 &= -4.93 \\
B_1 &= -2.14 \times 10^{-5} \\
\varpi_1 &= +0.144 \\
A_2 &= +0.932 \\
B_2 &= +0.777 \times 10^{-5} \\
\varpi_2 &= -0.087 \\
A_3 &= -0.285 \\
B_3 &= -0.396 \times 10^{-5} \\
\varpi_3 &= +0.0622 \\
A_4 &= +0.107 \\
B_4 &= +0.245 \times 10^{-5} \\
\varpi_4 &= -0.0494
\end{align*}
\]

By inspection of (49) it is readily seen that the maximum temperature, as might be expected, is to be found along \( z = 0 \), the bottom surface of the generator. Two points at the bottom of the shield were chosen for a temperature calculation, \( T(0,0) \) next to the fuel cylinder, and \( T(L,0) \) at the outer wall of the uranium shield.

Using the first five terms of (49), the results are:

\[ T(0,0) = T_b + 141^\circ \text{ F.} \]
\[ T(L,0) = T_b + 139^\circ \text{ F.} \]

The sum of the first five terms is estimated to be well within 1 per cent of the infinite series solution for \( 0 \leq x \leq L \) and \( z = 0 \).

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Thus the maximum possible temperature in the uranium shield will be about 140\(^\circ\) F. above the temperature \(T_s\) of the cold face. From the most pessimistic bias of all of our assumptions, it is reasonable to expect the maximum temperature differential in the actual generator to be less than 100\(^\circ\) F. And finally, it should be mentioned that since the thermal conductivity of uranium actually increases with temperature, in contrast to most metals, this will tend to flatten the temperature gradient even more.
APPENDIX C

Shielding Calculations

Summary

The dose rate 4 ft. from the center of the cesium generator on a plane perpendicular to and bisecting the generator axis is calculated to be 105 mr/hr. The effects of the 1.5% Cesium-134 contamination, finite volume of fuel source with attendant self-absorption and geometrical considerations, plus dose build-up in the shield, have been included. The 1/8 in. Hastelloy C cylinder surrounding the fuel, thermal insulation, and aluminum pressure shell (exterior to the uranium shield), were neglected and should provide additional attenuation.

Nuclear Data

The latest available data on the energies, conversion coefficients and intensities of gamma rays arising from decay of Cesium-137 and Cesium-134 are shown below, together with the appropriate reference.

Cs\textsuperscript{137} - 25,500 curies

<table>
<thead>
<tr>
<th>Gamma Energy (Mev)</th>
<th>Total Conversion Coefficient</th>
<th>Unconverted Gammas/Disintegration</th>
</tr>
</thead>
<tbody>
<tr>
<td>.662</td>
<td>.114</td>
<td>.826</td>
</tr>
</tbody>
</table>

Cs\textsuperscript{134} - 383 curies

<table>
<thead>
<tr>
<th>Gamma Energies (Mev)</th>
<th>Total Conversion Coefficient</th>
<th>Unconverted Gammas/Disintegration</th>
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</thead>
<tbody>
<tr>
<td>.474</td>
<td>negligible</td>
<td>.013</td>
</tr>
<tr>
<td>.563</td>
<td>&quot;</td>
<td>.10</td>
</tr>
<tr>
<td>.569</td>
<td>&quot;</td>
<td>.14</td>
</tr>
<tr>
<td>.605</td>
<td>&quot;</td>
<td>.98</td>
</tr>
<tr>
<td>.796</td>
<td>&quot;</td>
<td>.80</td>
</tr>
<tr>
<td>.801</td>
<td>&quot;</td>
<td>.10</td>
</tr>
</tbody>
</table>
Cs\textsuperscript{134} - 383 curies (Cont'd)

<table>
<thead>
<tr>
<th>Gamma Energies (Mev)</th>
<th>Total Conversion Coefficient</th>
<th>Unconverted Gammas/Disintegration</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.960</td>
<td>negligible</td>
<td>0.006</td>
</tr>
<tr>
<td>1.040</td>
<td>&quot;</td>
<td>0.015</td>
</tr>
<tr>
<td>1.170</td>
<td>&quot;</td>
<td>0.022</td>
</tr>
<tr>
<td>1.370</td>
<td>&quot;</td>
<td>0.033</td>
</tr>
<tr>
<td>1.570</td>
<td>&quot;</td>
<td>0.0012</td>
</tr>
</tbody>
</table>

Shielding Calculations

Based on the actual generator configuration, a calculation was made of the dose rate at 4 ft. from the center of the cylinder on a plane perpendicular to, and bisecting its axis. The various aspects of the calculation are discussed below.

1. Fuel Cylinder.

The fuel cylinder was considered to be a uniform, cylindrical volume source 2.5 in. in diameter and 8.75 in. in height. Since Compton scattering predominates over photoelectric and pair production for the gamma energy ranges of interest here, it is safe to consider the chemically complex cesium fuel as a pure Compton scatterer. All materials, except hydrogen, contain roughly the same number of electrons per gram \( \times 4 \). The linear absorption coefficient of the fuel (for the purpose of self-shielding calculations) is, to a good approximation, only a function of the material density and primary gamma energy. The linear absorption coefficients for the fuel were calculated, using the data of White \( \times 5 \).

2. Uranium Shield.

The 2.1-in. thick cast uranium shield was assumed to have a density of 19.0 g./cm\(^3\) \( \times 6 \). The mass attenuation coefficients used were those of Grodstein \( \times 7 \).


Calculation of the uncollided energy flux at a point exterior to a finite cylindrical-volume source surrounded by an annular shield has been carried out for a wide range of problems and the results plotted graphically \( \times 8 \).
For the details and calculations, the reader is referred to reference *6. The calculated values of $I_0$ (Mev/cm²·sec.) are shown in the table below. For the 1.5 per cent Cesium-134 contaminant, only those gammas with energies in excess of the .662 Mev Cesium-137 gamma were considered and the .796 Mev and .801 Mev gammas were taken as an equal number of .800 Mev gammas.

4. Conversion of Uncollided Energy Flux to Dose Rate.

Although the ratio of uncollided flux (Mev/cm²·sec) to dose rate (r/hr) is usually assumed constant over the gamma energy range of interest here, the actual values *9 were used, producing the results as shown in the table.

5. Dose Build-Up Factor.

The method of moments has had considerable success in describing the penetration of gamma rays *4. Calculations of differential energy spectra and build-up factors have been carried out for many materials (incl. uranium) and for a wide range of primary energies and depths of penetration *10. The only source geometries, however, are point isotropic and plane monodirectional. Considering the distance of detector from source (4 ft.) in comparison to the fuel cylinder height (8.75 in.), it is evident that most of the gammas reaching the detector have traversed essentially the same thickness of shield. Thus, the approximation of point isotropic geometry for the purpose of build-up factor calculation introduces little error. Also, the build-up factors, although calculated for an infinite medium, are within a few per cent of those applicable to a finite medium for the number of mean-free paths under consideration here *4. An effective value of mean-free paths, which takes into account the finiteness of the fuel source, was calculated in the course of computing the uncollided energy flux. Since this contribution is only a few per cent of the total number of mean-free paths, the build-up factors were calculated by considering the penetration to be characteristic of the same total number of mean-free paths in uranium. For purposes of interpolation, the build-up factor data are presented in more convenient form by Moteff *8.
### Uncollided Flux and Dose Rate for Gamma Energies

<table>
<thead>
<tr>
<th>Gamma Energy (Mev)</th>
<th>Uncollided energy flux at 4 ft. (Mev/cm²·sec)</th>
<th>Uncollided flux dose rate D₀ (Mr/hr)</th>
<th>Dose rate build-up factor B₀</th>
<th>Dose Rate D (Mr/hr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>.662</td>
<td>$1.02 \times 10^4$</td>
<td>$4.94 \times 10^5$</td>
<td>20.6</td>
<td>2.4</td>
</tr>
<tr>
<td>.800</td>
<td>$2.13 \times 10^3$</td>
<td>$5.02 \times 10^5$</td>
<td>4.2</td>
<td>2.5</td>
</tr>
<tr>
<td>.960</td>
<td>$9.92 \times 10^1$</td>
<td>$5.17 \times 10^5$</td>
<td>9.6</td>
<td>2.6</td>
</tr>
<tr>
<td>1.040</td>
<td>$5.03 \times 10^2$</td>
<td>$5.24 \times 10^5$</td>
<td>3.62</td>
<td>2.6</td>
</tr>
<tr>
<td>1.170</td>
<td>$1.94 \times 10^3$</td>
<td>$5.36 \times 10^5$</td>
<td>12.05</td>
<td>2.5</td>
</tr>
<tr>
<td>1.370</td>
<td>$6.68 \times 10^3$</td>
<td>$5.54 \times 10^5$</td>
<td>2.5</td>
<td>2.5</td>
</tr>
<tr>
<td>1.570</td>
<td>$4.49 \times 10^2$</td>
<td>$5.72 \times 10^5$</td>
<td>.785</td>
<td>2.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td><strong>TOTAL 104.7 Mr/hr</strong></td>
</tr>
</tbody>
</table>

Thus, the calculated dose rate at a point 4 ft. from the center of the generator, is approximately 105 Mr/hr. Additional attenuation will be provided by the 1/8 in. Hastelloy C cylinder surrounding the cesium fuel and the thermal insulation and aluminum pressure shell exterior to the uranium shield, all of which were neglected in this calculation.

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*References*

1. Nuclear Data Sheets, National Research Council, Washington, D. C.

