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Synthesis and Characterization of Eu^{2+}activated $KNa_3Al_4Si_4O_{16}$phosphor

M.M.Kolte, V.B.Pawade[1] andS.J.Dhoble*
Department of Physics, R.T.M. Nagpur University, Nagpur-440033, India
1Department of Applied-Physics, Laxminarayan Institute of Technology, Nagpur-440033, India
*Corresponding author:vijaypawade003@gmail.com

Abstract
This work presents Eu2+ doped KNa3Al4Si4O16 phosphor synthesized by combustion synthesis method. X-ray diffractionpattern (XRD) and photoluminescence spectra were used to characterize the structure and luminescence properties of the as-synthesized phosphors.From the excitation and emission curve it can be seen that thephosphorcanbeefficiently excitedbytheultravioletvisiblelight, it emits the light in the region from400nmto600nm.The broad emission band of Eu2+ ions is observed due to 5d-4f transition of rare earth ions.
Keywords: Blue Phosphor: CS: WLEDs

1. Introduction:

Research on rare-earth (RE) ions activated inorganic phosphors have been widely carried out due to their potential applications in modern lighting and display field dueto the abundant emission colors based on their 4f–4f or 5d–4f transitions due to their strong emission .[1,2]
Eu^{2+}isoftenusedas activatorbecauseoftheintensebroadexcitation andemissionbands obtainedfromtheirdipoleallowed4f–>5d electronictransitions.Indifferenthosts,the Emissionwavelengthofthe Eu^{2+} ionscanchangefrom n-UVtoredregion according to the selection of the host.[3]. Research onnext generation ofsolid-stateilluminationdevices,whiteLEDshas become a hot cake now a days because oftheirdurability,low energy consumption,longlifetimeand less pollution featuresas compared to theincandescentandfluorescentlamps [4-7]. whiteLEDs can be made by basically two ways. [8,9]. Thefirst approach is by combining blueLEDchipandyellowphosphors (suchasYAG:Ce^{3+}) [10].But thiscombination has lowcolorrendering indexduetothe absence ofredcomponent . Inorderto achieve warmwhite light ,thesecond method involved to obtain whiteLEDs is by using tri-coloredphosphors(red,greenandblue)excited by a near ultraviolet(near-UV,around350–420nm)chip. Aluminosilicates based phosphor have been paid more attention as an optical and luminescent materials due to their high luminous efficiency ,water resistant property ,excellent durability ,low cost , good physical and chemical stability.Now a days, aluminosilicates compounds have been extensively studied as host lattices for phosphors activated by Ce^{3+}, Eu^{2+} and Tb^{3+}. However,tothebestofourknowledge,very few aluminosilicates phosphors have been reported.Here Eu^{2+} activated $KNa_3Al_4Si_4O_{16}$ phosphors have been reportedand its photoluminescenceproperties has been discussed. Thus,the presented phosphor would offer good candidature for solid state lighting.

2. Synthesis procedure-

Combustion synthesis is simple,less time consuming and inexpensive method. This route yields highly pure and homogeneous single phase compound as a result of high flame
temperature generation during the combustion reaction. Thus it is fast and effective method
to synthesize phosphors by varying the dopants and starting materials. A series of $KNa_3Al_4Si_4O_{16}$ phosphors was prepared via combustion route at 550^0C.The starting materials for the preparation of $KNa_3Al_4Si_4O_{16}$ phosphors were used as KNO_3(Merck's 99.9%),$NaNO_3$(Merck's 99.9%), $Al(NO_3)_3$ $9H_2O$ (Merck's 99.9%), SiO_2(A.R.), urea NH_2CONH_2 (Merck's 99.9%), $(NH_4)_2Ce(NO_3)_6$ .In this method metal nitrates were used as oxidizers, and urea (NH_2CONH_2) was used as fuel for combustion. The weight of all the ingredient used above was calculated using the concept of propellant chemistry. All the precursors were mixed together in mortal pestle and were crushed up to the form a homogeneous solution. While crushing, small amount of dilute nitric acid was added to the mixture to maintain the homogeneity of the solution. After crushing for about 15 min, precursor solution was transferred to the silica crucible and it was then placed into a vertical muffle furnace preheated at 550 ^{0}C. During the

exothermic reaction betweenmetal nitrates and organic fuel complexes at low temperature, sufficient heat gets generated to form the highly fluffy and porous crystalline materials which were then fully ground for the characterization.

3. Results and discussion

3.1 X-ray diffraction and SEM

The phase purities and the crystalline nature of the as prepared phosphor was checked by powder X-ray diffraction (XRD).Powder diffractogram of $KNa_3Al_4Si_4O_{16}$ phosphors prepared by combustion synthesis has been shown in Fig.1. X-ray diffraction pattern was recorded using Cu-kα(1.54060 nm) radiation with Step Size 2θ (deg.) 0.0190, scan step time (s) 31.8152, and measurement temperature (1C) 25.00. It is well agreed with standard JCPDS file no.741718. Surface morphology of the combustion synthesized powder particle of $KNa_3Al_4Si_4O_{16}$phosphor is analyzed using scanning electron microscopy (SEM). SEM is a versatile tool to give structural information over a wide range of magnification, the study of texture,topography and surface features of the prepared phosphors.Fig. 3 shows the SEM images of $KNa_3Al_4Si_4O_{16}$phosphor.From Fig.3, it is observed that $KNa_3Al_4Si_4O_{16}$phosphor particlesarenon agglomeratedwith irregular morpholgy with partical size under 1-3μm.Thus from the result it is inferred that by using combustionmethod there is an increaseinprocess flexibility andit has acceleratedthereactionbythepresenceofureaas a combustiblefuel.

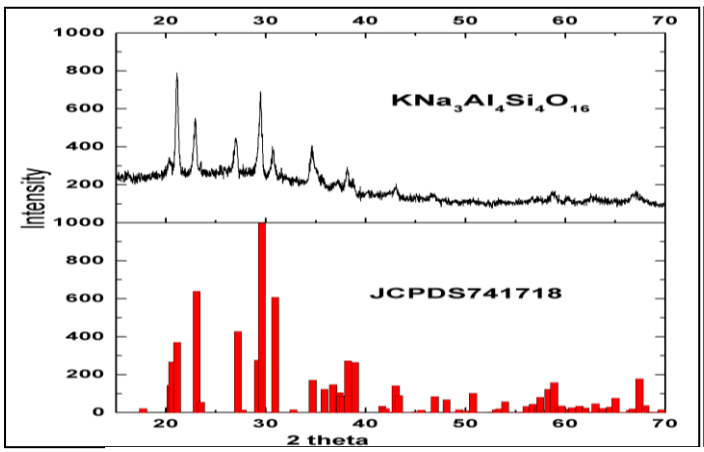

Fig.1 XRD of $KNa_3Al_4Si_4O_{16}$ phosphors

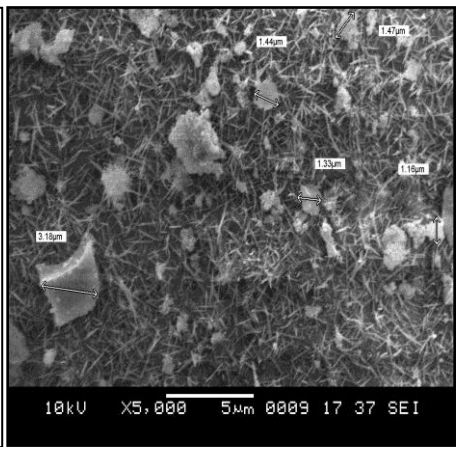

Fig.2SEM of $KNa_3Al_4Si_4O_{16}$

3.2 PL Characterization

Eu^{2+} luminescence spectroscopy

The excitation and emission spectra of$KNa_3Al_4Si_4O_{16}$:Eu^{2+}phosphor is shown in Fig.3.

From the excitation and emission spectra it can be seen that, under excitation of 357 nm, the emission spectrum is a broad band spreading over 400nm to 600nm due to the $4f^7 \rightarrow 4f^65d^1(^8S_{7/2})$ transition of Eu^{2+} ions as shown in the Fig.4

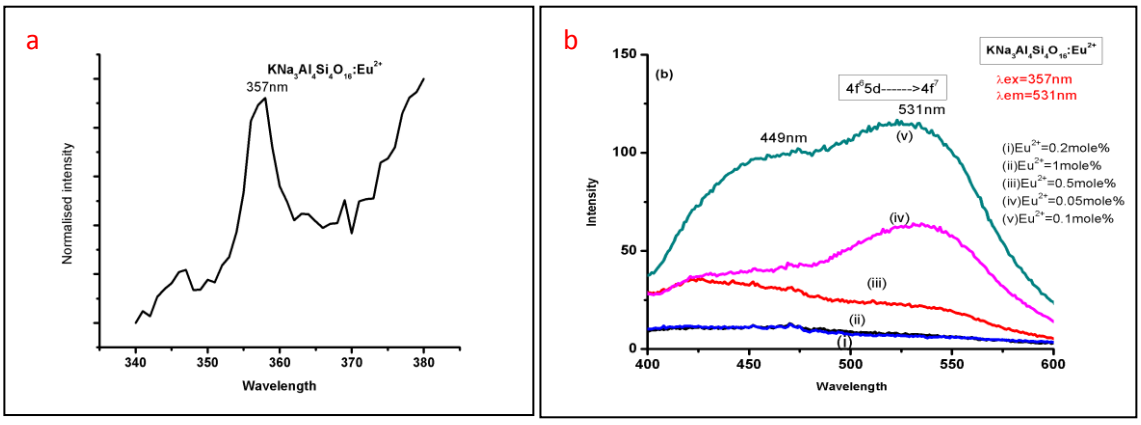

Fig.3(a)Excitation curve (b)Emission curve of $KNa_3Al_4Si_4O_{16}$:Eu^{2+}phosphor

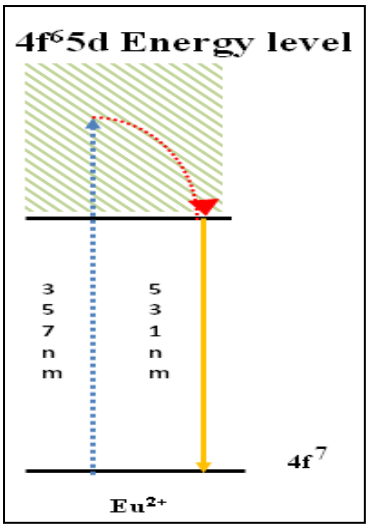

Fig.4Energy level diagram of Eu^{2+}

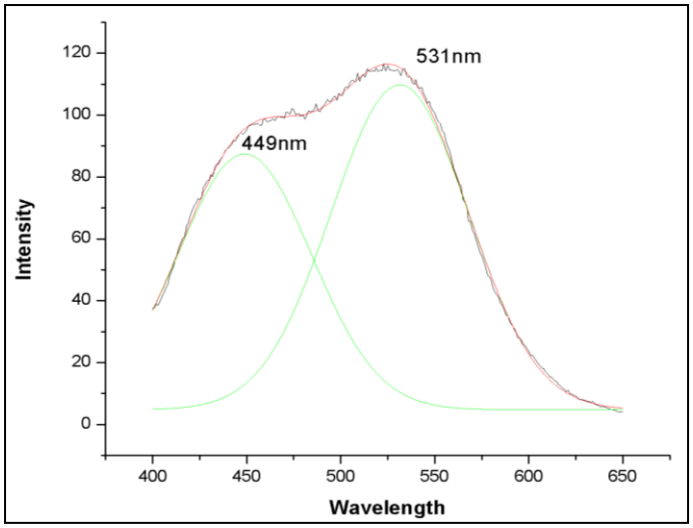

Fig.5 Gaussian fit Emission spectra of $KNa_3Al_4Si_4O_{16}$:0.1Eu^{2+}phosphor

The $4f^7 \rightarrow 4f^6$ 5d1 transition of Eu^{2+} strongly depends on the host lattice because the outermost 5d orbit is very sensitive to the crystal-field surrounding. Eu^{2+} emission can vary from UV to red region depending on the host lattice, the size of the cation, and the strength of the crystal field [11]. From the emission curve it can be found that the broad band shows two peaks ,one centred at 449nm in the blue region and the other highest intensity peak at 531nm in the green region .Gaussian fit of the emission spectra in to two peaks for 0.1mole %concentration of Eu^{2+} has been shown in Fig.5.With 351 nm UV light excitation, the Eu^{2+} ions are excited to the higher $4f^6$5d energy level .After a non-radiative process, the excited Eu^{2+} ions fall back to the lower energy region of $4f^6$5d state and then emit green light while falling back to the $4f^7$ ground state[12]. Also from emission spectra, it can be seen that the broad band emission is slightly asymmetricdue to the superposition of two emission bands of the same initial state (the lowest $5d^1$ crystal-field level) but different final states, i.e., $^2F_{7/2}$ and $^2F_{5/2}$ multiple of $4f^1$,as a result of spin-orbit interaction. Theconcentration of Eu^{2+}ions is increased from 0.05 mol% to 1 mol %, thenemission intensity maximum found at0. 1 mol % and further increase in concentration ofdopent it decreases, which may be assigned due to the non radiative energy transferbetween Eu^{2+} ions at different sites of the host . The role Eu^{2+}concentration insearching of optimal composition of phosphor is very crucial and important.Therefore, the variation of PL intensity with differentEu^{2+}concentrations for$KNa_3Al_4Si_4O_{16}$:Eu^{2+} phosphor has been graphically shown in Fig.6. The positions of the emission peak are not influenced bythe Eu^{2+} concentration. The luminescence intensity increases with Eu^{2+}doping increasing until a maximum intensity at x = 0.1 isattained, and then it decreases because of conventional concentrationquenching process.

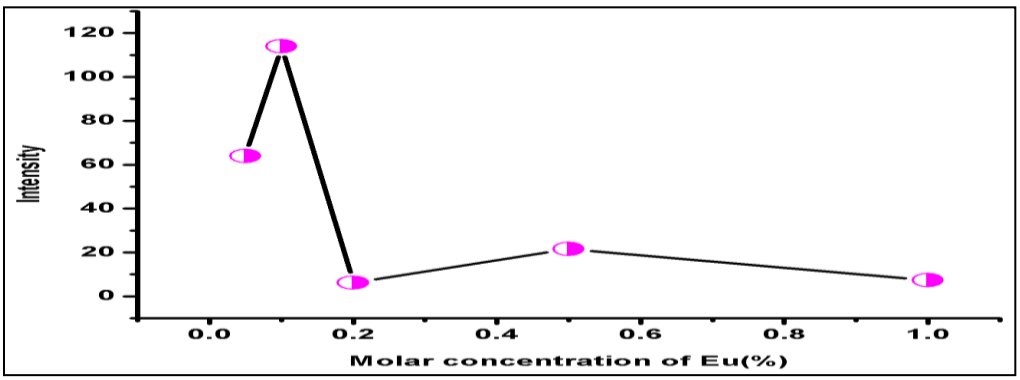

Fig.6 Concentration quenching of $KNa_3Al_4Si_4O_{16}$: Eu^{2+}

4.Conclusion:

Thus, a novel $KNa_3Al_4Si_4O_{16}$: Eu^{2+} phosphor was successfully synthesized by combustion method for the first time. Phosphor shows efficient green emission under nearer to UV excitation wavelength.Synthesized phosphor is characterized by XRD, SEM,analysis. Good crystalline natureof the phosphor is confirmed by XRD pattern. Thus, the observed broad band emission in $KNa_3Al_4Si_4O_{16}$: Eu^{2+} phosphor shows that it may be used for display and solid state lighting applications.

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