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Sol-Gel Synthesis and Photo-Luminiscence Study of $NaSr_{1-x}PO_4$:xDy^{3+} Phosphor for Use in PC-WLED

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***Abstract:** The series of Dy^{3+} doped $NaSrPO_4$ white light emitting phosphors were prepared by sol-gel method. In the XRD analysis structure characterization is done and it is found that the $NaSrPO_4$: Dy^{3+} sample is very well matched with the standard ICDD file no. (00-033-1282). Photoluminescence spectra having two main peaks were obtained at 483 nm and 575 nm, which corresponds to the blue and yellow color emission respectively. When these two peaks are combined then white light is formed with the chromaticity coordinates (0.298, 0.333).*
***Keywords:** Sol-gel, Phosphate phosphors, White light LED, Photoluminescence, Chromacity co-ordinates*

I. INTRODUCTION

Phosphor-converted LEDs are the most common LED which are based on white light sources. In the near future White Light-emitting diode (W-LED) lamps are expected to take the place of conventional incandescent and fluorescent lamps for general lighting applications in solid state lighting. Phosphor-converted white LEDs are abbreviated as pc W-LED such a light source has been mostly applied to general lighting. In general, the materials required for pc W-LEDs are oxides, nitrides, oxy-nitrides, sulphides and silicates. High luminous efficacy is the property of pc W-LEDs and due to this property light sources in most of the lighting systems are replace. For the purpose of global energy saving the topic of high luminous efficacy is essential. By using a semiconductor substrate such as sapphire makes a pc W-LED to perform luminous efficacy as high as 150 lm/W operated at 1 watt [1,2]. In recent years, there is a developing a interest in generation of white light sources for a various applications such as solid-state lighting, multicolor display technologies, back light, and so on. pc W-LEDs have some conventional lighting properties so that these properties may differ pc W-LEDs from the general lighting systems. Advantages of the pc W-LEDs are high energy efficiency, fast response, and low cost. The major challenges in pc W-LEDs have been to achieve high luminous efficacy, high chromatic stability, good color-rending properties, and less market value against fluorescent lamps [3]. We have made a meticulous survey to study the various light emitting phosphors for solid state lighting. Some of the phosphor material that are prepared by various researchers using different methods are described in tabular format in table (I).

TABLE I Phosphate phosphors for wled

Sr.no	Material name	Excitation (nm)	Emission (nm)	Method of synthesis	CIE	Ref
1	$KSrY(PO_4)_2$:Eu^{2+}	250 and 450	520	SSD	-	[4]
2	$KSrPO_4$:Dy^{3+}	351 and 388	570	SSD	(0.302, 0.360)	[5]
3	$LiSrPO_4$:Dy^{3+}	350	483 and 574	SSD	-	[6]
4	$NaSrPO_4$: Dy^{3+}	351	488 and 575	SSD	(0.30, 0.34)	[7]
5	$Sr_8MgLu(PO_4)_7$: Eu^{2+}	390	594	SSD	(0.336, 0.353)	[8]
6	$NaCaPO_4$:Dy^{3+}	382	482 and 575	SSD	-	[9]
7	$NaCaPO_4$:Eu^{2+}	400	505	SSD	-	[10]
8	$KMgPO_4$:Eu^{2+}	396 nm	470	SSD	-	[11]
9	$NaCaPO_4$:Dy^{3+}	386 nm	480 and 573	SSD	-	[12]
10	$LiSrBaPO_4$: Eu^{3+}	401 nm	595	SSD	-	[13]
11	$SrMg2(PO_4)_2$:Eu^{2+}	375nm	416	SSD	-	[14]
12	$Ca_9Lu(PO_4)_7$:Eu^{2+},Mn^{2+}	250 and 430 nm	480 and 645	SSD	-	[15]
13	$SrZn_2(PO_4)_2$: Tb^{3+}	369 nm	544	SSD	-	[16]
14	$Ca_3Mg_3(PO_4)_4$: Mn^{2+}	410 nm	610	SSD	-	[17]
15	$BaSrMg(PO_4)_2$: Eu^{2+}	350 nm	447 and 556	SSD	(0.291, 0.349)	[18]

*Solid state reaction (SSD)

White light-emitting diodes (WLEDs) as new solid-state light sources have a greatly promising application in the field of lighting and display. But the major challenges in WLEDs are to achieve high luminous efficacy, high chromatic stability, brilliant color-rendering properties, so the much more efforts have been made to develop single-phase white-light-emitting phosphors. The white light can be emitted by various single rare earth ions such as Eu^{3+}, Dy^{3+}. The rare earth elements are widely used as the luminescent centres or activators in various host materials in the recent years for the development of efficient phosphor materials. The trivalent Dy^{3+} can be used as an activator or luminescent canter, the emission of Dy^{3+} mainly shows two transitions, $^4F_{9/2} \rightarrow {}^6H_{15/2}$ (~470nm) and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ (~570nm). We have investigated the different phosphor materials based on borate, oxides, sulphate and phosphate. The phosphate based material is chosen for the study such as $NaSrPO_4$ is the good host material because the phosphate based materials are stable than the other host. Phosphates have strong emission of all the colors of wavelength. $NaSrPO_4$ is the orthophosphate and it has various advantages such as low cost, acceptable thermal stability, and potential applications in solid state lighting [7].

II. EXPERIMENTAL

The $NaSr_{1-x}PO_4$:Dy^{3+} phosphors were synthesized by using the sol-gel method. The starting reagents $Na(NO_3)_2$, $Sr(NO_3)_2$, NH_4-H_2PO_4 (AR), and $Dy(NO_3)_3$ and stearic acid were taken in a stiochiometric molar ratio and put into a crucible together. Then the raw materials were thoroughly grinded so that a homogeneous mixture was obtained. The mixture was heated at 500^0 C for 2 h in furnace and sintered at 900^0 C for 3 h to obtain the phosphors samples [5]. Complete synthesis process is given in flowchart figure(1).

The crystal structure of the as synthesized phosphors was characterized by powder X-ray diffractometer (XRD, RigakuD/Max-3B). The luminescent properties including photoluminescence excitation (PLE) and emission (PL) spectra were measured by a spectrometer (Hitachi F7000) with an excitation source 150 W Xenon lamp at room temperature.

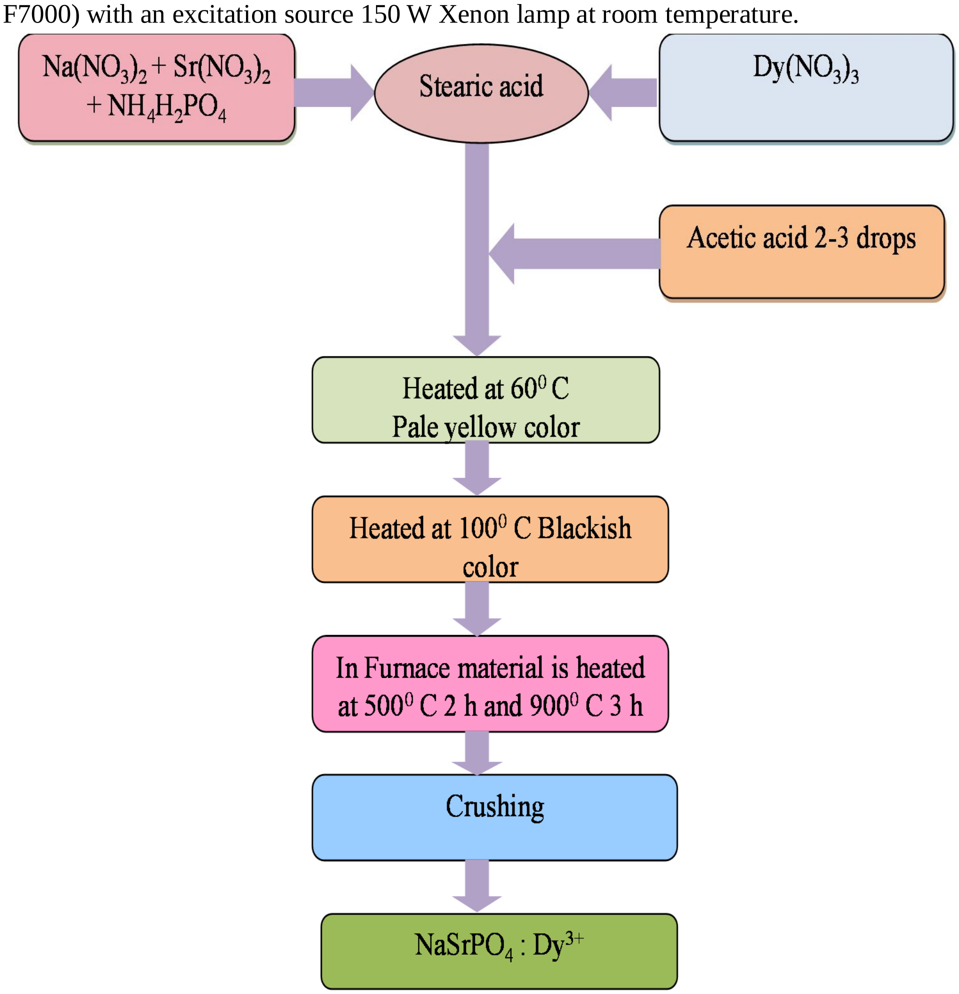

Figure 1 Flow-Chart of Sol-Gel method

III. RESULTS AND DISCUSSION XRD ANALYSIS

Figure(2) shows the XRD pattern of $NaSr_{1-x}PO_4$:xDy^{3+} sample for 0.05 mole of dopant. It can be observed that the XRD pattern of prepared sample is in good agreement with the available standard ICDD file no. (00-033-1282). X-ray diffraction pattern shows that the crystal structure was monoclinic and lattice parameters a= 20.4140 Å, b= 5.4290 Å and c= 17.2460 Å and α = 90, β = 101.760, γ = 90.

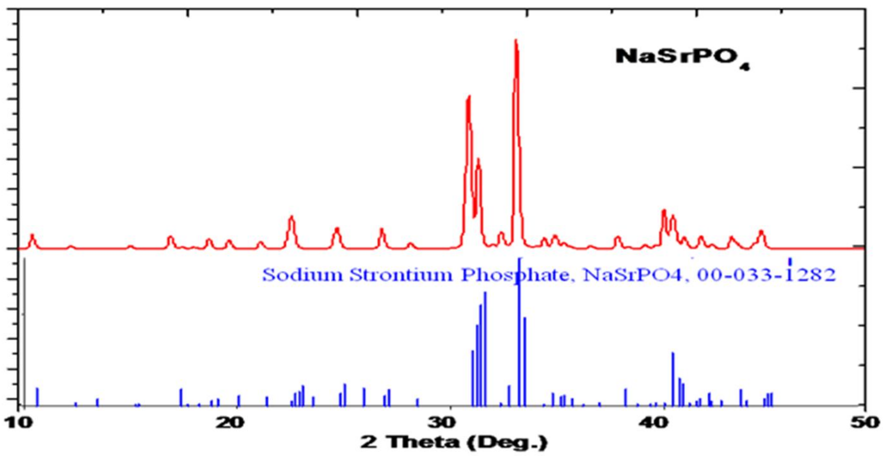

Figure 2 XRD pattern of NaSr1-xPO4 : xDy3+

A. Photoluminescence

The PL and PLE were measured for the phosphors $NaSr_{1-x}PO_4$: $0.05Dy^{3}$. The figure (3) show the PLE spectra of $NaSr_{0.95}PO_4$:$0.05Dy^{3+}$. The above phosphors exhibits the relatively strong and narrow absorption peaks within the wavelength range from 260 to 460 nm, when monitoring the emission at 483 nm. The absorption peaks obtained at 297 nm, 325 nm, 350 nm, 364 nm, 388 nm and 425 nm respectively are corresponding to transitions $^6H_{15/2}$ - $^4K_{13/2}$, $^6H_{15/2}$ - $^4K_{15/2}$, $^6H_{15/2}$ - $^4M_{15/2}$, $^6H_{15/2}$ - $^4P_{3/2}$, $^6H_{15/2}$ - $^4M_{21/2}$, $^6H_{15/2}$ - $^4G_{11/2}$. According to the spectral data the most intense absorption peak occurred at 350 nm. The excitation behaviour of Dy^{3+} doped $NaSrPO_4$ materials appreciably matches the excitation from the near-UV LED chips and converts the absorbed energy into visible light emission [6].

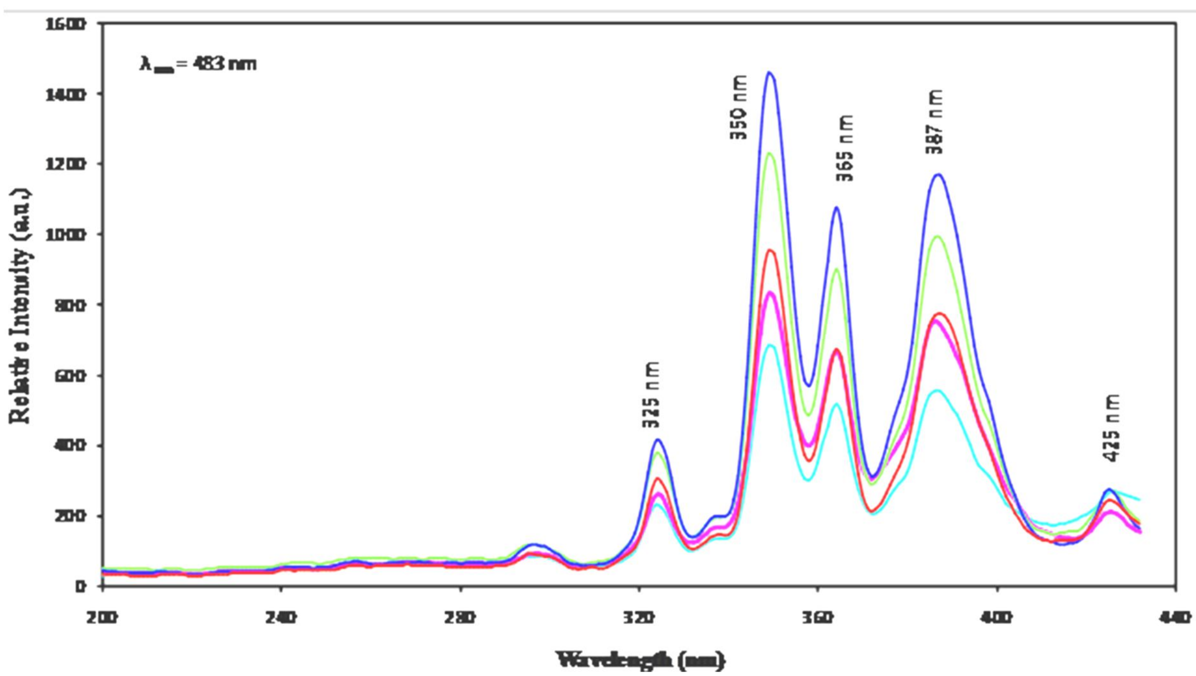

Figure 3 PLE spectra of NaSr0.95PO4:0.05Dy3+ monitored at 483nm

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The Figure (4) shows the PL emission spectrum of $NaSr_{0.95}PO_4$:$0.05Dy^{3+}$ under 350 nm excitation. Two intense emission peaks (blue and yellow) are observed, centering at 483 nm (blue) and 575 nm (yellow), related to the transitions for blue $^4F_{9/2}$ - $^6H_{15/2}$ and for yellow $^4F_{9/2}$ - $^6H_{13/2}$. Figure(4) also represents the PL spectra of $NaSr_{1-x}PO_4$: xDy^{3+} phosphors with different contents of Dy^{3+} dopant (x= 0.005,0.01,0.03,0.05). When the concentration (x) of Dy $^{3+}$ increased from 0.005 up to 0.05 mole then emission reached the maximum intensities for the concentration 0.03 mole of Dy^{3+} ions. It is well known that the combination of appropriate proportion of blue (483nm) and yellow (575nm) emissions generates white light with high color rendering index especially used in applications of the solid state lighting such as WLEDs.

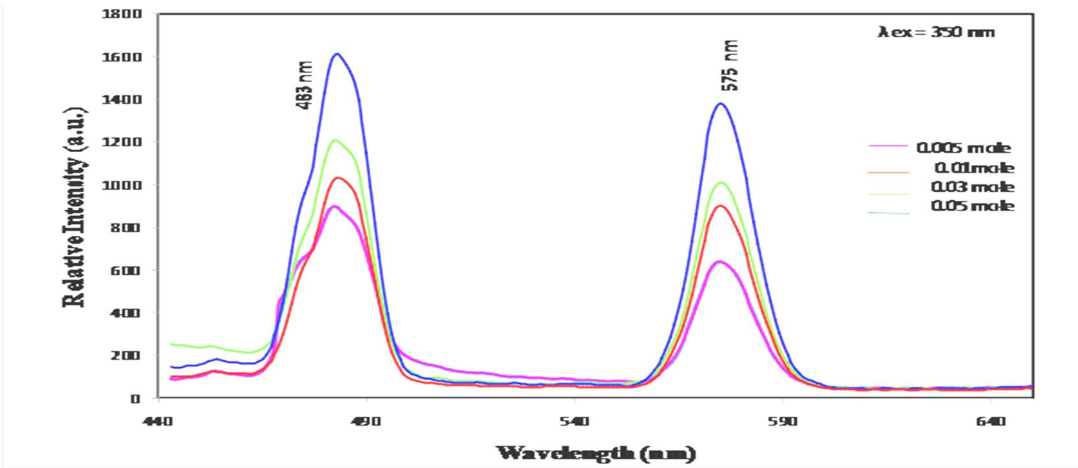

Figure 4 PL spectra of NaSr1-xPO4: xDy3+ phosphors

For evaluating the white light emission of $NaSrPO_4$:Dy^{3+} phosphor the CIE chromaticity coordinates are traced and represented in a Figure(5). These coordinates are close to white light cordinates [21]. In table (II) the chromaticity coordinates and B/Y intensity ratio of $NaSr_{1-x}PO_4$: xDy^{3+} phosphor excited at 350 nm is given.

TABLE II

Chromaticity coordinates and B/Y ratio of $NaSr_{1-x}PO_4$: xDy^{3+} phosphor excited at 350 nm.

Sr. no.	Sample composition	Chromaticity coordinates	Intensity ratio (B/Y)
1	0.005	(x=0.286, y=0.355)	1.19
2	0.01	(x=0.312, y=0.355)	1.16
3	0.03	(x=0.314, y=0.359)	1.14
4	0.05	(x=0.298, y=0.333)	1.35

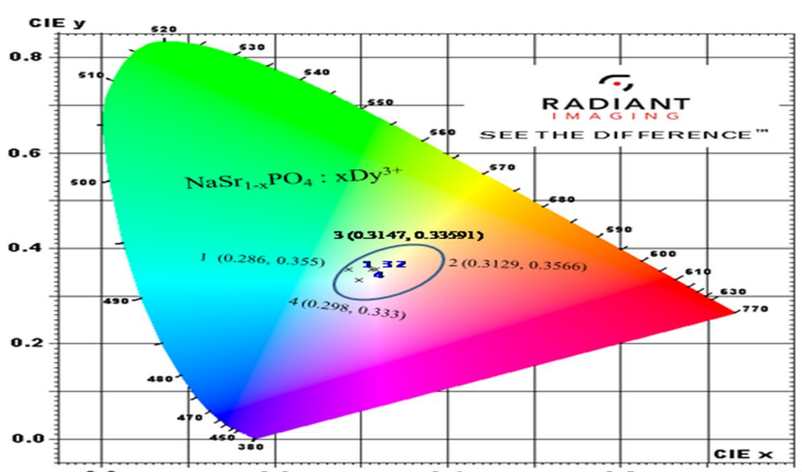

Figure 5 CIE chromaticity coordinates of NaSr1-xPO4:xDy3+phosphor samples prepared with various concentrations of 1) 0.005, 2) 0.01, 3) 0.03 and 4) 0.05 mole

IV. CONCLUSIONS

$NaSr_{1-x}PO_4 : xDy^{3+}$ x= 0.005, 0.01, 0.03, 0.05 phosphor, were prepared by sol gel synthesis method. This phosphor gave the strong emission at 483 nm and 575 nm monitored under 350 nm excitation. The critical concentration for $NaSrPO_4:Dy^{3+}$ was determined and it is 0.05 mole. XRD analysis is very well matched with the available standard ICDD file no. (00-033-1282). The intensity ratio (B/Y) of blue emission to yellow emission is 1.35, which is highest at 0.03 mole concentration with the CIE chromaticity coordinates (0.298, 0.333), which is closer to the white light coordinates (0.33, 0.33).

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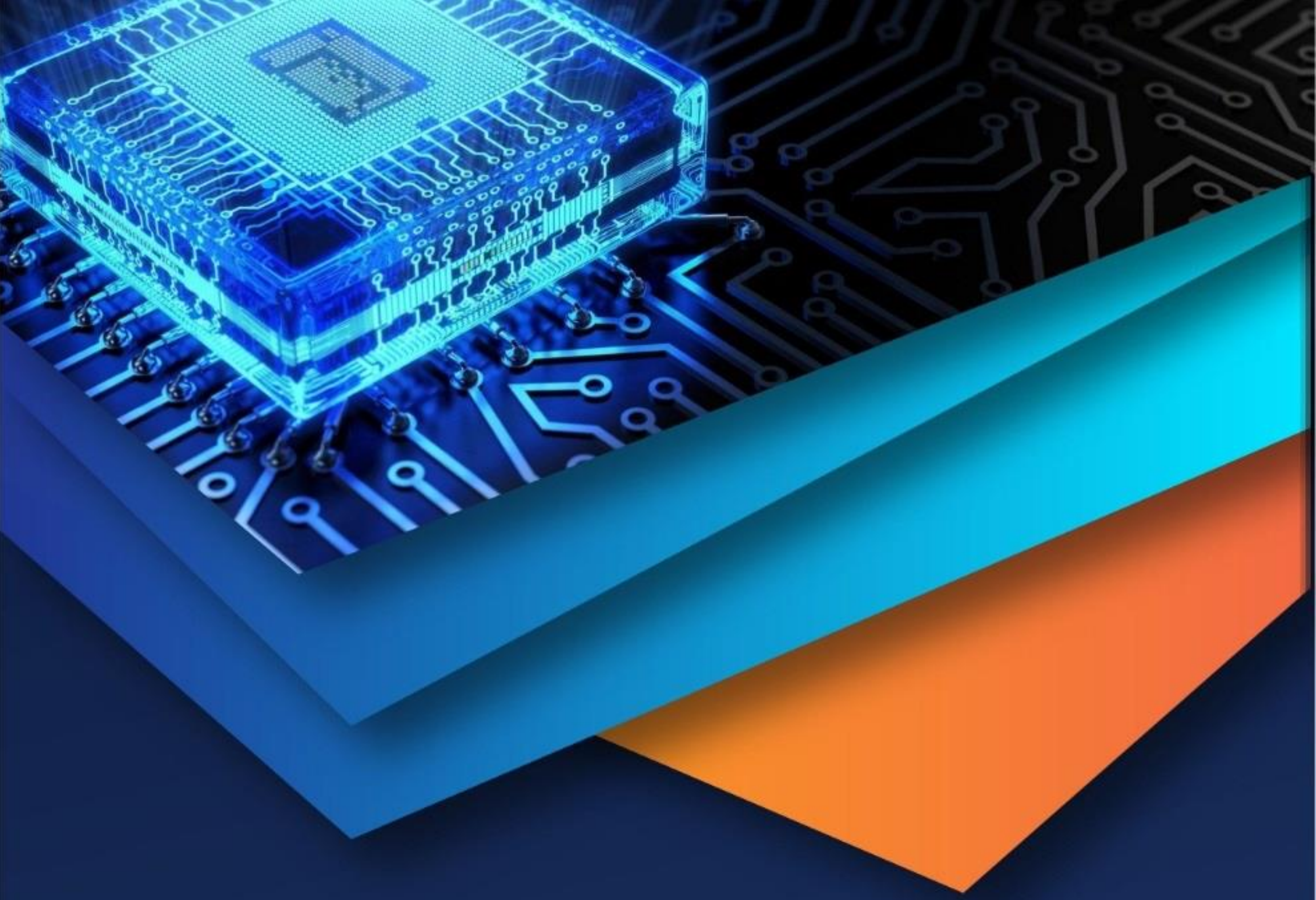

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