Research Progress on the Photoelectric Properties of Indium-Doped Cadmium Oxide Transparent Conductive Films

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Abstract. Indium-doped cadmium oxide (CdO: In) thin films have excellent photoelectric properties, and have broad application prospects in the fields of photoelectric devices, enhanced spectroscopy, and the like. In recent years, the influence law and internal mechanism of various preparation technologies and process conditions on the photoelectric performance of CdO: In thin films are still one of the research hotspots. The preparation methods of CdO: In transparent conductive thin films were reviewed, the effects of various preparation process conditions on the photoelectric performance of the thin films were discussed, and the future research directions of CdO: In thin films were given.

1. Introduction
Transparent conductive oxides (TCO) such as cadmium oxide, indium oxide, and zinc oxide have low resistivity and high light transmittance, and thus have broad application prospects in the fields of optoelectronic materials and the like. For example, transparent conductive electrodes, surface acoustic wave devices, varistor, etc. Among these TCOs, CdO is an ideal transparent electrode material for photovoltaic solar panels due to its high transmittance and low resistivity in the visible region of the solar spectrum. Doping CdO thin films with other elements can significantly improve their optical and electrical properties. At present, the main elements of CdO thin film doping are Al, Ti, Sn, In, etc. According to Kawazoe's calculations, In-doped CdO thin films are likely to have superior photoelectric performance. Subsequently, the lowest value of the CdO: In film resistivity prepared by Gupta et al. Using the PLD method can reach $2.9 \times 10^{-5} \Omega \cdot \text{cm}$, and the average transmittance in the visible light range is up to 85%, which also experimentally confirms that CdO: In is expected to become high Quality TCO film material. CdO has a simple cubic crystal structure. Doped In$^{3+}$ replaces Cd$^{2+}$ in the CdO lattice and becomes a donor, releasing more free electrons in the conduction band, thereby reducing the resistivity and widening the optical band gap. Past studies have shown that the photoelectric properties of CdO: In are closely related to the process conditions such as In content, substrate temperature, and annealing treatment. This paper refers to a large number of domestic and foreign literatures in recent years, summarizes the research progress of the optical and electrical properties of CdO: In films, and gives prospects for future research.

2. Preparation Technology of CdO: In Nano Thin Film Materials
At present, there are many methods for preparing CdO: In nano film, including magnetron sputtering method, pulse laser deposition method, metal organic chemical vapor deposition method, filtered
cathode vacuum arc method, etc. Each method has its advantages and disadvantages, and the corresponding preparation method needs to be selected according to actual needs.

Magnetron sputtering (MS). Magnetron Sputtering (MS) is currently the most mature thin film preparation method. Because of its good repeatability and simple operation, it is widely used to deposit oxide thin films. The basic principle of the magnetron sputtering method is: under the action of the electric field and the magnetic field, the high-voltage electric field is used to accelerate the inert ions to strike the atoms on the surface of the target material, so that they escape from the lattice bond and escape, and the sputtering atoms or atomic groups are deposited on the substrate. The surface reacts with oxygen atoms to form an oxide film. According to the different power sources selected, the magnetron sputtering method can be divided into three types: DC sputtering, RF sputtering and intermediate frequency sputtering. Among them, DC sputtering and RF sputtering are the most common methods. The experimental procedure of preparing CdO: In by magnetron sputtering mainly includes the following three parts: (1) A baffle is set between the indium-cadmium alloy target and the substrate, the sputtering chamber is evacuated to the required vacuum degree, and a flow meter is set Display the number and fill with argon gas to the working vacuum; (2) After setting the power and working time of the working power supply, perform pre-sputtering; (3) After pre-sputtering for a period of time, open the baffle and use the mass flow controller to Oxygen is released into the room and coating begins.

![Figure 1. Schematic diagram of magnetron sputtering device.](image)

Babu et al. Used the DC magnetron sputtering method, using cadmium (34%)-indium (66%) alloy as the target, setting the target-base distance to 65mm, and using a sputtering pressure of 4pa on a glass substrate at a temperature of 532K A CdO: In thin film was prepared with a low resistivity of $1.2 \times 10^{-3}$ and a visible light transmittance of 87%. Coutts et al. Obtained a CdO: In thin film with excellent photoelectric properties by radio frequency magnetron sputtering. The minimum resistivity of the CdO: In thin film is about $2.3 \times 10^{-4}$, and the average transmittance in the visible light region is as high as 90%.

Compared with other preparation methods, the magnetron sputtering method has the advantages of uniform film thickness, high repeatability of film quality, and strong adhesion between the film and the substrate. However, in the process of depositing thin films by reactive sputtering, problems such as anode disappearance and cathode poisoning often occur, which makes it difficult to control the sputtering process, which limits its application.

Pulsed laser deposition (PLD). Pulsed laser deposition (Pulsed Laser Deposition, PLD) is a new type of vacuum physical deposition film technology. The basic principle of PLD technology is shown in Figure 2. The focused laser enters the vacuum chamber through the entrance window to irradiate the target at a 45° angle, so that the target sputters plasma, and the particles in the plasma (including many
particles, ions, atoms, and atomic clusters, etc.) form a Gaussian distribution along the target it expands to the substrate and deposits a film on its surface.

![Figure 2. Schematic diagram of PLD device.](image)

The experimental procedure of preparing CdO: In film by PLD technology mainly includes the following three parts: (1) pumping the air pressure in the vacuum chamber to a preset vacuum degree; (2) introducing oxygen to increase the air pressure to the working pressure and ionizing the oxygen; (3) Set the substrate temperature, deposition time, target material rotation speed and target substrate distance, and start deposition.

PLD technology has the following advantages: (1) It can ensure that the composition of the target and the film is highly consistent, and realize the transfer of the chemical stoichiometric ratio of the material; (2) The optical system and the vacuum environment are used for deposition without introducing impurities; (3) Deposit at low temperature.

Gupta et al. Used the PLD technique to deposit CdO: In films on quartz substrates, and studied the effects of growth temperature and oxygen partial pressure on the structure, optical and electrical properties of the films. Studies have shown that the optical transmittance of the film mainly depends on the growth temperature, and the oxygen partial pressure has almost no effect on the transmittance. In addition, the electrical properties of the film are closely related to the growth temperature and oxygen partial pressure. Zheng et al. Irradiated a Cd-In alloy target with pulsed laser in an oxygen atmosphere (10pa), and grown a CdO-In thin film on a quartz glass substrate at 300°C. The film shows a cubic crystal structure of CdO, but because cadmium is replaced by indium atoms, the lattice constant changes slightly. The results also show that when the mass fraction of indium in the film reaches 3.9%, the resistivity of the CdO: In film is lower ($5.95 \times 10^{-5}$) and the direct band gap energy ($2.97\text{eV}$) compared to undoped CdO Higher, higher transmittance in the visible region. Subsequently, Segura prepared CdO: In thin film using PLD technology to study the relationship between the photoelectric performance of the thin film and In doping concentration and substrate material. The results show that the optical band gap of the film can reach more than 3.2eV when doped with 0.5% In. The minimum film resistivity on c-axis sapphire substrate is $4.5 \times 10^{-5}$. On a single crystal MgO (100) substrate, the film resistivity is about $3.2 \times 10^{-5}$. Sun Xiaonu focused on the effects of In content and substrate temperature on the structure and photoelectric properties of CdO: In films. The study found that In doping weakened the dominant growth of the thin film (200) crystal plane. When the concentration of In reaches 3.9wt%, the photoelectric performance of the thin film is the best. When the concentration of In increases to 5.6wt%, In2O3 is formed in the thin film, and the photoelectric performance of the whole thin film decreases. The surface of the film deposited at a substrate temperature of 300 °C is the densest.

The above results show that the PLD technology can be used to prepare thin films with precise composition, simple operation and wide application range. However, PLD technology also has the
disadvantage of poor uniformity of large-area deposited films, which is difficult to achieve industrial production.

Metal Organic Chemical Vapor Deposition (MOCVD). Chemical vapor deposition (CVD) is a technology in which one or several gaseous reactants undergo a chemical reaction on the surface of a substrate to deposit a film. This technology has been widely used to prepare semiconductor films (crystalline and amorphous), insulators, and metals. Metal-Organic Chemical Vapor Deposition (MOCVD) is one of chemical vapor deposition methods. The organometallic precursor used in MOCVD technology (shown in Figure 3) will decompose to form a metal oxide when heated, and then deposited as a thin film. Under appropriate growth conditions, an epitaxial layer can be formed. Oxygen plasma assisted technology (OPA) MOCVD can achieve an oxide film with extremely low defect density and high chemical purity grown under high vacuum. In many cases, MOCVD does not necessarily require a high vacuum, and indeed deposition can occur at moderate pressures (2 to 100kPa). Currently, MOCVD has become the preferred method for manufacturing compound semiconductor films.

Figure 3. Collection of volatile organic metal precursors used for TCO thin film MOCVD processing.

Wang et al. Prepared a 150nm thick CdO: In thin film on a glass substrate and studied the effect of In doping concentration on the photoelectric properties of the thin film. The highest electron mobility of the film is 69cm2 / Vs, and the lowest resistivity is about 6 × 10-5. The CdO: In thin film deposited by Jin et al. On single crystals on MgO (100) has excellent optical transparency, with an average transmittance in the visible range > 80%. When the In doping concentration is 2.6%, the room temperature film conductivity is 5 × 10-5, and the optical band gap is 3.18eV.

The films obtained by MOCVD technology can be of high quality both chemically and structurally, but this technology also has limitations, which are related to the purity of the required metal organic precursor and the cost of preparation. In many cases, metal-organic precursors tend to be extremely reactive, and therefore require careful handling and purification through thorough recrystallization or sublimation procedures. In addition, most metal organics are volatile and often highly toxic, so special handling procedures are required.

Filter cathode vacuum arc method (FCAD). Filtered Cathodic Arc Deposition (FCAD) is a technique for preparing various high-quality metal oxide thin films. The principle of FCAD technology is: first generate and excite plasma on the surface of the cathode target, then use the magnetron elbow to filter the uncharged large particles, after filtering the plasma with a certain energy reaches the surface of the substrate and finally condenses into a film (As shown in Figure 4).
Yuankun Zhu used FCAD technology to prepare high-quality CdO: In films. The effects of substrate temperature, In doping concentration, oxygen partial pressure and other factors on the film structure and photoelectric properties were studied in detail. The films prepared under the doping conditions of 230 °C, 7mtorr and 1.2at.% In have the best photoelectric performance.

FCAD technology has the advantages of fast deposition rate, low substrate temperature, easy adjustment of composition, and large area deposition. The prepared film has good surface flatness, uniform thickness and uniform doping, and is suitable for preparing high-quality films.

The above results indicate that CdO: In films prepared by different methods have different photoelectric properties. Among these methods, the films prepared by PLD technology and FCAD technology have the best performance, but these two technologies also have their own shortcomings. Therefore, it is of great significance to further study the influence of the preparation process on the performance of the film for the application of CdO: In film.

3. Effects of process conditions on the photoelectric properties of CdO: In thin films

The photoelectric properties of CdO: In films are closely related to the process conditions such as In content, substrate temperature, and substrate materials in the films.

Effect of In doping amount on the photoelectric properties of CdO: In thin films. During the preparation of CdO: In thin films, changing the amount of In doping can significantly affect the photoelectric properties of the thin films. Studies have shown that with the increase of In doping content, In$_3^+$ replaces Cd$_2^+$ in the lattice to provide extra electrons, so the carrier concentration continues to increase. When the In doping concentration is low, the grain boundary scattering in the film dominates, and free electrons accumulate in the grain boundary to form a charge shield, which in turn reduces the barrier height in the grain boundary and the electron mobility gradually increases. When the content of In doping is high, ionization scattering in the film replaces grain boundary scattering, and the interaction between ionized impurities and electrons hinders the movement of free electrons, which reduces the electron mobility.

As the In doping content increases, the carrier concentration increases, and the Fermi level of the degenerate semiconductor enters the conduction band, and the Burstein-Moss effect occurs, and the forbidden band width of the film becomes larger.

Zheng et al. Used high-purity cadmium (99%) and indium (99%) powders with indium contents of 3wt%, 5wt%, and 7wt%, respectively, and fully mixed them to make targets for PLD deposition. The film was deposited at a fixed temperature of 300 °C and an oxygen partial pressure of 10pa. Studies have shown that the resistivity of the thin film decreases to $5.95 \times 10^{-5}$ as the indium content increases to 3.9wt%, and slightly increases as the indium content increases to 5.6wt%. The change in resistivity can be explained by the doping concentration of In and its presence in CdO. When the doping concentration of In$_3^+$ is less than the solid solution limit of In$_3^+$ replacing Cd$_2^+$, the doped In$_3^+$
replaces Cd2+ and releases more electrons to increase the carrier concentration, and the resistivity decreases accordingly. When the indium doping concentration exceeds the solid solution limit, excess In will exist in the gap In3+ or form an In-O bond (such as In2O3). The increase of the gap In3+ (acting as the donor center) will cause the receptor vacancy V2-cd to increase significantly to maintain the charge balance in the crystal, which is one of the reasons for the decrease in carrier concentration. In addition, non-conductive In2O3 clusters as carrier traps will also reduce the carrier concentration of the film. Combining the above two effects, when the concentration of doped In exceeds the solid solution limit, the carrier concentration decreases and the film resistivity increases.

Le et al. Prepared CdO: In thin films on quartz substrates by RF magnetron sputtering at room temperature. The effect of In doping concentration on the photoelectric properties of thin films was studied in detail. Studies have shown that compared with undoped CdO films, when the In doping concentration is low (0.012 at.%), The carrier concentration and electron mobility of the film increase simultaneously. The experiment confirmed that the increase in mobility is attributed to the decrease in oxygen vacancy (VO) concentration and the increase in grain size in the film. At the same time, it was found that the optical band gap of CdO: In film increases with the increase of In doping concentration, which is the increase of carrier concentration (Burstein-Moss effect) and the composition of CdO: In film (Vegard rule).

Zhu et al. Prepared CdO: In thin films on borosilicate glass using pulsed filter cathodic arc deposition (PFCAD). The substrate temperature was maintained at 230°C, the oxygen pressure was maintained at 0.931pa, the film was annealed in air and nitrogen atmosphere, and maintained at 450°C for 1 hour, and then naturally cooled to room temperature. Studies have shown that compared to undoped CdO thin films (refractive index of 2.52 at 500nm), the refractive index of CdO: In films decreases (refractive index of 2.28 at 500nm), and extinction coefficient also decreases Due to the increase in optical band gap.

Flores et al. Used sol-gel technology to mix CdO and In2O3 precursor solution to obtain CdO: In thin film, and explored the effect of In doping concentration on the photoelectric properties of the thin film. The experimental results show that at a sintering temperature of 450°C, the resistivity of the film decreases first and then increases with the increase of the doping amount. The resistivity is the smallest when the In doping amount is 5at%. The transmittance of the thin film in the wavelength range greater than 600nm is kept above 85%.

Effect of substrate temperature on the photoelectric properties of CdO: In thin films. In the preparation of CdO: In thin films, the temperature of the substrate is a very important process parameter. The substrate temperature will change the crystallinity, surface roughness, Cd oxidation degree and other properties of the film, and then affect the film carrier concentration and mobility changes, and ultimately determine the photoelectric properties of the film. At present, the influence of the substrate temperature on the CdO: In film is a hot issue.

Gupta et al. Studied the effect of substrate temperature on the optical and electrical properties of thin films. Studies have shown that the resistivity increases with the substrate temperature, the carrier concentration decreases with the substrate temperature, and the electron mobility increases with the substrate temperature. The decrease in carrier concentration may be caused by annealing of point defects and interstitial impurities. The annealing of point defects and interstitial impurities leads to a decrease in impurity scattering and an increase in mobility.

Using PLD technology, Zheng deposited a transparent CdO: In thin film on a quartz glass substrate and studied the effect of substrate temperature on the carrier concentration and Hall mobility of the thin film. The results show that with the increase of the substrate temperature, the resistivity tends to decrease first and then increase, showing the minimum resistivity (1.15 × 10-4) and the maximum carrier concentration (5.35 ×) at 300 °C 1020cm-3) and electron mobility (101.43cm2 / (V·s)). When the substrate temperature is greater than 300°C, the decrease in carrier mobility may be related to the increase in surface roughness and particle size of the film with increasing temperature, which leads to an increase in structural defects. In addition, as the substrate temperature increases from 100 to 300 °C
C, the optical band gap increases, reaching a maximum (2.91 eV) at 300°C, and then decreases as the substrate temperature increases, which can be explained by the Burstein-Moss effect.

Zhu Yuankun used filtered cathode vacuum arc technology (FCAD) to prepare CdO: In thin films on quartz glass substrates. Studies have shown that as the substrate temperature increases, both the resistivity and surface resistance of the film decrease, the electron mobility gradually increases, and the carrier concentration decreases first and then increases. The lowest resistivity of the 125nm thick film prepared at 425°C was $5.9 \times 10^{-5}$, the highest electron mobility was 112cm² / (V·s), and the corresponding carrier concentration was $9.4 \times 10^{20}$cm⁻³. As the substrate temperature increases, the energy of the atoms on the surface of the film increases, and these atoms are more likely to migrate to the optimal growth position, so that it is easier to crystallize to form larger grains, which reduces the grain boundaries in the film. This will facilitate the transport of free electrons, so the electron mobility of the thin film increases with increasing temperature. The carrier concentration of the film is related to the concentration of internal defects. When the substrate temperature increases from room temperature to 230°C, the defects in the film decrease, and the carrier concentration also decreases. When the temperature is further increased, the gap In³⁺ is more likely to replace Cd²⁺ to provide excess electrons, and the carrier concentration increases accordingly. In addition, with the change of the substrate temperature, the transmission curve of the film also shows changes in the absorption cut-off blue shift and near infrared plasma resonance wavelength shift. These changes are closely related to the changes in carrier concentration.

Effect of Annealing Treatment on the Photoelectric Properties of CdO: In Thin Films. Studies have shown that the photoelectric properties of CdO: In films are closely related to the crystal structure and crystal quality of the films. Appropriate annealing treatment can make the crystallinity of the film improved to a certain extent, thereby improving the photoelectric performance of the film. At present, the research on the annealing process mainly focuses on the annealing atmosphere and annealing temperature.

Flores et al. Used two solutions of CdO and In₂O₃ as precursors to prepare CdO: In thin films on glass substrates at room temperature using sol-gel technology. The effects of annealing temperature and annealing atmosphere on the photoelectric properties of thin films were studied. Studies have shown that after annealing, the electrical resistivity of the film is significantly reduced, which may be related to the increase in the oxygen vacancy concentration of the film. Compared with the annealing treatment in three different atmospheres of H₂, N₂ / H₂, and vacuum, the resistivity of the film annealed in vacuum is the lowest, reaching $2 \times 10^{-3}$; the direct band gap energy is the highest, reaching 3.6eV (the annealing temperature is 600°C, the annealing time is 1h, the indium concentration in the precursor solution is 55 at%).

Yang et al. Used DC magnetron sputtering technology to prepare CdO: In films on glass substrates, and studied the effect of annealing process on the structure and electrical properties of the films. The results of the study showed that the resistivity of all samples decreased after annealing. The resistivity of the film prepared at an oxygen concentration of 4.29% was $2.95 \times 10^{-4}$, and after annealing treatment, the resistivity decreased to $2.00 \times 10^{-4}$. On the one hand, the annealing process can provide energy for grain aggregation and reduce the grain boundaries in the film; on the other hand, the annealing process can increase the oxygen vacancy concentration in the film and release dissolved residual oxygen, thereby capturing free electrons on the film surface. Therefore, the decrease in resistivity of the film after annealing is the result of the combined action of these factors.

4. Summary
The transparent conductive film has both good conductivity and high transmittance in the visible light region, so it has great research significance and broad application prospects. CdO: In film is one of the candidates to occupy the transparent conductive film market due to its superior photoelectric performance. At present, the photoelectric performance of CdO: In thin films obtained by various preparation methods can reach the following levels: the transmittance in the visible light region is more than 80%, and the resistivity is on the order of 10⁻⁴.
However, there are still some deficiencies in the current research on CdO: In thin films. The main problems are as follows:

1. There is a lack of systematic research on the relationship between the structure of CdO: In film and the photoelectric performance. The nucleation and growth process of the thin film should be studied more deeply to prepare CdO: In thin film with better performance.

2. The optical band gap values of CdO: In films obtained by different methods are large. The extended misuse of some images in related research makes the obtained optical band gap value lack physical significance.

3. Lack of low-cost and high-quality CdO: In thin film preparation technology. Other preparation technologies should be actively explored to meet the requirements of new optoelectronic devices.

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