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Synthesis of Novel $KLa(WO_4)_2:Pr^{3+}$ Phosphor and Investigation of their Luminescent Properties

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Abstract—

Highlights

- *Novel $KLa(WO_4)_2:Pr^{3+}$ phosphor was synthesized by solid state diffusion*
- *The phases and luminescent properties of $KLa(WO_4)_2:Pr^{3+}$ has been discussed.*
- *It finds potential applications as UV detectors, red emitting phosphor for colour television and scintillators.*

$KLa(WO_4)_2:Pr^{3+}$ phosphors have been synthesized by the simple solid state diffusion method at 800°C for the dopant concentrations 0.05 mole%, 0.1 mole%, 0.2 mole%, 0.5 mole% and 1 mole%. The spectroscopic and laser properties of various Pr^{3+}-doped hosts proves it as a functional material for applications such as fiber amplifiers, fiber lasers and up-conversion lasers [1-4]. X-ray powder diffraction (XRD) analysis ensured the formation of the compound in a crystalline and homogeneous form. There is no standard JCPDS file for this compound. The XRD pattern indicated that no traces of reactants were present in the prepared compound $KLa(WO_4)_2$ and it is in the homogeneous form. The morphological characterization was carried out using the Scanning Electron Microscopy (SEM). The photoluminescence excitation and emission spectra of the phosphor were investigated. Out of the three-excitation wavelengths, the most intense emission spectra were found to be at an excitation wavelength of 448 nm. Corresponding to this wavelength, the prominent emission bands were observed around 478, 488, 499, 533, 558, 605, 620 and 648 nm in the blue, yellow, green and red regions of visible range. As the excitation is mercury free, the prepared phosphor is environment friendly too. The cost considerations of the production of these phosphors would also put them on preferred position, ahead of their existing counterparts. With very prominent red emission under blue excitation, it can be used for white light production.

1. INTRODUCTION

Recently, light emitting diodes (LEDs) are rapidly replacing the conventional sources of white light containing mercury. Mercury containing fluorescent lamp seems to be harmful to the environment when exposed to air. Also the LEDs are efficient, power saving, small size and have long lifetime, so the white LEDs are regarded as next generation solid state lighting source. White light can be obtained when semiconductor chip such as InGaN is used to excite the tricolour phosphor with excitation wavelength λ=380 nm to give red, blue and green colour which combines to give a white light. $BaMgAl_{10}O_{17}:Eu^{2+}$ is commercially produced blue phosphor with low absorption efficiency in near-UV region. To avoid the difficulties with this phosphor research is going on to develop more advanced materials with potential applications in eco-friendly solid state lighting [5-12].

However, Pr^{3+} has good luminescent properties and low cost, it is the less studied rare earth ion. Emission from Pr^{3+} is the host dependent when originate from $^3P_0 \rightarrow ^3H_4$ transition it is green in colour and when originates from $^3P_0 \rightarrow ^3H_6, ^3F_2$ is red in colour. Pr^{3+} has emission range from UV to IR region and has so many applications such as fiber amplifiers, fiber lasers and up-conversion lasers. Here we are investigating luminescence from Pr^{3+} ion doped double tungstate group. Tungstate has been used since last 75 years for the X-ray photography. These are the excellent luminescence materials in which WO_4^{2-} group is itself acting as a luminescent centre. Tungstate and double tungstate is resulted in promising host for all types of rare earths activators due to their excellent optical, mechanical and thermal properties [13-15].

In this paper we have reported Pr^{3+} doped $KLa(WO_4)_2$ phosphor prepared by solid state diffusion technique. X-ray diffraction (XRD) characterisation of the phosphor was done to ensure the formation of the phosphor and to check its crystallinity and homogeneity. Phosphor was investigated by the scanning electron microscopy (SEM) characterisation in order to study its morphology. Photoluminescence spectrum of the phosphor was obtained by keeping excitation wavelength at 448 nm. Phosphor is found to be useful in various applications

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such as white light production by LED, as a red phosphor for colour television.

2. EXPERIMENTAL

$KLa(WO_4)_2$:Pr was synthesized by solid state reaction. All the precursors of analytical grade with 99.9% purity were used. Stoichiometric quantity of KNO_3, La_2O_3, H_2WO_4 and $Pr(NO_3)_3$ were taken in the agate mortar pestle and grinded thoroughly for the period of one hour. Concentration of dopant was taken 0.05 mole%, 0.1 mole%, 0.2 mole%, 0.5 mole% and 1 mole%. The mixture is taken in silica crucibles, firstly dried in oven at 200 ^{0}C for 2 hours, and then kept in muffle furnace for 24 hrs at 640^0 C, and then cooled slowly till the room temperature is achieved. Obtained material is crushed again and characterised by XRD, SEM and PL. The chemical reaction that took place in the process is represented by the following equation.

$$4\ ANO_3 + 2\ (1\text{-}x)\ La_2O_3 + 2x\ Pr(NO_3)_3.5H_2O + 8\ H_2WO_4 \rightarrow 4\ ALa_{(1\text{-}x)}(WO_4)_2{:}Pr_x^{3+} + (8+10x)\ H_2O + (4+6x)\ NO_2 + O_2$$

The phase purity and crystallinity of the prepared samples were examined by X-ray powder diffraction method using PAN-analytical diffractometer (Cu-K$\acute{a}$ radiation) at a scanning step of 0.01^o, continue time 20s, in the range of 2θ from 20^0 to 80^0. The wavelength of X-rays was 1.54 nm. The scanning electron microscope (SEM) was used to study surface morphology and particle size of the undoped and Pr^{3+} doped phosphor samples prepared in the present investigation. Pl characterisation is taken on RF-5301 PC Series Spectrofluorophotometer. Excitation and emission spectrum was obtained.

3. RESULT AND DISCUSSION

3.1 XRD Characterisation

$KLa(WO_4)_2$ was synthesized by solid state method, white powder obtained, which was characterized XRD technique. Standard JCPDS data of this compound is still not reported, we are presenting here XRD data of $KLa(WO_4)_2$ in table (1). The XRD pattern of these compounds did not indicate the presence of traces of reactants which is an indirect evidence of formation of desired compounds. The patterns also indicated that final compound is in homogeneous form. Doping of Pr^{3+} does not change the crystal structure.

Table 1: XRD data of $KLa(WO_4)_2$

Pos. [°2Th.]	*Height [cts]*	*FWHM [°2Th.]*	*d-spacing [Å]*	*Rel. Int. [%]*
27.9879	583.69	0.2448	3.18543	100.00
30.0590	79.22	0.2244	2.97050	13.57
33.4269	106.51	0.2856	2.67850	18.25
45.3299	191.19	0.2448	1.99900	32.76
47.7568	67.77	0.1632	1.90293	11.61
51.6753	100.83	0.1632	1.76746	17.28
56.0869	109.17	0.3264	1.63844	18.70
57.2154	106.03	0.2244	1.60878	18.17
72.6296	71.89	0.2652	1.30069	12.32

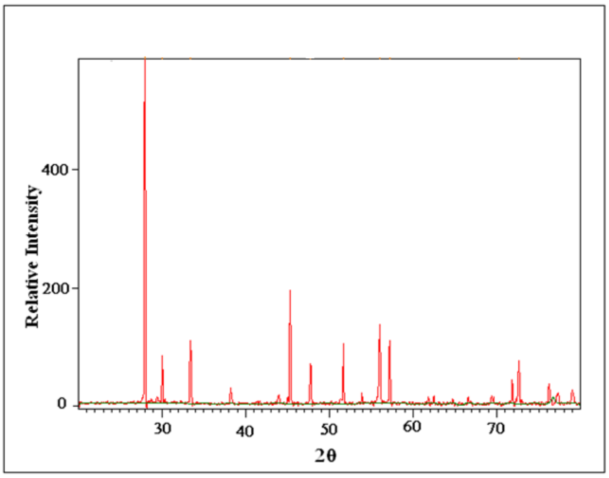

Fig.1: XRD pattern of $KLa(WO_4)_2$

3.2 SEM Study

A scanning electron microscope (SEM) is a type of electron microscope that produces images of a sample by scanning it with a focused beam of electrons. The beam of electron is attached with signal detector scans the surface of material in raster scan pattern. Specimen coated with gold or platinum, was mounted on holder. SEM image of $KLa(WO_4)_2$ is shown in fig 2.

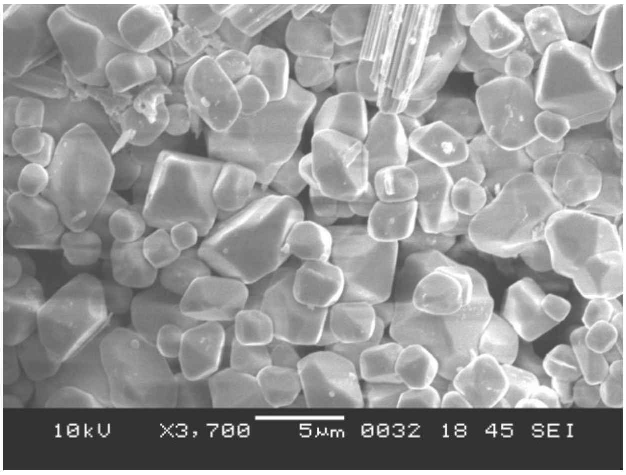

Fig. 2: SEM Image of $KLa(WO_4)_2$ showing the particles of prismatic and elongated rod like shapes.

Figure shows different sizes and shapes of particles with some definite structure. It can be seen that some of the particles are in rod shape. Doping of Pr^{3+} does not change the crystal size and shape of compound. It is observed that size of doped and undoped particle is in the range of 1micrometer to 5 micrometer.

3.3 PL Characterisation

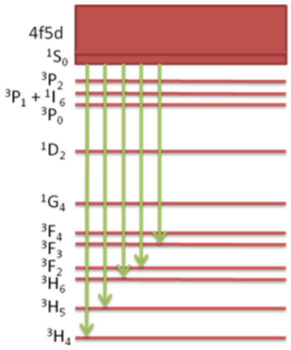

Fig. 3: Energy level diagram of Pr^{3+} rare earth ion

Excitation spectra of $KLa(WO_4)_2$ was recorded by monitoring emission wavelength at 648 nm which is its characteristics emission. Excitation spectra shows three peaks at 448 nm, 473 nm and 486 nm corresponds to transition $^3H_4 \rightarrow {}^3P_2$, $^3H_4 \rightarrow {}^3P_1+{}^1I_{6,}$ $^3H_4 \rightarrow {}^3P_0$ respectively. Spectrum shows that 486 nm is the most intense band among the other excitation band.

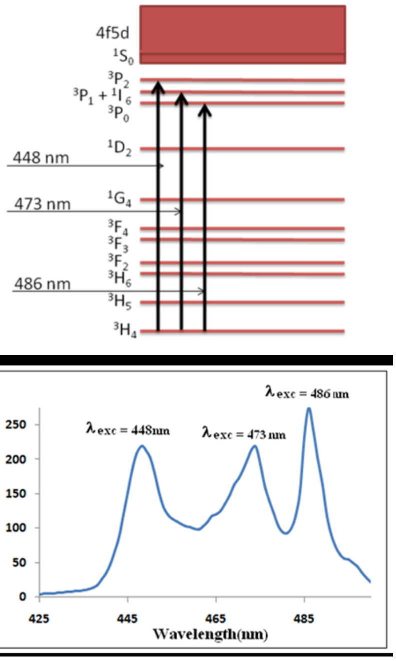

Fig. 4: Excitation spectra of $KLa(WO_4)_2:Pr^{3+}$

By keeping excitation wavelength at 448 nm emision spectra was recorded. Number of emission peaks were obtained ranging from 488 nm to 645nm which includes blue, green and red emission. At 488 nm,499 nm,530 nm,547 nm,556 nm,602 nm,618 nm,645 nm emission peaks were observed. The most intense emission peak was 488 nm which is corresponds to blue colour and second prominent emission found at 648 nm belongs to red colour. Highest intensity corresponds to 1 mol% followed by 0.2 mol %, 0.5 mol%, 0.05 mol % and 0.05 mol %.

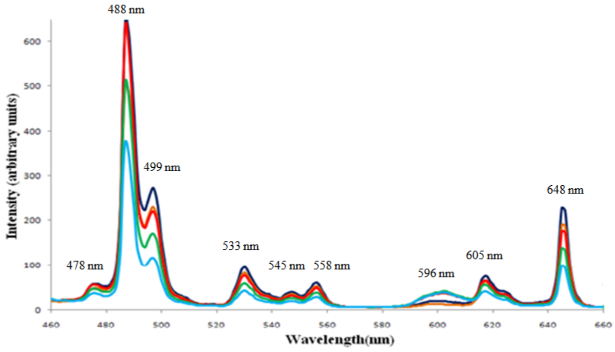

Fig. 5: The characteristic emission spectra of $KLa(WO_4)_2$: Pr^{3+}

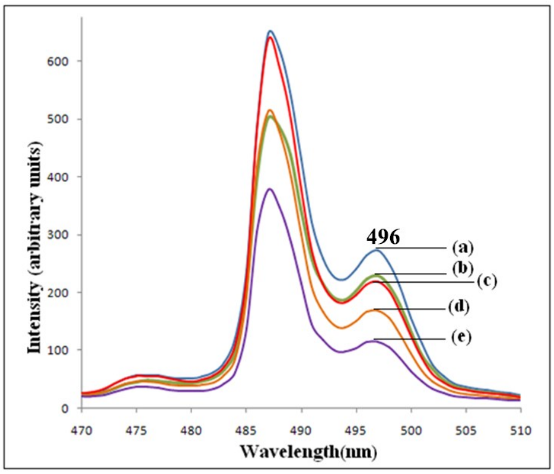

Fig. 6: The 488 nm and 496 nm emissions of Pr^{3+} ion in $KLa(WO_4)_2:Pr^{3+}$. Here the curves correspond to (a) 1 mole % (b) 0.2 mole % (c) 0.5 mole%

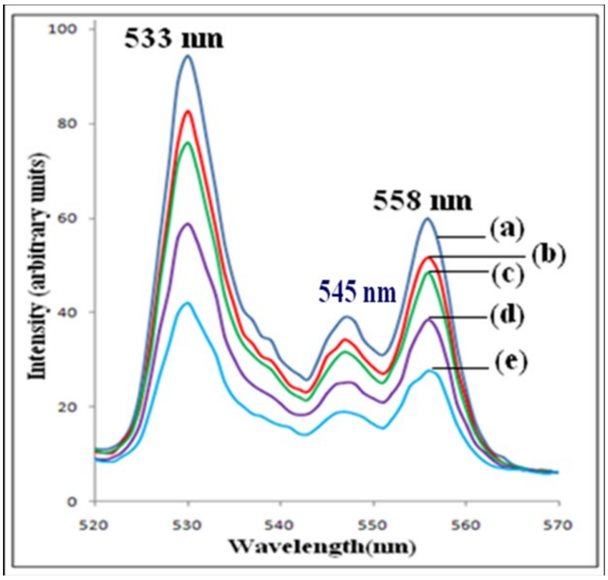

Fig.7: The 533 nm, 545 nm and 558 nm emissions of Pr^{3+} ion in $KLa(WO_4)_2:Pr^{3+}$

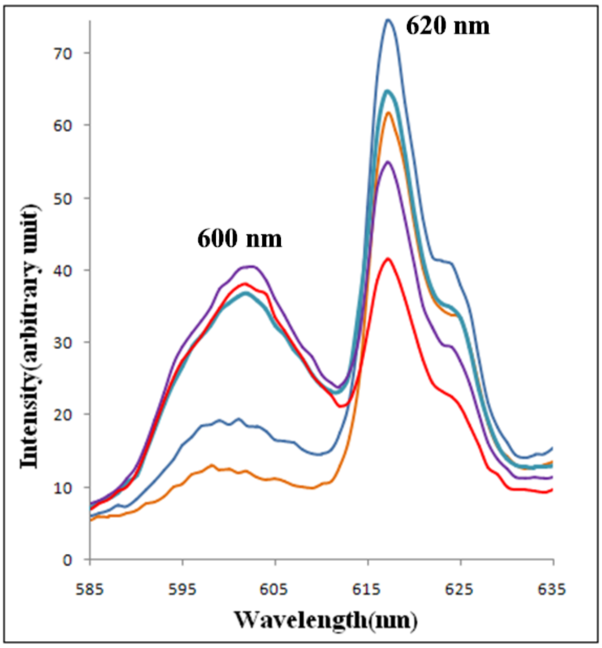

Fig. 8: The 600 nm and 620 nm emissions of Pr^{3+} ion in $KLa(WO_4)_2:Pr^{3+}$

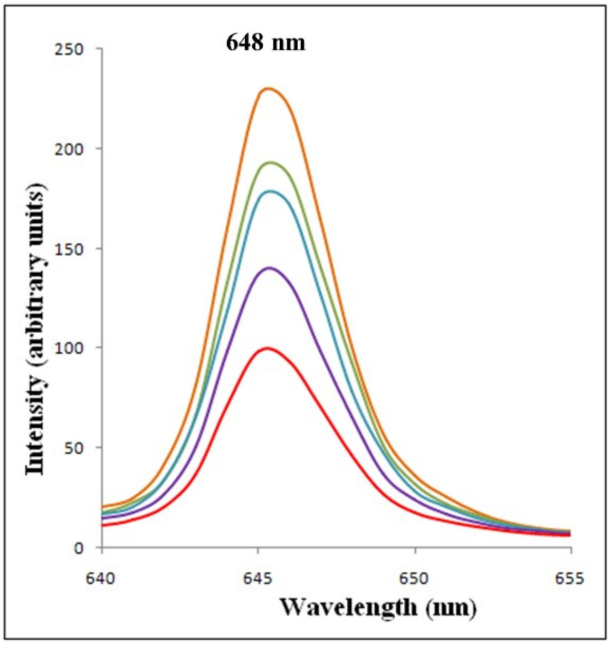

Fig. 9: The 648 nm emission of Pr^{3+} ion in $KLa(WO_4)_2:Pr^{3+}$

4. CONCLUSION

Series $KLa(WO_4)_2$ doped with various concentration of Pr^{3+} was synthesized by solid state synthesis. Obtained powder is homogeneous and crystalline form. Formation of compound and phase purity was confirmed by XRD pattern. The morphological characterisation were carried out by SEM. Photoluminescence study of this phosphor shows that excitation wavelength is situated at 448 nm which is in blue region and gives intense emission at 488 nm and 648 nm corresponds to yellow and red colour. Other emissions were observed at 499 nm,530 nm,547 nm,556 nm,602 nm,618 nm. This wavelength covered the yellow, green and red region. Hence the phosphor can be use to produce monchromatic LEDs by filtering other emitted wavelengths. It can be used to produce white light by mixing the desired wavelength. For colour telivision it can be used as red phosphor.

REFERENCES

[1] P.K. Mohan, U. Elman, M.Z. Tian, S. Kroll, Opt. Lett. 24, (1999) 37

[2] S. Kuck, I. Sokolska, M. Henke, T. Scheffler, E. Osiac, Phys. Rev. B 71, 165(2005) 112

[3] A. Richter, E. Heumann, E. Osiac, G. Huber, W. Seelert, A. Diening, Opt. Lett. 29, (2004)2638

[4] E. Osiac, E. Heumann, G. Huber, S. Kuck, E. Sani, A. Toncelli, M. Toncelli, Appl. Phys. Lett. 82(2003) 3832.

[5] J.K. Park, C.H. Kim, S.H. Park, H.D. Park, S.Y. Choi, Appl. Phys. Lett. 84 (2004)1647–1649.

[6] A. Xie, X.M. Yuan, F.X. Wang, Y. Shi, J. Li, L. Liu, Z.F. Mu, J. Alloys Compd. 501(2010) 124–129.

[7] Y.B. Chen, J. Wang, M.L. Gong, Q. Su, J. Solid State. Chem. 180 (2007) 1165–1170

[8] D.A. Steigerwald, J.C. Bhat, D. Collins, R.M. Fletcher, M.O. Holcomb, M.J. Ludowise, P.S. Martin, S.L. Rudaz, IEEE J. Sel. Top. Quant. 8 (2002) 310.

[9] J.K. Sheu, S.J. Chang, C.H. Kuo, Y.K. Su, L.W. Wu, Y.C. Lin, W.C. Lai, J.M. Tsai, G.C. Chi, R.K. Wu, IEEE Photonic Tech. Lett. 15 (2003) 18.

[10] M.S. Shur, A. Zukauskas, Proc. IEEE 93 (2005) 1691.

[11] M.R. Krames, O.B. Shchekin, R. Mueller-Mach, G.O. Mueller, L. Zhou, G. Harbers, M.G. Craford, J. Display Technol. 3 (2007) 160.

[12] M. Yamada, Y. Narukawa, H. Tamaki, Y. Murazaki, T. Mukai, IEICE Trans. Electron. E88C (2005) 1860.

[13] A. P. Vink, P. Dorenbos, C.W.E. Van-Eijk, J. Solid State Chem., 171(2003) 308

[14] S. Kück, I. Sokólska, M. Henke, M. Döring, T. Scheffler, J. Lumin., 102–103(2003) 176

[15] A. M. Srivastava, D.A. Doughty, W.W. Beers, J. Electrochem. Soc., 144(1997)L190