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REVIEW OF BETAVOLTAIC ENERGY CONVERSION

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Betavoltaic energy conversion refers to the generation of power by coupling a beta source to a semiconductor junction device. This paper briefly reviews the theory of betavoltaic energy conversion and some past studies of the subject. Calculations of limiting efficiencies for semiconductor cells versus bandgap are presented along with specific studies for Pm-147 and Ni-63 fueled devices. The approach used for fabricating Pm-147 fueled batteries by the author in the early 1970's is reviewed. Finally, the potential performance of advanced betavoltaic power sources is considered.

INTRODUCTION

Betavoltaic energy conversion refers to the generation of power by coupling a beta source to a semiconductor junction device. Some interest has been shown in this approach to energy conversion at the last two SPRAT meetings. As a result, it seemed timely to review the subject. This paper briefly reviews the theory of betavoltaic energy conversion, past studies in the field and discusses the potential performance of betavoltaic systems based on the availability of beta sources and currently available solar cell materials.

PAST BETAVOLTAIC STUDIES

The first report of an electron-voltaic effect was given by Ehrenberg, et al., in 1951 (ref. 1). They were primarily interested in the current magnification that resulted when selenium photocells were bombarded by an electron beam. Rappaport was the first to describe betavoltaic studies, that is, investigations involving beta sources coupled to semiconductor junction devices (ref.2). He reported in 1953 on characteristics of silicon alloy junctions coupled to a 50 milliecurie Sr^{90}-Y^{90} radioactive source. One cell produced 0.8 microwatts with an overall efficiency of 0.2 % being attained. The overall efficiency is based on the total power produced by the radioisotope source. Pfan and Roosbroeck reported on similar studies about the same time (ref. 3). They discussed the general problem of betavoltaics and gave experimental results for Sr^{90}-Y^{90} sources combined silicon and germanium devices.

A more detailed report on the work by Rappaport and coworkers at RCA was given in 1956 (ref. 4). Further results were described for silicon and germasnium alloy junctions coupled to Sr^{90}-Y^{90}beta sources. In addition, the theory of betavoltaic devices was formulated. The interdependence of beta source parameters such as self absorption coefficient, beta energy

spectrum and actitivity, and semiconductor parameters such as energy gap and minority carrier properties were emphasized. The RCA group also identified the potential of Pm-147 betavoltaics in the 1956 paper. This was partially motivated by the negative results obtained with Sr^{90}-Y^{90} sources coupled to Si devices. As a result of radiation damage, the maximum power produced by a Si/Sr-90 system was found to decay to one-tenth of its initial value within one week of life. The final contribution of the RCA group is contained in a 1964 paper by Flicker, et al. (ref. 5). Si and GaAs diffused junction devices were coupled to Pm-147 sources. Beta sources were made by precipitating Pm-147 as the hydrated oxide ($Pm_2O_3 \cdot 6H_2O$) onto a substrate. Betavoltaic studies with GaAs cells yielded very poor results. Studies with silicon cells included the fabrication of prototype power sources consisting of a Pm-147 source combined with one or two silicon cells. Overall efficiencies of 0.4 % and 0.77 % were achieved. Lifetime studies with these prototypes showed only a slight effect due to radiation damage.

The most extensive effort concerning betavoltaic energy conversion appears to have occurred in a program led by the author at Donald W. Douglas Laboratories , Richland, WA, from 1968 to 1974 (ref. 6 and 7). This effort was based on the use of Pm-147 beta sources combined with Si n/p cells to produce nuclear batteries that were utilized as power sources for heart pacemakers. A brief description of this effort is discussed in a subsequent section.

PRINCIPLES OF BETAVOLTAIC ENERGY CONVERSION

The basic entity in a betavoltaic power source consists of a beta-emitting material coupled to a junction device as depicted in Figure 1. Some of the key aspects of betavoltaic energy conversion are described by Figure 2. An equivalent circuit for a betavoltaic cell is essentially the same as that for a solar cell, except that the current source is due to collection of electron-hole pairs generated by high energy beta particles. The importance one places on the series resistance R_S and the shunt resistance R_{Sh} are reversed when comparing betavoltaics and photovoltaics. The value of $R_S$can be relatively large in the case of betavoltaics since the value of J_{SC} will typically be in the range of 1 μA/sq.cm.to 100 μA/sq.cm., whereas in photovoltaic applications J_{SC} is typically in the range of 10 to 40 mA/sq.cm..Thus, R_S can be 100 ohms in a betavoltaic cell and cause a ptoblem. On the other hand, it is important to minimize the shunt conductance -- that is, maximize the shunt resistance. Since a loss current of 1 μA may be significant, it is necessary to utilize devices based on single crystal material. In the remainder of this section, a synopsis of the theory of betavoltaics will be presented, and then utilized to calculate the maximum efficiency of betavoltaic power sources versus semiconductor band gap.

The current supplied to a load by a betavoltaic cell is given by

$$J = J_{SC} - J_{Loss}(V) \qquad (1)$$

$J_{SC}$is the short circuit current and J_{Loss} is the loss current given by

$$J_{Loss} = J_o \exp(qV/kT) + J_{or} \exp(qV/2kT) + J_{ot} \exp(BV) + V/R_{sh} \qquad (2)$$

where the loss terms refer to minority carrier injection, depletion layer recombination, tunneling and current loss through the effective shunt resistance. Since betavoltaic power sources typically provide low current , the dominant loss mechanism is typically tunneling or depletion layer recombination.

The short-circuit current is given by

$$J_{sc} = (1-r)\, Q\, J_{max} \tag{3}$$

where 'r' is the reflection coefficient for beta particles from the semicnductor surface, Q is the collection efficiency and $J_{max}$is the maximum possible current. The beta particle reflection depends primarily on the atomic number of the semiconductor . The collection efficiency is the fraction of electron-hole (EH) pairs collected as current relative to the total number of EH created by the beta particle flux that enters the semiconductor device. Since the decrease of beta particle flux within the semiconductor is proportional to exp(-αx), where α is the absorption coefficient, analytical expressions for Q for a given device structure are essentially the same as those derived for solar cells. In particular, the beta flux passing through a material can be written as

$$N(x) = (1-r)\, No\, \exp(-\alpha x) \tag{4}$$

If the beta particles penetrate only on the order of a minority carrier diffusion length, then values of Q can approach 1.0. For example, since betas from Pm-147 only penetrate silicon to a depth of 60 μm, Q-values can approach 0.8 to 1.0. If Pm-147 is coupled to a direct bandgap material, however, the collection efficiency will be significantly less since the diffusion length will be much smaller . Thus the value of Q depends on properties of both the source and the semiconductor.

In order to calculate the maximum efficiency of a system, we must know the maximum possible current. The key considerations concerning the calculation of Jmax are described in Figure 3. One can define an effective ionization energy ε which is the average amount of energy expended to create one electron-hole pair. An empirical relationship exists which relates ε to semiconductor bandgap, namely,

$$\varepsilon = (2.8)\, E_g + 0.5 \qquad eV \tag{5}$$

If N_β and E_β are the incident beta flux and the average beta particle energy, respectively, then the maximum possible current that one can derive from a betavoltaic device is given by

$$J_{max} = q\, N_\beta\, (E_\beta / \varepsilon) \tag{6}$$

The maximum power delivered by a cell can be written as

$$P_{max} = J_{sc} Voc\, FF \tag{7}$$

Once a beta source and device structure are defined, J_{max} , r and Q can be calculated. Finally, Voc and FF can be calculated if the dominant loss current term(s) is identified. The overall efficiency is defined by

$$\eta = (P_{max}/P_{in}) \times 100 \quad (8)$$

where

$$P_{in} = q N_o E_\beta \quad (9)$$

where N_o refers to the number of beta particles emitted by the source per second, per square cm. of device area.

It is convenient to write the overall efficiency as a product of three terms, or efficiencies,

$$\eta = \eta_\beta \eta_c \eta_s \quad (10)$$

$$\eta_\beta = N_\beta / N_o \quad (11)$$

$$\eta_c = (1-r) Q \quad (12)$$

$$\eta_s = [Voc\, FF / \varepsilon] \times 100 \quad \% \quad (13)$$

The term η_β expresses the fraction of all betas created that are actually emitted from the source and directed towards the device, and is therefore referred to as the beta source efficiency. η_c is a coupling efficiency since it involves properties of both beta source and the semiconductor device. The term is designated as the semiconductor efficiency, since it determines the maximum possible efficiency that can be attained with a given semiconductor coupled to a particular beta source.

The maximum possible efficiency of a given betavoltaic system as a function of bandgap if one assumes that the semiconductor device is an ideal homojunction. In this case, the current loss term is dictated by minority carrier injection. Following Green (ref.8), we estimate that Jo can be written as

$$J_o = 1.5 \times 105 \exp(-E_g / kT) \quad A/sq.cm. \quad (14)$$

The fill-factor can be accurately calculated as follows (ref.5)

$$FF = [v_{oc} - \ln(v_{oc} + 0.72)] / [v_{oc} + 1], \quad voc = Voc/kT \quad (15)$$

Thus, once a given beta source is selected and an ideal device is assumed, the semiconductor efficiency (η_s) becomes a fuction only of bandgap. The potential efficiency of some systems will be examined after possible beta sources are considered. One can calculate an upper limit to betavoltaic device efficiency that is independent of the beta source, however. In particular, it can be shown that

$$\eta_s \leq Eg / \varepsilon(Eg) \quad (16)$$

This limiting value of ns is plotted in Figure 4 versus bandgap. Due to the functional dependence of the effective ionization energy, the limiting value of efficiency rises with bandgap and then levels off at a value slightly over 30 %. Thus, in principle, it is advantageous to utilize large bandgap devices. One must remember, however, that ideal cell behavior is being assumed.

BETA SOURCE CONSIDERATIONS

The process of selecting a beta emitter involves simultaneous consideration of isotope, half-life and the effects of radiation damage of semiconductor devices. To fabricate long lived power sources, it is clearlly desirable to utilize isotopes with long half-lives. On the other hand, since the beta flux derivable from a source material is inversely proportional to the half-life, the value of J_{max} and thus P_{max} are inversely proportional to the half-life. One must also consider the beta particle energies relative to the semiconductor radiation damage threshold (E_{th}). In general, it is preferable to have the maximum beta particle energy (E_{max}) less than E_{th}. Typically, E_{th} is on the order of 200 keV to 400 keV. Other key considerations are the availability of the radioisotope and the potential dose rate that might exist near the power source. Table 1 lists some possible beta emitters that meet some of the criteria that have been identified. Availability has become a key issue. The only isotopes that are readily available are tritium and Kr-85. Both are available in gaseous form, and tritium can be obtained in the form of tritiated Ti foils. If one were interested in one of the other isotopes, the Department of Energy would need to be consulted.

THEORETICAL EFFICIENCY OF Pm-147 AND Ni-63 BETAVOLTAICS

Considerable attention has been given to the use of Pm-147 and Ni-63 in betavoltaic systems. As noted above Pm-147 fueled batteries were actually reduced to practice. Ni-63 has been considered in the past because of its long half-life. The use of both of these isotopes is hindered because of the complex processes required to generate the isotope. Calculated efficiencies are considered here because of interest shown in these materials in the past, and for the purpose of illustration.

Figure 5 gives a plot of theoretical efficiency of Pm-147 fueled devices versus bandgap assuming ideal semiconductor junctions and bidirectional sources. By bidirectional sources, it is implied that the beta flux from both sides of a slab of beta emitting material is utilized. Due to the ideal cell assumption, the efficiency vs bandgap curve has a similar shape as the limiting efficiency curve given in Figure 4.

Figure 6A abd 6B describe calculated results for Ni-63 fueled cells. The device efficiencies are much lower in this case because of the beta source efficiency. As a result of the low beta energy, Ni-63 sources would suffer from effects of self absorption. The low values of current and power are results of the long half-life and low beta particle energy. Similar results are obtained when one considers properties of tritium fueled betavoltaic devices.

Pm-147 FUELED BETAVOLTAIC BATTERIES

The author led a program to develop Pm-147 fuelled betavoltaic batteries at the Donald W. Douglas Laboratories, Richland, WA , from 1968 to 1974. Pm-147 was available in the form of Pm_2O_3 from the U.S. Government. Custom made silicon cells were obtained from Heliotek (now Spectrolab) and from Centralab (now ASEC). The cells had n/p junctions with a mesa around the device periphery to minimize leakage currents at low voltages. The author benefited from interactions with Gene Ralph at Heliotek and Peter Isles at Centralab.

The basic approach to battery construction is illustrated by Figure 7. The n/p cells and beta sources were stacked in tandem so that the devices were connected in series. The Pm-147 sources actually consisted of Pm_2O_3 deposited onto Ta sheet. Thus the sources were unidirectional. Self-standing bidirectional Pm_2O_3 sources were under development when the program was terminated. Properties of a typical silicon cell coupled to a unidirectional source are described in Figures 8A and 8B. Batteries were typically designed with 5 mg/cm^2 Pm_2O_3 sources. Figure 9 shows a picture of three of the batteries that were made in reasonable quantities. Their properties are summarized in Table 2. These batteries were referred to as Betacel batteries.and were nominally 2 % efficient (overall efficiency). With bidirectional sources, they would have had efficiencies of 4 %. The Model 400 Betacel was considered seriously for powering heart pacemakers by companies in the United States and Germany. The short-circuit current and maximum power versus time for a typical Model 400 Betacel are plotted in Figure 10. Since the power required by the pacemaker circuitry was approximately 10 μwatts, the potential lifetime was ten years. Over 100 people received Betacel powered pacemakers, and many of the units lasted 10 years. Although the potential use of Betacel batteries for pacemakers appeared very promising, the lithium battery was developed about the same time. Lithium batteries lasted only 7 years, but since they were non-nuclear they were preferred by the pacemaker industry. The Betacel batteries were also utilized to a limited extent for military purposes.

CONCLUSIONS

Interest in the use of betavoltaic energy conversion seems to 'pop up' every few years. When the right application emerges, it may finally be utilized extensively. Many more choices are now available for the semiconductor cell than were available when the Pm-147 fuelled batteries were developed. Unfortunately, the choice of beta emitting material would appear to be more limited. To place the potential use of betavoltaic power sources in perspective, it is useful to estimate the power density versus time for some possible advanced systems. Figure 11 describes results of calculated properties of some advanced concepts. The thin film AlGaAs cells are assumed to be self standing devices. There are many other devices that one could consider. For example, GaP with an indirect bandgap, and thus potentially long diffusion length, would certainly be of interest for coupling to Pm-147. InP with its radiation resistant properties could be interesting for coupling to high energy beta emitters such as Tl-204. Nevertheless, the power density curves shown in Figure 11 can be used to make a few key points. Betavoltaic sytems should only be considered for low power applications. For example, if one is intersted in power levels on the order of one watt for ten years, it is clear that on the order of 1000 cm^3 of Pm-147 fueled devices must be considered. The size may not be a problem, but the cost might be

prohibitive. If one considers an application for which 1 milliwatt or 10 microwatts are required, tritium or Pm-147 fueled systems seem reasonable.

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TABLE 1 -- POSSIBLE BETA SOURCES

ISOTOPE	E_{max} (MeV)	$\tau_{1/2}$ (yr)	Maximum J_{SC} for Si (arbitary units)
H^3	0.018	12.3	3×10^{-3}
Ni^{63}	0.067	92	10^{-3}
Pm^{147}	0.230	2.62	1
Tl^{204}	0.765	3.75	17
Kr^{85}	0.670	10.9	28

TABLE 2 -- BETACEL CHARACTERISTICS

Characteristic	Model 50	Model 200	Model 400
Performance characteristics†			
Maximum power (μW)	50	200	400
Voltage at maximum power (V)	1·3	3·3	4·0
Open circuit voltage (V)	1·7	4·7	4·9
Short circuit current (μA)	45	72	112
Curies Pm^{147}	12	73	66
Efficiency (%)	1·0	0·7	1·7
Physical characteristics			
Diameter (cm)	1·52	2·03	2·29
(in)	0·60	0·80	0·90
Overall height (cm)	1·02	1·65	2·44
(in)	0·40	0·65	0·96
Mass (g)	17	55	98
Radiation dose rate at 2·5 cm from battery center‡			
BOL (mrem/h)	2·3	8·1	6·1
At end of 5 years (mrem/h)	1·2	4·3	3·3
5 year time averaged (mrem/h)	1·7	5·9	4·4

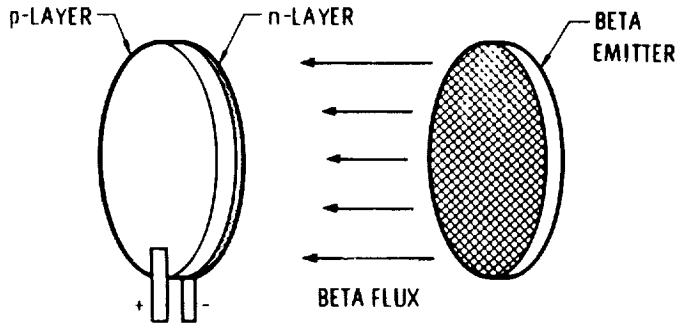
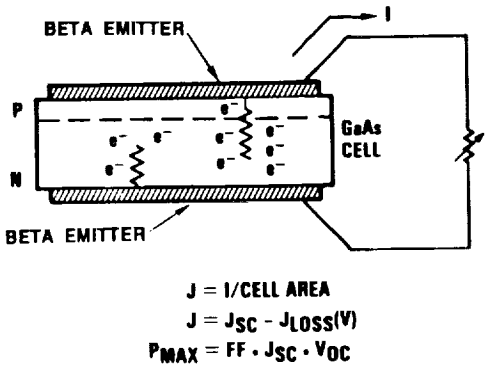
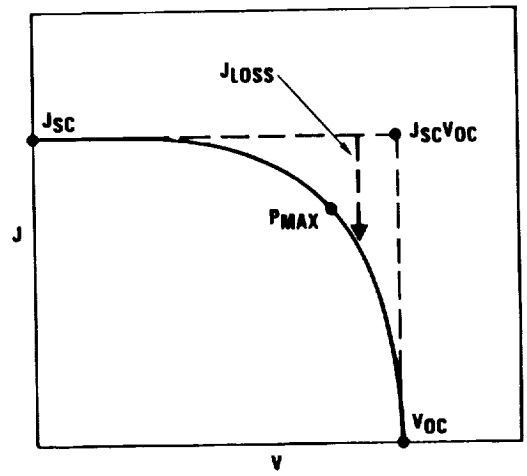
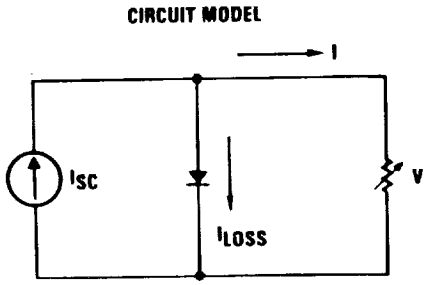


Figure 1. Basic Approach To Betavoltaic Energy Conversion.

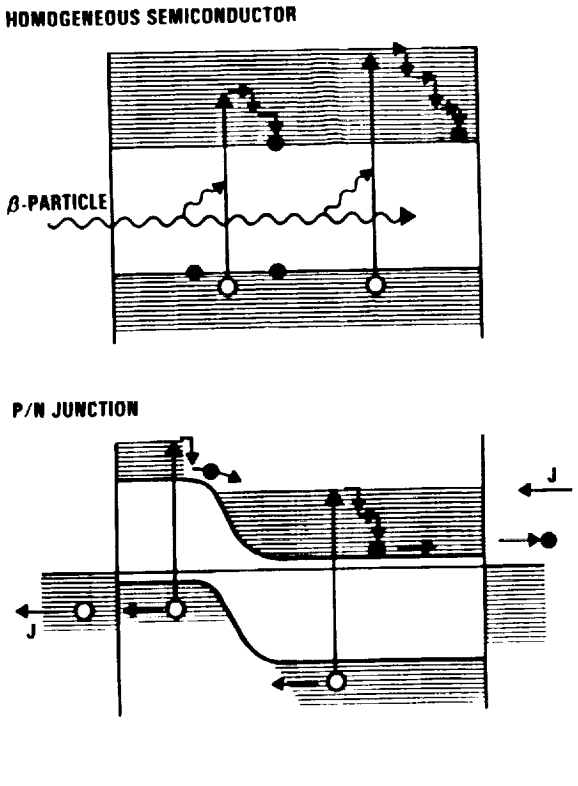






- High energy electrons (beta rays) produce electron/hole pairs in semiconductor cell
- Diode characteristics, J_{LOSS} (V), of junction determine the current, I, to external load
- J_{LOSS} (V) must be small in order to produce useful power

Figure 2. Betavoltaic Principles.



Electron-hole pairs

N_{EH} = (No electron-hole pairs/cm^2/sec)

$= N_\beta \left(\frac{\bar{E}_\beta}{\varepsilon}\right)$

ε = Effective ionization energy

$= (2.8)\, E_g + 0.5$ eV

N_β = Beta flux entering junction device

Maximum current

$J_{max} = I_{max}$/cell area

$= eN_{EH} = eN_\beta \left(\frac{\bar{E}_\beta}{\varepsilon}\right)$

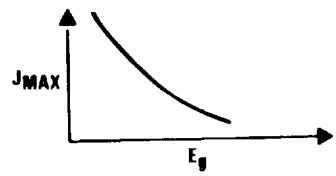


Figure 3. Considerations For Maximum Current.

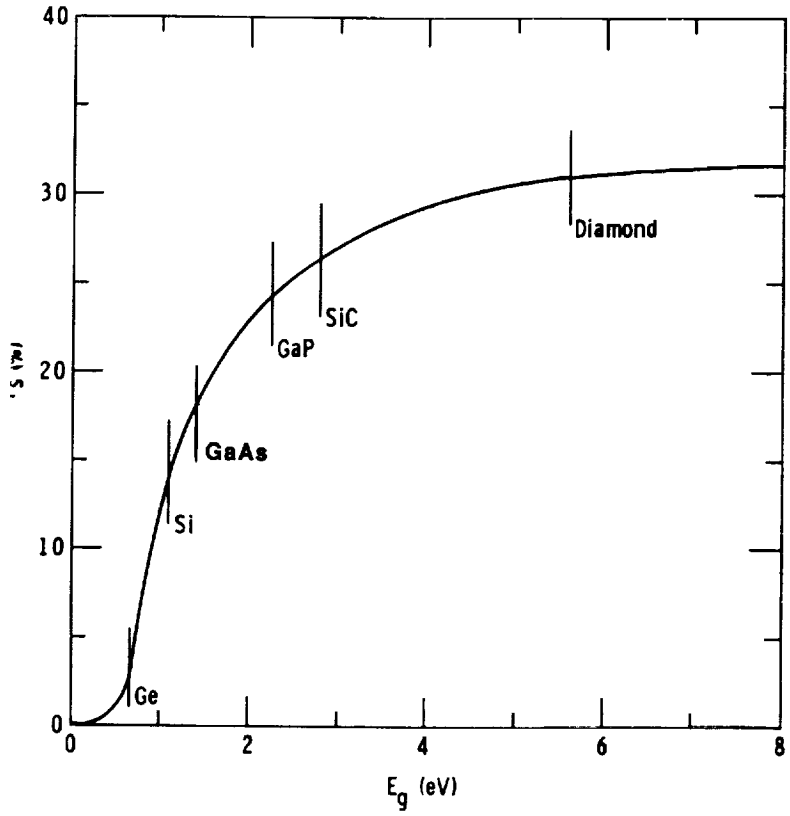


Figure 4. Limiting Betavoltaic Efficiency Versus Semiconductor Bandgap.

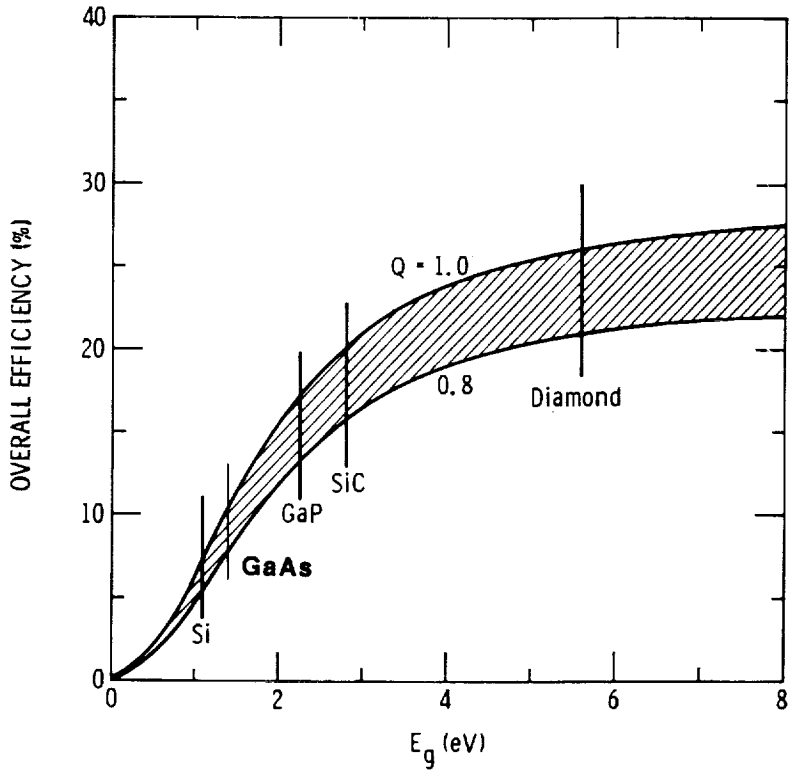
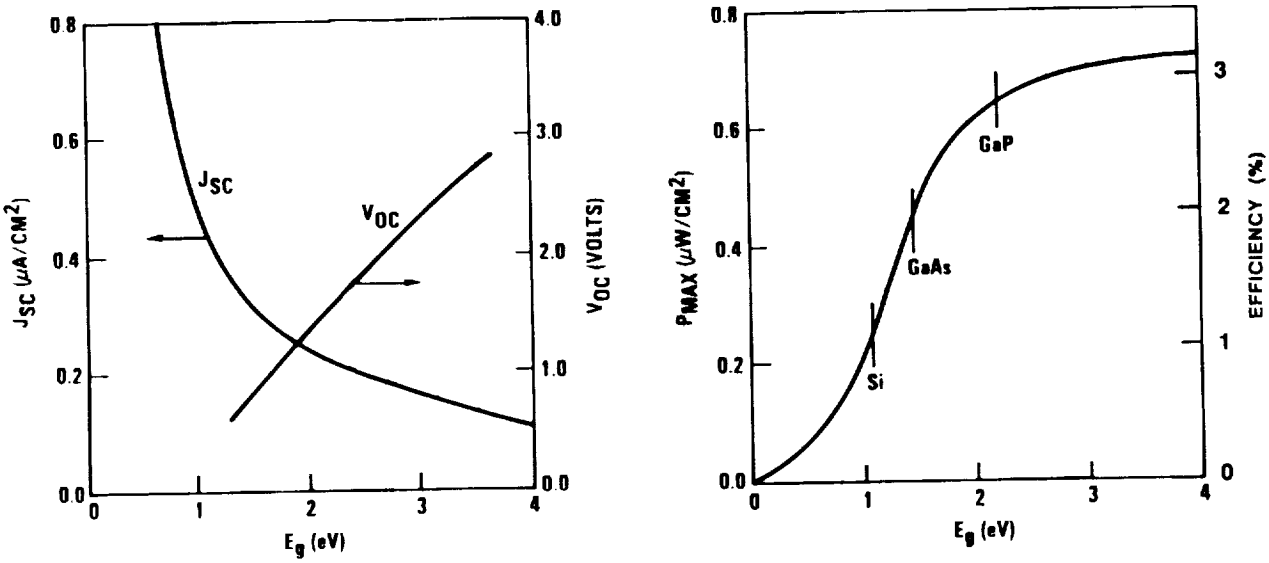


Figure 5. Calculated Efficiency For Pm-147 Betavoltaic Systems Versus Bandgap.



Assumptions: 1. Ideal Diode Characteristics
2. Two-Sided Beta Source With 100% Nickel 63

Figure 6. Calculated Properties Of Ni-63 Betavoltaic Systems Versus Bandgap.

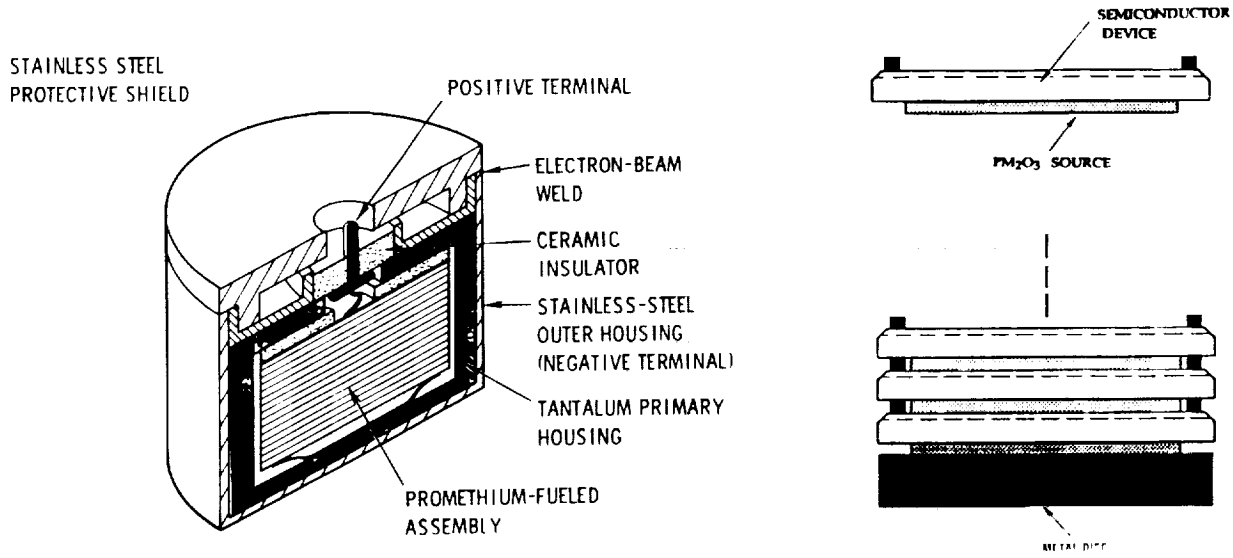


Figure 7. Approach Used For Betacel Construction.

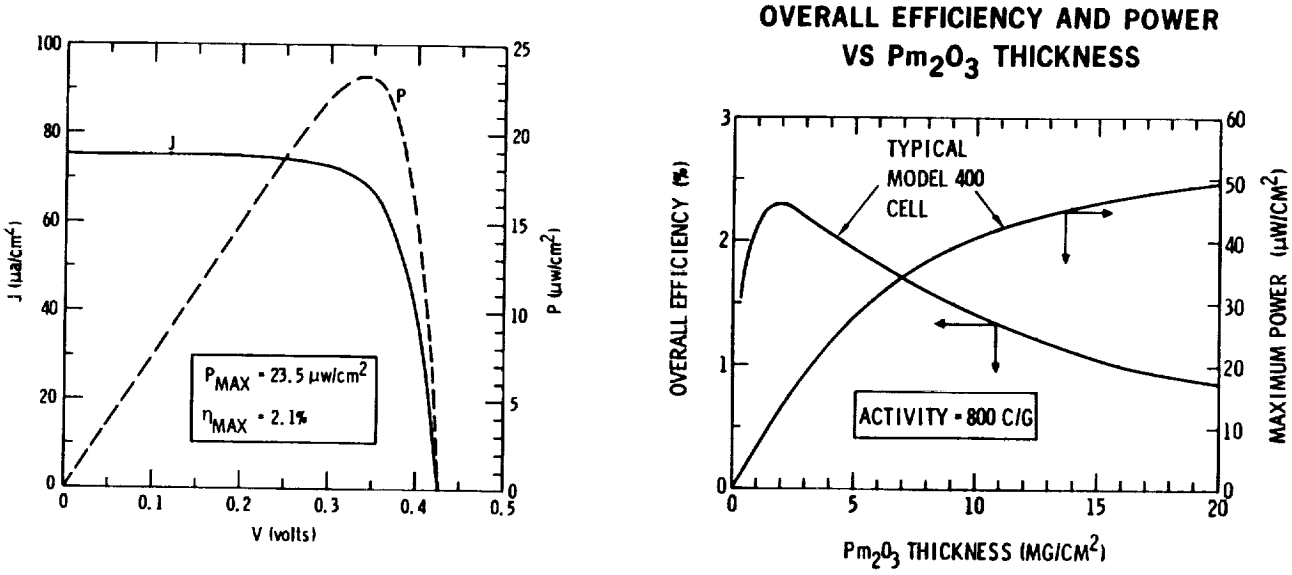


Figure 8. Betavoltaic Properties Of Silicon n/p Cells Coupled To Pm-147 Sources.

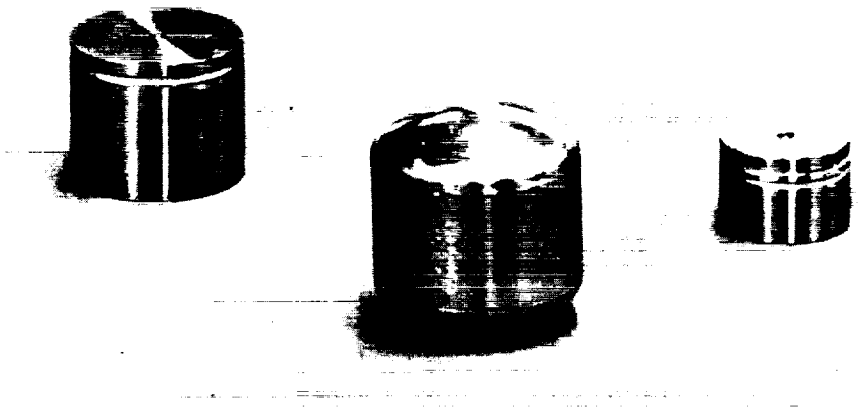


Figure 9. Betacel Batteries Developed At Donald W. Douglas Laboratories. Left to Right: Model 200, Model 400 and Model 50.

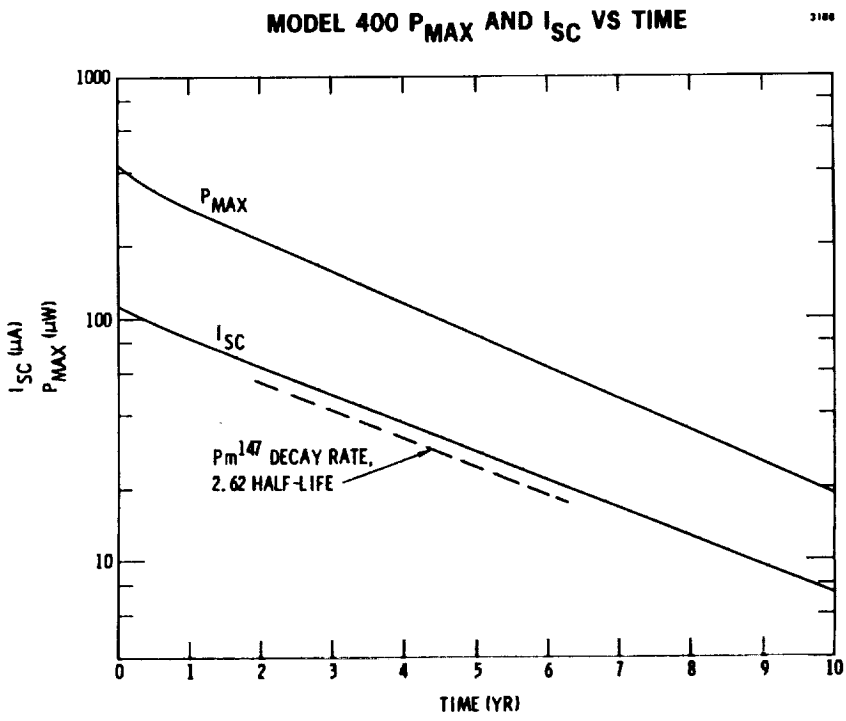


Figure 10. Short Circuit Current and Maximum Power Versus Time For a Model 400 Battery.

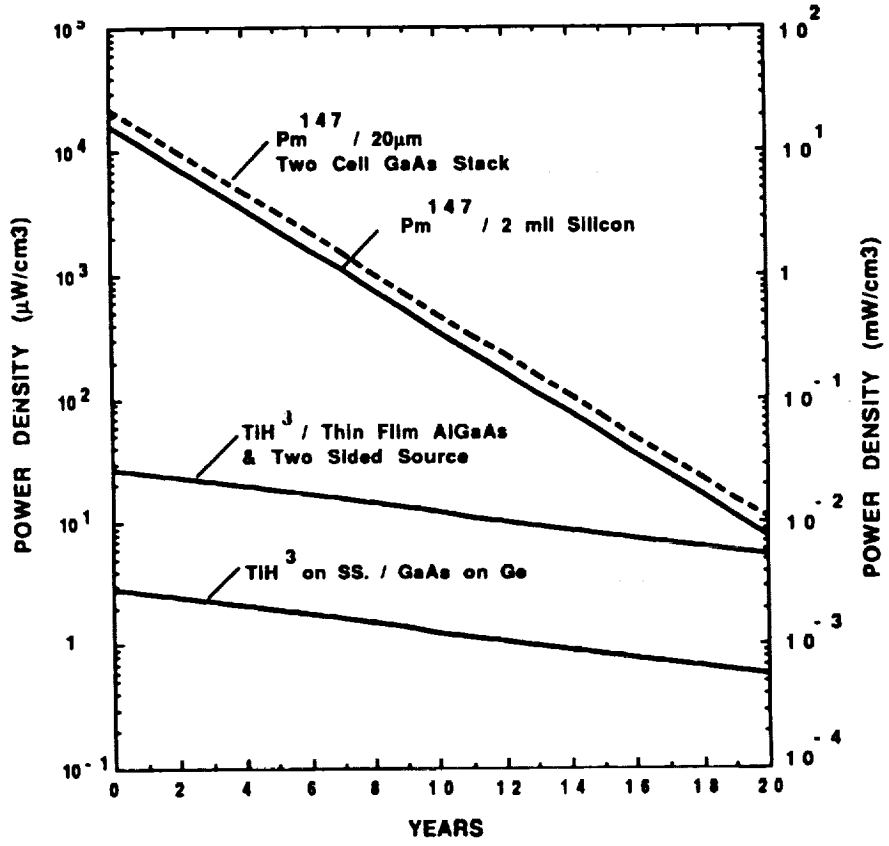


Figure 11. Power Density Versus Time For Advanced Betavoltaic Concepts.