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NUCLEAR ELECTRIC POWER SOURCES

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ABSTRACT

Measurements on radioactive commercial p-n junction silicon cells show that these units are capable of delivering several hundred microwatts per curie of Am^{241}-alpha source, indicating their usefulness in such electronic devices as hearing aids, heart pacemakers, electric watches, delay timers and nuclear dosimeter chargers. It is concluded that the Am^{241} sources are superior to the beta sources used previously, because of higher alpha specific ionization and simultaneous production of low energy photons which are easily converted into photoelectrons for additional power.

INTRODUCTION

There are a number of ways in which the kinetic energy of radiations emitted by the radioisotopes can be used to produce electric power. (1) The particle energy could be degraded into thermal energy by collisions with the stopping medium. The heat thus produced in the stopping medium can be transformed into electricity by dynamic heat engines such as Rankine and Brayton cycle turbogenerators. Or, the heat could be used to drive direct conversion devices such as thermoelectric converters. (2) The charged particles emitted by the radioisotopes could do work against an impeding electrostatic field, producing what might be called a primary nuclear battery. This arrangement, however, will require an independent power supply to provide the impeding field. (3) The particle kinetic energy could be used up in producing electron-hole pairs near p-n junctions in semiconductors or in a gaseous medium between two dissimilar materials. In both cases, the existence of a natural voltage gradient will produce a current flow. Such devices belong in the class of secondary nuclear batteries by virtue of the secondary processes that constitute the ultimate source of energy. (4) The particle kinetic energy could be converted to light pulses in phosphors such as CsI, NaI, and ZnS, etc. These light pulses could be used to activate solar cells in much the same way as sunlight. This technique of obtaining power constitutes what will be called tertiary nuclear battery.

Of all these energy conversion techniques, the first one has been developed the most, mainly due to the powerful momentum built up by the heat engines during the Project Feedback studies, the Prototype Mound Laboratory Experiments, and the early SNAP work. The nuclear batteries - primary, secondary, and tertiary - have all so far been little more than laboratory curiosities, mainly due to their low output power. Yet the first radioisotope generator was a direct charging type of nuclear battery! There is no fundamental technical reason why the nuclear batteries must be confined to the low efficiency and power levels. The researchers have always felt somewhat frustrated because they have not been able to make more effective use of the high energy, electrically charged particles emitted by most of the radioisotopes. It is

the purpose of this report to explore the possibilities of developing more efficient nuclear power supplies based on the principles of secondary and tertiary batteries mentioned in techniques 3 and 4 above.

PRINCIPLES OF OPERATION

Secondary Nuclear Battery

(a) (p-n) junction cell.- The energy band diagram of a (p-n) junction is shown in figure 1. The Fermi level which is the electrochemical potential of electrons must be constant throughout the crystal, so that at the junction, the bands follow the shape given in the figure. A potential difference across the junction acts as a barrier to both electrons and holes. When nuclear radiations strike the p-n cell, they create electron hole pairs in the junction region. The junction separates the electrons and holes to their proper terminals constituting a cell current through the external circuit and thus making useful electrical energy available. For the most efficient collection of the current carriers, it is necessary that they be produced in the vicinity of the junction and that the junction region be free from defects which may act as traps. In contrast to the solar cells for use with the sunlight, the junction of the p-n cell for use with nuclear radiation does not have to be close to the incident surface, thereby reducing I^2R internal loss. As a matter of fact, the junction can be located at the end of the range of the incident radiation. However, the radiation which produces current carriers also causes damage to the cell thereby reducing the output power. The damage can be eliminated if the nuclear radiation interacts with a phosphor whose output light then activates the junction cell. This usually costs appreciably in power output available from a given radioisotope. However, a judicious choice of the radioisotope - such as Am^{241} - can partially offset this shortcoming as will be discussed later under tertiary batteries.

(b) Ionization-chamber nuclear cell.- This type of power supply involves two dissimilar materials - say copper and zinc - embedded in a gaseous medium which can be ionized by the nuclear radiation. The contact potential between the two dissimilar materials provides an electrostatic field in the region between the plates. Figure 2 shows a schematic diagram of this type of cell. In order that the incident particle spend its entire energy in producing electrons and ions in the region where electrostatic field exists to separate them, it is necessary to properly adjust the gas pressure as well as the spacing between the electrodes. It is also necessary to choose a gas with low electron-ion recombination loss coefficient. This type of battery has the advantage of freedom from radiation damage which plagues the p-n junction cell.

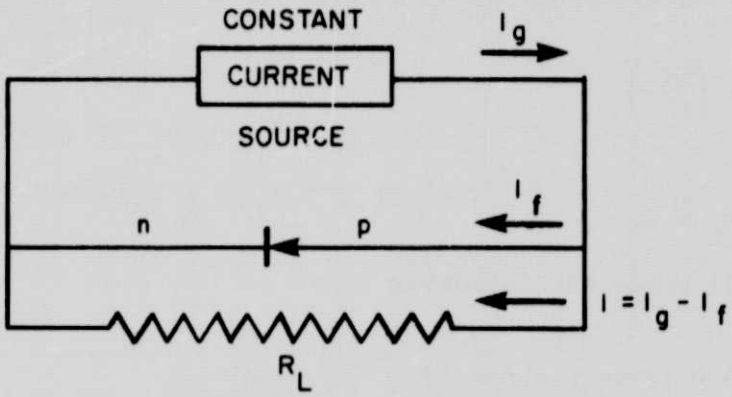
Tertiary Nuclear Battery

In this technique, use is made of the fact that nuclear radiations produce scintillations in certain materials called phosphors. These scintillations, when allowed to fall on solar cells, produce a current through an external

circuit in much the same manner as the sunlight. Since the solar cell is not directly exposed to the nuclear radiation, this arrangement involves no radiation damage to the cell. However, the efficiency in this arrangement is necessarily less than that for a solar cell alone since only about 15 percent of the incident particle energy can be converted into light pulses inside the phosphor. Choice of a suitable phosphor and a radionuclide such as Am^{241} can partially remove this loss of efficiency. As seen from figure 4, Am^{241} decays by α-emission to excited states of Np^{237} thereby producing a 60 keV gamma ray at the same time. By choosing a high atomic number phosphor - such as CsI(Na)(*) it can be assured that most of these γ-rays will produce photoelectrons inside the phosphor. Some of these photoelectrons can produce scintillations inside the phosphor in the same way as the alpha particles do. The escaping photoelectrons will strike the p-n junction and produce a current in the manner analogous to a beta cell. Because of the low energy of these photoelectrons, the radiation damage to the junction caused by them will be minimal. Figure 4 shows a schematic diagram for this type of cell.

ANALYSIS OF THE JUNCTION CELL

The three concepts of nuclear cells described above are characterized by a region where a strong electrostatic field exists to separate the electron-hole pairs produced by the incident nuclear radiation. An equivalent circuit for such a high field region can be drawn as follows:



(*)The use of this phosphor was decided upon because its light output is the highest amongst high Z phosphors.

The following equation describes this circuit adequately (ref. 1).

$$\frac{kT}{eR_o} R_L \left\{ \left(\exp \frac{eV}{kT} \right) - 1 \right\} + V = I_g R_L \qquad (1)$$

(Various symbols have their usual significance.)

Maximizing the power in the external circuit, one obtains the following expression:

$$I_g = \frac{kT}{eR_L} \ln \left(\frac{R_o}{R_L} \right) + \frac{kT}{e} \left(\frac{1}{R_L} - \frac{1}{R_o} \right) \qquad (2)$$

where R_o = zero voltage junction resistance obtained from its I-V characteristics in the absence of radio-activity.

$\simeq 10^5 \rightarrow 10^8$ ohms-cm^2 for silicon junctions

If we put $I_g/I_o = G$

where I_o = reverse saturation current

$$= \left(\frac{kT}{eR_o} \right)$$

and $R_o/R_L = Z$,

equation (2) takes the following form:

$$G = (Z \ln Z + Z - 1) \qquad (3)$$

The accompanying figure gives the relationships between G and the various relevant parameters of the Alpha Voltaic Cell (AVC). From this figure, it is evident that the junction cell works most efficiently only for G in excess of 10^4. For a zero voltage junction resistance of 10^8 ohms-cm^2, this corresponds to an Am^{241} alpha source of about 270 μc incident on the cell.

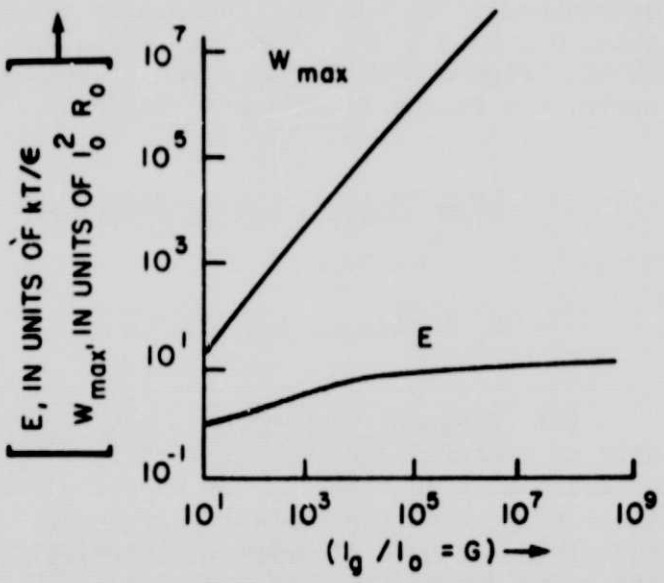
$$W_{max} = I_o^2 R_o Z (\ell nZ)^2$$

E = Ideal Efficiency

$$= \left(\frac{V^2/R_L}{I_g \frac{\varepsilon}{e}}\right) = \left(\frac{eV}{\varepsilon}\right)\left(\frac{I}{I_g}\right)$$

$$= \left(\frac{kT}{\varepsilon}\right)\left(\frac{Z}{G}\right)(\ell nZ)^2$$

where ε = energy required to produce one electron-hole pair



DEPENDENCE OF POWER OUTPUT AND EFFICIENCY OF THE JUNCTION CELL ON G.

The maximum power delivered to the external load under those conditions would be about 200 μwatts. Two important limitations to these estimates should however be pointed out:

1. To provide 270 $\mu c/cm^2$ on the cell, the source concentration will have to be 540 $\mu c/cm^2$ which will necessarily result in self-energy degradation.

2. Zero-voltage resistance of 10^8 ohms-cm^2 is an upper limit on the silicon solar cells. Impedances of $10^5 \rightarrow 10^7$ ohms are more commonly available.

In view of these practical limitations, it is unlikely that one would attain the maximum ideal efficiency of about 8 percent(*) as indicated in the above figure for G in excess of 10^4.

EXPERIMENTAL RESULTS

(a) p-n junction cell.- A 360 microcuries Am^{241} alpha source of surface area 1 cm^2, was brought against the face of a commercial p-n (n/p) silicon solar cell of planar area 1 cm^2 in the manner shown in figure 5. From the I-V characteristics of the cell, its zero voltage resistance, R_o, was

(*)Rappaport et al. (ref. 2) have analyzed electron voltaic effect in silicon p-n junctions and calculated a maximum conversion efficiency of about 5 percent for incident (Sr 90 - Y 90) beta particles.

determined to be 220 kΩ. The source alpha particles ranged in energy from about 0.1 to 3.5 MeV. The load resistance, R_L, was varied from 10 kΩ 10 MΩ. Figure 6 shows the relation between R_L and V^2/R_L. From this curve, the following data are obtained:

$$V^2/R_L \text{ (max)} = 7 \times 10^{-9} \text{ watt/cm}^2$$

$$R_L \text{ (optimum)} = 45 \text{ k}\Omega$$

(b) Ionization-chamber nuclear battery.- A 270 μc Am-241 α source, with an average alpha energy of ~1.80 MeV, was located between two semi-cylindrical electrodes in the manner shown in figure 7. On the basis of their high contact potential difference, one electrode was made of zinc while the other was made of copper. The electrodes were first located in air and the power across the load resistor, (I^2R_L), was measured as a function of R_L when the Am^{241} source was moved between them. Figure 8 shows the relationship between R_L and I^2R_L. From the data in this figure, the following results are obtained:

$$I^2R_L \text{ (max)} = 2 \times 10^{-11} \text{ watt/cm}^2$$

$$R_L \text{ (optimum)} = 1200 \text{ M}\Omega$$

Next, air was replaced successively by nitrogen, methane, and argon and the above measurements were repeated. The maximum external power was obtained with argon at a pressure(*) of about 10 lbs/in.^{2}.

$$I^2R_L \text{ (max)} = 5 \times 10^{-11} \text{ watt/cm}^2$$

$$R_L \text{ (optimum)} = 1200 \text{ M}\Omega$$

(c) p-n junction cell with CsI(Tℓ) crystal.- The experimental arrangement for this case was similar to that for p-n junction cell except for the presence

(*)This value of the gas pressure was dictated by the electrode size and the range of the alpha particles at the reduced pressure. For maximum power output, the alpha particles should be stopped between the electrodes.

of a CsI (Tℓ) crystal coupled to the solar cell face. Figure 9 shows the experimental arrangement used to obtain external power versus external load resistance graph shown in figure 10.

From figure 10 the following results are obtained:

$$V^2/R_L \text{ (maximum)} = 1.25 \times 10^{-11} \text{ watt/cm}^2$$

$$R_L \text{ (optimum)} = 65 \text{ k}\Omega$$

From the measured values of maximum power obtained from the three types of batteries, power conversion efficiencies have been calculated and are given in the table below:

Table I.- Summary of Experimental Results on Nuclear Batteries

These results are applicable to p-n junction silicon cells with R_o = 220 kΩ and power conversion efficiencies of 7.5 and 15.0 percent for sunlight, respectively.

No.	Type of battery	Relative efficiency with respect to solar cell	W (max)/curie	
			p-n junction efficiency 7.5%	p-n junction efficiency 15.0%
1	(p-n) function cell	4.70 percent	120 μW	240 μW
2	Ion-chamber cell (argon)	0.04 percent	1.0 μW	1.0 μW
3(a)	(p-n) junction + (CsI(Tℓ))(*)	0.012 percent	0.3 μW	0.6 μW
3(b)	(p-n) junction + CsI(Na)(**)	0.10 percent	2.5 μW	5.0 μW

(*)CsI(Tℓ) crystal efficiency of 5 percent, giving source current, $I_g = 2.4 \times 10^{-8}$ amp.
(**)CsI(Na) crystal efficiency of 15 percent, giving source current, $I_g = 1.4 \times 10^{-7}$ amp.

It should be pointed out that these batteries are not intended to compete with the solar cells. They are intended for use where the solar cells cannot be used for lack of sunlight or other reasons such as miniaturization requirements and continuous uninterrupted operation.

DISCUSSION

Results on secondary and tertiary nuclear batteries using a (120 μc) effective Am^{241} alpha source as the ionizing radiation have been described. For a silicon junction with R_o = 220 kΩ, the radioactive junction cell output is about 4.7% of the photovoltaic cell under sunlight and a factor of 400 lower for the (junction + CsI(Tℓ) crystal). (This factor of 400 reduction arises from the fact that the light conversion efficiency (LCE) of the CsI(Tℓ) crystal used was only 5 percent and no attempt was made to prevent the loss of scintillation light from the sides and the front face of the crystal. A factor of 3 → 4 increase in LCE and an almost total light collection using a selected and properly mounted CsI(Na) crystal is easily possible, thereby increasing the relative efficiency to about a quarter of that of the radio-junction alone.) The relative power output of the radioactive junction and its matching load resistance are strong functions of the source current and the junction impedance. Using a junction resistance of - say 10^7 ohms - one expects the relative junction efficiency to rise to about 28 percent of the photovoltaic cell, i.e., 1440 microwatts/curie of Am^{241}. A (junction + CsI(Na) crystal) combination will need less than 4 curies of Am-241 to produce comparable power. For an ionization chamber battery, the efficiency is only about 1 percent of the junction cell - mainly due to recombination loss at high pressures used in the present study. Using appropriately low pressures, the recombination coefficient could be lowered by a factor of about 5 (ref. 3) thereby increasing the source current correspondingly. However, the source impedance for this arrangement is extremely high, making it only suitable as a low voltage reference source.

CONCLUSIONS

Measurements on radioactive commercial p-n junction silicon cells indicate that these units are capable of delivering several hundred microwatts per curie of Am^{241} alpha source, indicating their usefulness for such electronic applications as hearing aids ($\simeq$ 1 milliwatt), heart pacemakers (50 → 200 microwatts) and electric watches ($\simeq$ 10 microwatts). Other areas of application include dosimeter chargers, delay timers, and electricity storage systems. The dc-dc conversion techniques can be used to increase the nuclear battery output voltage to the levels required for the device operation. It should of course be recognized that nuclear batteries are suited to some but not all battery applications. They are vastly superior to chemical batteries in energy storage potential; they can have much longer life time and be much more reliable than other batteries and they are ideal for relatively inaccessible installations. For low power level, reference voltage sources in remote, inaccessible places, the ion-chamber cell is recommended. These cells can deliver approximately 3 millivolts per microcurie of Am^{241}.

The use of Am^{241} radiation source is superior to the commonly used beta sources (refs. 4, 5) in tertiary nuclear batteries, because of higher specific ionization by alpha particles, simultaneous production of low energy photo-electrons and easier shielding requirements. The use of an alpha source in conjunction with an efficient CsI(Na) crystal minimizes the radiation damage that necessarily occurs in radioactive junction cells. The reduction in power due to less than 100 percent light conversion efficiency of CsI(Na) crystals can be made up by using a stronger alpha source. The shielding requiremenents even for these stronger alpha sources are such that these batteries will be light enough for easy implantation. An additional gain in power can be obtained by using a CdS solar cell whose spectral response matches better with the CsI(Na) scintillation spectrum.

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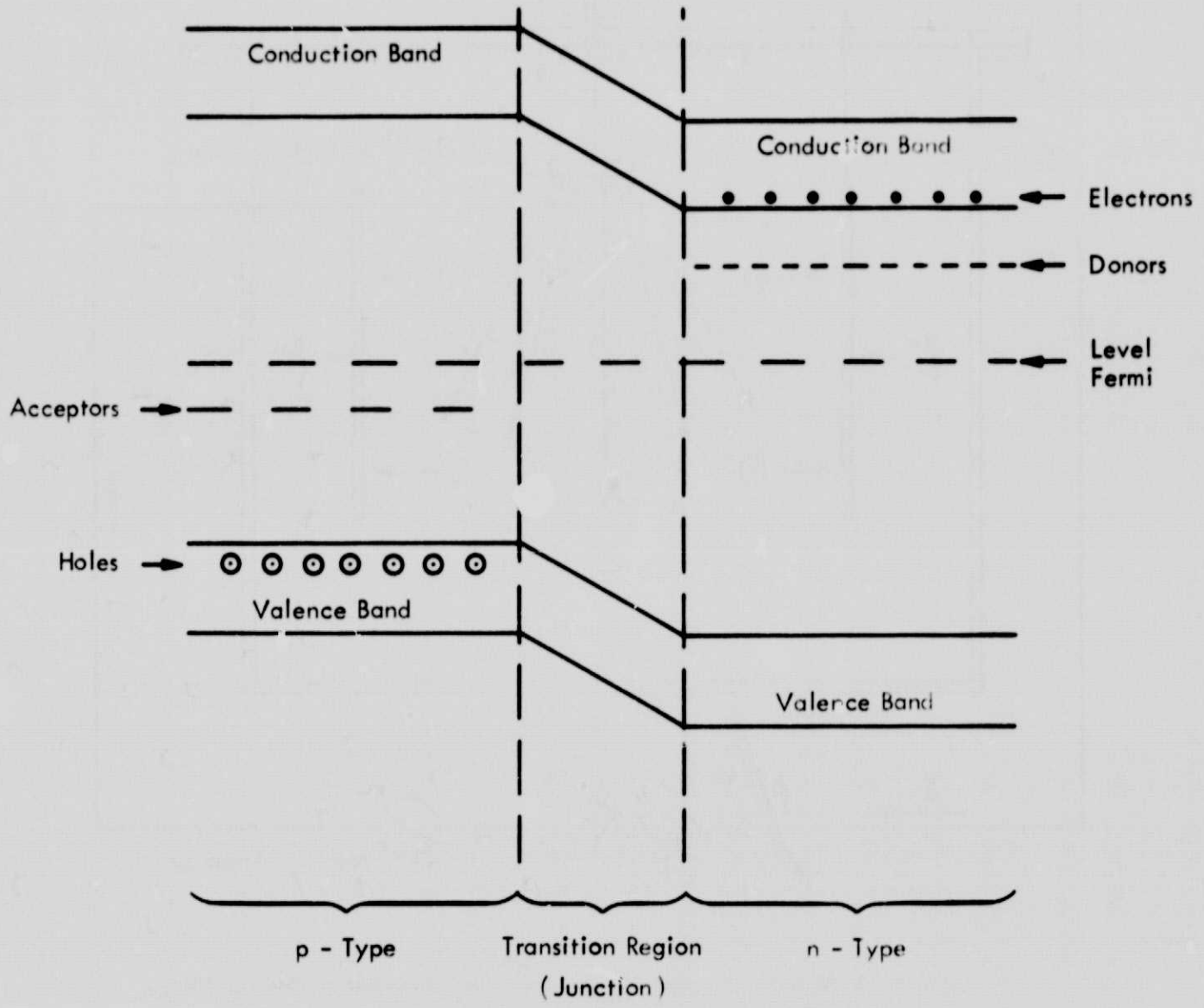


Figure 1. - Band diagram of a p - n junction at room temperature.

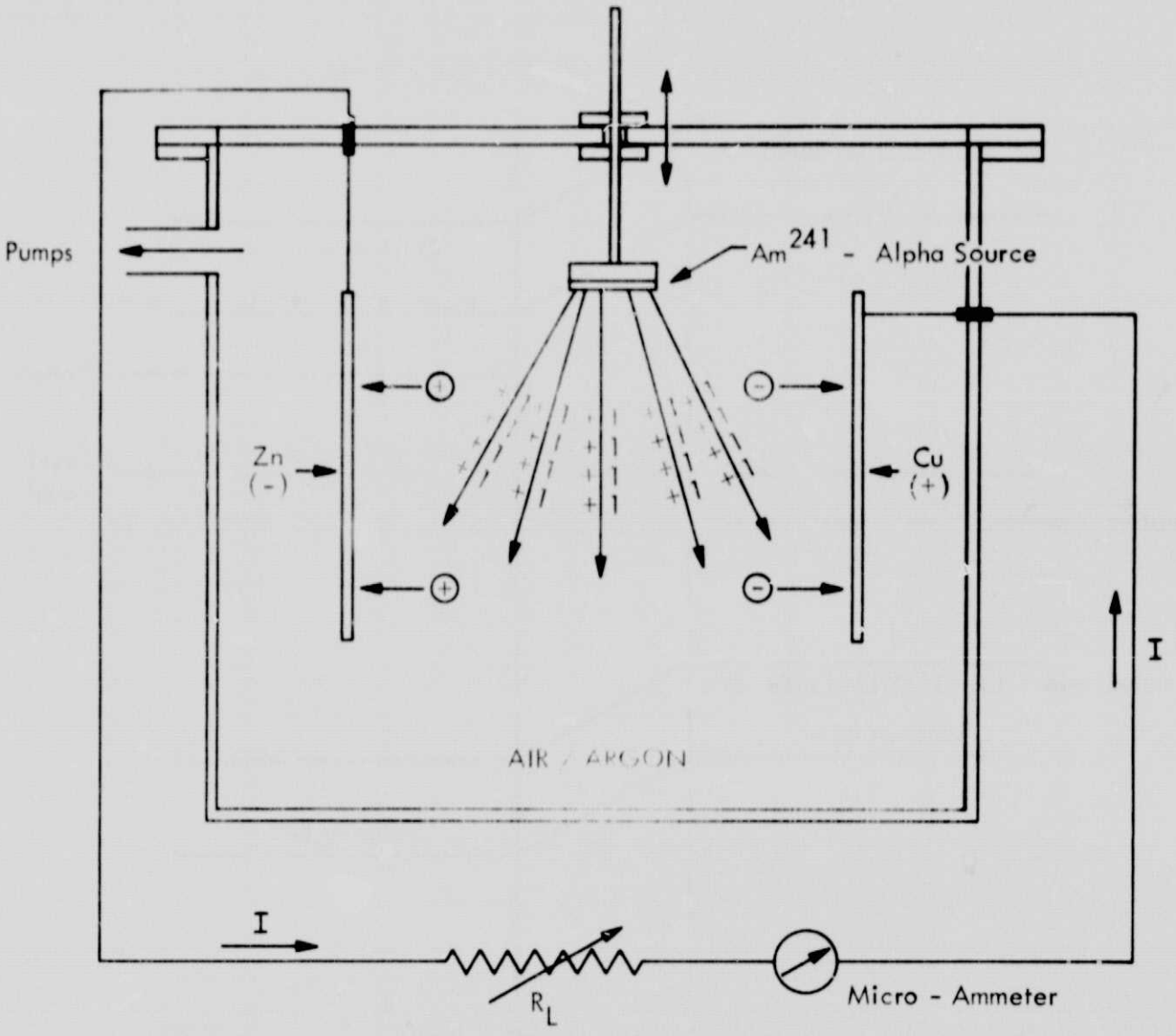
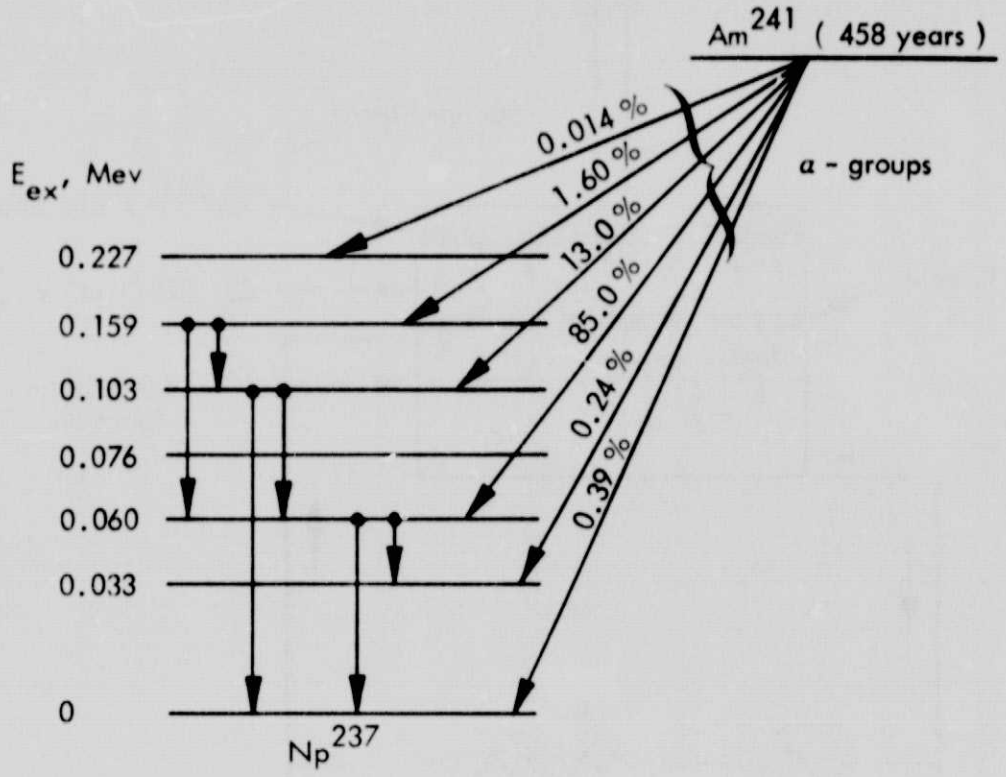


Figure 2. - Schematic diagram of an ionization chamber nuclear battery.



(Energy Levels Are Not Drawn to Scale).

Major Alpha Groups:

5.477 Mev - (85.0 %)
5.435 Mev - (13.0 %)
5.378 Mev - (1.6 %)

Major Gamma Groups:

60 Kev - (91.3 %)
27 Kev - (7.4 %)

Figure 3. - Energy Level Diagram of ($Am^{241} \xrightarrow{\alpha} Np^{237}$) Decay.

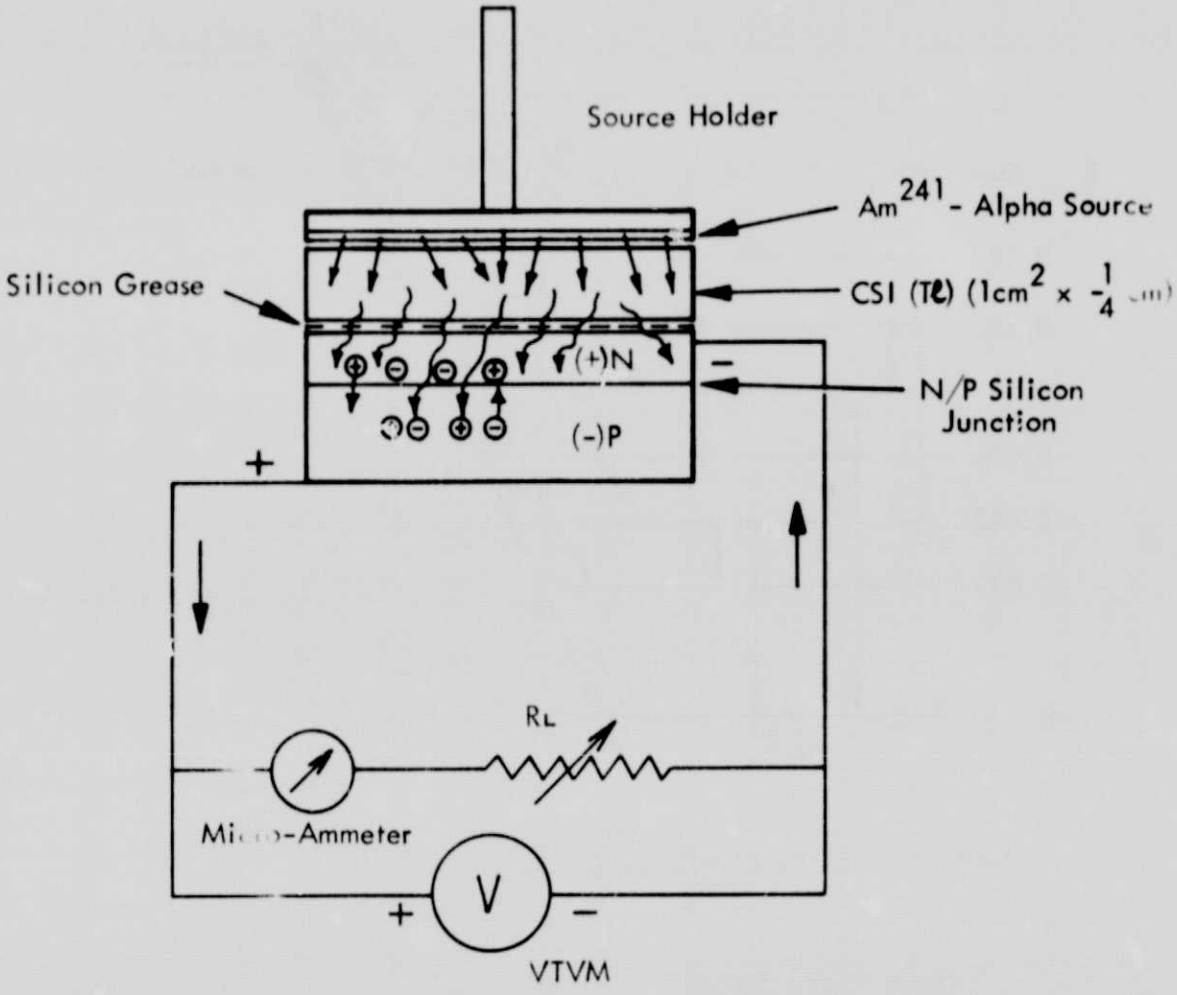


Figure - 4. Schematic Diagram of (N/P Junction + CsI) Nuclear Cell.

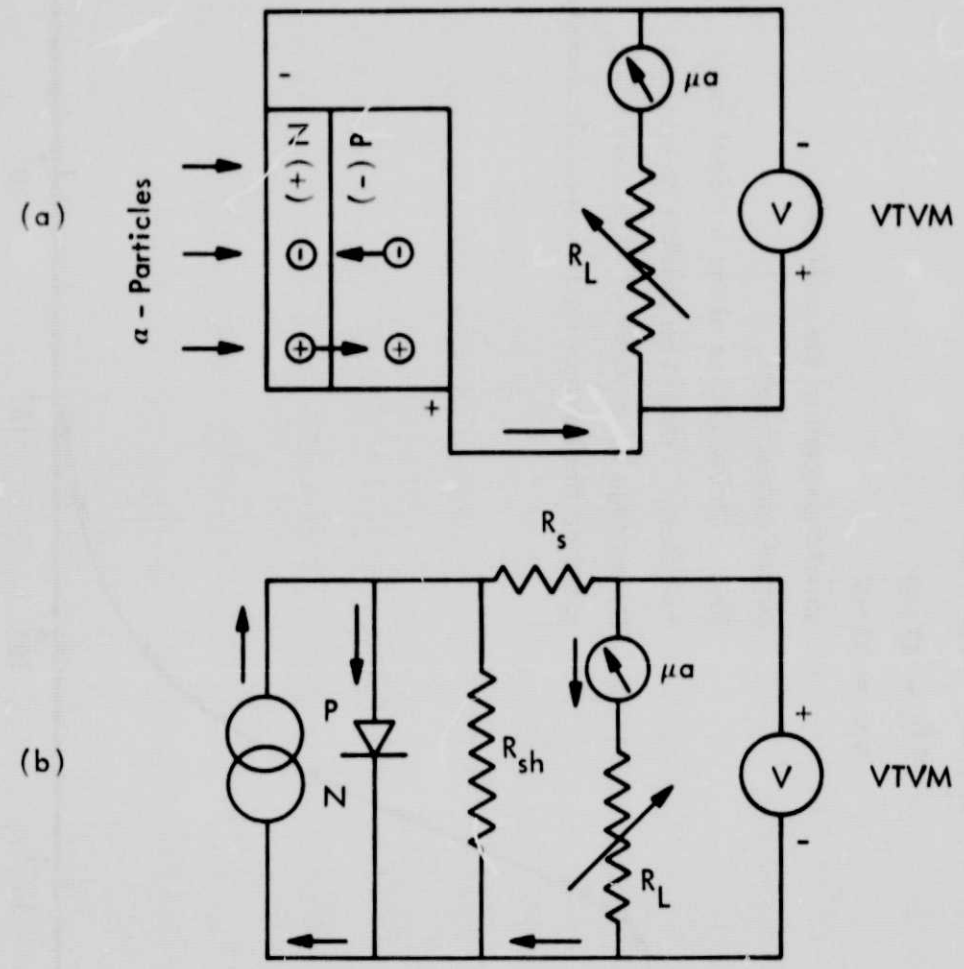


Figure 5. - (a) Experimental arrangement for measuring external power output from N / P junction cell.

(b) Equivalent circuit diagram for junction cell. (R_{sh} = Shunt Impedance; R_s = Series Resistance)

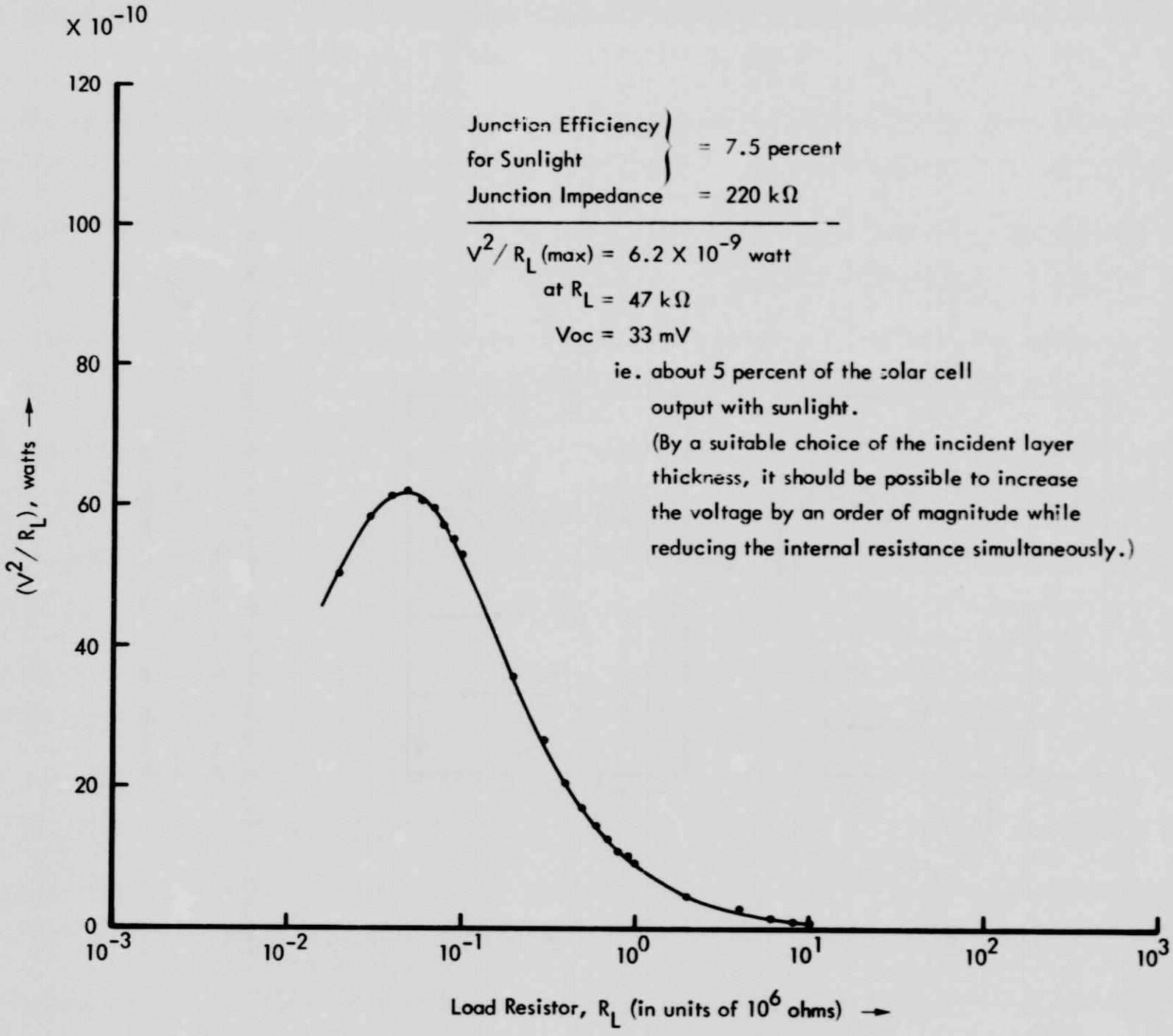


Figure 6. - External power output as a function of load resistor in P - N junction cell.

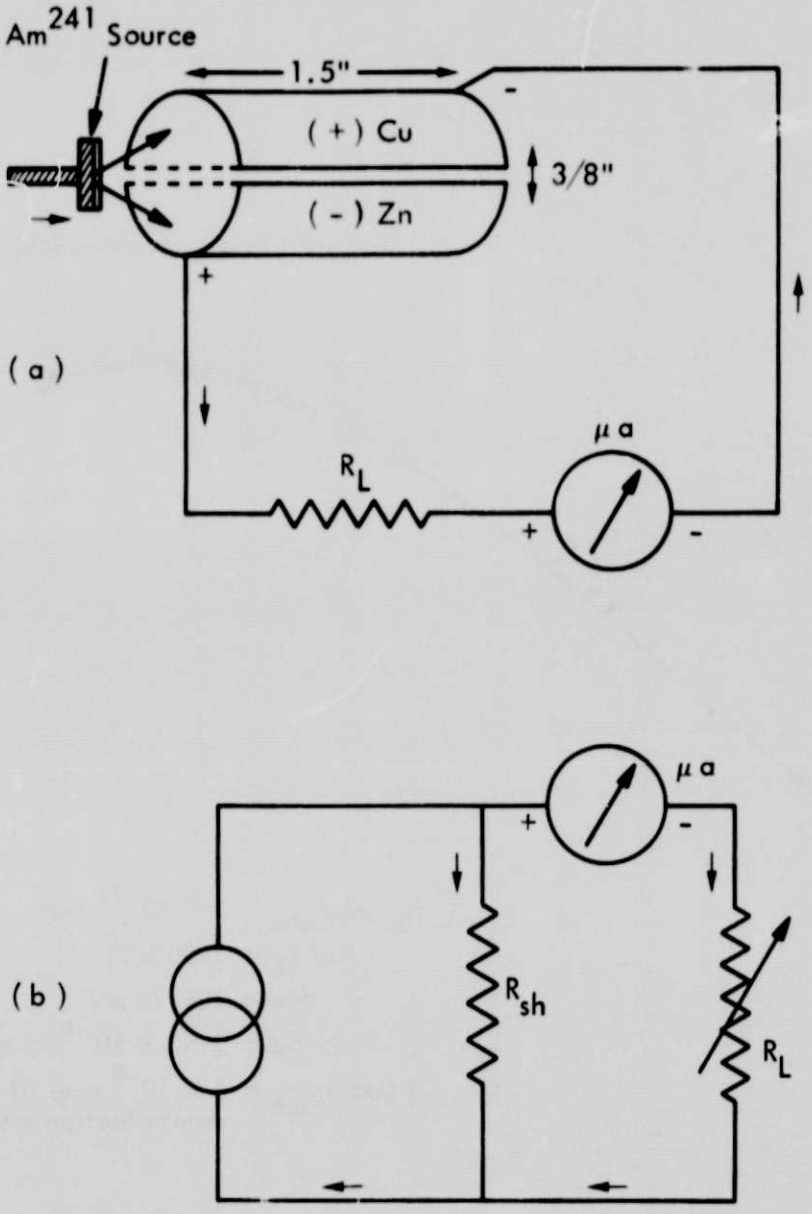


Figure 7. - (a) Experimental set up for measuring external power from an ion - chamber battery.
(b) Equivalent circuit for (a). (R_{sh} = Shunt impedance due to ion - chamber.)

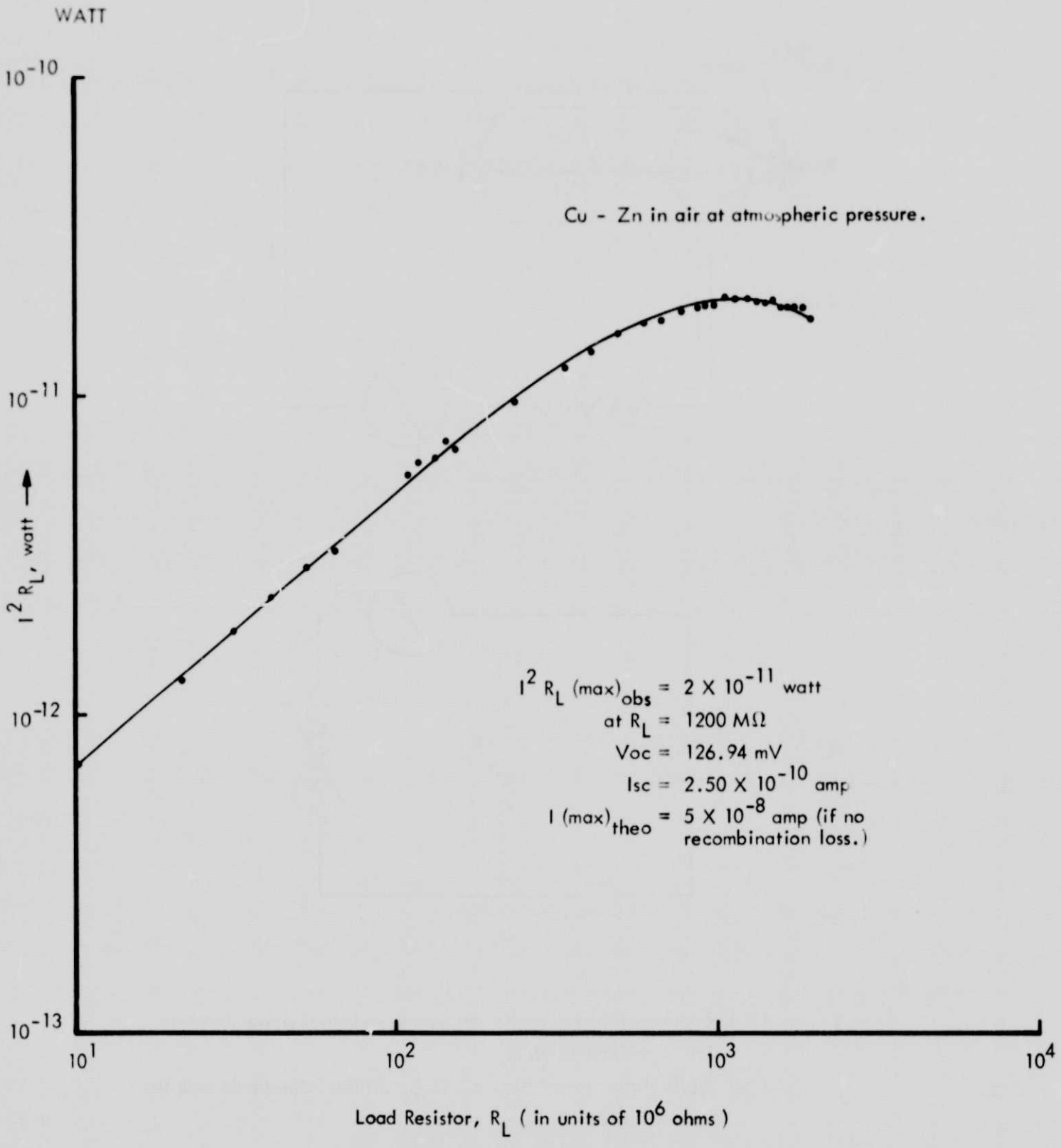


Figure 8. - External power output vs load resistor in the ionization chamber battery.

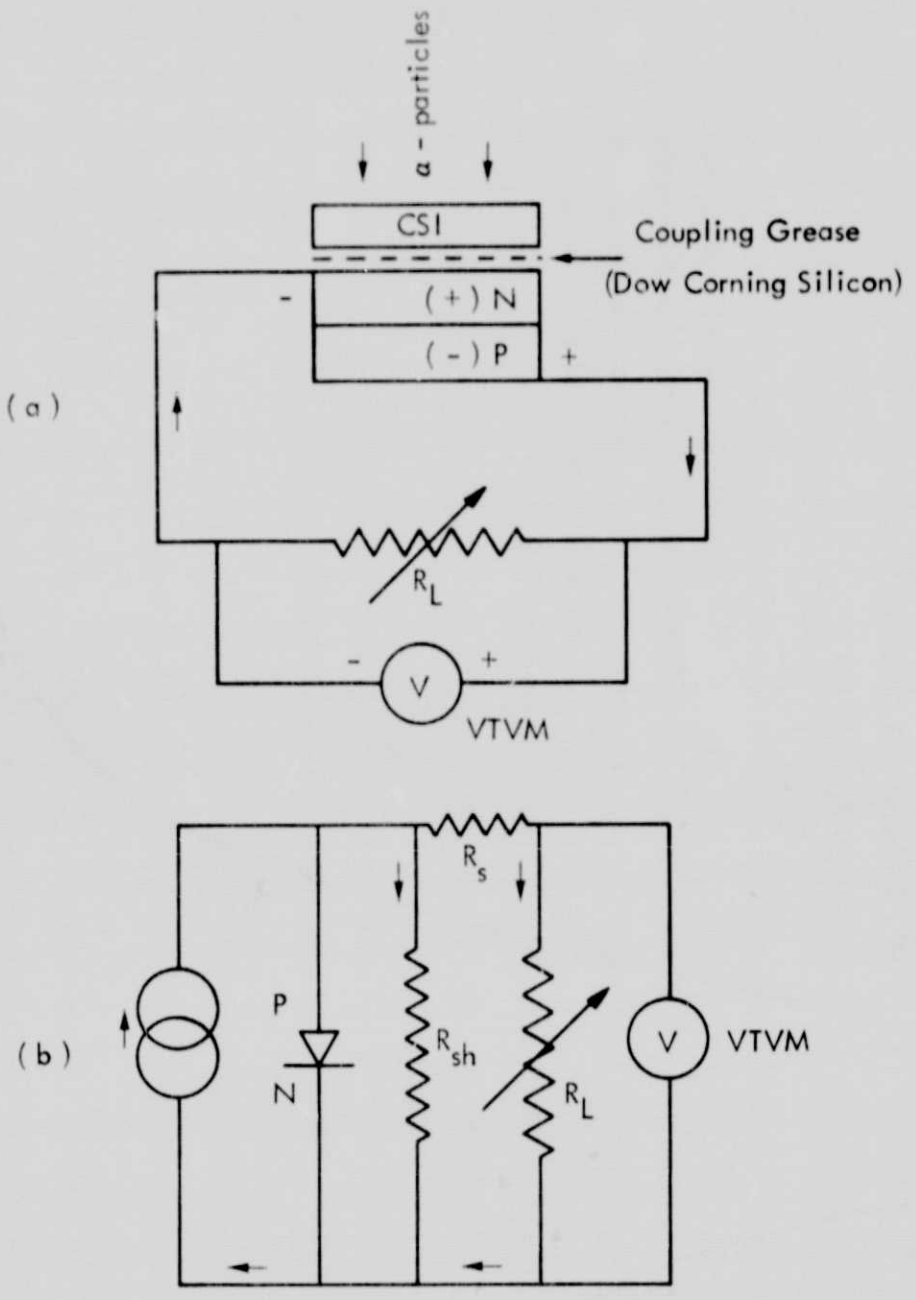


Figure 9. - (a) Experimental arrangement for measuring output power from a tertiary battery.

(b) Equivalent circuit for 9 (a).

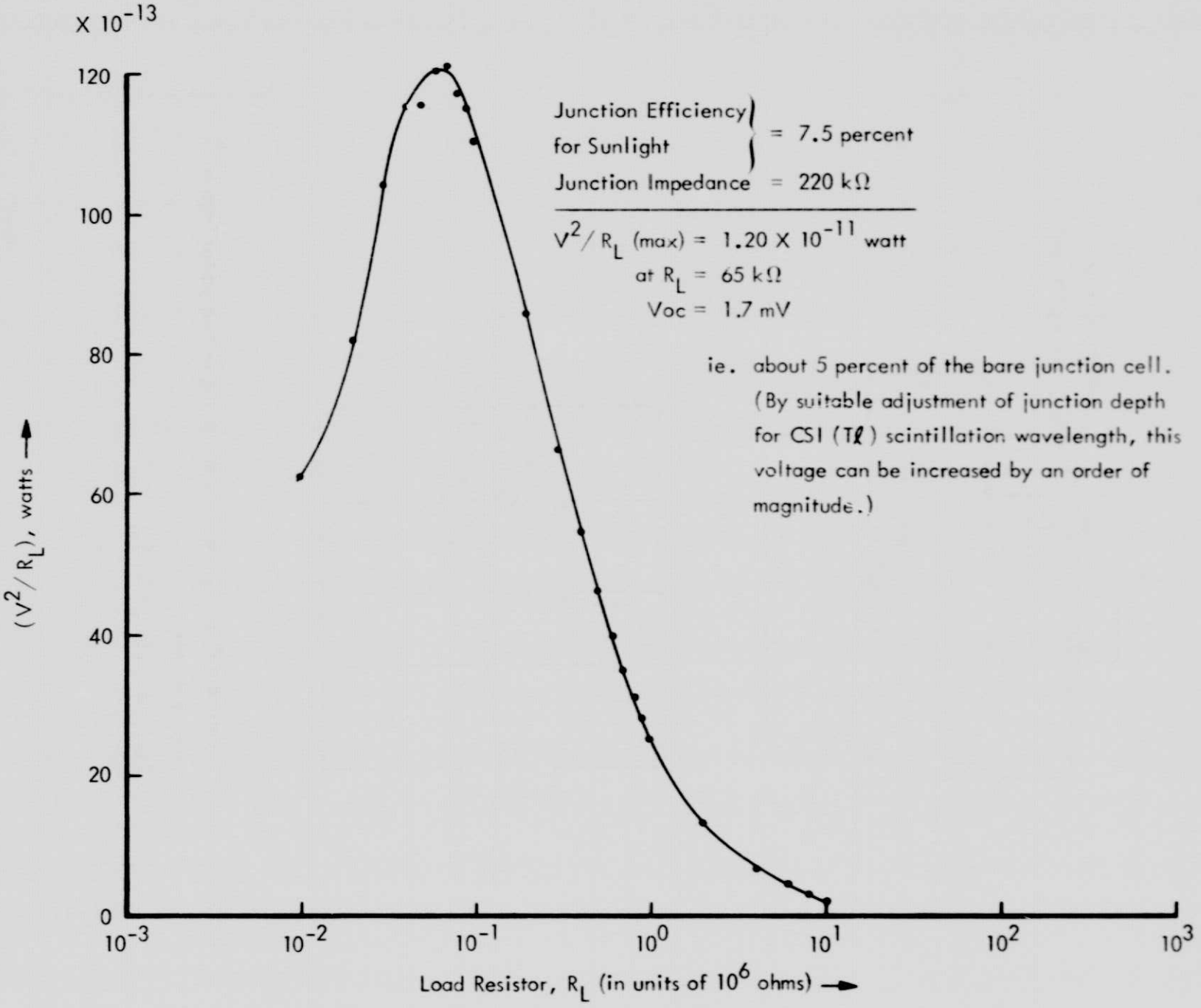


Figure 10. - External power output vs load resistor in the junction cell with CSI (Tl) crystal.