Radiative Performance of Rare Earth Garnet Thin Film Selective Emitters

Roland A. Lowe*, Donald L. Chubb[+] and Brian S. Good[+]

* Kent State University, Kent, Ohio 44242

[+] NASA Lewis Research Center, Cleveland, Ohio 44135

Abstract. In this paper we present the first emitter efficiency results for the thin film 40% Er-1.5% Ho YAG (Yttrium Aluminum Garnet, $Y_3Al_5O_{12}$) and 25% Ho YAG selective emitter at 1500 K with a platinum substrate. Spectral emittance and emissive power measurements were made ($1.2 < \lambda < 3.2$ μm). Emitter efficiency and power density are significantly improved with the addition of multiple rare earth dopants. Predicted efficiency results are presented for an optimized (equal power density in the Er, $^4I_{15/2} - {}^4I_{13/2}$ @ 1.5 μm, and Ho, $^5I_7 - {}^5I_8$ @ 2.0 μm emission bands) Er-Ho YAG thin film selective emitter.

INTRODUCTION

Efficient thermophotovoltaic (TPV) energy conversion is strongly dependent on emitter performance[1]. The most promising solid state selective emitters are compounds containing elements in the Lanthanide Series (rare earths). Nelson and Parent[2,3,4] using the rare earth oxide Welsbach mantle emitter have been the most successful. A new class of rare earth solid state selective emitters, the rare earth garnets[5], has the potential for being efficient, as well as, more durable than the mantle type emitter. In this paper we present the emitter efficiency results for the thin film YAG (Yttrium Aluminum Garnet, $Y_3Al_5O_{12}$) doped with holmium and erbium.

RARE EARTH GARNET SELECTIVE EMITTER

The atomic structure of the doubly and triply charged rare earth ions such as Yb, Er, Ho, Tm, Dy, and Nd accounts for their unique spectral emission characteristics. The orbits of the valence 4f electrons, whose electronic transitions determine the spectral emission properties, lie inside the 5s and 5p electron orbits. The 5s and 5p electrons "shield" the 4f valence electrons from the surrounding ions in the crystal. As a result, the rare earth ions in the solid state have radiative characteristics much like if they were an isolated atom in a gas and emit in relatively narrow bands rather than in a continuum as do most solids.

As already mentioned, the first successful rare earth selective emitters were the rare earth oxides. Recently rare earth doped YAG has shown excellent emissive properties[5]. Doping limits, dependent on the particular ion and crystal host may be

---

*NASA Resident Research Associate at Lewis Research Center.

as high as 100 at. % (complete substitution of the yttrium ion with the rare earth ion). These materials have high melting points (1940 C for undoped YAG) making them excellent candidates for a high temperature thin film selective emitter. For most of the rare earths the emission spectra is dominated by a single emission band in the near infrared ($0.9 < \lambda < 3.0$ µm) with low emission outside this band. Since YAG can be doped with more than a single rare earth it is possible to make a thin film selective emitter with multiple emission bands. As a result, greater power density over a wider wavelength interval can be achieved than with a single rare earth dopant.

The efficiency, η_E, of a selective emitter is defined as

$$\eta_E = \frac{Q_b}{Q_T}$$

(1)

where Q_b is the emitted power in the emission band and Q_T is the total emitted power. Also

$$Q_b = \int_{\lambda_u}^{\lambda_l} q_\lambda d\lambda$$

(2)

$$Q_T = \int_0^\infty q_\lambda d\lambda$$

(3)

where q_λ is the spectral emissive power of the emitter, λ_u is the short wavelength limit for the emission band and λ_l is the long wavelength limit for the emission band. In the experiment to be described in the next section q_λ is measured with a spectroradiometer and Q_b and Q_T are measured with a pyroelectric radiometer.

Experimental Procedure

Quantities such as emissivity and absorptivity are usually thought of as properties of an isothermal surface. However, for the thin film rare-earth YAG emitter emission from throughout the film contributes to the spectral emittance. Since significant temperature gradients (230 K) exist between the front and back emitter surfaces the "emitter temperature", used to calculate emittance from the intensity measurements, is not a surface temperature but rather the average of the front and back surface temperatures, T_{Eavg} in the center of the sample. A radial temperature gradient of 5 K was also measured on the sample in the field of view of the pyroelectric radiometer. This variation, small in comparison to the front-back temperature gradient and other sources of experimental error, was neglected in the emittance calculations. Temperature measurements, made with type R thermocouples, had an accuracy of +/- 6K. Total power and power in the Ho YAG

emission band was measured with a Laser Precision Rk-5100 radiometer calibrated to an NBS traceable standard. Normal spectral emittance, ε_λ, measurements were made with a spectroradiometer constructed from a 1/8 meter monochrometer, a temperature controlled PbS detector, and an 800 Hz chopper calibrated with a 1270 K blackbody reference. Energy in the emission band, Q_b, is measured with the spectroradiometer and calculated using eq. 2. Normal spectral emittance measurement accuracy and repeatability with the blackbody reference at 1270 K was typically within 2% from 1.2 < λ < 3.2 μm. A series of precision optical pinholes was used to limit the field of view at the specimen surface for both the spectroradiometer and the pyroelectric radiometer. Custom software developed with LabView for the Macintosh was used to calculate spectral emittance and emissive power, and emitted power/wavelength-interval from the intensity measurements.

Specimens were cut from Czochralski grown crystals and polished on both sides with 1μ diamond abrasive. Platinum foil was placed behind the samples as a low emittance substrate. The configuration shown in fig. 1 was used to eliminate
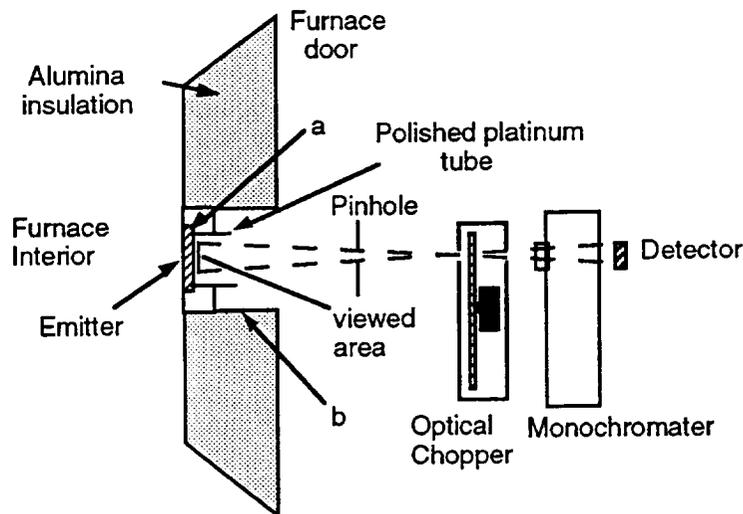
FIGURE 1. Schematic diagram of experimental setup

reflected radiation from the furnace interior, a near blackbody radiator, from reaching the detector. Two sources of measurement error were identified. Alumina insulation, a nearly perfect diffuse reflector, reflects radiation originating from the perimeter of the sample (a), which is at a higher temperature than the viewed area, to the surface of the sample in the field of view of the detector. Also, emission from the hot alumina (b) is reflected directly from the viewed area to the detector. A polished (specular) platinum tube was inserted in the hole through the insulation near the specimen to reduce the measurement error from these combined effects.

Comparison of ε_λ measurements of polished platinum foil at 1648 K and data from ref. 6 show the increase in ε_λ due to reflected radiation and temperature measurement error is +0.06 @ 2.0 μm and +0.07 @ 1.5 μm. This will be the

maximum measurement error for materials with high spectral reflectance (~80 %) and low emittance such as platinum . However, in the Ho and Er-YAG specimens, reflectance in the emission band is less (20-70% for Ho-YAG) and the actual measurement error will be lower than the error for platinum.

Efficiency Results

Thin film selective emitter performance is dependent on factors such as emitter thickness and temperature, substrate emittance[7] and rare earth dopant type and concentration. For maximum emitter performance it is essential that these parameters be optimized. The experimental results presented here for the Ho and Er YAG thin film selective emitters do not include efforts to maximize performance through optimization of these parameters. The test temperature (1500K) is high enough to produce reasonable emission levels for optical detection but is too low to achieve the maximum predicted efficiency for Ho (1800 K) and Er (2400 K) emitters. The thickness and doping level of the test specimens reflects the commercial availability of material developed for solid state lasers. Even without optimization results indicate that rare earth thin film selective emitters have potential as elements in a high efficiency TPV system.

A co-doped Er-Ho YAG emitter was used to investigate the efficiency of Er YAG at 1500 K. Since the Ho doping concentration, 1.5%, and hence the emission at 2.0 μm, is significantly less then the Er doping concentration, 40%, efficiency results are expected to be similar to that of the 40% Er YAG emitter in ref. 5. Fig. 2 shows the normal spectral emittance of a 40% Er-1.5% Ho YAG emitter 1.5 mm
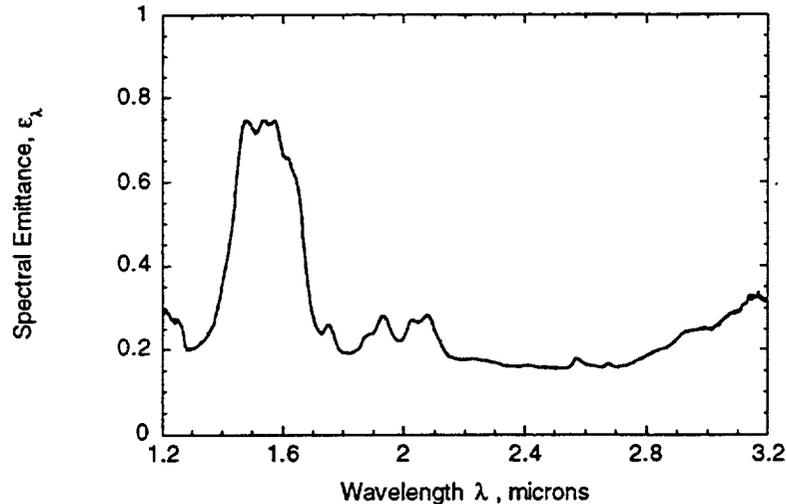
FIGURE 2.   Spectral emittance of the 40% Er - 1.5% Ho YAG
thin film selective emitter

thick at 1472 K from 1.2< λ < 3.2 μm. Relatively strong emission is present at the

characteristic laser transitions for Er YAG ($^4I_{15/2}$ - $^4I_{13/2}$ @ 1.5 μm) and Ho YAG ($^5I_7$ - $^5I_8$ @ 2.0 μm).

Several methods were used to calculate values for emitter efficiency using eq.1. First, the power in the Er emission band, Q_b, from λ_u = 1.3 to λ_l = 1.8 μm is divided by the total measured power, Q_T. Second, the power in the Er and Ho emission bands from λ_u = 1.3 to λ_l = 2.2 μm, is divided by the total measured power. Third, the power in the Er emission band, from λ_u = 1.3 to λ_l = 1.8 μm, is divided by the total measured power reduced by the power in the Ho emission band. To estimate the power in the Ho emission band the integrated power using a constant emittance of 0.16 from 1.8 to 2.53 μm @ 1472 K was subtracted from the integrated power over the same wavelength interval from the experimental results. The contribution of the Ho emission band to the total radiated power using this method is 0.26 watts/cm^2.

The normal spectral emittance of a 25% Ho YAG thin film selective emitter 0.65 mm thick @ 1477 K is shown in fig. 3. Two methods were used to calculate
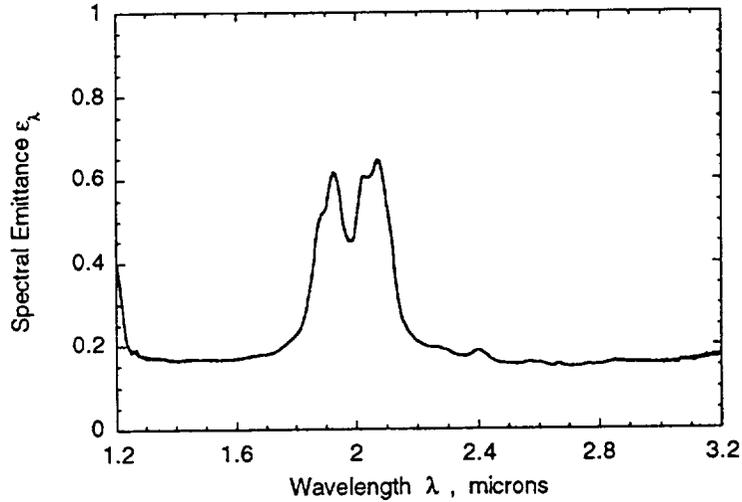
FIGURE 3. Spectral emittance of a 25% Ho YAG thin film selective emitter

the efficiency of this emitter. First, the power in the emission band, λ_u = 1.75 to λ_l = 2.2 μm, measured with the spectroradiometer is divided by the total measured power. Second, the power in the emission band, λ_u = 1.85 to λ_l = 2.25 μm, is measured with an interference type bandpass filter and the pyroelectric radiometer and divided by the total measured power. Table 1 summarizes the results of the efficiency calculations for the Er-Ho and Ho YAG emitters.

The experimental results suggest that co-doping the emitter with more than one rare earth significantly improves the power density and radiative efficiency of the emitter. Using spectral emittance data for the Er-Ho and Ho emitters the spectral emissive power is calculated at T_{Eavg} = 1477 K and shown in fig. 4 simulating the

5

TABLE I. Radiative Efficiency of the YAG Thin Film Selective Emitter

Type	$T_{E_{avg}}$	Method	Q_b (watts/cm^2)	Q_T (watts/cm^2)	η_E (%)
40% Er - 1.5% Ho	1472 K	1	1.74	9.33	18.7
		2	2.55	9.33	27.3
		3	1.74	9.07	19.2
25% Ho	1477 K	1	1.72	9.75	17.7
		2	1.92	9.75	19.7
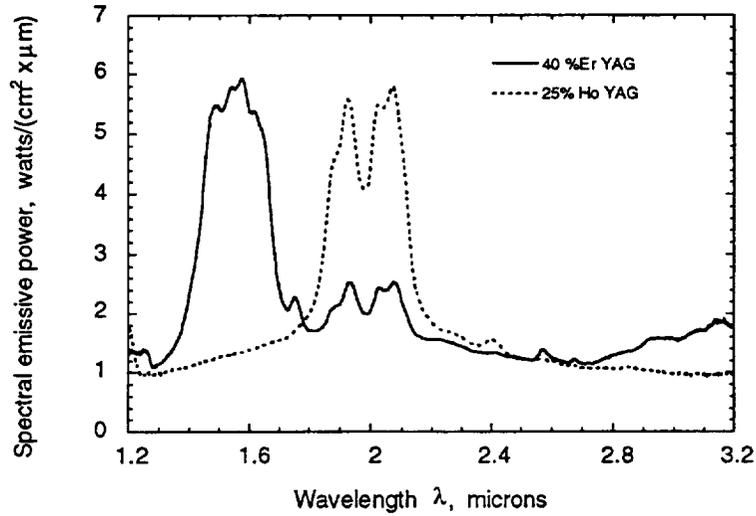
FIGURE 4. Spectral emissive power of an optimized Er-Ho YAG emitter

performance of an "optimized" Er - Ho emitter. Using the combined power in Er emission band, $Q_{Er} = 1.75$ watts/cm^2 ($\lambda_u = 1.3$, $\lambda_l = 1.78$ μm), and Ho emission band, $Q_{Ho} = 1.67$ watts/cm^2 ($\lambda_u = 1.78$, $\lambda_l = 2.2$ μm), the total power in the emission band $Q_b = 3.42$ watts/cm^2. To estimate Q_T, the contribution of the Er emission band, Q_{Er} minus the integrated power from 1.3 to 1.78 μm assuming a emitter is estimated to be constant emittance of 0.16, is added to the total power of the Ho YAG emitter, and $Q_T = 10.9$ watts/cm^2. As a result, the efficiency of an "optimized" Er-Ho thin film selective is 31.4%.

Further improvement in emitter efficiency may also be achieved by reducing the front-back emitter temperature gradient. Since the extinction coefficient for rare earth doped YAG is relatively low outside the emission band (~ 0.2 cm^{-1}) the emission in this region is dominated by the platinum substrate. Reducing the temperature gradient, with the back surface temperature constant, will increase Q_b while the emission from the substrate remains constant, improving[8] η_E. In this

experiment convection loss contributes to the large emitter temperature gradient. Operating the emitter with a front surface vacuum interface would eliminate this loss, reduce the temperature gradient, and improve η_E.

CONCLUSION

The first measured results for the "non optimized" emitter efficiency of rare earth YAG thin film selective emitters show that the emitter efficiency may be low, evenwith high emittance in the emission band (~0.7) and low out of band emittance (~0.15), if the emission band is relatively narrow. Multiple emission bands from additional rare earth species in the emitter material increase the overall emission band width significantly improving emitter efficiency and usable power density. Additional improvements in efficiency and power density should also be possible through optimization of emitter film thickness and temperature, rare earth doping level, and reducing the emitter temperature gradient.

REFERENCES

1. Chubb D.L., "Reappraisal of Solid Selective Emitters", in *Proceedings of the 21st Photovoltaic Specialists Conference* IEEE, New York 1990 pp 1326-1342, also NASA TM103290, 1990
2. Nelson R.E., in *Proceedings of the 32nd International Power Sources Symposium*, Electrochemical Society, Pennington, NJ, 1986 pp 95-101
3. Parent C.R. and Nelson R.E., in *Proceedings of the Twenty First Intersociety Energy Conversion Engineering Conference>*, American Chemical Society, Washington DC, 1986 vol. 2, pp 1314-1317
4. Nelson R.E. and Iles P.A., "Space Solar Power for Orbital and Lunar Missions", *ASME/ASES/SOLTECH Joint Solar Energy Conference*, 1993
5. Lowe R.A., Chubb D.L., Farmer S.C. and Good B.S., *Appl Phys. Letters*, **64**, 3551 1994)
6. Touloukian Y.S. and Ho C.Y., *Thermophysical Properties of Matter vol. 7* "*Thermal Radiative Properties Metallic Elements & Alloys*", (IFI/Plenum, New York, 1970, p. 542 (curve 24)
7. Chubb D.L. and Lowe R.A., *J. Appl Phys*, **74**, (1993)
8. Good B.S., Chubb D.L. and Lowe R.A.,"Temperature-Dependent Efficiency Calculations foe a Thin Film Selective Emitter", in *First NREL Conference on Thermophotovoltaic Generation of Electricity*, American Institue of Physics, AIP Press, Woodbury NY, 1994

REPORT DOCUMENTATION PAGE

1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	August 1994	Technical Memorandum

4. TITLE AND SUBTITLE

Radiative Performance of Rare Earth Garnet Thin Film Selective Emitters

6. AUTHOR(S)

Roland A. Lowe, Donald L. Chubb, and Brian S. Good

5. FUNDING NUMBERS

WU–233–01–0A

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

National Aeronautics and Space Administration
Lewis Research Center
Cleveland, Ohio 44135–3191

8. PERFORMING ORGANIZATION REPORT NUMBER

E–9088

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

National Aeronautics and Space Administration
Washington, D.C. 20546–0001

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

NASA TM–106722

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT

Unclassified - Unlimited
Subject Categories 44 and 76

12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)

In this paper we present the first emitter efficiency results for the thin film 40% Er-1.5% Ho YAG (Yttrium Aluminum Garnet, $Y_3Al_5O_{12}$) and 25% Ho YAG selective emitter at 1500 K with a platinum substrate. Spectral emittance and emissive power measurements were made ($1.2 < \lambda < 3.2$ μm). Emitter efficiency and power density are significantly improved with the addition of multiple rare earth dopants. Predicted efficiency results are presented for an optimized (equal power density in the Er, $^4I_{15/2}$ - $^4I_{13/2}$ @ 1.5 μm, and Ho, $^5I_7$ - $^5I_8$ @ 2.0 μm emission bands) Er-Ho YAG thin film selective emitter.

14. SUBJECT TERMS

TPV; Thermophotovoltaic; Selective emitter

15. NUMBER OF PAGES
9

16. PRICE CODE
A02

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
Unclassified	Unclassified	Unclassified	