

# Selective Emitters for Thermophotovoltaic Power Systems for Use in Aerospace Applications

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Thermophotovoltaics is the term applied to the technique for energy conversion whereby the energy emitted by an incandescent source is converted to electrical energy by a photovoltaic cell. There are three approaches currently being investigated for practical devices. These are 1) selective filtering, 2) multibandgap conversion, and 3) selective emission. This article will concentrate on selective emission. The selective emitters are made from oxides of the rare Earths such as erbia, holmia, ytterbia, and neodemia. Large area emitters are made through a specialized series of processes that begin with nitrates of the rare Earth and end with rare-Earth oxide filaments. A special requirement for these applications is the need for a robust large area emitter that could take the shock and vibration of space launch or those associated with Army applications. Conventional paper-making techniques have been used to combine materials suitable as binders with the radiating material. As a result, this technique allows the fabrication of large area robust emitters that were heretofore unobtainable. These emitters will be described in some detail, both for fabrication and performance.

## Nomenclature

- $c_1$  = first radiation constant,  $3.742 \times 10^8 \text{ W } \mu\text{m}^4/\text{m}^2$   
 $c_2$  = second radiation constant,  $1.439 \times 10^4 \text{ } \mu\text{m K}$   
 $P$  = power density,  $\text{W}/\text{m}^2$   
 $P_\lambda$  = spectral emissive power/ $\text{m}^2/\text{m}$  wavelength interval  
 $T$  = temperature, K  
 $\epsilon$  = emissivity  
 $\lambda$  = wavelength  
 $\sigma$  = Stefan–Boltzmann constant

## Introduction

There is an enormous interest in finding clean sources of energy for a multitude of applications. Of particular interest in recent years is the possibility of utilizing photoconversion with fossil-fueled radiators. The applications contemplated range from electric automobiles to small auxiliary power for the military. Key to any application is the need for special emitters that can effectively and efficiently convert energy from a heat source to the format necessary for photoconversion. Adair and Rose<sup>1,2</sup> have shown that selective emitters can be made reproducibly that are cheap and have a large radiating area. This emitter technology will be discussed in this article.

It has been shown theoretically<sup>3</sup> that certain rare Earth oxides are capable of emitting as much as 70% of their total radiated energy when heated at high temperatures in a narrow band characterized by their electronic structure. At short wavelengths, in the uv region of the spectrum, the rare Earth oxides tend to have a high emittance. Fortunately, these modes are only excited efficiently at extremely high temperatures. On the other end of the spectrum, in the far infrared (IR), similarly, there is little energy emitted, even though there is a high emittance for these materials. For the temperatures

contemplated for thermophotovoltaics, the emittance of these materials is effectively low, except at the line frequency. As a result, in crystalline solids of these materials, the radiative characteristics of the elements are narrow-band emissions, rather than a more broadband continuum superimposed upon a line spectrum. The rare Earths in oxide form that have received significant interest are  $\text{Nd}_2\text{O}_3$ ,  $\text{Ho}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$ , and  $\text{Yb}_2\text{O}_3$ .

All matter is continually emitting radiant thermal energy as a result of the vibration of its constituent particles. By definition, a blackbody radiator absorbs all of the radiant energy incident upon it and emits the maximum possible amount of flux per unit area at any given wavelength or wavelength interval for any body at its temperature. All real materials exhibit less than this ideal behavior. The radiant power density from a blackbody is given by the Stefan–Boltzmann equation as

$$P = \sigma T^4 \quad (1)$$

For a nonblackbody source, the previous equation is modified by  $\epsilon$ , a factor between 0–1, which describes the deviation from an ideal blackbody. The  $\lambda$  distribution of this flux is given by the Planck equation as

$$P_\lambda = c_1 \lambda^{-5} (e^{c_2/\lambda T} - 1)^{-1} \quad (2)$$

It is this spectrum against which the performance of selective emitters must be compared with respect to spectral content and efficiency.

Clearly, the intensity at a given wavelength is a function of the temperature and it is this temperature that will also determine the efficiency of selective emitters. If the short wavelength cutoff for the blackbody spectrum is at a wavelength longer than that required to excite a specific band, there will be little or no emission from the sample. However, the intensity at a particular wavelength is exponential in temperature, which should result in a strong temperature dependence for emission. This is in fact what is observed.

The primary reason for using selective emitters in a thermophotovoltaic (TPV) system is to increase the total system efficiency. Because these emitters radiate in a single narrow band instead of a broadband continuum [Eq. (2)], it takes less energy to heat them to a given temperature than it does to heat a blackbody or graybody to the same temperature.

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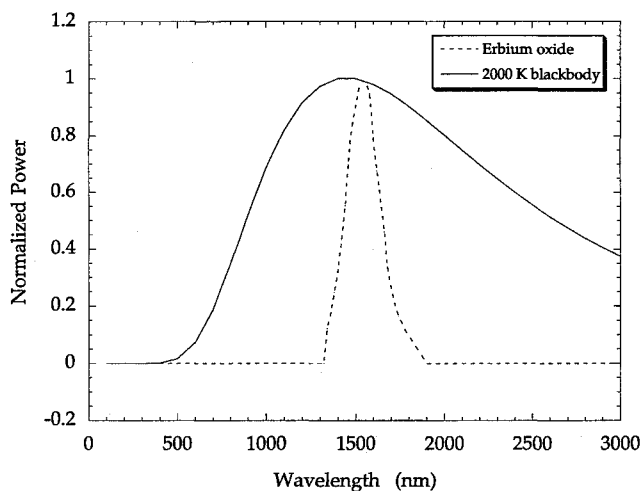


Fig. 1 Idealized emission from erbia and a blackbody operating at a temperature of 2000 K. Both curves have been normalized with respect to the peak intensity.

Therefore, a TPV system utilizing a selective emitter needs less fuel input to heat the selective emitter when compared to the fuel needed to heat a blackbody emitter to the same temperature. Figure 1 illustrates the power emitted as a function of wavelength for a theoretical blackbody and an idealized erbium oxide selective emitter at a temperature of 2000 K. Both curves have been normalized with respect to the peak intensity and an emittance of 1.0 is assumed. The emittance on either side of the emission band is small for oxides in fibrous form. This figure clearly shows the importance of using a selective emitter that can be optimally matched to a photovoltaic cell. Nearly all of the out-of-band energy, radiating from the blackbody, is unusable by a single bandgap photocell. For total energy efficiency, the waste energy must be utilized in preheat, or at worst, must be rejected through a thermal management system.

Guazzoni demonstrated the phenomena of selective emission in his early work on selective emitters.<sup>4</sup> He used the rare Earth oxides in a monolithic ceramic form. This work resulted in emitters that were severely limited by thermal shock. Further, these monolithic radiators did not minimize the offband radiation on either side of the desired band. The spectral characteristics of these emitters were best described as "graybody" with a superimposed selective band.

Nelson<sup>5</sup> later showed that by creating the rare Earth oxides in a fiber form the emitters were less prone to thermal shock. This fibrous form (approximately 10  $\mu\text{m}$  in diameter) limited off-band radiation. The rare Earth oxides in fiber form also coupled well thermally to a combustion heat source due to the large surface area associated with fine fibers. These emitters are very similar to the Welsbach-type mantles that are used in Coleman lanterns. Unfortunately, they also demonstrated the fragility that is common to this mantle type.

## Experimental

### Emitter Description

The fundamental problem that has plagued the use of selective emitters in TPV systems has been structural integrity. This problem was addressed by constructing a composite emitter that contains the desired rare Earth oxide and a structural material that adds considerable strength to the final selective emitter. The basic technology used in the fabrication of these emitters is covered by U.S. Patents 5,080,963, 5,096,663, and 5,102,745, with significant additions applicable to the unique materials requirements placed on selective emitters. The detailed procedure for fabricating is described in Ref. 2. The preliminary form of the radiator consists of a

suitable precursor for the rare Earth oxide, cellulose, and structural/binder fibers that have been blended and made into a standard sheet of paper. The structural material must have low emittance in the wavelength region of interest and must be able to maintain its structural integrity at the elevated temperatures that are required for selective emission.

There are several advantages to using composite emitter structures. In addition to controllable structural strength, composite emitters can be formed into any reasonable geometry because of a unique wet layup or paper process. By using this technique, highly uniform sheets can be made that can be molded into any form while still wet. When dry, the sample is oxidized to produce a composite structure. Sinterbonding, which occurs during the annealing process, provides an excellent mesh effectively holding the radiating material in place as well as providing structural strength.

Numerous structural fibers can be used in emitter fabrication. Quartz and aluminoborosilicate (Nextel 440) structural fibers were used in the samples described in this article. Quartz and Nextel are ideal and can be purchased in a number of sizes. The quartz fibers, 9  $\mu\text{m}$  in diameter, act as a binder for the Nextel fibers, which are between 7–13  $\mu\text{m}$  in diameter. During high-temperature annealing, quartz softens and bonds to the Nextel 440 fibers, creating a strong structural fiber matrix that holds the rare Earth oxides in the composite emitter structure. Quartz is molten at temperatures greater than 1900 K. Aluminoborosilicate (Nextel) is molten at temperatures greater than 2100 K. In general, emitters are operated at temperatures several hundred degrees less than these critical points.

A fibrous material has a higher emittance than the same material as a solid. Emittance is a surface phenomena. The emittance of a material can be increased by surface roughening or decreased by polishing. Because of this effective surface roughness, the emittance of layered fibrous materials is higher than that of the solid component and is a function of fiber porosity.<sup>6</sup> Fibrous emitters have emittance greater than 0.8 at the characteristic emission peak. Because the rare Earth oxides are in a fibrous form, there is more surface area per unit mass resulting in more radiated power per unit mass. By varying the constituents, thick paper can be made to the point of being essentially optically opaque over a wide range of wavelengths. We have used this technique to produce robust radiators from erbia, holmia, ytterbia, and neodymia.

### Experimental Setup

Figure 2 is a diagram of the experimental assembly used to characterize the emission from the emitter surfaces. It is possible to replace the combustion heater with an electrically heated element. To make accurate efficiency measurements it would be desirable to heat the samples electrically. In that mode, the input power to the heating element must equal the power radiated from the surface plus any losses due to convection and conduction.

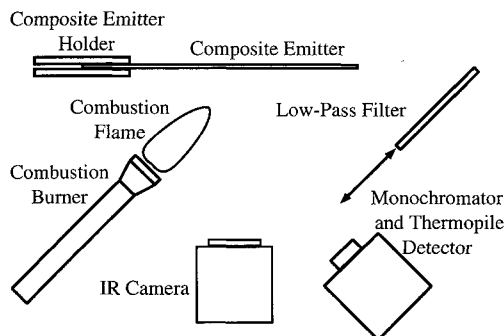


Fig. 2 Diagram of experimental test setup for measuring the properties of selective emitters.

The composite emitter structures were heated with a combustion flame source or by a silicon carbide heating element. The composite emitters were molded into a cylindrical shape to maximize the thermal coupling to the silicon carbide heating element or fabricated in square sheets for placing in a combustion flame. When heated, the spectral content of the composite emitter output was determined using a Jarrell-Ash Quarter-Meter Ebert Monochromator with the appropriate gratings. A thermopile detector was placed at the exit slit of the monochromator to determine the relative amount of radiation at each wavelength. A thermopile detector was used because of its uniform response over a wide range of wavelengths. The power incident upon the detector head at each scanned wavelength was logged using a data recorder. The measured power was corrected for nonlinearity of the gratings using calibration curves provided by Jarrell-Ash. To determine the percentage of the radiation in the selective line, the monochromator was removed and a low pass filter inserted to eliminate all radiation above the selective line. The thermopile was used to detect the total radiation from the sample and the radiation below the cutoff of the low pass filter. The ratio of the intensity below cutoff to the total intensity is a measure of the efficiency of the radiator.

### Results

The rare Earth oxides in whisker form are fragile in nature and cannot be used in large area emitters without the structural support of some type of reinforcing material. The strength is governed by the appropriate heat treatment of the mantle and the ratio of the constituents. For most applications, this issue is solved and strong mantles can be made in most geometries and if necessary with surface areas of several square feet. We have observed radiation from mantles excited both electrically and with combustion heat sources. Figures 3-5 show the emission spectra of rare Earth oxide composite emitters when heated with a combustion flame. For holmia, the peak-to-background ratio is about 6:1 and the bandwidth at half maximum is approximately 400 nm. A wide bandwidth may be good for TPV applications from the perspective of cell output power design. Under certain circumstances it may be desirable to broaden the emission profile or to have radiation at two distinct wavelengths. The technique described previously is readily adaptable to producing "two line" emitters. Figure 6 is the output spectrum of an erbia/holmia composite emitter.

Electrically heated samples also show the characteristic line, but superimposed upon a much weaker "graybody" spectrum. The spectrum is complicated due to the blackbody-like emission from the silicon carbide heating element that manages

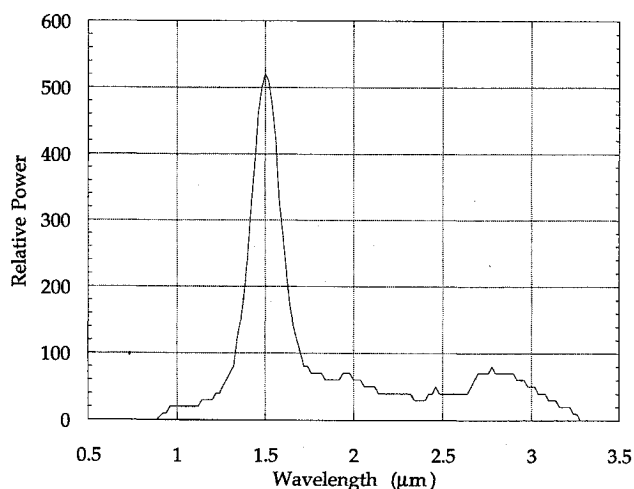


Fig. 3 Erbia composite emitter heated with a combustion flame.

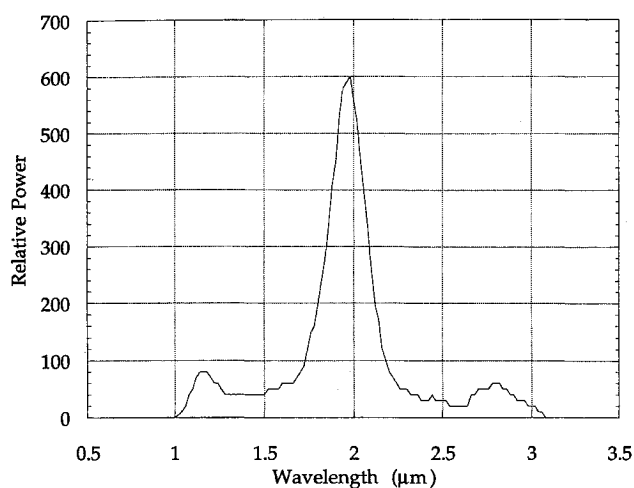


Fig. 4 Holmia composite emitter heated with a combustion flame.

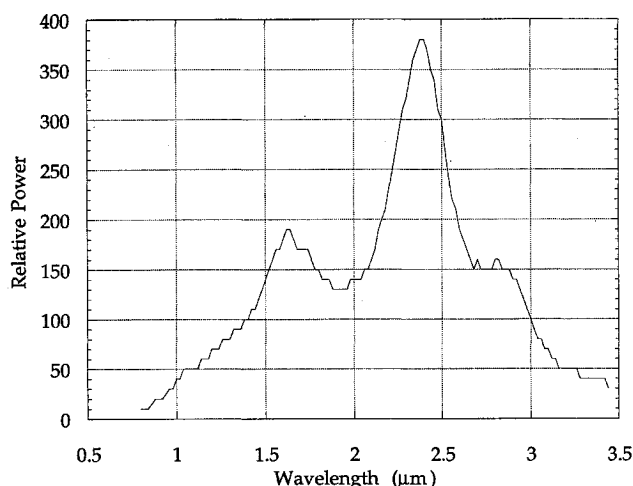


Fig. 5 Neodymia composite emitter heated with a combustion flame.

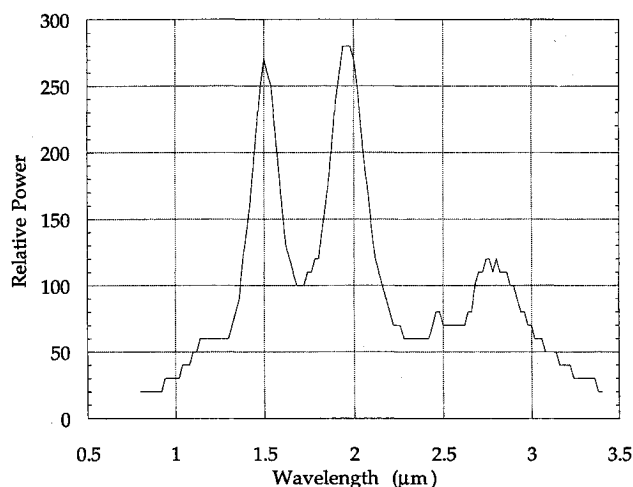


Fig. 6 Erbia/holmia composite emitter heated with a combustion flame.

to penetrate the fibrous mantle structure. Thicker emitter structures and the use of low emissivity undercoats should effectively eliminate that problem. Figure 7 illustrates the data from holmia. Note that a peak occurs at the predicted wavelength of the rare Earth oxide, but blackbody radiation penetrates the fibrous mantle structure lowering the peak to background ratio.

It is very difficult to accurately determine the temperature of an emitter. Because the composite emitters are of a fibrous

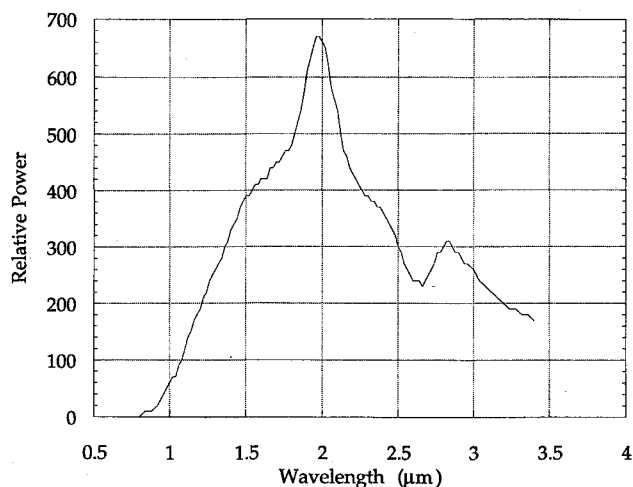


Fig. 7 Spectral characteristics for an electrically heated holmia emitter.

nature, good surface contact with the oxide fibers and a thermocouple junction is virtually impossible. Temperature is estimated by using a small dot of high-temperature paint on the radiator surface. If the dot is small, it is in approximate thermal equilibrium with the radiator. In Fig. 2, the IR imaging system is used to measure the temperature of the dot using the manufacturers specifications for the emissivity of the paint at high temperature. The high-temperature paint, Pyromark 2500<sup>®</sup>, has been described in Ref. 7. The emittance over the temperature range 1000–1400 K varied less than 5% for a variety of substrates. Using this technique, the samples discussed in this article were heated to temperatures greater than 1200 K.

The paint dot was placed on all composite emitters allowing the relative temperature of each composite emitter to be determined by assigning an emittance value (0.85) to the paint dot. The temperature of the paint dot was then assumed to be the same as the temperature of the radiating surface. This assumption may be in error by as much as 10%, but it is assumed that the error is constant since the emittance does not change significantly. Relative temperatures should be accurate. The emittance of the paint is in the process of being accurately determined by standard laboratory techniques.

The radiated power for each composite emitter, with paint dot, was measured with and without the low pass filter in place. Prior to making both of these measurements, however, the flame spectrum, corrected for grating nonlinearity, was measured with and without the filter in order that the output radiation values for the composite emitter could be properly scaled. An assumption was made that the radiation from the flame would not change greatly when the composite emitter is placed in the flow path of the combustion gases. The measurements show that essentially all of the flame radiation occurred above 2.7  $\mu\text{m}$ . Holmia, ytterbia, and erbia composite emitters were heated to three different temperatures as determined by the paint dot technique. The flow rates for the combustion burner were adjusted to the same temperature for each composite emitter. The flame emissions for each experimental test were also recorded knowing that the difference in flow rates affected the amount of radiation emitted from the flame. The flame emission intensity was subtracted from the intensity obtained from the total amount of radiation for each composite emitter.

The short wavelength radiation, below 2.7  $\mu\text{m}$ , was corrected for losses through the low pass filter. Laboratory experiments determined that the filter transmitted 90% of the short wavelength radiation incident on its surface, which is in excellent agreement with the manufacturer's specification. The filtered and unfiltered radiation intensities were used to cal-

culate the percentage of the radiation associated with the spectral region containing the selective line. Since very little radiation below 2.7  $\mu\text{m}$  was due to flame or structural material emissions, this value can be considered to be totally due to selective emissions. This ratio can also be considered to be the composite emitter selective band efficiency, which represents the percentage of total composite emitter radiation in the selective band. Figure 8 is a compilation of these data for holmia. The samples were made with sufficient binder material to represent a good compromise between radiation efficiency and strength of the emitter structure.

As expected, structural materials used in a composite emitter significantly affect the efficiency. For the temperatures investigated in these experiments, holmia has a greater percentage of radiation below 2.7  $\mu\text{m}$  than the other two rare Earth oxide composite emitters. This agrees quantitatively with the theory of Chubb.<sup>3</sup> The maximum theoretical efficiencies are a function of temperature. The selective bands can only approach an emittance of 1, i.e., not greater than that of a blackbody, then the maximum emission of the selective band should be a function of temperature and should be related to the change in the point on the blackbody curve where selective band emission occurs. The optimum temperature is related in part to the shift in the peak of the blackbody spectrum as a function of temperature. All other factors being equal, holmia should have a higher emission at a lower temperature than erbia or ytterbia, which have peaks at 1.5 and 0.955  $\mu\text{m}$ . As temperature increases, the blackbody peak shifts to shorter wavelengths, providing more thermal energy to excite the electronic states in erbia and with a further temperature increase approaches the maximum in the ytterbia emission spectrum. In agreement with Chubb's<sup>3</sup> theoretical treatment, the temperature dependence of the emission process for these rare Earth oxides appears to be linear in the temperature regime investigated in these experiments.

Referring to Fig. 8, for erbia and holmia, almost 50% of the radiation is in the selective band, using only 13% by mass of the structural materials at 1640 K. For comparison, a theoretical 1500 K blackbody has nearly 50% of its radiation below 2.7  $\mu\text{m}$ , which is spread in a continuum instead of centered at one band matched to a photocell. Also, a much greater amount of energy or fuel would be required to heat a blackbody to this temperature.

In the introductory comments, it was stated that an advantage for a selective emitter lies in the fact that less fuel would be needed to achieve a given temperature, thereby increasing the overall efficiency. An experiment was conducted to verify the fact that for a given flow rate in a combustion flame, it is possible to heat composite emitters to a higher temperature

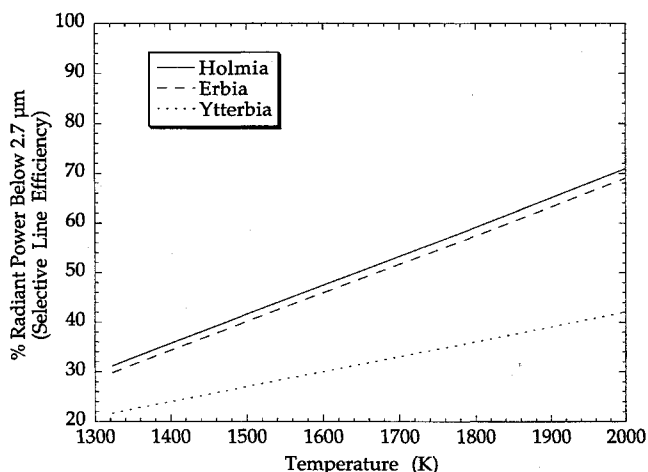


Fig. 8 Temperature dependence of the emitter efficiency for holmia, erbia, and ytterbia.

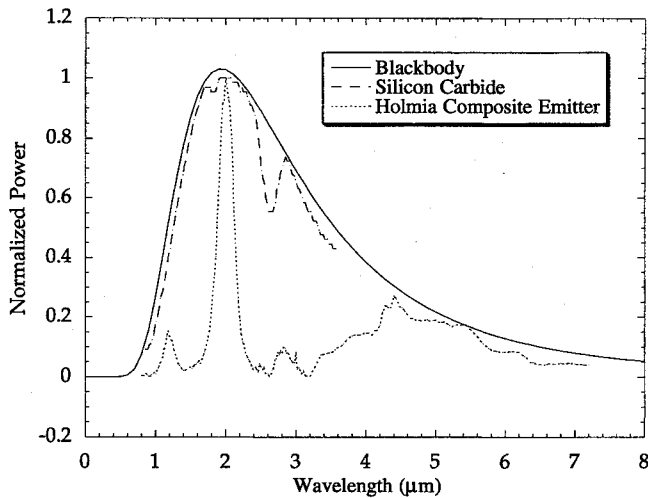


Fig. 9 Comparison of blackbody to silicon carbide and holmia composite emitters.

than a similar blackbody-like emitter. Both selective and composite silicon carbide emitters, which approximate a blackbody, were constructed with the same geometry for this experiment. The selective composite emitter was heated to 1420 K. The selective emitter was removed and the silicon carbide emitter placed in the same position with respect to the combustion flame. The blackbody-like emitter achieved a temperature of 1070 K. This simple experiment tends to verify the fact that selective emitters could improve the system efficiency of a TPV system by allowing the system to use less fuel per unit time than would be required to heat a blackbody emitter.

Figure 9 shows the normalized spectra of silicon carbide and holmia composite emitters referenced to a blackbody curve all at the same temperature. The band of radiation that is missing around  $2.7 \mu\text{m}$  in the silicon carbide emitter is due to absorption bands in the air. The area under these curves is proportional to the energy radiated and in equilibrium, equal to the energy input to the radiating structure. In the absence of other loss mechanisms, it is obvious that the energy needed to heat a selective emitter to a given temperature is much less than that needed to heat a blackbody to the same temperature. This factor is of great interest within a systems context. The energy not photoconverted must be reflected back to the source, heat exchanged into the incoming fuel-air stream, or disposed of through a thermal management scheme.

### System Concepts

Figure 10 shows conceptually how these selective emitters might be used in the conversion mechanism for a radioisotope thermal generator (RTG). Schock<sup>8</sup> has examined this concept in detail and predicts a substantial reduction in the amount of isotope necessary for a given power output when compared to that needed for thermoelectric conversion. Conceptually, the fuel could be the standard configuration used in the general purpose heat source (GPHS). The GPHS unit has an aeroshell made of carbon-carbon composite with high thermal conductivity. Unfortunately, the carbon-carbon composite would also have a high emittance. To limit the spectral output, it is necessary to have a low-emittance, high-thermal conductivity coating on the surface of the aeroshell. Reference to the *CRC Handbook of Chemistry and Physics*<sup>9</sup> indicates that copper, silver, gold, and platinum have low emittance, less than 0.2, for highly polished surfaces. Platinum with a melting temperature of approximately 2000 K is the only one of the metals with the necessary high melting temperature. Lowe et al.<sup>11</sup> have observed selective emission for thin-film rare Earth yttrium aluminum garnet emitters with platinum substrates.

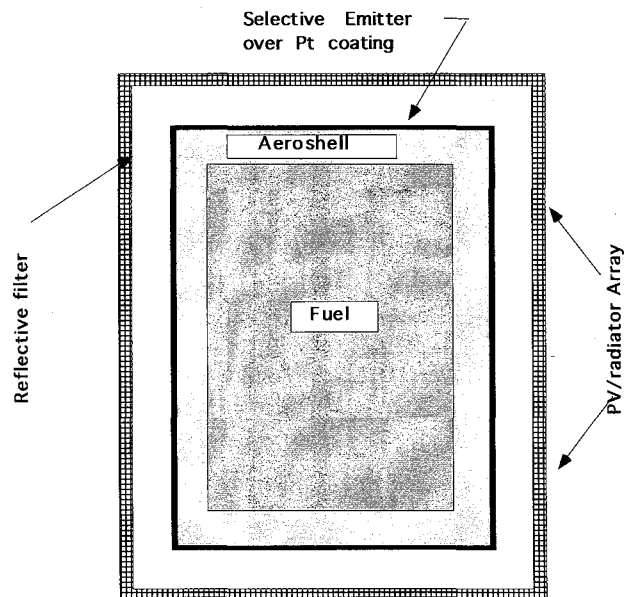


Fig. 10 Conceptual design of a nuclear-powered TPV system.

The selective emitter could be sized by knowing the thermal output of the RTG fuel, which in the vacuum of space must be radiated from the emitter surface. Referring to Fig. 10, it would be desirable to operate at the maximum temperature compatible with fuel and aeroshell structural stability. The melting temperature of the  $\text{PuO}_2$  is on the order of 2500 K. If it is possible to operate the unit at 0.75 of the melting temperature, a selective emitter temperature of approximately 1900 K would be achievable. However, it would make the use of a platinum substrate doubtful. Extrapolating the results from Fig. 8, a temperature of 1900 K would correspond to a selective emitter efficiency of about 72% for holmia, 68% for erbia, and 40% for ytterbia. From the standpoint of thermal management, it would be desirable to work with ytterbia, which is a good bandgap match to silicon. Green<sup>10</sup> has demonstrated silicon photovoltaic (PV) cells at 45% efficiency when illuminated by a laser matched to the silicon bandgap. If these parameters could be realized, the resulting system would have an efficiency approaching 20%. We have measured 20% conversion efficiency for InGaAs cells matched to erbia. At the same temperature, approximately 14% efficiency could be realized. If InGaAs could be made to perform as well as the silicon cells reported by Green,<sup>8</sup> efficiencies greater than 30% could be expected. On the surface of the PV array, it would be advantageous to employ a low pass reflective filter that would reflect the long wavelength radiation, unusable by the cell, back into the emitter-fuel assembly. This would allow a given temperature to be maintained for less fuel. The final system efficiency would be determined by the filter efficiency and any loss in PV performance as a result of operation at elevated temperature. It is conceivable that system efficiencies greater than 30% could be realized with a breakthrough in both InGaAs PV cells and reflective filter materials technology.

Figure 11 illustrates schematically a similar TPV concept powered by fossil fuels such as propane or methane. An optimum preheated fuel-air mix is injected into the combustion chamber and combusted. The hot gases from the combustion process are allowed to flow through a porous selective emitter, separated from the PV array with a window that can pass the selective band. Quartz or Nextel are good candidates for this application. The hot gases are next passed to a heat exchanger/recouperator to exchange combustion energy into preheat of the incoming fuel/air mix. It is estimated that between 40–60% of the waste heat from the combustion products can be recovered by these techniques. Combustion temperatures are

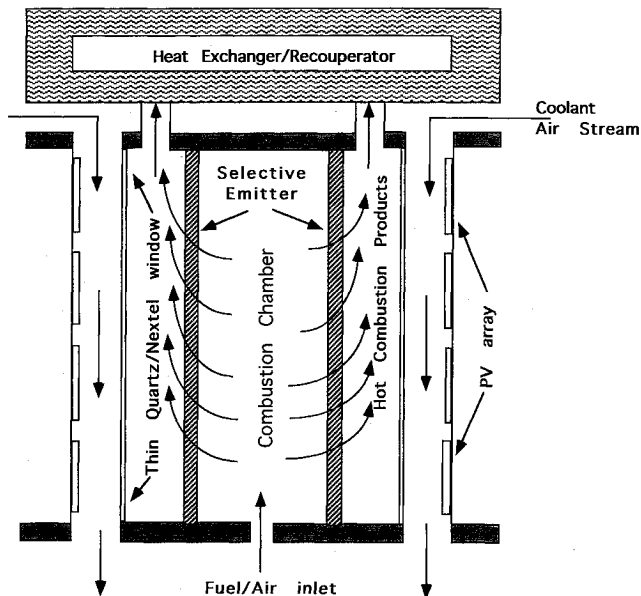


Fig. 11 Fossil-fueled TPV concept.

on the order of 2500 K and can readily heat the selective mantle to a temperature on the order of 1700–1800 K.

Quartz and Nextel as low pass filters allow the selective emission to pass to the photocells with minimal absorption. These materials absorb rather than reflect the longer wavelengths. As the filter materials heat, they subsequently emit as a gray body. This requires that the windows be cooled to prevent secondary long wavelength radiation from reaching the photocells. It would be ideal to achieve a low pass reflection filter that could withstand the requisite temperatures. In the concept shown in Fig. 11, it is necessary to keep the temperature of window and cells to less than 500 K if silicon PV cells are used and less than 400 K if InGaAs PV cells are used. To maximize the total system efficiency, it would be desirable to also recoup the heat lost in the coolant stream.

Preliminary measurements indicate that it is possible to achieve intensities greater than  $1 \text{ W/cm}^2$  of emitter. Based upon the previous estimates, a 300-W power system would need approximately a  $1000\text{-cm}^2$  photovoltaic array.

### Conclusions

This research has shown that it is possible to build low-cost robust radiators that are efficient and capable of being constructed in an arbitrary geometry. Efficiencies greater than 30% appear possible if the most optimistic improvements in

the technologies associated with the components are successful. Erbium and ytterbium emitters would appear to be the best radiator systems to use in that there is proven photocell technology waiting for applications. Ytterbium, however, needs higher temperatures for efficient radiation. It is easy to show<sup>2</sup> that selective emission offers substantial advantages in that less fuel is needed to achieve a given operating temperature if the radiator is a selective emitter and substantial out-of-band radiation cannot be recuperated. The technology is flexible and appears to have potential for both terrestrial and space applications. The most challenging problems remaining to be solved are engineering within a systems concept.

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