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TRITIUM-FUELED BETACELLS

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ABSTRACT

Betavoltaic power sources operate by converting the nuclear decay energy of beta-emitting radioisotopes into electricity. Since they are not chemically driven, they could operate at temperatures which would either be too hot or too cold for typical chemical batteries. Further, for long lived isotopes, they offer the possibility of multi-decade active lifetimes.

Two approaches are being investigated: direct and indirect conversion. Direct conversion cells consist of semiconductor diodes similar to photovoltaic cells. Beta particles directly bombard these cells, generating electron-hole pairs in the semiconductor which are converted to useful power. When using low power flux beta emitters, wide bandgap semiconductors are required to achieve useful conversion efficiencies. The combination of tritium, as the beta emitter, and gallium phosphide (GaP), as the semiconductor converter, was evaluated.

Indirect conversion betacells first convert the beta energy to light with a phosphor, and then to electricity with photovoltaic cells. An indirect conversion power source using a tritium radioluminescent (RL) light is being investigated. Our analysis indicates that this approach has the potential for significant volume and cost savings over the direct conversion method.

INTRODUCTION

Sandia National Laboratories (SNL) has been investigating low level, multi-decade lifetime, nuclear power source concepts which use no special nuclear material (e.g. plutonium) or other environmentally sensitive isotopes. The objective is to build a 1 milliwatt (mW) power source fueled by an abundant and affordable beta-emitting radioisotope with a lifetime of at least 12, and preferably 20^{+} years.

Our initial approach was to use the direct conversion betacell technique first demonstrated at Sarnoff Laboratories [1] in the mid 1950's, and further developed in the early 1970's [2]. Shown schematically in Fig. 1, a beta-emitting radioisotope is placed in close proximity to a solid state converter, consisting of a large area p/n junction analogous to that used in solar cells. Beta particles penetrating the converter create electron/hole (e/h) pairs in the semiconductor. Those carriers, generated close enough to diffuse across the p/n junction, can provide useful power. ^{90}Sr and silicon cells were used in the Sarnoff cell.

As we became more familiar with the limitations of this technology, we began to look at alternatives. The most promising alternative initially converted the beta energy to light with a phosphor, and then to electricity with photovoltaic cells. The Elgin-Kidde nuclear battery, shown in Fig. 2, first demonstrated this approach [3]. It used ^{147}Pm mixed with a powdered phosphor (yielding a ^{147}Pm RL light) and silicon cells.

The choice of beta emitter and semiconductor are determined by a number of factors, including power source safety (i.e. external radiation levels), lifetime, output, cost, volume, weight, etc. ^{90}Sr was not acceptable because of the radiation hazard from its ^{90}Y daughter product, and ^{147}Pm had too short a half-life. Low energy beta emitters with a half-life of at least 10 years, and no other nuclear or significant Bremsstrahlung radiation were the most desirable. It became apparent that wide bandgap semiconductors were necessary to efficiently convert the low beta power flux into useful power.

DIRECT CONVERSION

Tritium Beta Emission Characteristics

Tritium is a negative beta emitter with an average energy of ~6 keV, a cutoff (maximum) energy of 18.6 keV and a 12.3 year half-life, sufficient for a 20 year device. As a gas, tritium poses a relatively small health risk since it is not readily absorbed by the human body [4]. It emits no other nuclear radiation, and would generate only weak Bremsstrahlung x-rays easily shielded by a thin (0.25 mm) sheet of stainless steel. Tritium is also available in quantity at a reasonable cost.

Self-absorption of the beta energy limits the maximum power flux from tritium gas to ~11.5 microwatts/cm^2. This is illustrated in Fig. 3 where the computer-predicted [5] beta particle power flux emitted by a two dimensional slab of tritium gas is plotted as a function of the gas thickness. The squares correpond to pure tritium gas @ 0.103 MPa, the circles to 50% tritium and 50% ^{3}He @ 0.154 MPa, and the triangles to 25% tritium and 75% ^{3}He @ 0.180 MPa. These are expected gas mixtures and pressures for 0, 1 and 2

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tritium half-lives, respectively. Note that the maximum power flux decreases by more than 50% at the end of each half-life. This is due to increased electron scattering by the ^3He decay product as it builds up in the remaining tritium.

Semiconductor Converter

The maximum short circuit current density obtainable from a semiconductor converter bombarded by beta particles (assuming no backscattering and that all the resulting e/h pairs are collected) can be estimated from:

$$Jsc = P_{(beta)} / E_c, \quad (1)$$

where Jsc is the short current density (A/cm^2), $P_{(beta)}$ is the beta power flux (W/cm^2), and Ec is the energy cost (eV) required to create an e/h pair in a semiconductor. Ec can be estimated using:

$$Ec = (14/5)\ Eg + K, \quad (2)$$

where Eg is the bandgap of the semiconductor and K ~ 0.75 eV [6].

With good e/h transport properties, a wide bandgap semiconductor will insure a high open-circuit voltage, and a correpondingly high efficiency at low beta power fluxes. Gallium phosphide was chosen because of its large indirect bandgap (2.24 eV), good carrier diffusion lengths, and its moderately advanced state of development [7]. Based on equations (1) and (2), the maximum current density expected is ~1.6 microamps/cm^2. Fig. 4 is a computer prediction of the normalized integral of the beta energy, deposited in GaP from 2 cm of tritium gas at 0.103 MPa, as a function of GaP thickness [5]. Note that the maximum range of the tritium beta particles is only ~1 micron. This plot was used to help optimize cell structures.

EXPERIMENTAL

Three GaP p/n cells were selected for test in the Sandia Tritium Research Laboratory (TRL), in Livermore, California. The cells were made by AstroPower, Inc. using Liquid Phase Epitaxy (LPE), and consisted of 1 cm^2 p/n structures on GaP substrates.

Sample F5926 consisted of a 0.075 micron AlGaP emitter anti-recombination (AREC) layer, a 0.13 micron GaP emitter, and a 21.2 micron base. Since it is a shallow junction cell, it was expected to be the most susceptible to: 1) beta-induced radiation damage, 2) tritium diffusion/permeation, and 3) the transmutation of tritium to ^3He within the lattice of the cell. Radiation damage was of particular interest. Many experiments have demonstrated that 1 MeV electrons can damage solar cells. 200 keV electrons were also shown to damage GaP cells [8]. But, our accelerated aging studies showed that electron energies as low as 10 to 20 keV can damage many semiconductor materials, even though this energy is not high enough to displace semiconductor atoms. The damage is not normally detected where doses are <100 megarads. However, over the life of a betacell we estimate the dose will be ~100 gigarads in the top few tenths of a micron. Our tests show that doses at these levels can result in reduced cell output.

Sample F6103 consisted of a 0.75 micron AlGaP emitter AREC layer, a 0.91 micron emitter and a 9.3 micron base. The junction of this cell was near the maximum range of tritium beta particles, and radiation damage to the junction was expected to be significantly less than to sample F5926. Tritium diffusion, permeation or transmutation effects would still be observeable.

Sample F6016 had no emitter AREC layer, a 0.85 micron emitter, and a 19 micron base. This configuration was tested to determine whether a separate AREC layer was necessary. It is known that chemically bound hydrogen will passivate silicon surfaces and some gallium arsenide (GaAs) traps, reducing carrier recombination losses at these sites. Since tritium gas contains a significant amount of atomic tritium, it was speculated that it might chemically react with and passivate the emitter surface. This would have had a significant impact on cell efficiency, since ~40% of the beta energy is lost in the 0.075 micron AlGaP AREC layer according to Fig. 4.

Prior to test in the TRL, two additional tests were performed on each cell. First, their dark (no light or beta irradiation) current density-voltage (J-V) characteristics were measured, and are shown in Fig. 5. These curves are an indication of the overall quality of the converters as well as their suitability for use with the available beta power flux. The projected intercept with the J axis (J_0) is a measure of the electrical leakage across the junction and at the cell edges. Note that this value must be lower (the lower the better) than the expected beta-induced short circuit current density in order to produce useful power. The shapes of these curves are an indication of e/h recombination effects and the presence of series resistance or reverse diode structures within the cell. The curves should follow the standard diode equation:

$$J = J_0\ (\exp(qV/nkT) - 1), \quad (3)$$

where J is the current density with the diode biased forward by voltage V, J_0 is the saturated current density described above, q is the electron charge in coulombs, k is Boltzman's constant, T is the absolute temperature, and n is the "ideality factor". At low current densities, n should equal 2, and at higher current densities, n should equal 1 [9].

The second test consisted of measuring the cell short circuit current density as a function of electron energy from 2.5 to 20 keV at a constant beam power density of 1 microwatt/cm^2. These response curves, shown in Fig. 6, were then convoluted with the computer-predicted tritium spectrum, also shown in Fig. 6 [5], and corrected for the reduced beta flux due to the buildup of ^3He. The result was a prediction of the cell short circuit current as a function of tritium exposure time. This procedure was necessary to verify the predictability of cell response to the tritium beta flux since our computer simulation [5] was not originally intended for use at this low an electron energy.

All cells were subsequently mounted in fixtures with a 2 cm open space above each cell, filled with tritium gas to assure a saturated power flux to the cell surface. The tritium gas pressure was 1.03 MPa over sample F5926, and 0.103 MPa over samples F6103 and F6016.

RESULTS

The power J-V characteristics (cell current density vs. cell voltage) at the beginning of test for each cell are shown in Fig. 7. The intercepts of the characteristics with the J and V axes are the short circuit current density (Jsc) and open-circuit voltage (Voc), respectively. The peak power point (Pmax) is the maximum value of the product of the cell current density and voltage. Note that the current density of F6016 is ~10 times lower than for F5926 or F6103.

Fig. 8 compares the measured short circuit current density with the predicted values as a function of time for each cell. Testing began with sample F5926. From day 0 to 57, data was acquired by computer, and the cell was short circuited between measurements. On day 57, F6103 and F6061 were added to the test schedule. The data was then taken manually until day 133 when a multiplexer was installed to return the data acquisition to computer control. From day 57 to 133, the cells could have been either open or short circuited between measurements. But, from day 133 on, the multiplexer circuit switched the cells to resistor loads designed to keep the cells biased at their peak power point between measurments. The data on days 250-300 was missed due to data recovery schedule problems arising from the San Francisco earthquake.

Figs. 9 and 10 show the comparable Voc and Pmax behavior, respectively. The cells were visually inspected when the fixtures were opened. There were no obvious differences compared to an unexposed sample. Fig. 11 shows the optical spectral response of the cells at the end of the test using narrow band optical filters and light intensities ranging from 2.5 to 60 microwatts/cm^2. The peak response for F5926 and F6103 is at ~450 nanometers. F6016 had essentially no light response at these microwatt light intensities.

DISCUSSION

Jsc

The results from F5926 and F6103, which have an effective emitter AREC layer, indicate that we can accurately predict the short circuit current of these cells using the computed tritium spectrum and the cell response to mono-energetic electrons. The long term rate of decrease of Jsc with time closely matches the expected degradation rate due to tritium decay. This implies that neither permeated tritium, its ^{3}He decay product nor the radiation flux from the tritium, significantly degraded the transport properties of the cells.

The short circuit current of sample F6016, which does not have an emitter AREC layer, is ~3 X that of the predicted value. Either our prediction technique is not applicable to this type of cell, or the tritium may have done some passivating of the cell surface as described above. If it were passivation, it was unfortunately not as effective as the AREC layers on F5926 and F6103. However, note that the Jsc of F6016 initially decreased, but then recovered and was still increasing when the experiment ended. Perhaps the passivating effect was not yet complete.

There is an anomalous peak in Jsc during the first 60 days in cell F5926 which is not seen in F6103. The two main differences between these tests were that F5926 was exposed to 10 times the tritium pressure of F6103, and the junction of F5926 was much shallower than that of F6103. One of our speculations is that the higher tritium pressure on F5926 might have increased the concentration of tritium and/or ^{3}He in the AREC layer, the emitter, the depletion zone or the near base region. If the effectiveness of any of these sensitive regions is strongly dependent on the concentration of tritium or ^{3}He, it could explain the absence of the peak in the F6103 deep junction, low pressure test, compared to its presence in the shallow junction, high pressure F5926 sample. Changes in concentration with time, first below and then above, an "optimum" level could produce a peak similar to what is observed.

Note that Jsc in F5926 appears to be bi-modal during days 57 thru 133. This time period was when the data was being taken manually, and any cell could have been either in an open or short circuit condition between measurements. The higher values appear to correspond to the open-circuit condition, and the lower values to the short circuit condition. After day 133, all the cells were resistively biased at their peak power points between measurements. In F5926, the degradation curve after day 133 appears to be an extension of the higher Jsc values. Similar trends can be seen in F6103 and F6016, but not as clearly. It is speculated that the higher current density after an open-circuit condition is due to a reduced field-assisted carrier recombination rate in the junction. The higher Jsc's, measured when the cells were biased at the peak power point, is evidence of this effect since the field within the junction at the peak power point is more comparable to an open-circuit rather than a short circuit condition.

Voc and Pmax

All three cells show steadily decreasing Voc values until day 133. Voc then either becomes flat or begins to increase with time.

The Pmax value of F5926 rapidly decreases until it is peak power biased, and then it decreases more slowly toward the end of the test. F6103 decreases and levels off before being biased. Thereafter, it increases slightly and then decreases slowly. In F6016, Pmax rapidly decreases and is nearly level before bias. After bias, Pmax decreases slightly and then increases steadily.

These results illustrate the importance of the bias condition of the cells between measurements (or when not in use!). Some of the initial degradation may have been caused by short circuiting the cells for

significant fractions of the test time. It is known that amorphous silicon cells can be damaged by the energy released from carrier recombination if the cells are kept open-circuited. The bias condition was just the opposite for these cells. Perhaps a field-enhanced recombination rate in the junction region itself is responsible for the degradation seen here.

Spectral response curves were not taken on these cells before tritium exposure. However, the responses of F5926 and F6103 are not significantly different from those of similar un-irradiated cells. It appears that the tritium exposure did not significantly degrade their optical properties. The poor response seen in F6016 might be attributable to either a shunt within the cell, or to the absence of the emitter AREC layer. Note that none of these cells had an optical anti-reflection layer.

Based on the performance of F5926, projections were made for a power source with a 1 mW output at the end of 12 years. We estimate it would have a volume of 64 cc, and a mass of 125 grams, exclusive of the tritium containment structure and electrical feedthrough. It would require 3.3 kCi of tritium gas at 10.3 MPa, and ~1 m^2 of GaP converter on a thin silicon substrate. The potential cost of the converter was estimated to be ~$60K.

INDIRECT CONVERSION

The basic problem with direct conversion using tritium is that the tritium self-absorbs too much of its own beta energy. Since the beta flux cannot be concentrated, the low power density requires a large converter area to extract a reasonable amount of power. Indirect conversion may allow us to overcome this limitation.

Note that a power source simply based on the Elgin-Kidde design, using a standard tritium gas tube RL light, would require the same (or larger) area of converter. This is because phosphor conversion efficiencies are typically less than 25%. The brightest tritium gas RL lights only have surface intensities ~2.3 microwatts/cm^2. Even if photovoltaic cells could be fabricated with 20% conversion efficiencies at this intensity, the net power out would be 0.46 microwatts/cm^2, compared to ~0.5 microwatts/cm^2 demonstrated in our direct conversion results.

An efficient volumetric RL source is needed to concentrate the light generated in a small volume of tritium gas, thereby reducing the area of photovoltaic cells required. A cathodo-luminescent phosphor, dispersed in a low density silica aerogel and filled with 10.3 MPa of tritium gas, may be one such light source [10]. Preliminary measurements indicate that a light power flux of ~23 microwatts/cm^2 is possible. GaAsP cells have been fabricated which have projected efficiencies approaching 20% under these conditions.

Assuming we can achieve a 22% converter efficiency at this intensity, we are projecting a 12 year, 1 milliWatt power source with a volume of 32 cc and a mass of 51 grams, exclusive of the tritium containment and electrical feedthrough. We estimate that ~3.3 kCi of tritium, about the same as for direct conversion, would be required. However, we would only need ~400 cm^2 of converter. At $25/$cm^2$, the converter cost would be ~$10K. Even greater cost, volume and weight savings would be realized if the light could be made even more intense and/or if the converter could be grown on thin silicon substrates.

CONCLUSIONS

A direct conversion power source using tritium gas and GaP semiconductor converters is technically feasible. There was no significant degradation due to radiation damage and/or tritium permeation after one year of exposure to tritium gas in any of the cells studied here. It is estimated that the GaP converter for a 1 milliWatt, 12 year direct conversion power source would cost ~$60K. In contrast, the converter cost for a comparable indirect conversion power source is projected to be ~$10K. In addition, significant reductions in volume and weight are also projected for the indirect conversion device.

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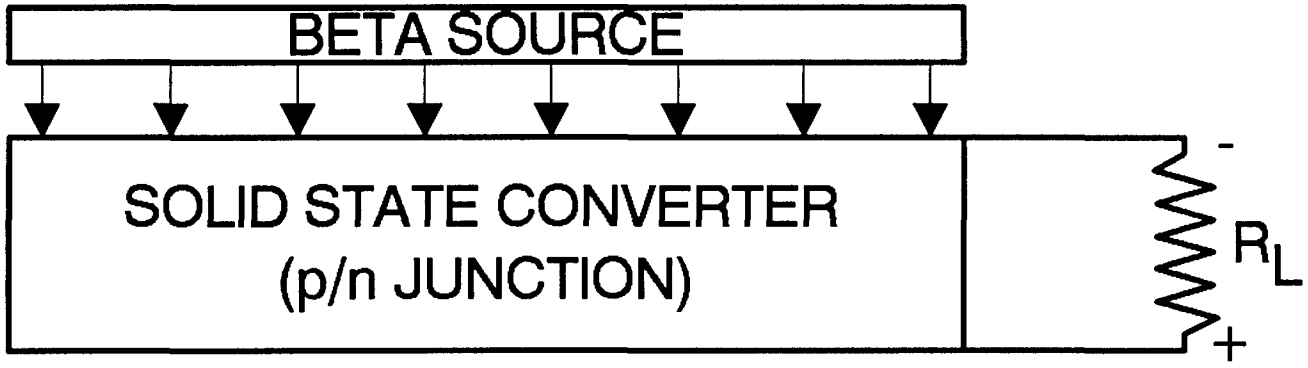

Figure 1. Direct conversion betacell.

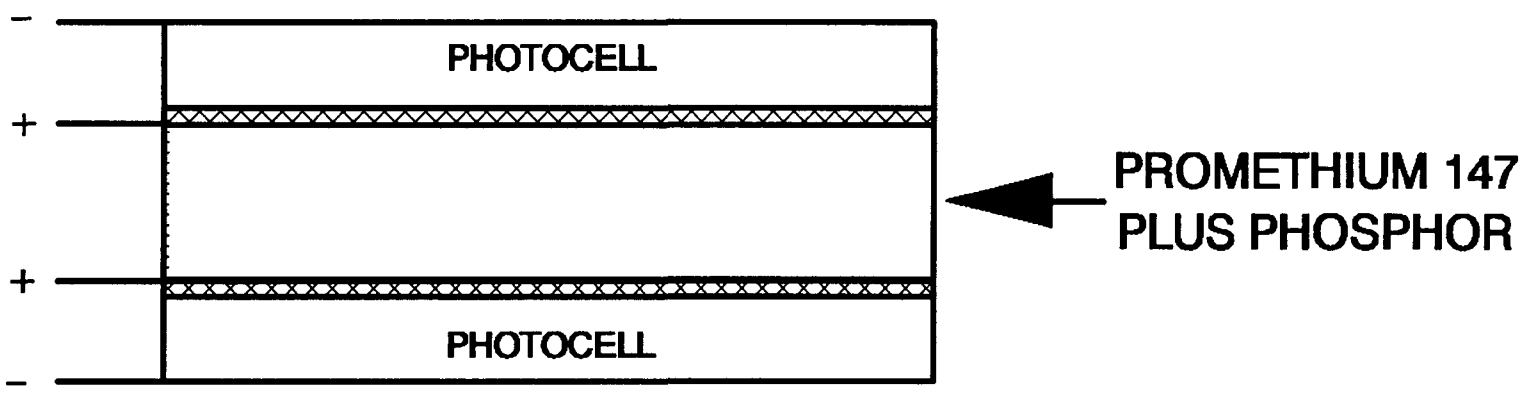

Figure 2. The Elgin-Kidde nuclear battery.

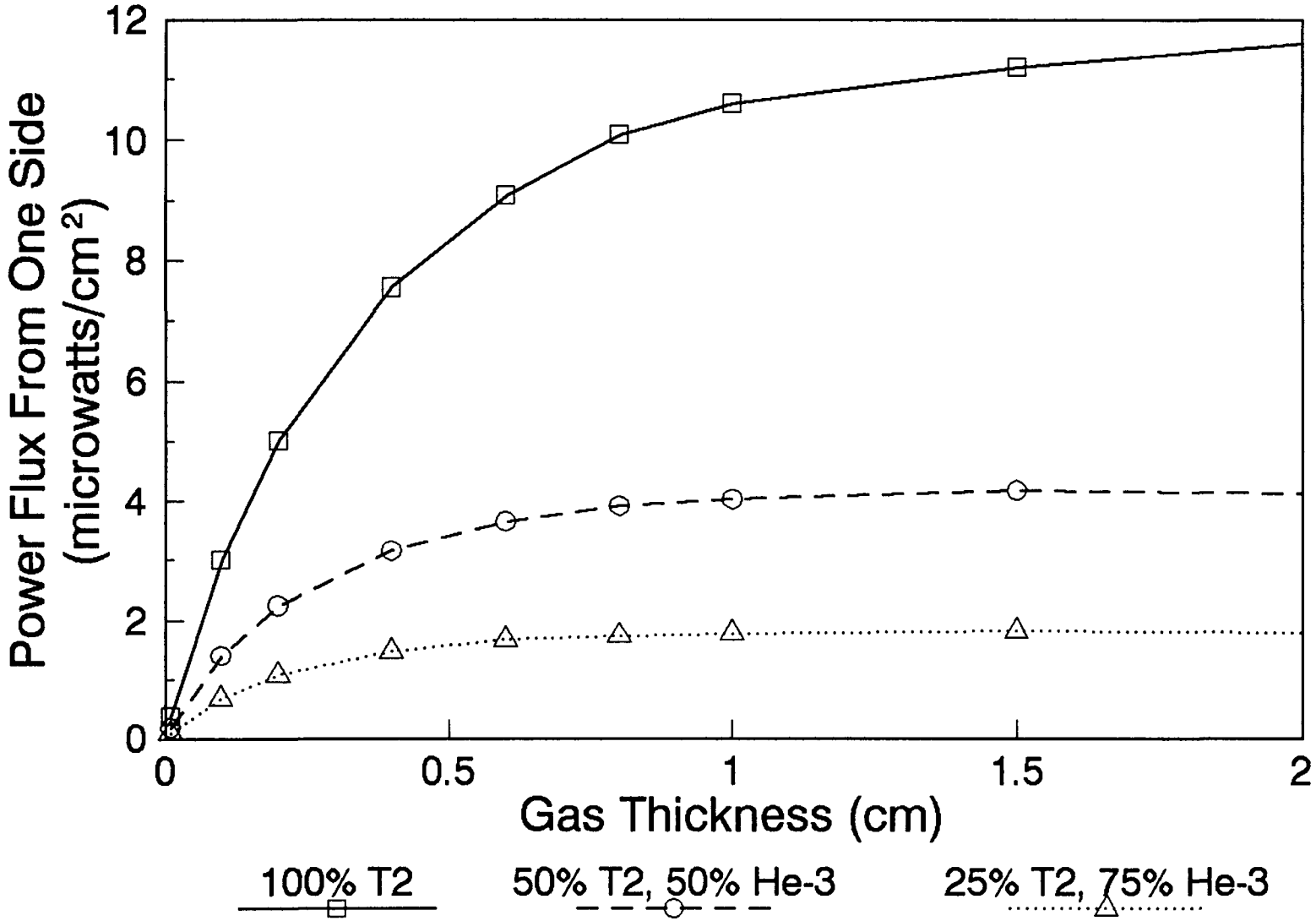

Figure 3. Computer predicted power flux vs. gas thickness for tritium gas sources.

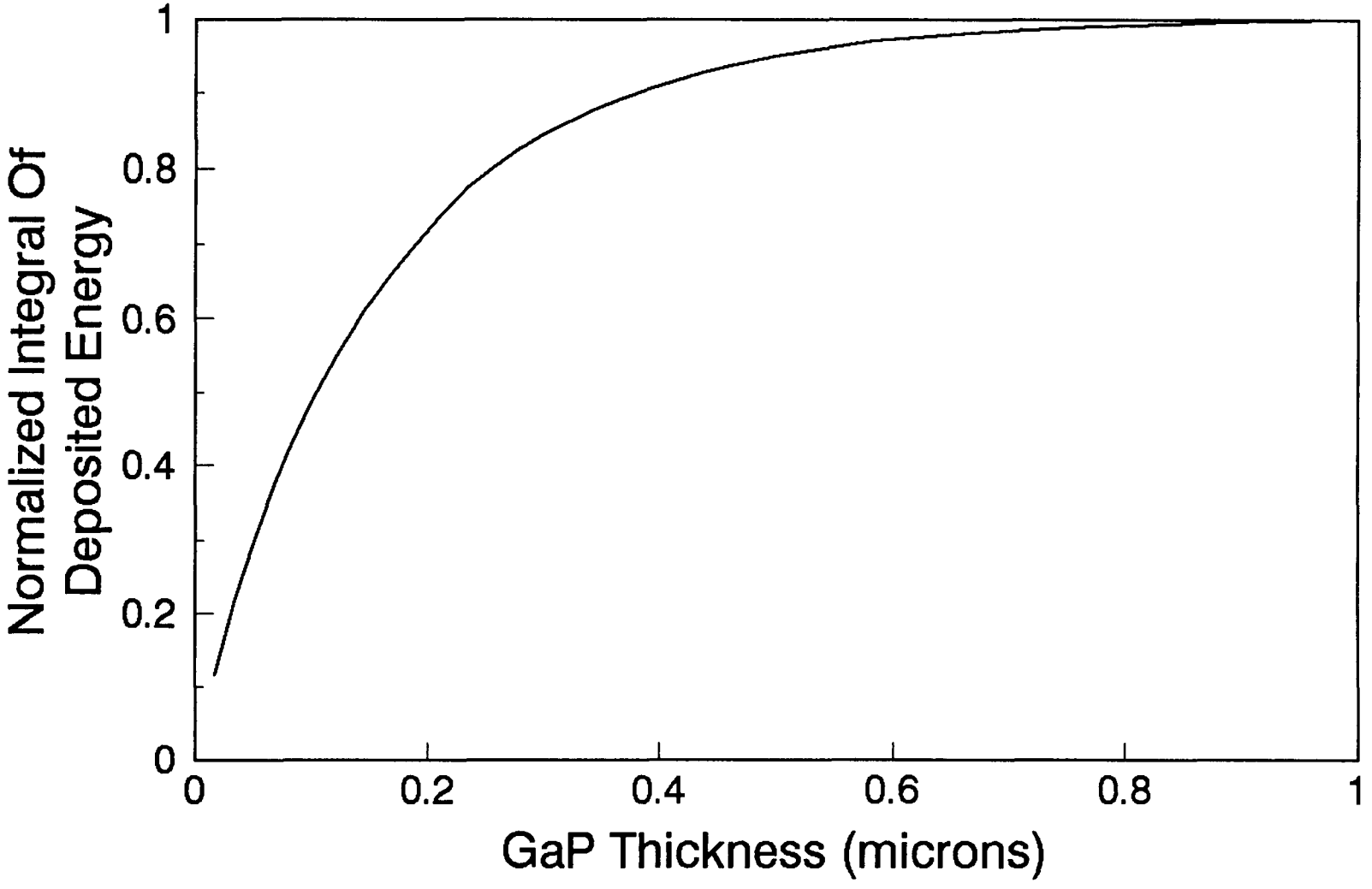

Figure 4. Normalized integral of tritium beta energy deposited in GaP vs. GaP thickness.

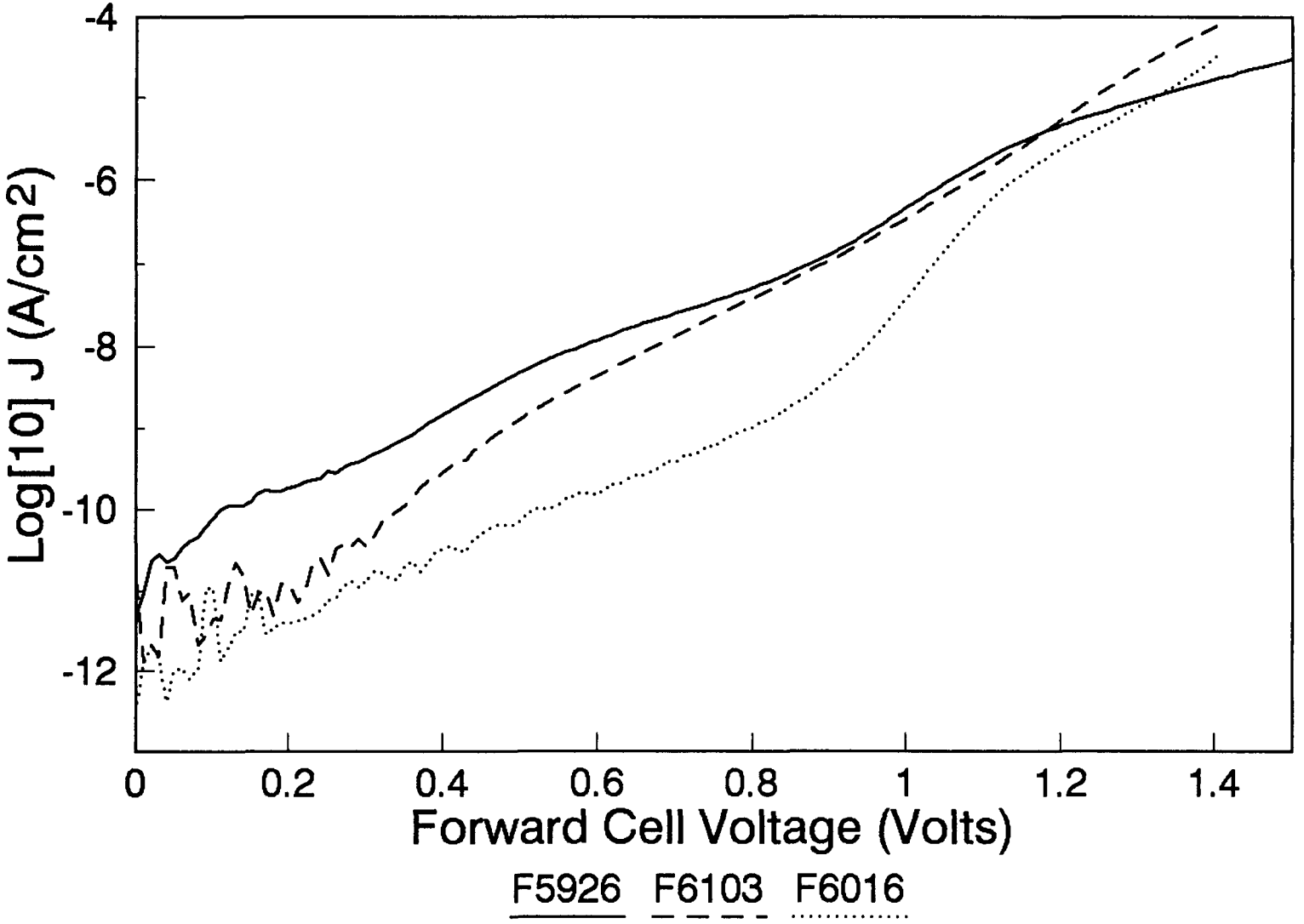

Figure 5. Forward dark J-V characteristics.

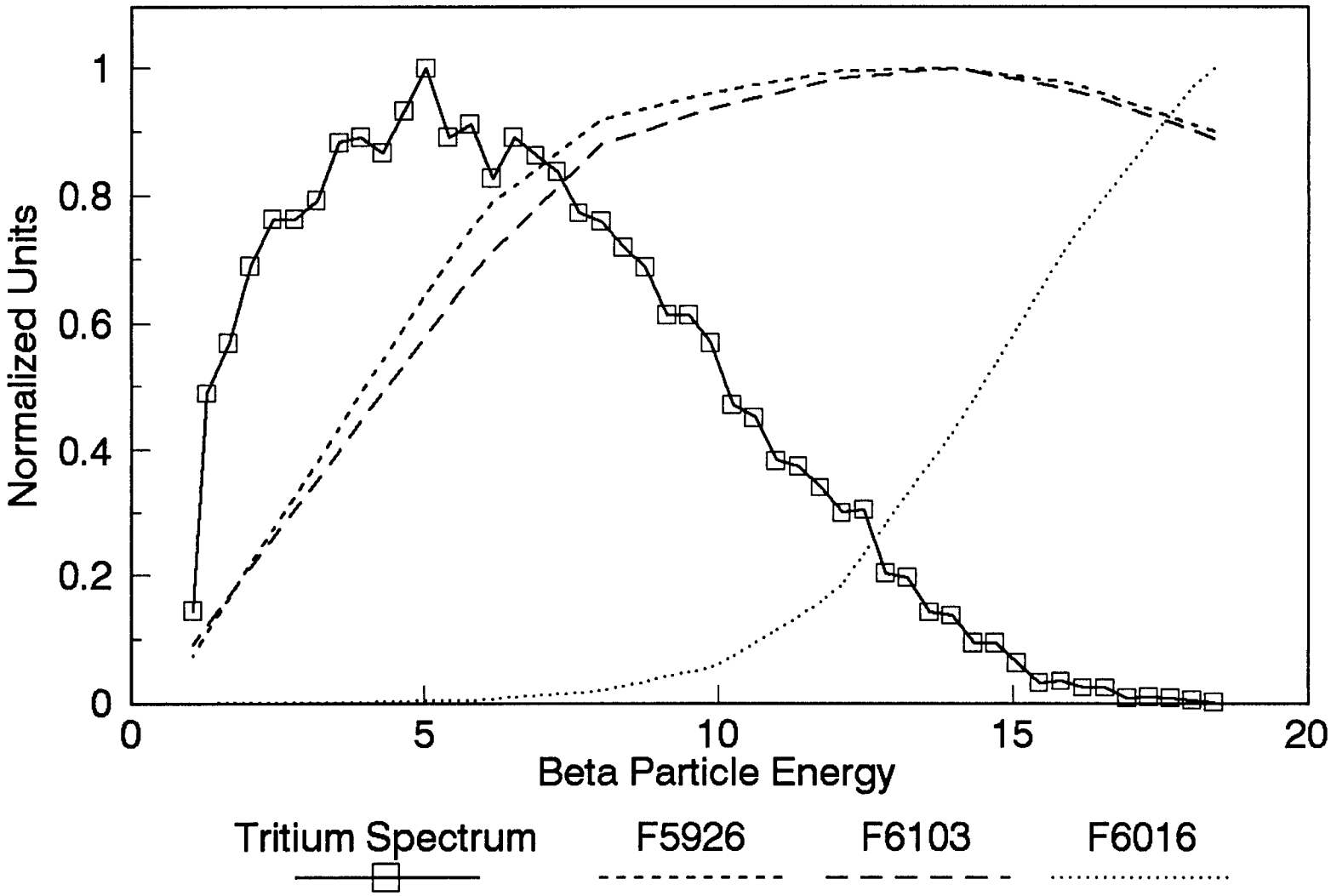

Figure 6. Overlay of computer-predicted tritium spectrum with measured cell short circuit current responses to mono-energetic electrons.

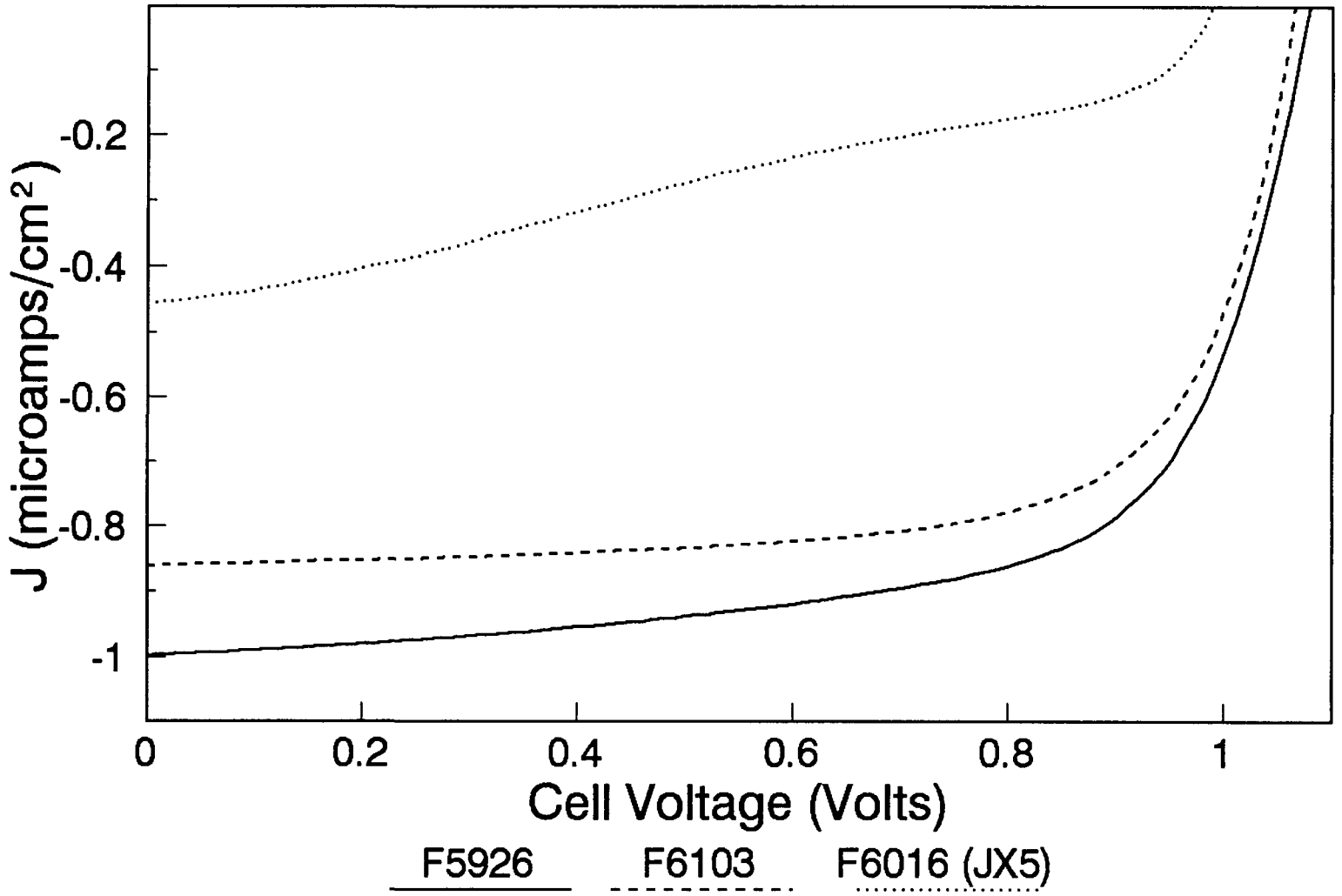

Figure 7. Power J-V characteristics at beginning of test. The F6016 plotted J values are 5 times the measured values for clarity.

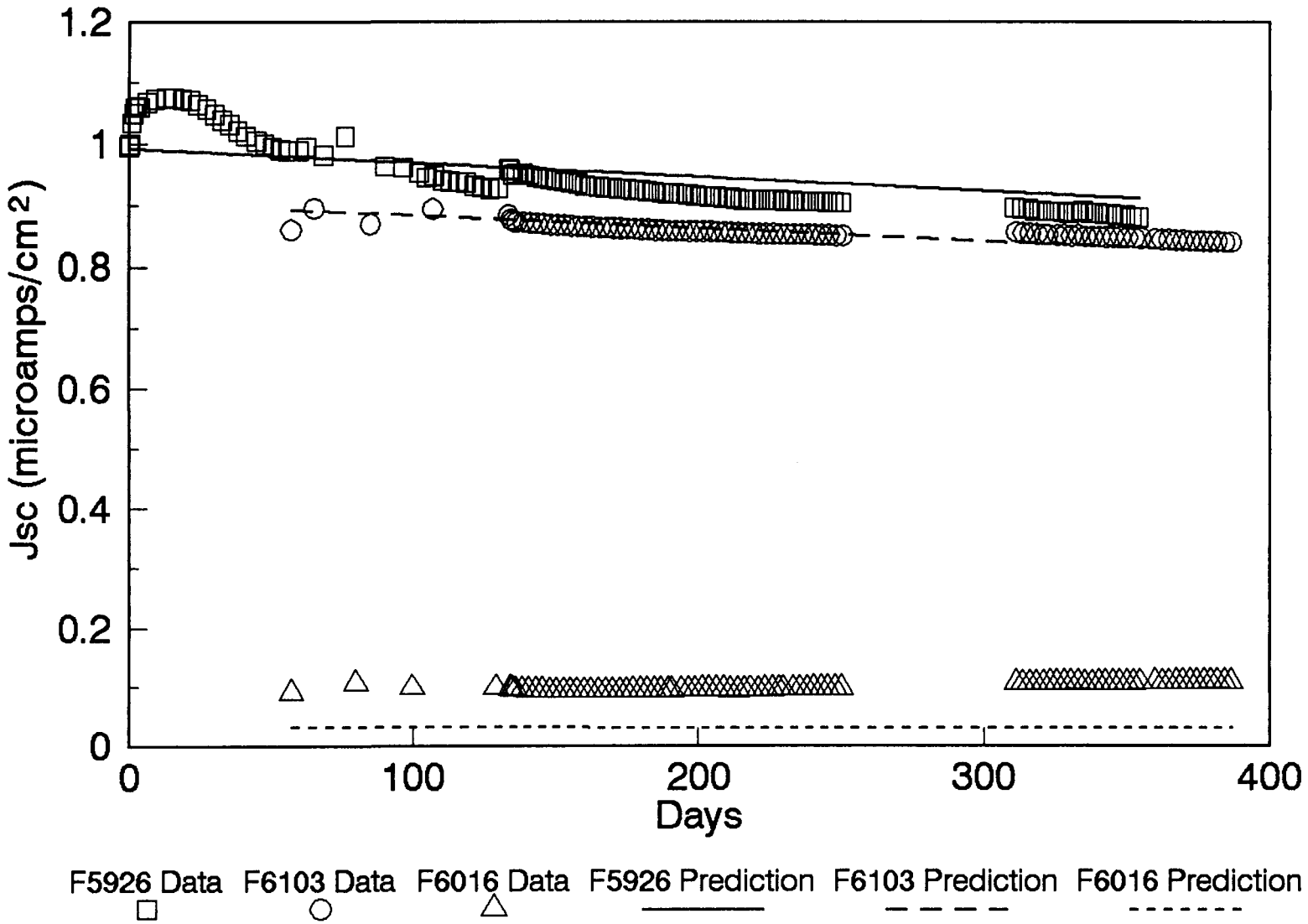

Figure 8. Comparison of measured and predicted short circuit currents as a function of time.

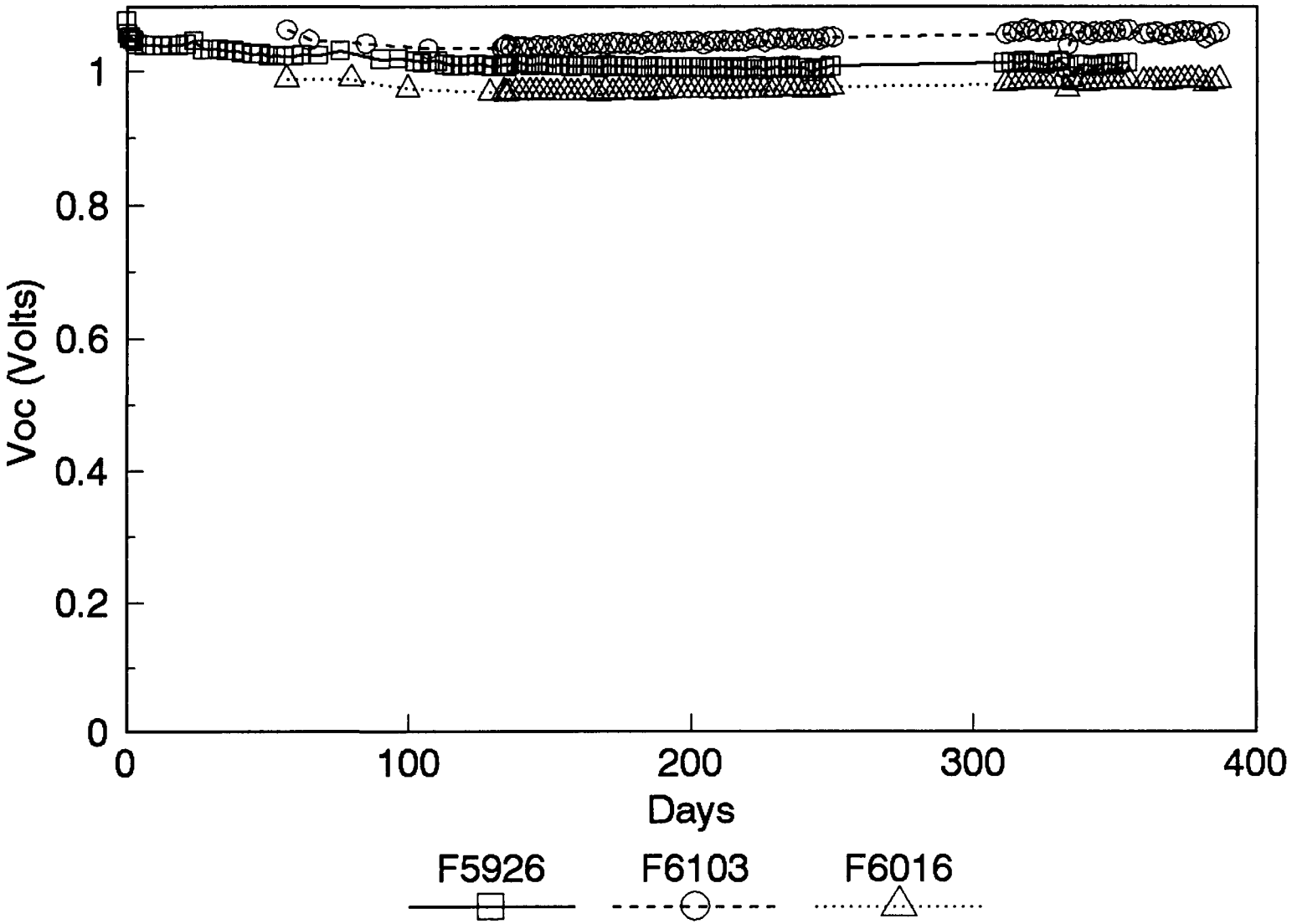

Figure 9. Open-circuit voltage as a function of time.

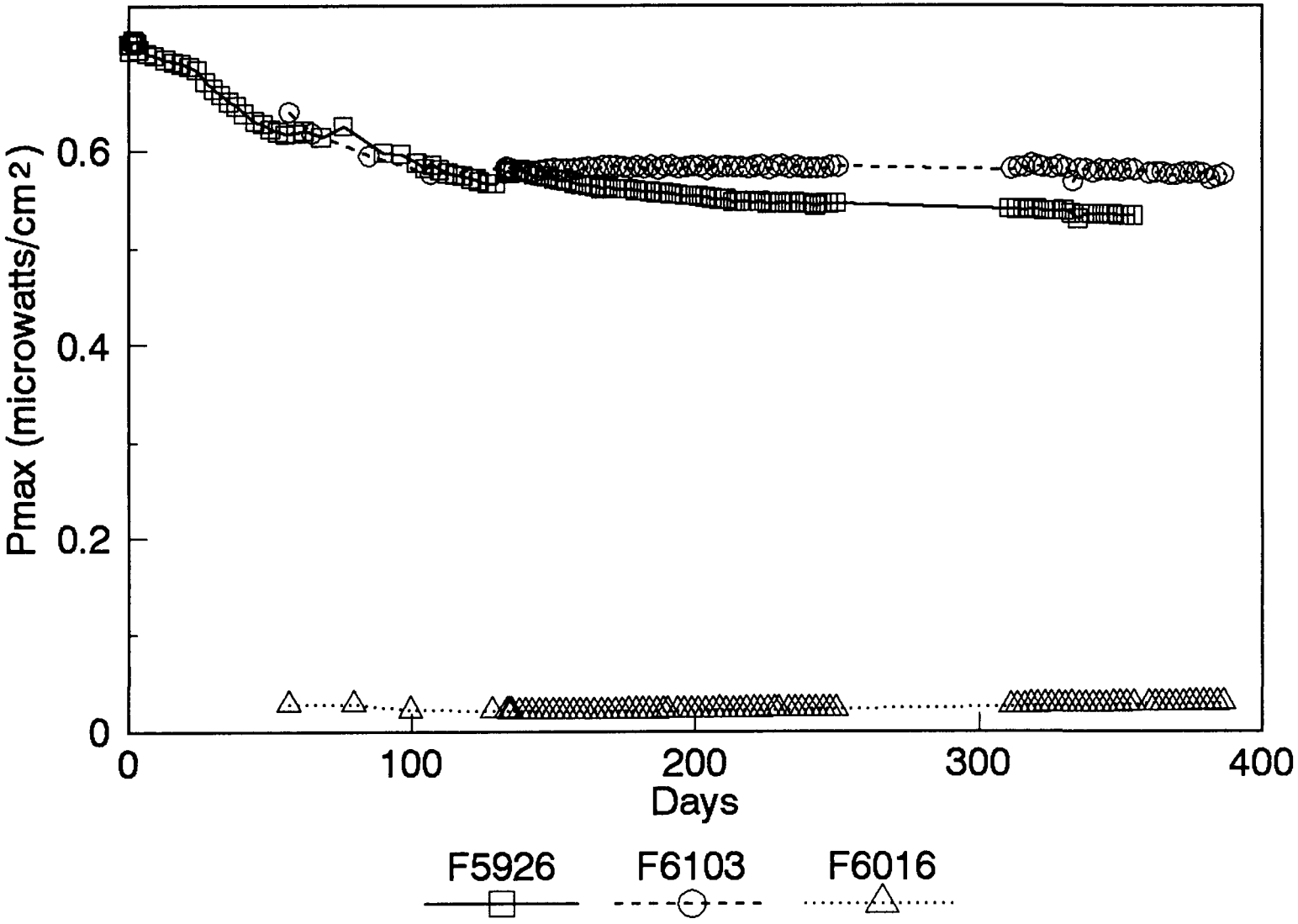

Figure 10. Peak power density as a function of time.

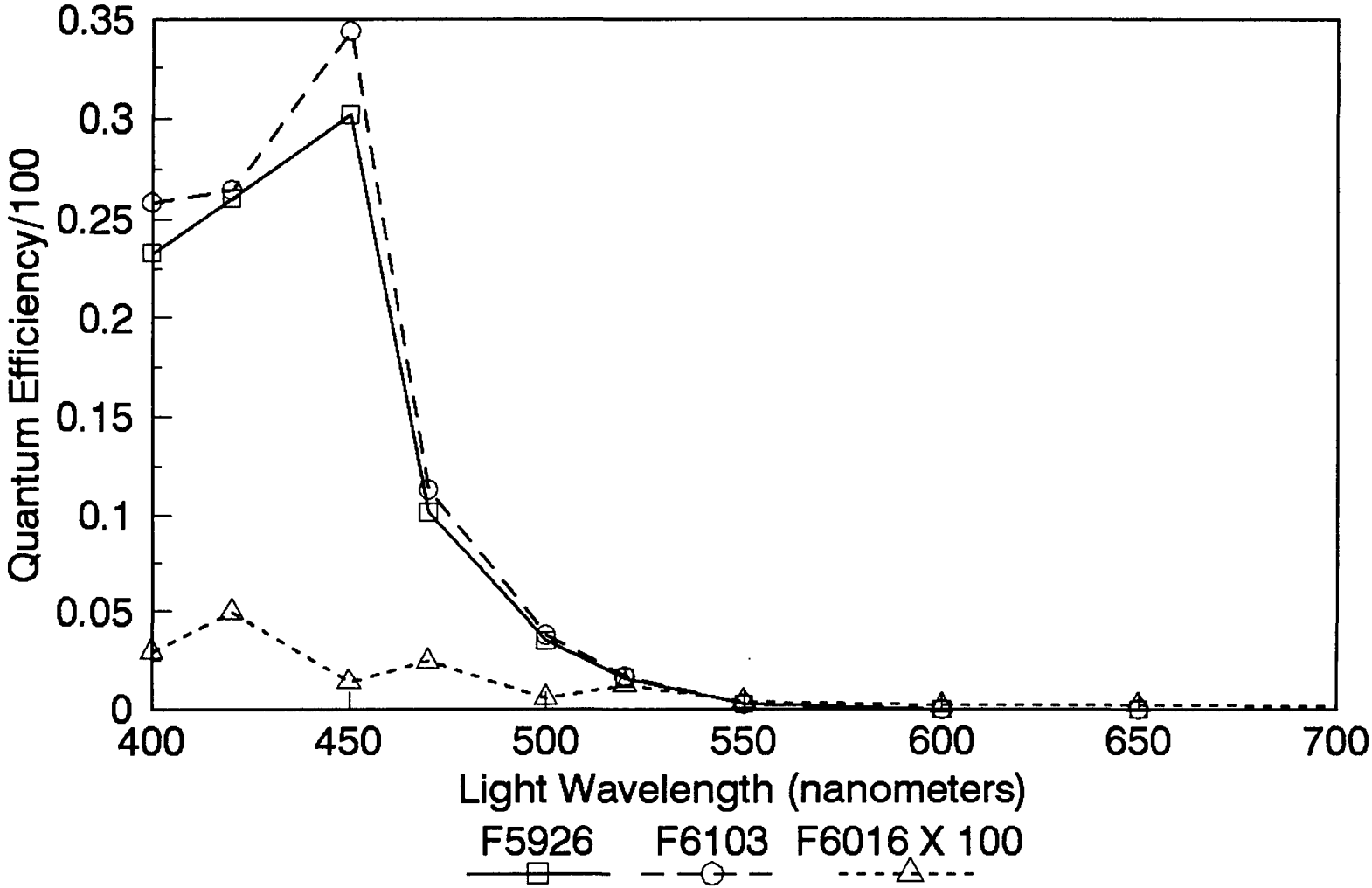

Figure 11. Cell quantum efficiency vs. light wavelength after tritium exposure. The plotted values for F6016 are 100 times the measured values for clarity.