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A Comprehensive Review on The Effect of Doping and Co-doping onto Optical and Electrical Properties of ZnO Thin Film as P Type TCO in Various Optoelectronic Applications

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Abstract

ZnO stands out as a promising candidate over ITO for TCOs due to its large excitation binding energy (60meV) and direct wide band gap of approximately 3.37 eV .It inherent some intrinsic defect such as O vacancies and Zn interstitials, cause ZnO thin films to only naturally have n-type conductivity, making it challenging to achieve p-type conductivity. This review explores the impact of group IA, IB, and VA doping and co-doping on achieving stable p-type conductivity in ZnO thin films, focusing on their optical and electrical properties with the latest advancements and condensed findings. This study has shown that doping and co-doping strategies improve optical transparency significantly, modifies the band gap and enhances photoluminescence properties ,also causes an anticipated shift in electrical resistivity and carrier concentration with increase in mobility due to fewer lattice defect. Lastly, a brief discussion on issues and difficulties that will need to be resolved rounds off the piece.

Keywords: ZnO thin film, P-type conductivity, Doping, Co-doping.

1. Introduction

Due to their broad use transparent conducting oxide having both excellent electrical conductivity and optical transparency have drawn an abundance of study interest recently. Because of its excellent electrical conductivity and high transparency, indium tin oxide (ITO) is the most widely used TCO. But indium is prohibitively priced, scarce and toxic, ZnO is a very desirable substitute for ITO with an exciton binding energy (60 meV),wide energy band gap (3.37 eV) [1]. With regard to its intrinsic donor shortcomings notably O vacancies and Zn interstitials, ZnO naturally arises as n-type conduction. However, ZnO has issues forming strong-quality, reproducible p-type material via the dopant's restricted solubility, high self-compensation, high acceptor level, and low acceptor dopant dispersion[2].The fabrication of low-resistance p-type ZnO with high hole concentration can be achieved by doping (Li, k; Cu, Ag; N, P) and co doping (N-Al; N-Ga; N-In [3], [4], [5], [6]) of material of various group. For instance, most of Li-doped ZnO films are either n-type or semi-insulated, and conceptually substitutional group-I elements behave as shallower acceptors [7]. Group V element nitrogen (N) is believed to be the right dopant to gain p-type conductivity in ZnO as it is easy to substitute oxygen (O) in the ZnO lattice due to the same ionic radius and small ionization energy. While mono dopant is a useful method for producing p type ZnO films, producing high-quality p-type ZnO films is still challenging due to issues with dopant solubility and ionization energy that can be easily addressed with co dopant. For instance Ga and N co-doping by the sol-gel spin-coating process results hole concentration of 3.96×10^{17} cm^{-3} and resistivity of 5.09 Ω·cm [5] whereas N doping by Rf magnetron sputtering results resistivity of 0.2 Ω cm,hole concentration of 1.95×1017 cm^{-3} [8].Rf magnetron sputtering, Sol gel spin coating, Pulsed laser deposition, Co sputtering,and spray pyrolysis were some of the methods used to prepare P-type ZnO thin films. Of all the techniques accessible for forming p-type ZnO thin films, Rf magnetron sputtering tends to be the most acknowledged method. This method is applied as it delivers high-quality thin films with consistency and robust compositional supervision. In this review paper, all kind of dopant and co dopant with the process of forming the TCO ZnO film has been discussed.

2. Introducing mono doping

2.1. Group IA

Group-I elements, including Li, Na,k and Cs are consistently employed as dopants to replace Zn sites in ZnO and stating acceptor energy band levels.Tsai et al [2] deposited Li doped ZnO film using Rf magnetron sputtering and for each sample observed that the crystals had strong crystallinity and a high preferred orientation with a trans mittance about 87%, resistivity of 0.22 Ω cm, hole carrier concentration of 2.47x1018 cm-3 and mobility of 0.22 cm2 /V s.The Li atoms cause the nearby O atoms to significantly relax since they are located on the substitutional Zn site.when a hole is excited by light it gets trapped into O atoms [9].Xiao et al.[7] also realized the p-type behavior of Li doped ZnO using pulsed laser deposition, While the understanding of carrier type might be regu lated in pulsed laser deposition by altering the growth condition, the relative concentrations of defects such inter stitials and Li substitutions as well as the quantity of Li added to ZnO may have a significant impact on the conductivity of the films [10]. Heliang fan et al.-[11], [12] fabricated Na doped p-type ZnO thin film using sol gel and chemical vapor deposition. The film by sol gel show transmittance above 80% with a blue shift at UV ab sorption edge. The electrical and morphological characteristics were shown to be strongly influenced by the con centration of Na in CVD. Large size impurities like K are substantially better p-type dopants as the interstitial sites are preferably occupied by group I element [13]. The findings of k doped ZnO film prepared by Wu Jung et al. [14] using rf magnetron sputtering indicate a higher hole carrier concentration of 8.92× 1017 cm−3 and a lower resistivity of 1.8 Ω cm at 500 °C and 30% O2 pressure. B.Dey et al. [15] observed an increase in transmittance from 87% to 92% by doping 9% k in ZnO using solid state synthesis.Green synthesized Cs doped ZnO fabricated by Veena et al. [16] confirmed that Zn sites can effectively substituted by Cs resulting a p-type conductivity with a transmittance 86% and carrier concentration of 1.3 × 1018 cm-3.Since Cs act as a deep donor, p-type conductivity of Cs doped ZnO film depend on doping concentration,an increase in dopant concentration may attribute limited solubility in the lattice, leading to a decrease in hole concentration [16]

2.2. Group IB

Cu, Ag and Au belong to Group IB having low formation energy can effectively substitute in Zn sites, which are their preferred acceptor sites. Cu is a preferable option among the IB elements as Cu fills the band gap with electrical levels and replaces Zn due to their comparable ionic radii [9], [17]. Mohammad Suja et al [18] explained that the p-type conductivity of Cu-doped ZnO films using plasma-assisted molecular beam epitaxy, results from Zn being substituted by Cu. Experimental analysis shows a shallow acceptor state located 0.15 eV above the valence band edge possessing lowest resistivity of 0.6 Ω cm and a maximum hole concentration of 1.54 × 1018 cm−3.A photoluminescence spectroscopy result revealed by P. Khosravi et al [17] show that 2.7% Cu doped ZnO thin film prepared using co sputtering method have low defect density along with a improved UV emission. Also, the crystal structure was restored by the Cu dopants by filling the Zn vacancies leading to an enhanced p-type conductivity with a hole concentration of 2.6 x1015 cm-3 and mobility 0.286 cm2 /Vs. Given its size and energy level, Ag also seems like a viable fit as a dopant in ZnO thin film, inducing a 3.6 ev shallow acceptor state over head the valence band maximum with a minimum transition energy.A theoretical low formation energy of Agzn over Agi makes Ag a great choice for p-type doping,also inhibiting the creation of compensatory defects [19], [20]. A under substrate temperature of 500 ◦C and a partial O2 pressure of 250–300 mTorr.,Hong Seong Kang et al [21] observed a neutral bound electron hole pair acceptor with a hole density 6x1017 cm−3.The p-type conduc tivity of an Ag doped ZnO thin film formed using the electron beam evaporation was investigated by Soo Kim [20] as a function of annealing temperature. The results showed that p-type conductivity decreased when temper ature rose over 350 c.Ag:ZnO film at 350C annealing temperature showed the best p-type conductivity a hole concentration of 5.09x10^19 cm3, minimum resistivity of 7.25x10-2 Ω cm, and mobility of 1.69 cm2 /Vs. Au/ZnO nanoparticles exhibiting excellent physical and photoluminescence properties have synthesized by L.Ouarez et al. [22] using sol gel. All the films were preferably c-axis oriented with a increment in visible emissions by an increase in Au concentration. A maximum intensity was observed at 385 nm for the10 at% doped film.

2.3. Group VA

In recent research, group VA elements as N, P,As, Sb has successfully doped into zinc oxide applying various methods. N is typically a shallow acceptor in ZnO, facilitating easier p-type conductivity whereas P,As and Sb are generally deep acceptors, with energy levels farther from the valence band, making them less effective for p type doping at room temperature. N-doped ZnO thin films synthesized by means of PLD reveals that when N concentration rises from 0.2% to 0.7%, the band gap of ZnO:N films drops from 3.3 eV to 3.1 eV. Both the N inclusion and the development of N-induced defects in the ZnO:N films are responsible for this decrease in the band gap [23]. In comparison with the PLD, the film evolved via RF magnetron sputtering has the least electrical resistivity, highest carrier concentration, and mobility of 0.2 Ω.cm, 1.95×1017 cm−3, and 58 cm2 /V.s respec-tively. Additionally, sputtering results in a film formed on a quartz substrate shows transmittance for the 400–800 nm wavelength range, revealing a high optical transparency in the visible region with a predicted band gap of 3.24 eV [8].In MOCVD-prepared film, temperature is a major factor. Sample produced at 400 °C has the maximum

hole concentration, and its emission at 387.6 nm becomes dominant. The emission at 387.6 nm becomes mixed with 376 nm and loses its dominance as the growth temperature drops from 350 to 250 ℃ [8], [24]. Additional group-V elements with large atomic radii P, As, and Sb, are believed that to occupy the Zn sites rather than the O sites [25]. Pankaj et al. [26] reported strongly conductive and secured p-type phosphorus doped ZnO (PZO) thin films fabricated by dual ion beam sputtering and subsequent thermal annealing with maximum hole concentration 8.62×1019 cm−3,mobility 1.08 cm2/Vs and resistivity of 0.066 Ω.cm. In another report Kim et al. [27] stated a PZO film grown by radio frequency sputtering and rapid thermal annealing (RTA) has shown the hole concentration, carrier mobility, and resistivity 1.73x1019 cm-3,3.51 cm2/V s and 4.4 Ω.cm, respectively. Park et al. [28] found that As-doped ZnO films prepared by sol-gel spin coating under O-rich conditions achieved over 70% transmit tance from 380–800 nm and exhibited a blue shift, indicating not only high transparency also a shift to shorter wavelengths. Additionally As concentration of 6.7 at% showed a carrier concentration of 1.62×1019cm-3, mobility of 94.0 cm2/Vs, and resistivity of 3.99×10−3 Ω cm. P-type ZnO thin films doped with 3 at.% Sb and produced at 600 °C in a N2 environment were preserved by Feng et al. [29] via PLD, with resistivity 84.51 Ω.cm, hole con centration 1.17×1017 cm−3,mobility 0.63 cm2/V·s and reduced band gap value was around 2.9 eV but annealing at 650°C changed ZnO films from p-type to n-type, highlighting that Sb-doping and annealing control defects and conductivity. Dong Hun Kim et al [30] studied when the resistivity of Sb-doped ZnO thin films reduces as Sb doping concentrations rise because extra holes act as carriers and have a hole concentration of 4.412×1017 cm−3 with 1.5 at.% of Sb using RF magnetron sputtering and RTA.

Table 1. Effect of single doping and synthesis method on the optoelectronics properties of ZnO thin film.

Author	Year	Dopant	Synthesis method	Optoelectronics properties	Ref.
Shu-Yi Tsai	2011	Li	RF magnetron sputtering	Resistivity 0.22 Ω cm Avg transmittance 85% Hole Carrier concentration 2.47×10^{-18} cm^{-3}	[2]
Shuang Wu	2017	Na	Chemical vapor deposition	Hole concentration in order of 10^{15} cm^{-3}. Shallow acceptor level ∼132 m eV	[12]
Wu Jung	2008	K	Rf magnetron sputtering	Hole concentration 8.92×10^{17} cm^{-3}, resistivity 1.8 Ω cm, and Hole mobility of 3.89 cm^2/V s	[14]
Veena Ragupathi	2014	Cs	Sol gel spin coating	Increase crystalline size to 44.54 nm Hole carrier concentration 7.35×10^{18} cm^{-3} Average transmission 88 %	[16]
P.Khosravi	2019	Cu	Plasma assisted molecular beam epitaxy	High hole concentration of 1.54×10^{18} cm–3 Low resistivity of 0.6 Ω cm Moderate mobility of 6.65 cm^2 /V.s	[17]
Hong Seong Kang	2006	Ag	Pulsed laser deposition	Hole concentration 2.29×10^{18} cm^{-3} Resistivity is 0.9 Ω cm Mobility is 3.03 cm^2 /V.s	[21]
I.Ka	2014	N	Rf magnetron sputtering	Resistivity of 0.2 Ω cm, Hall mobility of 58 cm^2/V s Hole concentration of 1.95×10^{17} cm^{-3}	[23]
Pankaj Sharma	2017	P	dual ion beam sputtering	Hole concentration of 3.77×10^{19} cm^{-3} Resistivity of 0.066 Ω cm Mobility of 1.08 cm^2/V s	[26]
Chanhyoung Park	2013	As	Magnetron sputtering	Hole concentration of 2.16×10^{17} cm^{-3}, Mobility of 1.30 cm^2/V.s and Resistivity of 22.29 Ω-m	[28]

L.P.Purohit	2016	Sb	Pulsed laser deposition	Hole concentration of 1.17×10^{17} cm^{-3}, Hole mobility of 0.63 $cm^2/V\cdot s$ Resistivity of 84.51 Ω·cm	[3]

3. Introducing Co-doping

3.1. Acceptor-Donor pair

Donor-acceptor pair interactions can improve p-type behavior by stabilizing the generated holes trapping free electrons at donor sites, which would otherwise recombine with the holes. Kim et al.[3] noted a pronounced band emission and green emission in Al, N co-doped spin coated films, exhibiting a maximum hole concentration of 8.11x 10^{16} cm−3 and a carrier mobility of 9.965 $cm^2/V\cdot s$. The N incorporation was improved by the presence of In in the N-In doped ZnO film developed by Zhao et al.[5] resulting in a steady p-type conductivity, according to secondary ion mass spectrometry.3.12 Ω·cm was determined to be the lowest dependable room-temperature resistivity, accompanied by a 2.04 x10^{18} cm–3 carrier concentration and 0.979 cm^2/V S Hall mobility. Compared to N-doped p-type ZnO, B. Yao et al.[31] found that N-B co-doped ZnO has superior electrical characteristics, with a room-temperature resistivity of 2.3 Ω cm, Hall mobility of 11 cm^2/ V.s , and carrier concentration of 1.2 × 10^{17} cm^{-3}. Due to donor-acceptor pair recombination, the low-temperature photolumin escence spectra of the codoped ZnO film differs from that of the N-doped ZnO in that it has two dominating peaks at 3.096 eV and 3.251 eV, respectively. Using pulsed laser deposition, Xing et al. [6] describe the formation of Li–Ni codoped p-type ZnO thin films and find a remarkable drop in electrical resistivity, from 3.6 × 103 to 0.15 Ω cm by adding 2 mol% of Ni in the Li-doped ZnO film. Furthermore, when Ni is included into the Li:ZnO lattice, the activation energy decreases to 6 meV from 78 meV.

3.2. Acceptor-Acceptor pair

Acceptor-acceptor pair can lead to stronger hole generation by suppressing donor defects and by shifting their energy levels closer to the valence band, acting as shallower acceptors. Li–N dual acceptor activation in N-Li:ZnO thin films yields excellent p-type properties with a high carrier concentration of 2.64 × 1017 cm−3 and a relatively low resistivity of 0.09 Ω cm. Based on this, Yang et al. [32] used PLD to create the p-ZnO:(Li, N)/n-ZnO homojunctions on Si(100) substrate. The impact of post-annealing on the electrical characteristics and stability of the p-type Ag,N:ZnO thin film was illustrated by Ortega et al [33]. Following annealing at 300 and 400 °C, the highest electrical characteristics were discovered, showing a very high hole concentration of 3.17×10^{19} cm^{-3}, a low resistivity of 8.56×10^{-3} Ωcm, and a Hall mobility of 23 cm^2/Vs. Tanaka et al. [34] describe a stable and repeatable p-type conduction in ZnO by N and P doping with a hole concentration and Hall mobility of 5.3×10^{17} cm^{-3} and 0.94 cm^2/ V.s, respectively at an ideal annealing temperature of 600 ∘C. Huang et al. [35] reported that p-type single-crystalline ZnO films without the appearance of any secondary phase may be formed by dual ion implantation of Na,N in ZnO thin films followed by dynamic annealing, with hole concentration of 1.34 × 10^{17} cm^{-3} and hole mobility of 8.37 cm^2/ V.s.

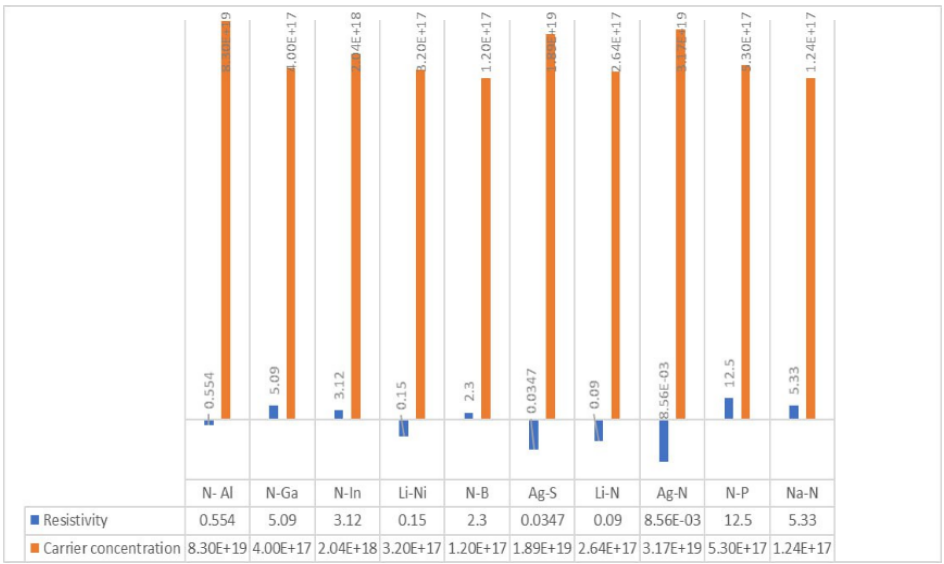


Fig.1 : Comparison of resistivity and carrier concentration of different co-dopant

4. Difficulties in achieving p-type ZnO and possible solutions

The complexity in achieving p-type conductivity originates by;1) limited solubility and formation of precipitate of p-type acceptor dopant,2)formation of low energy residual impurities and intrinsic defect leading to self-compensation of p-type dopant,3)unionized deep acceptor levels and 4)formation of AX and DX bistability structures [13], [17], [25], [36]. O vacancies, Zn interstitial, and Zn anticities, which are known to be the main barrier of p-type ZnO,owing to their limited formation energy [36], [37]. First-principles calculations and EPR experiments consistently show that the O vacancy is a deep, negative-U donor, where the 1+ charge state is thermodynamically unstable. In p-type ZnO, Vo can recompense for the acceptor impurities since they possess a truncated formation energy [9], [36]. Zn interstitials act as unstable superficial donors. Zni have a shallow donor state and large formation energy, but when the Fermi level gets closer to the valence band, it drastically decreases and impeding attempts to produce p-type conductivity and cause of an inadvertent n-type conductivity .Zn anticities are unlikely to function as significant compensatory centers in p-type ZnO, nevertheless, due to their high formation energy and act as deep donors [36]. Acceptors are compensated for when the AX center forms a deep defect complex ny releasing two electrons which is equal to the capture of two holes [13]. Numerous scholars have initiated research to examine potential solutions for the p-type doping challenge. In their investigation, Donald et al. [36] have concluded several approaches based on estimations of defect formation energies from first principles;1)By modifying their chemical potential, acceptor impurities' solubility can be raised by nonequilibrium growth techniques (MOCVD,MBE etc.),2)ionization potential of acceptor state can be lowered by designing dopant complexes or shallow dopant,3)Modifying the host band configuration at the band boundaries can lower the defect compensation. Research has demonstrated that altering the chemical potential of the acceptor dopant by the use of various nonequilibrium deposition techniques like CVD [12], Plasma assisted MBE [18], [38], Plasma assisted metal organic CVD[20] enhance the solubility of acceptor dopant. In terms of preparation techniques, some studies have adopted to grow ZnO in an O2 rich atmosphere [7], [17], [39] and at a high temperature annealing [2], [7], [11], [14], [16]. This is because the oxygen atmosphere replaces more oxygen vacancies with oxygen atoms and the post-annealing activates the acceptor [9], [36].To produce high-quality film, the right substrate that matches the lattice, appropriate deposition techniques, and a high-purity deposition procedure are also required [25].

5. Conclusion

In summary, doping and co-doping significantly influence the optical and electrical properties of ZnO thin films, enhancing their potential as p-type transparent conducting oxides (TCOs) for optoelectronic applications. Effective incorporation of dopants such as group V elements (e.g., N, P) or co-doping strategies with dual elements improves carrier concentration, mobility, and transparency, enabling better conductivity and tunable bandgap. These advancements address the inherent challenges of p-type ZnO, facilitating its integration into devices like light-emitting diodes, solar cells, and photodetectors. However, achieving stable and reproducible p-type conductivity remains a critical focus for future research to optimize ZnO's practical application in advanced optoelectronics.

6. References

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