

Spectroscopic Study of Al Doped Zinc Oxide (AZNO) Thin Films for Optoelectronic Devices

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Citation: Prasad, S. & Agrawal, V. (2026). Spectroscopic Study of Al Doped Zinc Oxide (AZNO) Thin Films for Optoelectronic Devices. International Journal of Global Research Innovations & Technology, 04(02(II)), 10–18.

[https://doi.org/10.62823/IJGRIT/4.2\(II\).9084](https://doi.org/10.62823/IJGRIT/4.2(II).9084)

ABSTRACT

Recently, large band gap semiconductor (ZnO) is among the largest studied material because of a wide range of applications in photodetectors, light emitting diode, blue and ultraviolet laser diodes, transparent conductive oxides (TCOs), transparent field effect transistors, gas sensors, solar cell and dilute magnetic semiconductors (DMSs). This paper reports 1 at% Al doped ZnO (AZNO) thin films prepared by sol-gel spin coating method at 1000rpm to 6000rpm spin coating speeds. The sol was prepared from zinc acetate dehydrate, 2-methoxyethanol and mono-ethanolamine as precursor, solvent and stabilizer respectively. Aluminium chloride hexahydrate was used for doping of Al. The effect of spin coating speed on absorbance, transmission, Raman shift and fluorescence spectra of Al-doped ZnO thin films is studied in this paper. The absorbance of thin films decreases corresponding to increase in spinning speed. The transmittance of all thin films in the range 400nm to 700nm is greater than 90%. Raman and fluorescence spectra of thin films show that the intensity of both spectra decreases with increase in the spin coating speed.

Keywords: Al Doped ZnO Thin Films, Sol-Gel, Spin Coating, Raman Spectroscopy, Fluorescence Spectroscopy.

Introduction

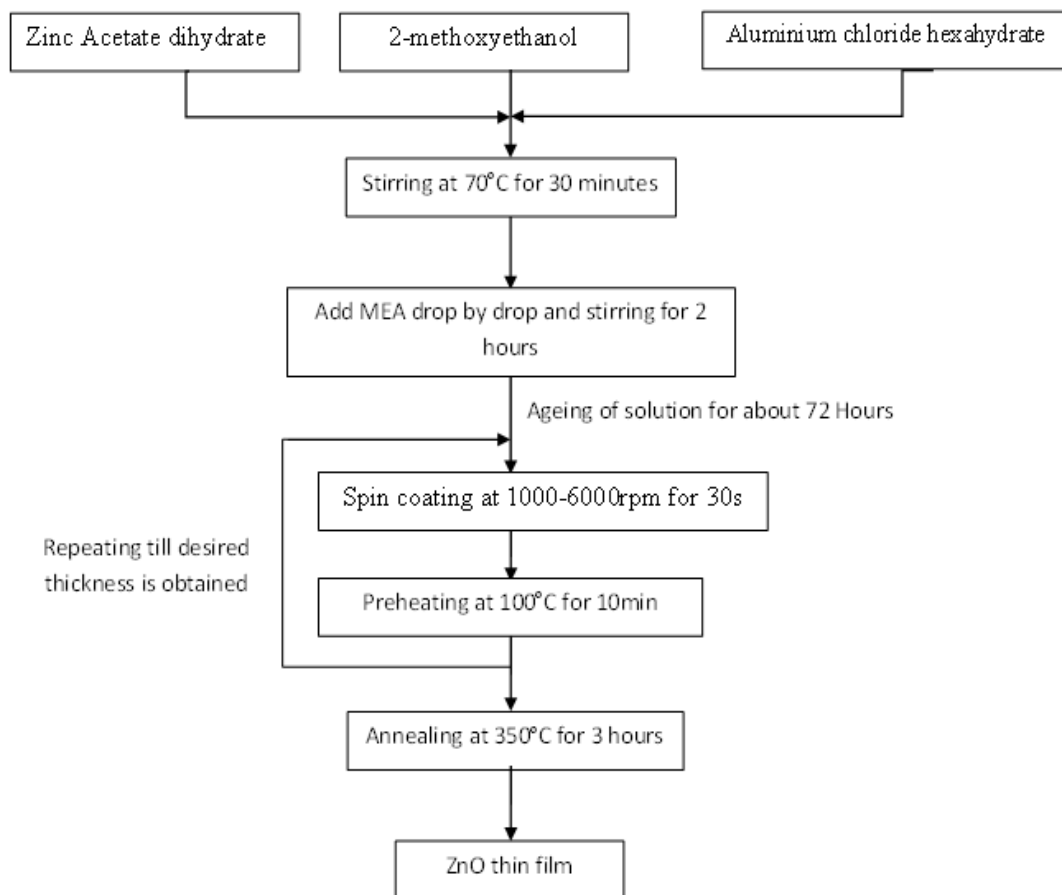
Transparent conducting oxide (TCO) films are prerequisites for most photovoltaic applications and flat panel displays (FPDs) which have been subjected to a large number of investigations for decades [1]. Practically, indium-tin-oxide films (ITOs) are the most widely used TCO materials industrially. Nevertheless, the increasing price and limited supply of indium has promoted the developing of other TCO alternatives. One of the promising substitutes are the Al and Ga doped ZnO (abbreviated as AZO and GZO, respectively) [2]. Zinc oxide is a direct, wide band-gap ($E_g \approx 3.37$ eV) semiconductor which has many applications in optoelectronics, electronics, spintronics and sensor materials [3, 4]. Group III-doped ZnO film is transparent and electrical conductive with extraordinary properties such as non-toxic, less expensive, resistant to defects and hydrogen plasma [5]. Moreover, AZO and GZO could be deposited at lower temperature by magnetron sputtering [6, 7]. It has been demonstrated that by DC magnetron sputtering, the Al-doped polycrystalline ZnO films with very low resistivity of $2.7 \times 10^{-4} \Omega \cdot \text{cm}$ could be realized at $T_s = 250^\circ\text{C}$ [8,9,10]. For a pilot scale deposition, Szyzka [7] demonstrated an AZO film with resistivity of $3.0 \times 10^{-4} \Omega \cdot \text{cm}$ by mid-frequency reactive magnetron sputtering over the borosilicate glass substrates at 200°C . Most ZnO has n-type character, even in the absence of intentional doping. Native defects such as oxygen vacancies or zinc interstitials are often assumed to be the origin of this n-type character. Controllable n-type doping is easily achieved by substituting Zn with group-III elements like Al, Ga, In or by substituting oxygen with group-VII elements like chlorine or iodine. Reliable p-type doping of ZnO remains difficult. This problem originates from low solubility of p-type dopants and their compensation by abundant n-type impurities, and it is pertinent not only to ZnO, but also to similar

compounds GaN and ZnSe. Current absence of p-type ZnO does limit its electronic and optoelectronic applications which usually require junctions of n-type and p-type material.

In present work, Al doped ZnO thin films were deposited on glass substrate via acetate dihydrate, aluminium chloride hexahydrate inorganic precursors by sol gel spin coating method. The UV-visible (UV- Vis) spectroscopy, Raman spectroscopy and fluorescence spectroscopy were utilized to describe the glass substrate deposited ZnO thin film. The optical, vibration and fluorescence properties of the thin film are critically observed.

Experimental

The 0.5M Al doped ZnO solution were prepared by dissolving zinc acetate dihydrate, aluminium chloride hexahydrate in 2-methoxy ethanol by using magnetic stirrer at 70 °C for 30 minutes. The solution becomes turbid after 30 minutes of constant stirring then monoethanolamine (MEA) was added drop by drop to stabilize the solution until solution becomes transparent by adjusting molar ratio of Zn^{2+}/MEA equal to unity. The solution was stirred continuously for another 2 hours until a colorless, homogeneous and transparent solution was obtained. The prepared sol was kept for aging in airtight bottles at room temperature for 72 hours. For depositing the thin films, the glass substrate (2.5 cm x 2.5 cm) were rinsed and washed with deionized water(DI), acetone and then ultrasonically cleaned with acetone, methanol and deionized water(DI) to remove any oil, dirt or grease. The glass slides were dried in an oven at 150 °C for 30 minutes. Aged solutions were then deposited on pre cleaned glass substrates by spin coating method [11,12,13,14]. ZnO, Ni doped ZnO, Al doped ZnO thin films were deposited at 1000-6000 RPM speed followed by drying on hot plate at 100 °C for 10 minutes. This process of depositing and drying were repeated several times for the proper synthesis of the films. The samples were then annealed at 350 °C for 3 hours in a tube furnace to remove organic species and improve the crystallinity and properties of films[15].



Flow Chart for fabricating 1 at% Al doped ZnO thin films

Result and Discussion

The figure 1, 2 and 3 shows the absorption spectra, raman spectra and fluorescence spectra of 1 at% Ni doped ZnO sol respectively. The absorption spectrum is examined by double beam UV visible spectrophotometer in the wavelength range from 200-600 nm. The figure 5.3 clearly shows that there is an absorption peak at 370 nm. The Raman spectrum is studied by Enspectr Enhanced Spectrometry at a wavelength of 532 nm. Figure 5.5 shows the fluorescence spectrum which is examined by Fluorescence Spectrometer in the wavelength range of 300-750 nm.

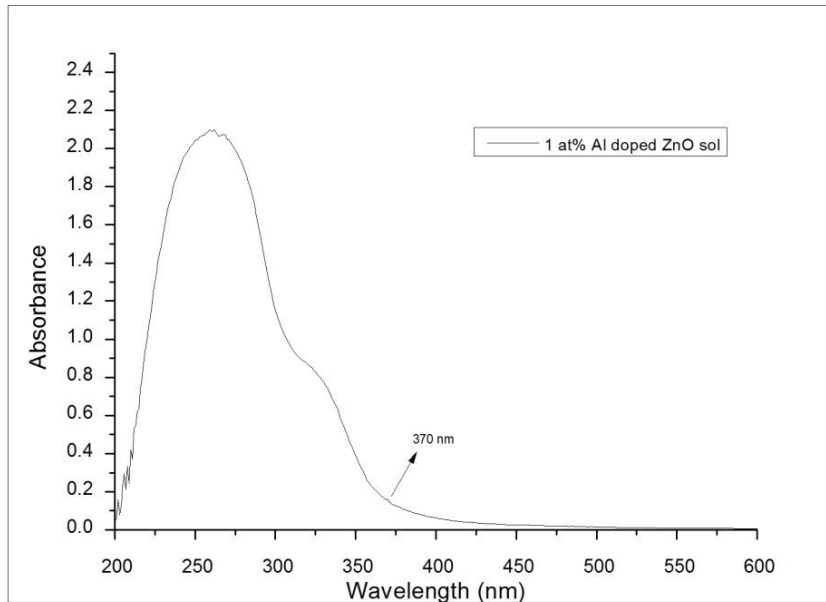


Fig 1: Absorption spectra of 1 at% Al doped ZnO Sol

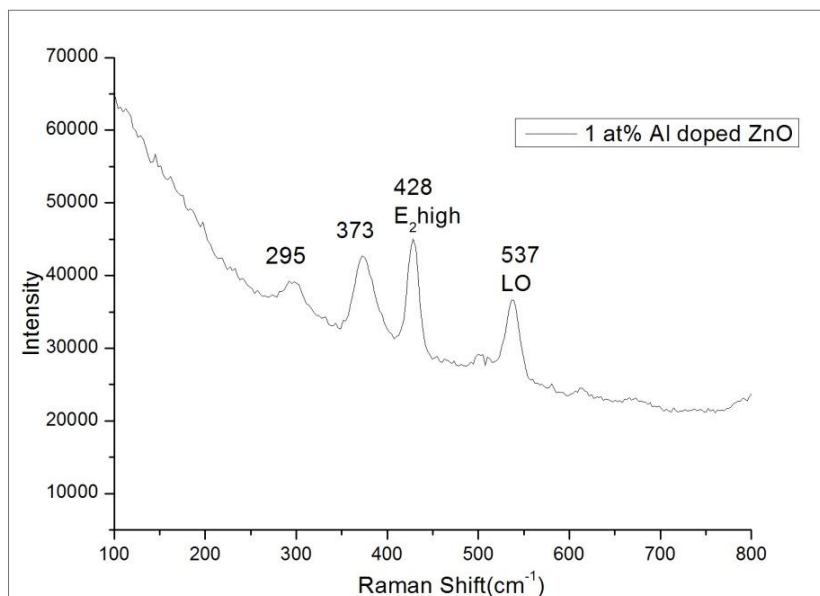


Fig 2: Raman spectra of 1 at% Al doped ZnO Sol

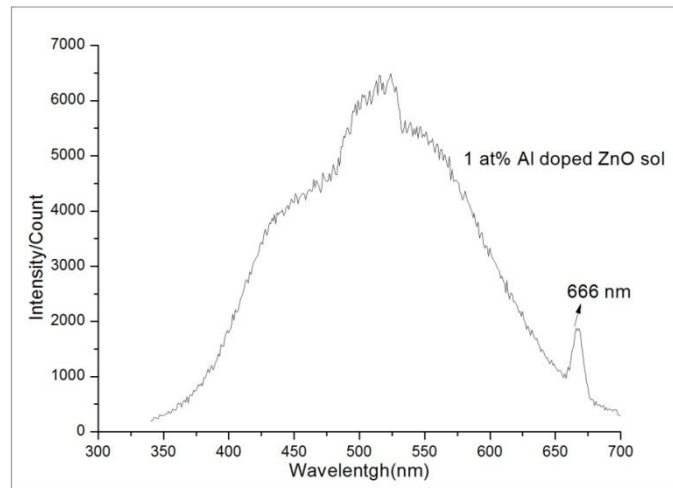


Fig 3: Fluorescence spectra of 1 at% Al doped ZnO Sol

UV Visible Spectra

The absorbance spectra of the synthesized 1 at% Al doped ZnO thin films at different rotating speed from 1000-6000 rpm were investigated in the wavelength range from 300-700 nm as shown in figure 4. Figure 4 shows the absorption spectra for the Al doped ZnO thin films at different rotating speed, the absorbance of the films decreases with increasing wavelength. The absorption spectrum also reveals that the films have low absorbance in the visible and near infrared region, while absorbance is high in the ultra violet region[16].

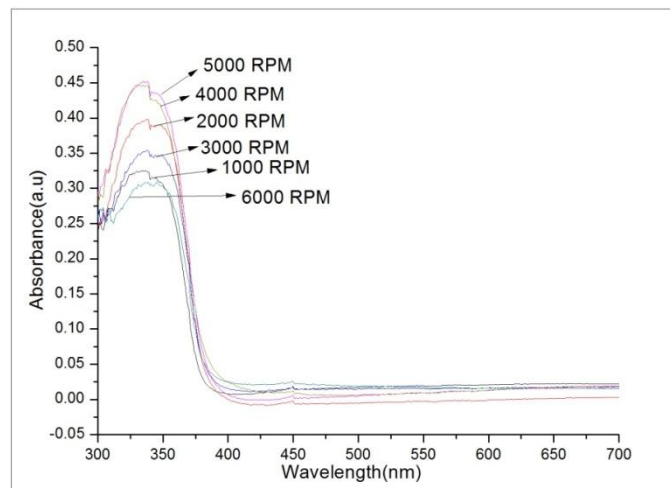


Fig 4: Absorption spectra of 1 at% Al doped ZnO thin film synthesized at 1000-6000 RPM.

Table 1: Variation of absorbance with spin coating speed from 1000-6000 rpm.

S. No.	Spin Coating Speed(rpm)	Absorbance(a.u)
1.	1000	0.3251
2.	2000	0.3975
3.	3000	0.3532
4.	4000	0.3088
5.	5000	0.4514
6.	6000	0.4462

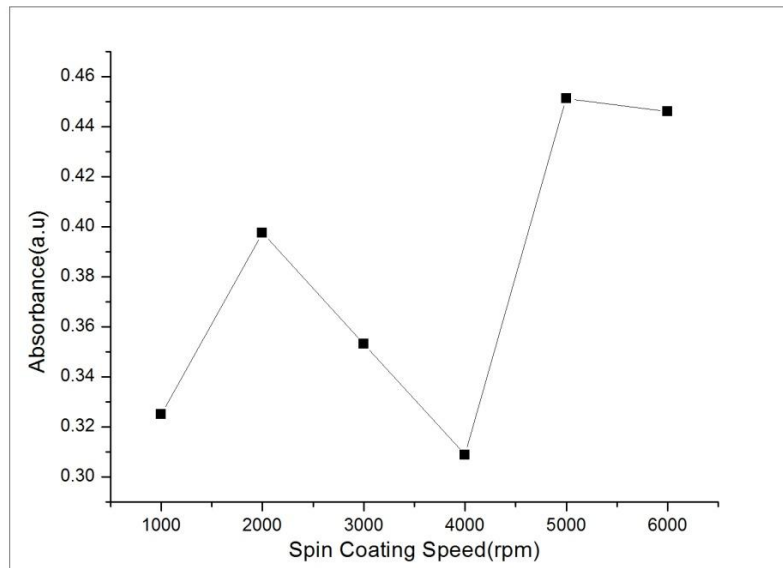


Fig 5: Variation of absorbance with spin coating speed from 1000-6000 rpm.

It is observed from the fig 4 that the rotating speed affects the quality as well as the uniformity of the thin films. The table 1 shows that there is lower absorption at the lowest rotating speed (1000 rpm) and highest absorption at the rotating speed (5000 rpm). At the lowest rotating speed (1000 rpm) there is a discontinuity in the films due to the lack of enough centrifugal force while at higher speed the discontinuity is possibly due to the excessive centrifugal force which spreads out liquid instantly and causes non uniform films. Figure 5 represents the Variation of absorbance with spin coating speed from 1000-6000 rpm.

Figure 6 shows the optical transmittance spectrum of ZnO thin films synthesized at 1000-6000 rpm using double beam UV-spectrophotometer from 320-700 nm. It has been observed that the transmission value of the films are low at short wavelengths (<370 nm) and high at long wavelength (>420 nm). This means that the films behave as a transparent material at longer wavelengths.

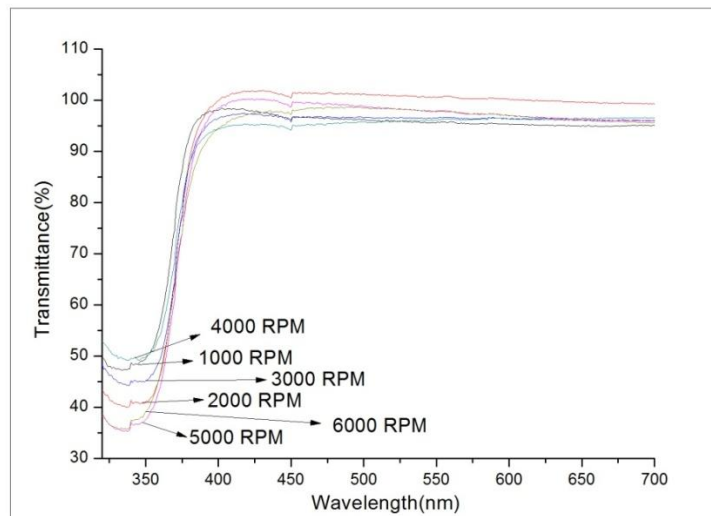
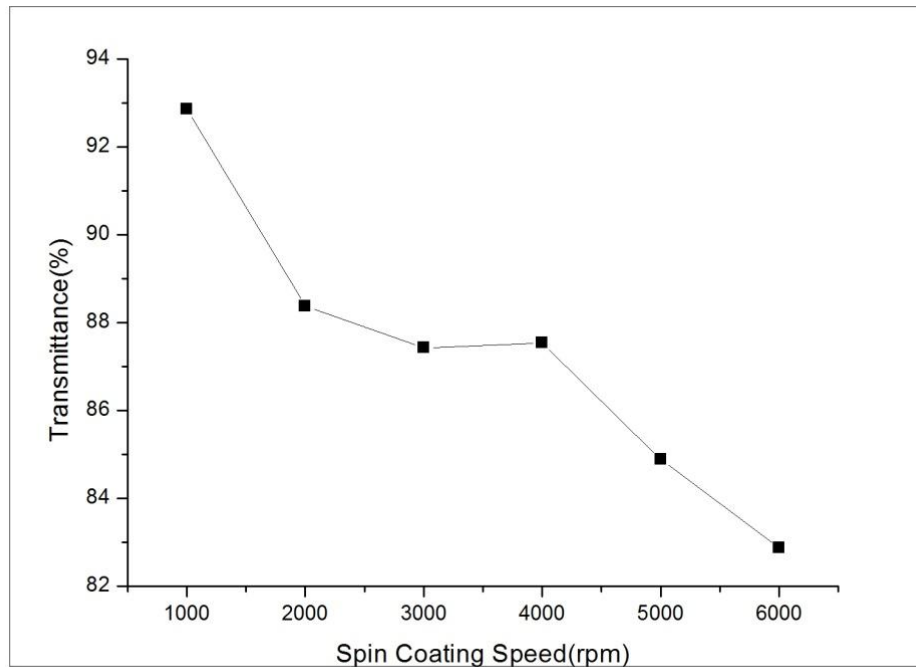


Fig 6: Transmission spectra of 1 at% Al doped ZnO thin film synthesized at 1000-6000 RPM.

Table 2: Variation of Transmittance with spin coating speed from 1000-6000 rpm

S. No.	Spin Coating Speed(rpm)	Transmittance (%)
1.	1000	92.87
2.	2000	88.38
3.	3000	87.43
4.	4000	87.54
5.	5000	84.89
6.	6000	82.87

**Fig 7: Variation of Transmittance with spin coating speed from 1000-6000 rpm.**

The transmission value increases from higher rotating speed to lower rotating speed which indicates that the thin films can be used as transparent conducting oxide for optoelectronic devices. But in case of 6000 rpm, the transmission value is lowest which can be explained by the interference pattern in the transmission spectra. The interference through the film happens when the light reflected from top at specific wavelength and from bottom at other wavelength as it is explained by D. A. Ajadi et al. [9]. Table 2 and figure 7 represents the variation of Transmittance with spin coating speed from 1000-6000 rpm.

Raman Spectra

Figure 8 shows the raman spectra of Al doped ZnO thin film at 1000 rpm - 6000 rpm rotation speed, synthesized by the sol-gel process, are reported herein. ZnO crystallizes in the wurtzite structure which belongs to the C_{6v}^4 ($P6_3mc$) space group. The group theory predicts that the phonon modes belong to the $2E_2$, $2E_1$, $2A_1$ and $2B_1$ symmetries. The $2B_1$ symmetry modes are Raman active. At the Γ point of the Brillouin zone, the theory of group predicts the existence of the following optical modes: $\Gamma_{opt} = A_1 + 2B_1 + E_1 + E_2$. The frequency from the B_1 (low) and B_1 (high) silent mode has been calculated as 260 cm^{-1} and 540 cm^{-1} respectively. In addition, A_1 and E_1 are infrared active, and therefore they split into longitudinal and transverse optical component (LO and TO)[17].

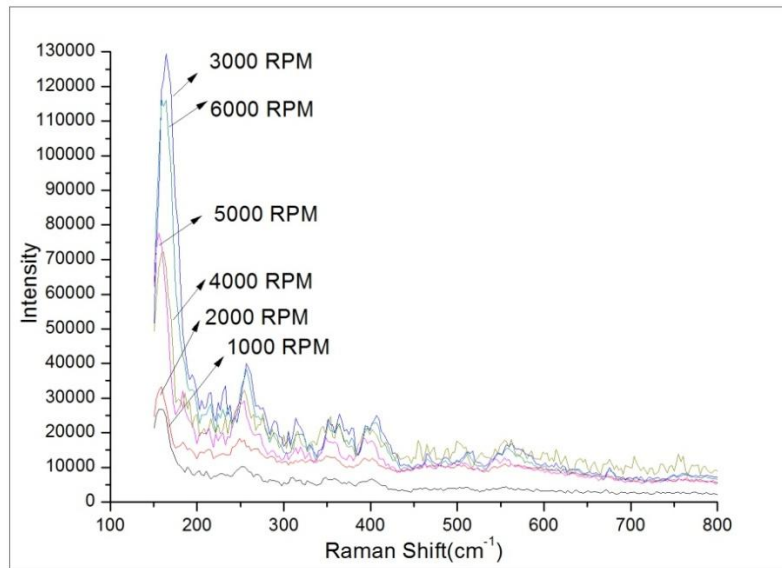


Fig 8: Raman spectra of 1 at% Al doped ZnO thin film synthesized at 1000-6000 RPM.

In the low-frequency region, the peak at 161 is attributed to E_2 (low) mode. The low-frequency E_2 mode, involve mainly Zn motion and E_2 (high) mode represents characteristic of ZnO crystallinity. The peaks at 254, 405 and 561 cm^{-1} may be attributed to 2TA, E_1 (TO) and A_1 (LO) modes. The appearance of peak at about 561 cm^{-1} (A_1 (LO) modes) may be due to lattice defects, namely either oxygen vacancies or zinc interstitials or their combination.

Fluorescence Spectra

The fluorescence spectroscopy is an excellent intensive technique for the investigation of the exact band edge transition levels of a material. A well-defined fluorescence spectrum of the synthesized ZnO thin films ranging from 1000 to 6000 rpm is depicted in Fig 9.

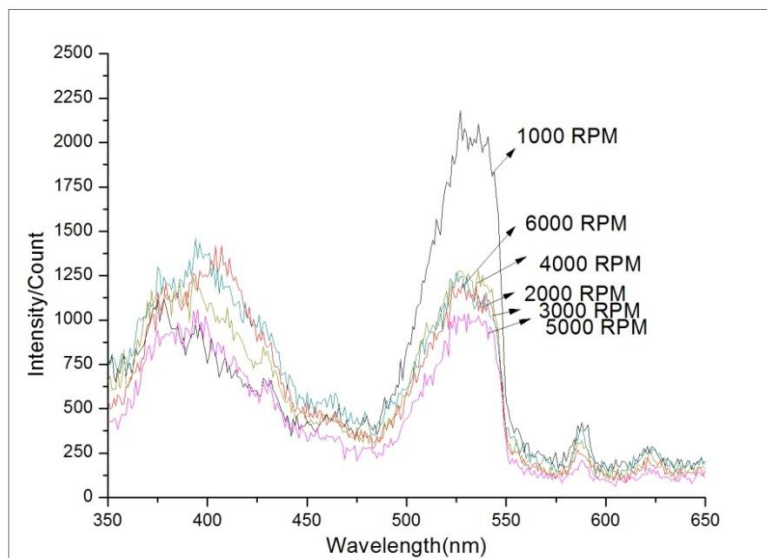


Fig 9: Fluorescence spectra of 1 at% Al doped ZnO thin film synthesized at 1000-6000 RPM.

The fluorescence spectrum showed an emission peak at about 397nm, originated from the exciton recombination corresponding to the near band edge transition (NBE) of ZnO [10]. The visible emission peak at about 527 nm corresponds to the green emission. The green emission peak is attributed to the oxygen vacancies (V_o) and antisite oxygen (O_{zn})[18, 19, 20]. A broad illumination peak is observed over 565 to 620 nm which is attributed to the non-stoichiometry, producing intrinsic defects in the material that may originate from Zn vacancy and antisite defect in the ZnO.

Conclusion

A high quality ZnO thin film on glass substrate has been prepared by sol-gel method using spin coating technique and also optical properties are verified. The average optical transmittance for 1000 rpm to 6000 rpm samples are greater than 80%. The present research shows that high transmittance can be achieved at the spin coating speed of 5000 rpm to 6000 rpm which can be used as transparent conducting oxide electrodes. The fluorescence spectrum emission peak at about 385nm, originated from the exciton recombination corresponding to the near band edge transition (NBE) of ZnO. The visible emission peak at about 521 nm corresponds to the green emission.

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