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COMPARATIVE STUDY BETWEEN MICROWAVE HEATING TECHNIQUE AND CONVENTIONAL HEATING TECHNIQUE IN PREPARING $Sr_4Al_{14}O_{25}:Eu^{2+}$ POWDER SAMPLES

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ABSTRACT

Comparative studies were done on the $Sr_4Al_{14}O_{25}:Eu^{2+}$ samples prepared using the conventional heating technique and microwave heating technique. A simple microwave heating system was designed for firing the phosphor samples. The system consists of metal chamber fitted with an 800W magnetron operating at 2.45 GHz. The synthesis technique prior to the microwave heating will be described. In the conventional method, the samples were sintered in a tube furnace at a temperature of 1200^{o}C for three hours in flowing N_2/H_2 (90%/10%) environment. However for the samples sintered using microwave, only a quarter of the time is needed. The X-Ray diffraction (XRD) data indicates that the sample is polycrystalline and acquires the host structure. The Photoluminescence (PL) and Photoluminescence Excitation (PLE) spectra for the powder phosphor prepared show similar results as those prepared using the conventional method. The $Sr_4Al_{14}O_{25};Eu^{2+}$ powder samples doped with Eu^{2+} show strong blue green emission peaking at 538nm. The Scanning Electron Microscope (SEM) picture taken shows that the crystal size of the microwave irradiated samples is smaller in comparison to those prepared using the conventional method. These results indicate that the microwave heating technique is a reliable, fast and suitable technique to produce these powder phosphors. The characteristics of these phosphors are as good as those prepared using the conventional heating technique.

INTRODUCTION

The luminescence of europium ion in its divalent state (Eu^{2+}) has received considerable attention because of its importance in production of blue emitting phosphors for various applications, including fluorescent lamps, cathode ray tube and field emissive display [1]. However, it is well known that europium ion in its trivalent state (Eu^{3+}) emits a characteristics red light with a number of narrow line due to the $^5D_0 - {}^7F_j$ transition [2]. Various materials doped with Eu^{3+} such as $Y_2O_3:Eu^{3+}$, $YVO_4:Eu^{3+}$, $Sr_3(PO_4)_2:Eu^{3+}$, $Y_2O_2S:Eu^{3+}$, $Gd_2O_3:Eu^{3+}$ and $Gd_2O_2S:Eu^{3+}$ have been use in display application, fluorescent light and also in the medical field such as for the x-ray intensifying screen [3,4,5].

There have been various techniques reported to synthesis the $Sr_4Al_{14}O_{25}:Eu^{2+}$ phosphor. The conventional technique was to use the solid state reaction by heating up the starting material in a furnace. There have been other techniques reported such as spray pyrolisis, sol gel method and combustion method to produce the same phosphor [6,7]. However

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there was no report using microwave heating technique. In this paper the microwave heating technique will be describe and elaborated. The sample preparation prior to the firing process will be described.

EXPERIMENTAL TECHNIQUE

The $Sr_4Al_{14}O_{25}:Eu^{2+}$ were prepared from stoichiometric amount of $SrCO_3$, $Al(OH)_3$ and Eu_2O_3. All the starting materials were weight stoichiometricly with Europium 5 atomic percent relative to the host. The powder were put inside an agate mortar and mixed thoroughly. The samples were again mix inside a ball mill to ensure the particles were crushes and the distribution of the sample is homogenous. The sample were washed using distill water and acetone and left to dry in an oven overnight. The mixed samples were then divided into two, one for firing in the furnace and the other for the microwave processing.

The first sample was placed in an alumina crucible with its end covered with quartz wool. The sample was fired in a mixture of N_2/H_2 (90:10) reduced environment. The exhaust was bubble in water to dissolve any corrosive gas that might have been emitted by the sample.

The home made microwave system is equipped with a magnetron that produces 2.45GHz frequency (800W). The temperature of the microwave furnace was monitored using a thermocouple. A gas feeding line is located besides the thermocouple. The second sample was placed in a small alumina crucible. The sample was heated in the microwave system in 10 minutes succession in a reduce environment. The microwave heating furnace is capable to reach a temperature of 1200^{o}C in less than three minutes. High temperature was needed for the reaction to be complete.

RESULTS AND DISCUSSION

The crystal structure was determined using the Siemens D500 X-Ray Diffractometer. The XRD spectra shown in Figure 1 indicate that the samples have all attained polycrystalline structure. The main peaks at 19.4^o, 31.8^o, 33.3^o and 35.8^o indicate that all the samples have a hexagonal β-alumina structure. The XRD peak is highest for the samples prepared using the conventional heating method. However, the sample prepared using microwave heating technique shows lower peaks. This might indicate that the degree of crystallinity for the microwave heated sample is smaller.

The photoluminescence (PL) and photoluminescence excitation (PLE) spectra are shown in Figure 2. The main blue green peak was located at 538nm. The emission intensity is higher for the samples prepared by firing it in furnace. This might be due to the higher degree of crystallinity of the sample. The main excitation bands are located at 230nm and 310 nm.

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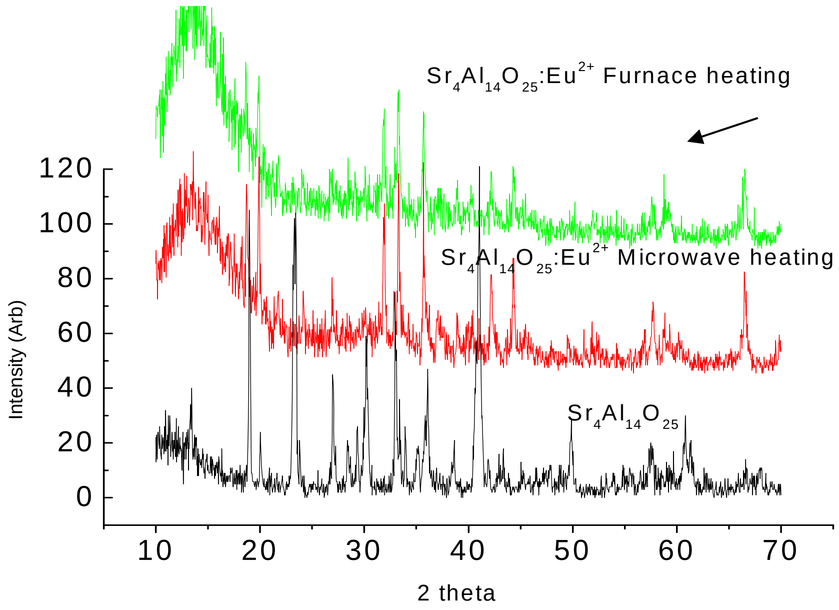

Figure 1: The crystal structure of the prepared samples

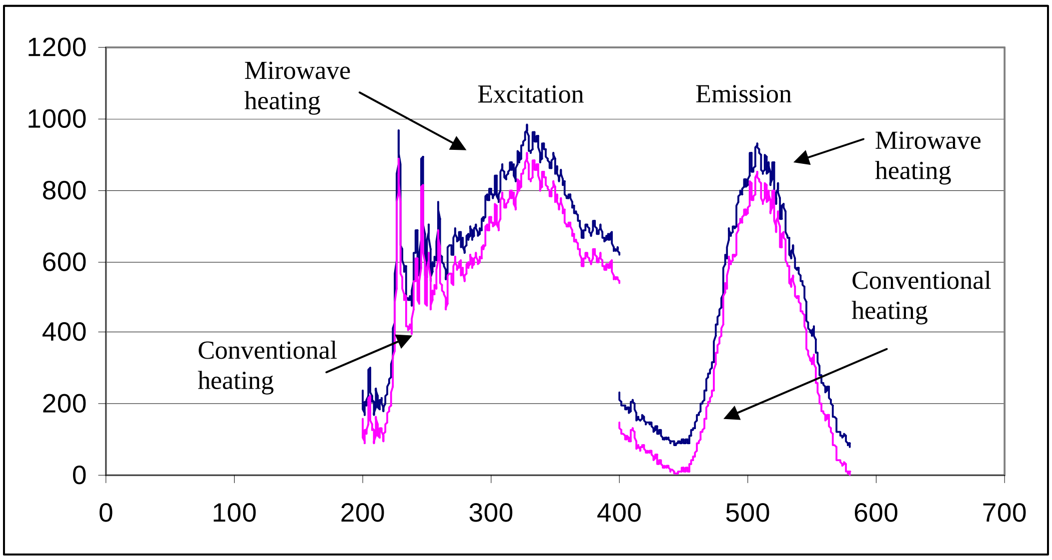

Figure 2: The PL and PLE spectra of the $Sr_4Al_{14}O_{25}:Eu^{2+}$ samples prepared using the conventional technique.

The SEM picture were taken by LEICA S5440 system. The SEM pictures clearly shows that the samples prepared using the microwave system are smaller compared to the samples prepared using the furnace. Figure 3 shows the SEM picture of $Sr_4Al_{14}O_{25}:Eu^{2+}$ samples prepared using microwave heating technique. The sample size is approximately

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4 micron. However the sample prepared using furnace heating, as shown in Figure 4 shows a bigger crystal size.

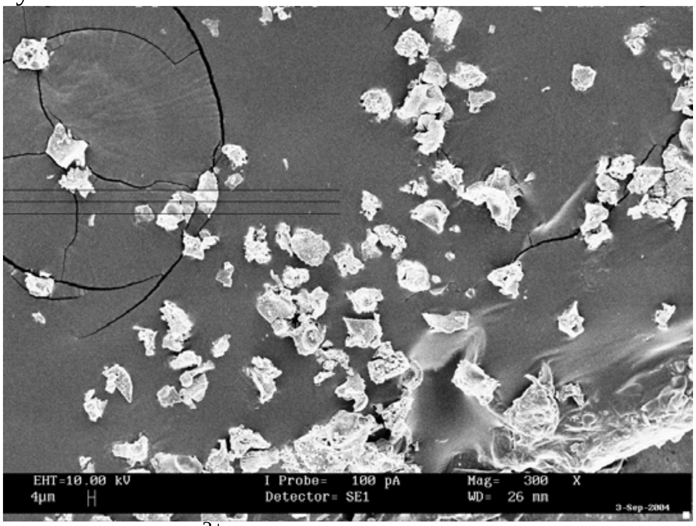

Figure 3: $Sr_4Al_{14}O_{25}:Eu^{2+}$ prepared using microwave heating technique

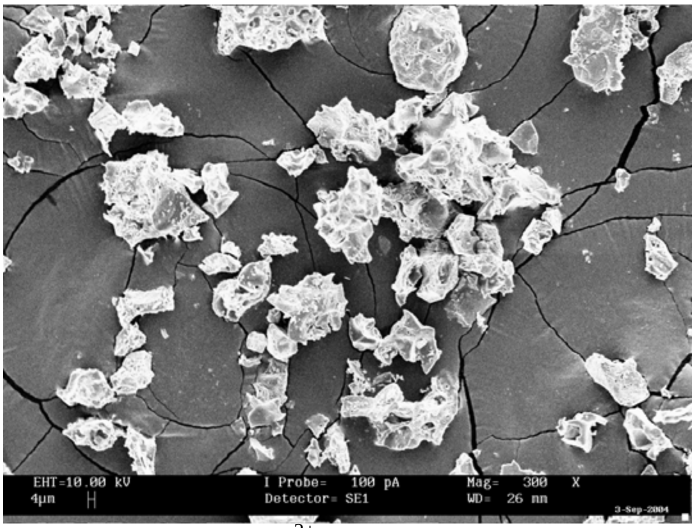

Figure 4: $Sr_4Al_{14}O_{25}:Eu^{2+}$ sample prepared using furnace

Energy Dispersive X-Ray analysis were done using the Oxford Model 5431. The results clearly show that the composition is as expexted. The summary of the EDX results is shown in Table 1 below:

Table 1: The EDX results for the BAM:Eu^{2+} samples

Heating technique	Sr%	Al%	O%	Eu%
Furnace	4.03	76.10	17.40	2.47
Microwave	7.41	77.71	13.12	1.75

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The results obtained shows that it is possible to synthesize the $Sr_4Al_{14}O_{25}:Eu^{2+}$ doped phosphor using microwave heating technique. Two-preparation technique was described, one using the furnace heating technique and the other using microwave heating technique. The XRD results clearly show that the entire sample exhibit the β-alumina structure. This structure is important because the europium ion have substituted the barium ion thus obtaining the Eu^{2+} charge state. The results also show that the sample prepared using the microwave heating technique obtain a higher degree of crystallinity in time. Only 20 minutes is needed to prepare the sample. The summary of the processing time is shown in Table 2. From the table it shows that the sol gel method requires less time compared to the conventional method.

Table 2: The summary of the processing conditions for sample $Sr_4Al_{14}O_{25}:Eu^{2+}$

Sample	Heating Technique	Duration	Condition
$Sr_4Al_{14}O_{25}:Eu^{2+}$	Furnace	3 Hours	N_2/H_2 (1200^{o}C)
$Sr_4Al_{14}O_{25}:Eu^{2+}$	Microwave	(10 min)x 6	N_2/H_2

The PL results show a strong broad blue green emission peak at 538nm. The luminescence intensity of the sample prepared using the furnace heating technique is higher compared to the sample prepared using the microwave heating technique. However the difference is small. For the PLE spectra, two broad peaks can be seen at 248nm and at 319nm. This is due to the host excitation of the $4f^65d$ band. The luminescence mechanism is shown clearly in Figure 5.

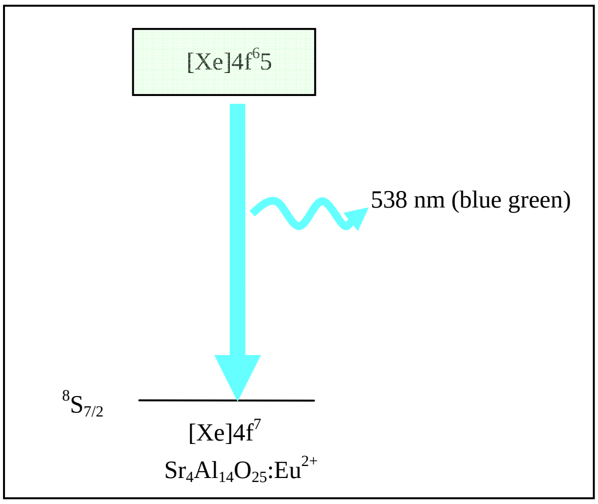

Figure 5: The luminescence mechanism of Eu^{2+} and Eu^{3+} doped phosphors.

$Sr_4Al_{14}O_{25}:Eu^{2+}$ sample shows a broad emission line because the 538nm originated for the $4f^65d$ to $4f^7$ transition of the Eu^{2+} ion. Because the 5d electron is involved in the

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excitation process, the emission spectrum is strongly influenced by the crystal field strength. The spectrum is broad because there is an overlapping between the $4f^6 5d$ and the $4f^7$ orbital. Since the distance between the bands is bigger, it will give rise to shorter wavelength, which will give rise to blue green emission.

The SEM picture shows that the sample prepared using the microwave heating method is smaller compared to the sample prepared using the furnace. The average size of the sample prepared using the furnace is around 6 micron and the sample prepared using the microwave heating is 4 micron respectively. This result is expected because the crystal tends to grow with prolonged heating time.

The EDX results also support that the $Sr_4Al_{14}O_{25}:Eu^{2+}$ phosphor that has been obtained is in agreement with the intended composition. It indicates clearly that all the composition of strontium, aluminum, oxygen and europium is in the intended value. However further study is needed to determine the optimized composition of the phosphor.

CONCLUSION

The results clearly show that microwave heating technique is another method that can be use to produce $Sr_4Al_{14}O_{25}:Eu^{2+}$ doped phosphors. The technique is not only fast but it is clean. Furthermore the heating is volumetric, where the entire sample is heated up simultaneously. The PL and PLE results show that all the samples are showing blue green emission at 538nm. The sample prepared using the microwave heating is emitting as bright as the sample prepared using the conventional heating technique. Due to the rapid heating of the sample prepared using the microwave heating technique, the crystal size obtained is smaller compared to the conventional heating technique.

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