Development of the Phosphor Thermometry Technique for Applications in Gas Turbines

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Abstract

The research described in the paper is concerned with the development of a surface thermometry technique, utilising thermographic phosphors, for application in gas turbine combustors and high temperature regions of the turbine were the application of conventional techniques is problematic. Thermographic phosphors consist of a ceramic host matrix with a lanthanide ion dopant. When illuminated with UV light they exhibit phosphorescence which is temperature dependent by virtue of variations in the relative intensities of distinct emission lines or in the time constant of the exponential decay which occurs once excitation has ceased. YAG:Dy, YAG:Tb and Y_2O_3:Eu have been selected as phosphors suitable for use in high temperature gas turbine applications. A calibration of their response is described herein and they are shown to be temperature sensitive over a range form room temperature to in excess of 1200^{o}C and to be suitable for the measurements with a precision of up to $\pm$0.2^{o}C.

The authors have proposed the concept of a smart thermal barrier coating with both insulation and temperature sensing properties. A candidate material (YSZ:Eu) has been manufactured, tested and shown to be temperature sensitive over a range from room temperature to 800^{o}C and suitable for measurements with a precision of up to $\pm$0.05^{o}C.

Further work will involve the application of thermographic phosphors for surface temperatures in a model gas turbine combustor. A new receiving optics module has been designed for this task and is reviewed in the paper.

Introduction

The trend in gas turbines development is for increasing turbine entry temperature due to the improvements in overall efficiency that can thereby be obtained. Temperature increases have been made possible by the use of advanced wall cooling schemes and of protective coatings made from refractory materials and turbine entry temperatures are already higher than superalloy materials, from which adjacent components are made, can withstand. The desire to increase temperature remains, but future designs will also have to take account of commercial constraints in terms of increased component lifetime and of more stringent emission regulations[2,4,15]. Schemes to limit emissions may reduce the amount of air available for component cooling thereby necessitating more efficient cooling and/or more extensive and more efficient use of protective coatings. The design of efficient cooling schemes and of effective, robust, protective coatings requires detailed knowledge of the temperatures, heat fluxes and flow regimes experienced by components under realistic conditions.

In combustors and high temperature regions of the turbine the application of conventional temperature measurement techniques is problematic. Infrared pyrometry is affected by interference from stray light (from flames for example) and by changes in surface emissivity whilst thermocouples may not survive the rigours of the environment, are intrusive, and are not suitable for use on rotating components. Research conducted by the authors, however, has concerned the development of a temperature measuring technique utilising thermographic phosphors for surface temperature measurement which may enable these problems to be overcome. It is well known that certain luminescent materials referred to as thermographic phosphors have emission characteristics that are temperature sensitive. They consist of a ceramic host matrix doped with rare earth (lanthanide) ions in a solid solution where the lanthanide concentration is typically between 1% and 10%. Examples are YAG:Tb, YAG:Dy and Y_2O_3:Eu. To make temperature measurements a thin layer of phosphor is deposited on the surface under investigation. When illuminated with UV light (typically from a pulsed laser) the coating exhibits phosphorescence which is temperature dependent by virtue of variations in the relative intensities of distinct emission lines or of the time constant of the exponential decay in emission which occurs once excitation has ceased. The former mode lends itself to the measurement of surface temperature distributions since a spatial variation in the emission intensity can be recorded, as an image, using a CCD video camera. The latter mode requires an exponential decay with a time constant of typically 100μs to be characterised. This can be best achieved using a photomultiplier and hence this mode is best suited to point temperature measurement. YAG:Dy is the only phosphor to date which has been shown the exhibit the intensity ratio response mode but all the phosphors mentioned, and indeed all those known to the authors, exhibit the lifetime decay response mode.

There are a number of features of thermographic phosphors which indicate that they should be well suited to the measurement of surface temperatures in gas turbine combustors and high temperature regions of the turbine. The phosphor emission lines are well defined and it is hence possible to distinguish them from broadband background emission using bandpass filters. The lifetime decay characteristics of the emission are independent of absolute intensity values and for both response modes emission is independent of blackbody radiation from the surface and of the surface emissivity. Furthermore, the phosphors are based on a ceramic host matrix and are hence resistant to high temperatures - the melting point of YAG:Tb, for example, being around 2100^{o}C.

The aim of the research conducted by the authors has been to evaluate thermographic phosphors for gas turbine temperature measurements and to develop a measurement system suitable for use with a laboratory based combustion rig. The work has included a review of a range of phosphors, including those mentioned above, which have been calibrated in a modified furnace. A review of coating techniques has also been carried out. In addition, the authors have investigated the possibility of developing new phosphors consisting of yttria stabilised zirconia (the material usually used to make the protective 'thermal barrier coatings' in gas turbines) doped with lanthanide ions. With this phosphor it is proposed to manufacture a smart thermal barrier coating offering both thermal insulation and instrumentation properties.

In the paper the basic physics of thermographic phosphors and the methods by which phosphor coatings may be made will be reviewed. Calibration data for a range of phosphors, including the yttria-stabilised zirconia (YSZ) based phosphor will be presented and the instrumentation developed for use with the model combustor rig will be discussed.

Phosphor Physics

Thermographic phosphors for high temperature applications consist of a ceramic host material doped with rare-earth ions (Lanthanides) at concentrations typically between 1% and 10%. The lanthanide ions produce well defined electronic states located in the band gap of the ceramic host. Excitation of the phosphor by a light source such as a laser generates electrons in these energy states. Depopulation may then occur by both radiative and non-radiative means. The radiative process causes the luminescence effects, which are observed for temperature measurement, but competes with a phonon quenching process which is non-radiative. The latter become increasingly dominant at high temperatures and it is this which influences the emission properties and leads to temperature sensitivity.

The exponential decay in phosphorescence which occurs when illumination of the phosphors ceases can usually be characterised by the decay time constant τ of a single exponential function of the form:

$$I(T) = I_0 e^{\left(-\frac{t}{t(T)}\right)} \qquad (1)$$

where I is the measured intensity of the luminescence and I_0 the intensity at time t=0. Sometimes a multi-exponential decay function must be applied when a single exponential approach is found to be inaccurate. Decay times of the phosphors employed in high temperature applications typically below 1ms and the lifetime decay method has been applied to measure temperature on rotating elements such as turbine blades with examples of previous work including Bird et al.[4], Allison et. al.[2] and Tobin et al.[20]. The lifetime decay method may also be used at low temperatures and an example is the work of Allison & Cates et al.[3] who made measurements below 100° C.

In the intensity ratio method the relative intensities of two distinct emission lines, at equilibrium, under constant illumination is measured and found to be temperature sensitive[14,19]. Relative intensities reflect the relative electron population of two adjacent energy states where the populations can be described using the Boltzmann equation

$$n_1 = n_2 . e^{\left(-\frac{\Delta E}{k_B T}\right)} \qquad (2)$$

which indicates the temperature sensitivity of the ratio. Here n_2 and n_1 are the electron populations of the lower and upper level respectively. ΔE the energy gap between them, T the temperature in Kelvin and k_B the Boltzmann constant. Experiments have confirmed the applicability of this equation as will be demonstrated in the results section. Successful use of the intensity ratio measurements was demonstrated by Goss[14], Bizzak & Chyu[5,11], Ervin[13] et al and Turley et al[21]. The method has been shown to be applicable for temperature measurements on curved surfaces[13] with the possibility of measuring two-dimensional temperature distributions and of the measurement of heat flux[5,11]. It has also been used to measure the temperature of a reacting surface[14]. A detailed review of the physics of phosphors can be found in the publications of Hüfner[18], Blasse[6] and Wybourne[22].

Coating Techniques

An essential component in the successful application of the thermographic phosphor thermometry technique in gas turbines is the production of a reliable phosphor coating, on the surface under investigation, capable of withstanding a hostile, high temperature environment possibly with combustion. Several methods have been investigated for the fabrication of phosphor films. Chemical binders can be used to prepare phosphor paints [4] and produce good quality films for use in development testing but are limited to application at a maximum temperature of around 1100 °C.

Other techniques, such as chemical vapour deposition[17] pulsed laser ablation[17] and physical vapour deposition (PVD) (e.g., RF-sputtering and electron-beam deposition[1]) are known to produce more robust films but require complicated and expensive equipment and a controlled atmosphere for their application. Moreover, the PVD methods are line-of-sight processes, which are not easily scaled up for films on large components of complicated shape such as combustion chambers. However, a new vapour deposition technique called electrostatic assisted chemical vapour deposition (EACVD) has recently been developed in the Materials Department at Imperial College by Choy et al[7,8]. This novel technology has been shown to be capable of depositing a wide range of oxide films in a cost-effective manner. The process involves spraying atomised precursor droplets across an electric field where the droplets undergo combustion and chemical reaction in the vapour phase near the vicinity of the heated substrate. This produces a stable solid film with excellent adhesion onto the substrate in a single production run. The process is capable of producing thin or thick strongly adherent films with well-controlled stoichiometry, crystallinity and texture and has already been applied to the deposition of simple, multi-component and doped oxide films.

In work carried out to date, the authors have primarily used the paint technique with paints prepared using proprietary chemical binders similar to those used in the production of thermal paint. However, a feasibility study has also been carried out using the new EACVD method. A Y_2O_3:Eu coating approximately 5μ thick was laid down on a Nickel alloy substrate. It was found to show temperature sensitivity similar to that of a painted coating but was resistant to a temperature of 1200°C with no signs of degradation. Details of the method by which the coating was produced and the experiments conducted with it can be found in Choy et al[10].

Experimental Procedures

The experiments described below include calibration of a range of phosphors thought suitable for high temperature applications and the production and testing of a new phosphor material suitable for smart thermal barrier coatings.

The lifetime decay characteristics of the various phosphor coatings was determined using the set-up shown in figure 1. A pulsed YAG:Nd laser (Spectra Physics; Model 201) was used to excite samples, which were housed in a furnace capable of reaching temperatures up to 1200°C and specially modified to provide optical access to the samples. The laser was operated at 266nm or 355nm (with Q-switch), a repetition rate of approximately 16 Hz and with output energy of about 60 mJ or 100mJ per pulse for the two wavelengths respectively. As shown in Fig. 1, an external beam dump was incorporated to avoid accidental irradiation of the samples by the 532 nm emission line of the laser, present due to leakage from the harmonic crystal assembly. The beam was steered into the furnace and onto the samples through a synthetic fused silica window of diameter 25mm. Subsequent luminescence was observed through a second similar window using a standard 50mm camera lens, which focused an image of the sample on to the entrance slit of a crossed Czerney-Turner spectrometer (Jarrell-Ash MonoSpec 18). The systems optical performance was limited by the diameter of the observation window and this was estimated to reduce the effective f-number of the collection optics from 3.8 to 12. A photomultiplier was placed at the exit slit of the spectrometer and used to measure the decay lifetime. An analogue to digital converter (PICO; ADC-200; 50MHz; 8-bit resolution) transferred data simultaneously from the PMT and the power meter to a personal computer. An exponential decay was fitted using custom written software to either a single shot or to the average of a series of pulses. The power meter data was used to monitor irradiation of the sample during testing and for triggering purposes.

For the intensity ratio measurements the apparatus was modified to allow the emission spectra of the phosphors to be observed. The PMT was replaced with a linear CCD array (Alton LS2000) which was linked to a grabber card housed in the computer. Spectra were obtained by integrating the response of the phosphor over approximately 0.5s corresponding to seven laser pulses

Using the above apparatus the temperature sensitive emission characteristics of coatings of the commonly used phosphors Y_2O_3:Eu, YAG:Tb and YAG:Dy were studied under the lifetime decay mode with YAG:Dy also studied under the intensity ratio mode. For the results discussed herein, all the coatings were produced by the paint technique with the paint produced by mixing commercially available phosphor powder (Phosphor Technology) of particle sizes between 1μm and 10μm into a high temperature binder. The resulting slurry was sprayed onto a Ni-alloy substrate and oven cured for several hours at temperatures up to 1000 °C. The thickness of the coatings were between 14μm and 50μm.

The concept of a smart thermal barrier coating has been proposed by the authors and Choy[9] and as a first stage in assessing its feasibility a sample of an appropriate phosphor has been prepared and tested. It consisted of yttria stabilised zirconia doped with europium. YSZ is the material most commonly used to make thermal barrier coatings due to a number of favourable properties including phase stability, and hence low thermal expansion coefficient, over a wide temperature range encompassing typical gas turbine operating conditions. Europium was chosen as the lanthanide dopant since the manufacturing process for YSZ:Eu was known from Dexpert-Ghys et al who investigated the luminescent properties of single crystals of the material at a fixed low temperature in order to obtain structural information. A sample of YSZ:Eu was obtained from a commercial source where it was manufactured in powdered form according the process described by Dexpert-Ghys and with composition $90ZrO_2$-$9YO_{1.5}$-$1EuO_{1.5}$. The lifetime decay characteristics of the YSZ:Eu powder were investigated using the set-up shown in figure 1 and described above with the sample housed in cuvette made from fused silica and placed on a ceramic stand to provide good thermal contact with the furnace.

Results and Discussion

Figure 2 shows the lifetime decay characteristics of the three established phosphors over a temperature range extending from 500°C to 1200°C for the main emission lines of each. Each measurement point corresponds to the average time constant derived from 10 individual laser pulses. From the figure it can be seen that the indicated dynamic range of Y_2O_3:Eu extends from about 500°C to 750°C. However, previous workers[20] have reported dynamic response for this phosphor at temperatures above 800°C. The shortfall in response was found to be due to limited frequency response in the PMT amplifier system which restricted the minimum lifetime decay time constant detectable to around 100µs. The system has subsequently been modified and can now resolve decay time constants down to 3µs although the measurements with Y_2O_3:Eu are yet to be repeated.

From the figure it can be seen that YAG:Tb and YAG:Dy both have a dynamic range greater than that for Y_2O_3:Eu and in the case of YAG:Dy it extends to temperatures in excess of 1200°C (which was the maximum temperature that could be obtained with the furnace used in the current investigation). The standard error was calculated for each measurement point but no error bars have been shown on graph. This is because, in practise, they are too small to be represented demonstrating the excellent repeatability of the results. As an example, at around 1100°C the YAG:Dy curve reaches a maximum gradient of approximately -2.2µs/°C. At this value, the standard error in the measured time constant implies a temperature uncertainty of ±0.2°C although it should be recognised that this represents the region of the distribution where errors are smallest.

The emission lines for YAG:Dy and YAG:Tb are in the blue-green part of the spectrum and at these wavelengths blackbody radiation is weaker than at the red end i.e. at the emission wavelength of Y_2O_3:Eu. This gives a better signal to noise ratio and, hence, these phosphors are better suited for applications in combustion chambers. Of the two, YAG:Tb shows the wider dynamic range, its dynamic range extends to lower temperatures and the response curve has the steepest gradient implying greater accuracy but YAG:Dy has a dynamic range extending to higher temperatures and indeed to temperatures in excess of 1200°C.

For the intensity ratio method the experimental arrangement was changed, as described previously, and spectra were obtained by integrating the response of the phosphor (YAG:Dy) over approximately 0.5s, corresponding to seven laser pulses. Figure 3 shows the spectrum of the phosphor scaled using the 493nm emission line as a reference. From the figure, the increase with temperature of the relative intensity of the 455nm line with respect to 493nm line can clearly be seen . The intensities of the 493nm and the 455nm line were calculated by integration of the lines over a region of width approximately 1nm. An Arrhenius plot (figure 4) presents the ratio versus the temperature and confirms an underlying Boltzmann distribution. Thus the slope determines the energy gap and from the data ΔE was calculated to be $1490cm^{-1}$ in units of the wave number. ΔE may also be calculated from the difference in wavelength between the emission lines. From 455nm ($21978cm^{-1}$) and 493nm ($20325cm^{-1}$) it follows that there is an energy gap of about $\Delta E=1653cm^{-1}$. The difference between the two ΔE values might be explained with additional line splitting since the model assumed only two electronic states. Figure 4 shows a linear response in the emission intensity ratio, in accordance with the Boltzmann equation, for temperatures from 600°C to 1200°C. Below 600°C the response of the phosphor departs from that described by the simple Boltzmann relation. However, by calibration, temperatures can be measured down to at least room temperature. Based on the linear region of the response function the error in temperature measurements made using the intensity ratio technique are currently estimated to be around ±5%. Part of this relatively large error is believed to be attributable to the continuous integration period used to record the spectra. Based on the emission decay time constant for YAG:DY it is estimated that there was no phosphor emission for over 99% of the integration period so that a significant amount of background emission was collected. The situation could be improved by using a gated detector to eliminate the background and it expected that, under these circumstances, uncertainty could be reduced.

From the results obtained with YAG:Dy it can be seen how this phosphor could be used to obtain surface temperature distributions. Two CCD cameras can be used to record images of the coated surface where each camera views the surface through a band pass filter one centred on the 455nm line and the other centred on the 493nm line. In this way intensities from corresponding pixels in the two recorded images could be used to form the emission intensity ratio and hence obtain the temperature. This principal has been demonstrated by Goss[14] who measured a one-dimensional temperature distribution on a reacting surface.

From the calibration data which has been obtained it is possible for the author to make recommendations regarding the selection of a phosphor for temperature measurements in gas turbine combustors and turbines stages and from those considered the phosphor which appears to be the most appropriate is YAG:Dy. This phosphor shows sensitivity in both the lifetime decay and intensity ratio modes and may therefore be used for measurement of surface temperature distributions and for high accuracy point measurements using the lifetime decay method. YAG:Dy has the widest dynamic range of the phosphors considered (if both response modes are considered) and its dynamic range extends to the highest temperature corresponding to values which may be expected for turbine entry temperatures in future generations of gas turbines. Excitation of YAG:Dy is at 355nm, as opposed to 266nm for the other phosphors considered, and at this wavelength transmission of laser pulses down optical fibres is considerably easier which bodes well for temperature measurement in enclosed rigs. The emission from YAG:Dy is at a relatively short wavelength (493nm as opposed to 543nm and 611nm for YAG:Tb and Y_2O_3:Eu respectively) and, since here interference from black body radiation diminishes, the prospects for surface temperature measurement on high temperature components are good.

An investigation of the characteristics of the new YSZ:Eu material revealed that phosphorescence occurs but only at the 266nm excitation wavelength with no emission observed for excitation at 355nm. An investigation of the emission spectrum at various temperatures revealed no intensity ratio type response. However, there was a lifetime decay temperature response and this is shown in figure 5. From the figure it can be seen that the dynamic range of YSZ:Eu is relatively large extending from room temperature to 800^{o}C. The upper limit was again restricted by the bandwidth of the PMT-amplifier system. The upgraded system was employed for this experiment but at 800^{o}C the decay time constant is approaching the 3μs limit. Each point in figure 5 represents 250 laser pulses and the standard error in the time constant indicates that, here, temperatures could be measured with a best precision ±0.05^{o}C. These results indicate that it should be possible to produce a smart thermal barrier coating with both insulation and temperature sensing properties and further proves that thermographic phosphors of the form YSZ:La (Lanthanide) are viable.

Further Work

Future work will be concerned with the development of further new phosphor materials suitable for the development of smart thermal barrier coatings and of the development of coating techniques suitable for laying down phosphor and smart TBC layers. Given that the viability of YSZ:La as a thermographic phosphor has been established, the possibility of using dopants other than Eu will be investigated and in particular, given the calibration results presented above, Dysprosium (Dy) will be considered. Of the coating techniques considered to date, the EACVD technique appears to offer the greatest benefits and hence work is planned to develop this technique for laying down both phosphor and smart TBC layers.

In addition to the above, work is currently underway to develop instrumentation suitable for the measurement of surface temperatures in a model gas turbine combustion chamber. It consists of a single sector of a research combustor (provided by Rolls Royce) operated at realistic air fuel ratios and atmospheric pressure. A full description of the rig can be found in Heyes et al[16]. An optically accessible region of the combustor wall has designated for the experiments and has been coated with samples of YAG:Tb, YAG:Dy and Y_2O_3:Eu using the paint technique. To facilitate temperature measurements in the combustor a new optics module has been constructed and is shown in figure 6. With this unit, light is collected using a camera lens and focussed onto a pinhole entry. Inside the box a dichroic mirror divides the light and delivers it via line filters to two PMT's. This device can be used for either lifetime decay measurements, where two lines can be simultaneously observed if desired, or for intensity ratio measurements on a point basis.

Concluding Remarks

The thermo-luminescent properties of selected phosphors (Y_2O_3:Eu, YAG:Tb, YAG:Dy) have been studied using laser irradiation over a temperature range from room temperature to 1200^{o}C. Both the lifetime decay and intensity ratio response modes have been considered and all three phosphors have been shown to be suitable for high temperature measurements. However, YAG:Tb and particularly YAG:Dy are preferred for applications in gas turbine combustors since the emission lines of these phosphors are further from the infrared end of the spectrum than those for Y_2O_3:Eu and since they have dynamic ranges extending to higher temperatures. YAG:Dy is particularly favoured since it is the only phosphor known to exhibit both response modes and since its dynamic range extends to more than 1200^{o}C.

The viability of producing smart thermal barrier coatings with both insulating and temperature indicating properties has been investigated by the production and characterisation of YSZ:Eu powder - a candidate material for the smart coating. YSZ:Eu was shown to be a thermographic phosphor with lifetime decay response and a dynamic range extending to 800°C. Further work is required to establish if other lanthanide dopants can be used in the YSZ matrix and the dynamic range extended.

Preparations have been made for a study of surface temperatures in a model gas turbine combustor. The combustor has been coated with samples of Y_2O_3:Eu, YAG:Tb and YAG:Dy laid down using the paint technique and a new receiving optics unit suitable for both lifetime decay and intensity ratio measurements has been designed and constructed.

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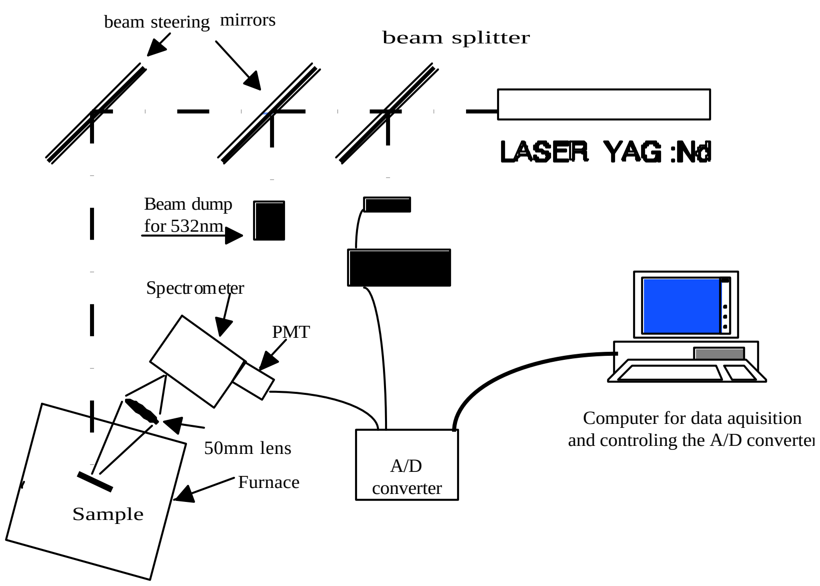

Figure 1: Experimental arrangement

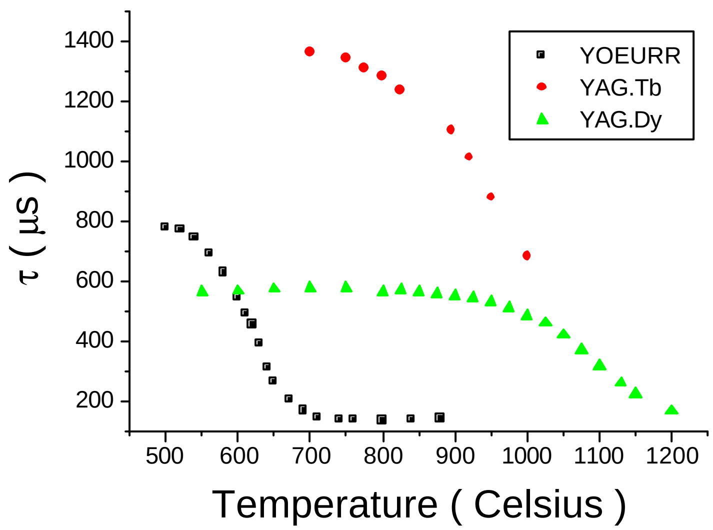

Figure 2 Lifetime decay response characteristics of selected phosphors

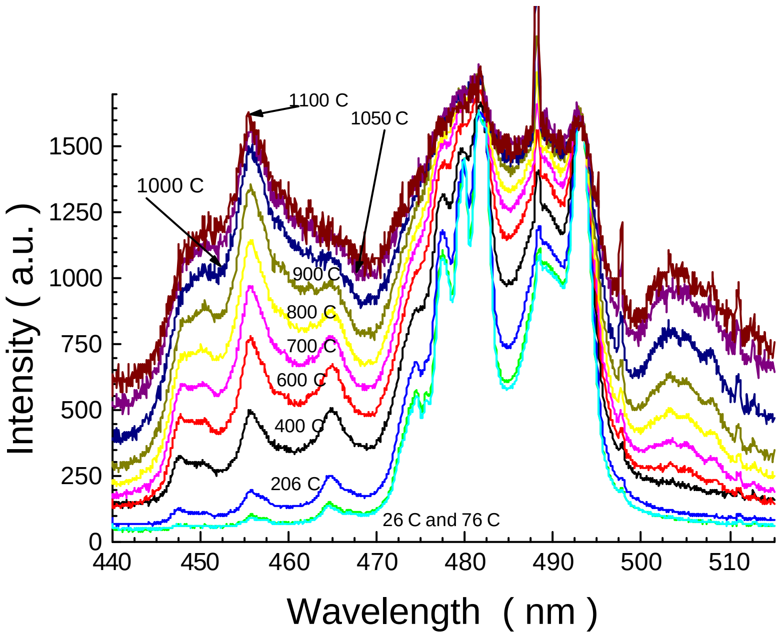

Figure 3: Scaled spectrum of YAG:Dy illustrating the increasing emission lines around 455nm.

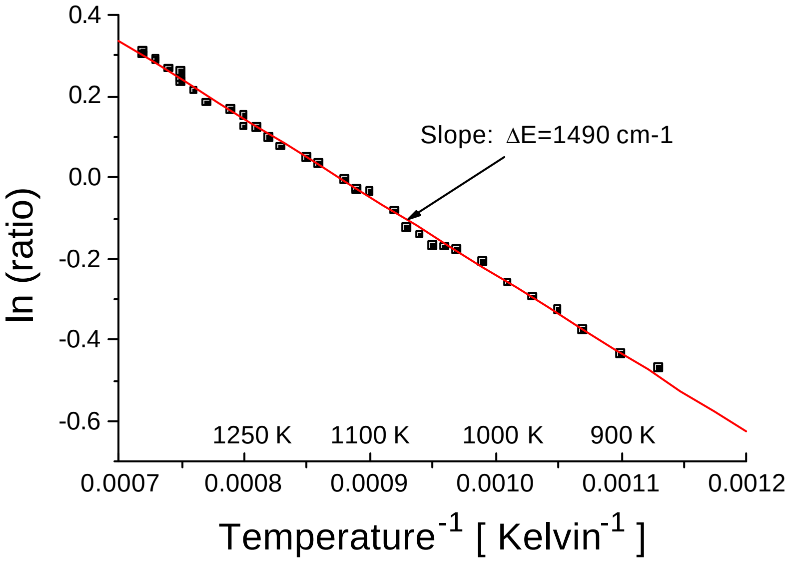

Figure 4: Arrhenius plot of the intensity ratio for YAG:Dy

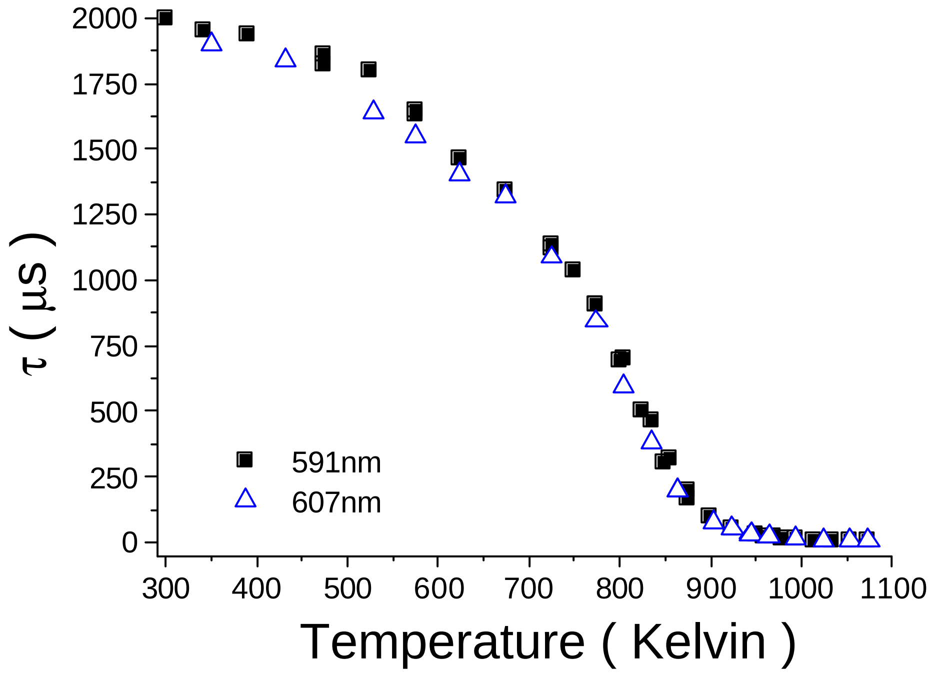

Figure 5 Lifetime decay response of YSZ:Eu

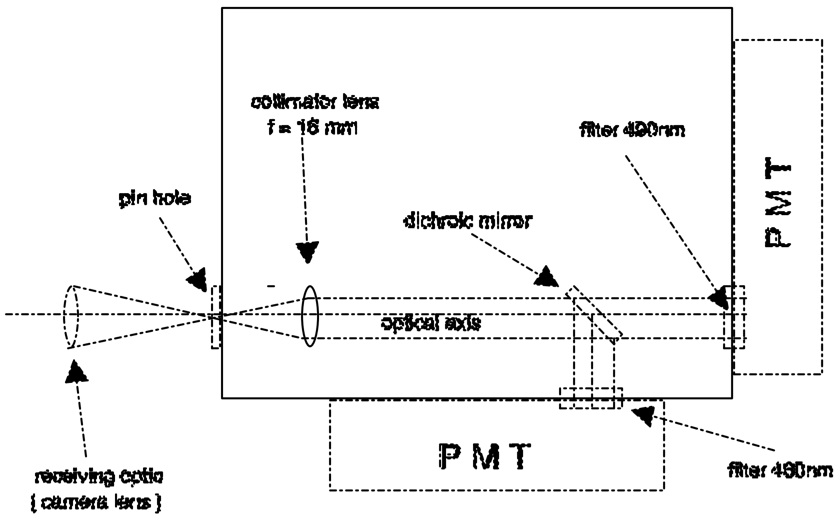

Figure 6 Receiving optics module