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Rare Earth Element Doped ZnO Thin Films and Applications

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Abstract

In this review, general information has been given about the recent studies on thin film coating techniques, zinc oxide (ZnO) thin films and zinc oxide films doped with rare earth elements (REE). As a thin film coating method, the sol–gel method is widely used due to its economical nature. However, it has been seen that the magnteron sputter technique is also very popular due to its ease of application. Zinc oxide is a semiconductor with a wide band gap and is easy to produce and to supply. In addition, it does not have negative effects on the environment and human health. All these properties have led to intensive studies on zinc oxide. Due to its optical and electrical properties, it has found many different uses such as solar cells, light emitting diodes, gas sensors and flat panel displays. Recently, various studies have been carried out to improve the properties of zinc oxide and to create new areas of use by doping with rare earth elements.

Keywords: Rare earth element, REE–doped, thin film, coating, ZnO

Nadir Toprak Elementleri Katkılı ZnO İnce Filmler ve Uygulamaları

Öz

Bu derleme çalışmasında ince film kaplama teknikleri, çinko oksit ince filmler ve nadir toprak elementleri (NTE) katkılandırılmış çinko oksit (ZnO) filmler konusunda son yıllarda yapılan çalışmalar hakkında genel bilgilendirme yapılmıştır. İnce film kaplama metodu olarak, sol–jel metodu ekonomik olması nedeniyle yaygın olarak kullanılmaktadır. Bununla birlikte magnteron sputter tekniğinin de uygulama kolaylığı nedeniyle oldukça popüler olduğu görülmüştür. Çinko oksit geniş bant aralığına sahip bir yarıiletkendir ve hem üretimi hem de temin edilmesi kolay bir malzemedir. Bunun yanında çevre ve insan sağlığı üzerinde olumsuz etkileri bulunmamaktadır. Bütün bu özellikleri çinko oksit üzerinde yoğun çalışmalar yapılmasına yol açmıştır. Optik ve elektriksel özellikleri nedeniyle, güneş pilleri, ışık yayan diyotlar, gaz sensörleri ve düz panel ekranlar gibi çok farklı kullanım alanları bulmuştur. Son zamanlarda, nadir toprak elementleri katkılandırma yapılarak çinko oksitin özelliklerini daha da üstün bir konuma getirme ve yeni kullanım alanları oluşturmak için çeşitli çalışmalar yapılmaktadır.

Anahtar Kelimeler: Nadir toprak elementi, NTE–katkılı, ince film, kaplama, ZnO

INTRODUCTION

Thin film is a layer of thicknesses from a few micrometers to several Å. Thin films, which have gained momentum among the research and development studies carried out in recent years, particularly form the basis of electronic device technology. With the development of technology, novel methods are developed in scientific studies and innovative materials and technologies can be produced as a result of these studies. Thin film coating techniques have also reached an important status with the development of technology in recent years. Thin films that can be produced in angstroms or nano thicknesses that are too thin to be seen with the naked eye can sometimes be used as a sensor, as a solar cell or as a functional element of optical systems. In addition to its wide band gap feature, its UV absorber feature and its antibacterial nature make zinc oxide a very interesting material (Manikandan et al., 2017).

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THIN FILM COATING TECHNIQUES

There are various taxonomies related to thin film coating techniques in the literature, the most commonly used methods are summarized in Figure 1.

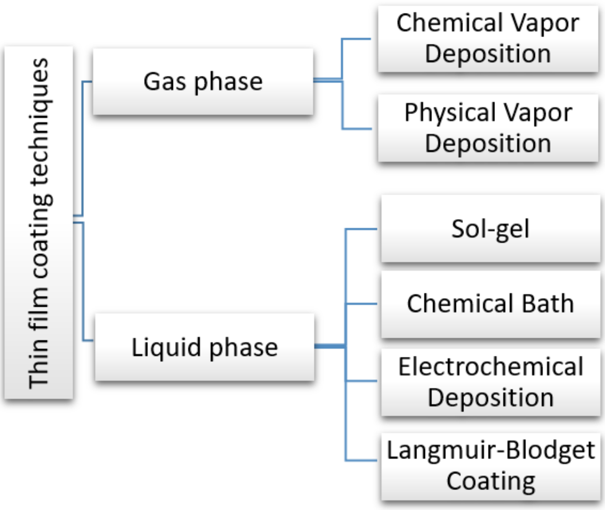

Figure 1. Thin film coating techniques.

Gas Phase

Among the thin film coating methods, when the material to be coated is in the gas phase, two types of coating techniques have been developed: Chemical vapor deposition and the physical vapor deposition methods.

Chemical vapor deposition

Chemical vapor deposition (CVD) method is the storage of vapor phase reactants on the substrate surface in the reactor by heat or magnetic effect. In this method, film formation can be done even at low temperatures. This technique is also used for high purity nanoparticle synthesis. Chemical vapor deposition involves flowing precursor gas or gases into a chamber containing one or more heated objects to be coated. Chemical reaction occurs on and near the hot surfaces resulting in the deposition of thin film on the surface. Chemical by–product gases and unreacted gases exhausted out of the reactor. Chemical vapor deposition is done in hot–wall reactors, cold–wall reactors, sub–torr pressured reactors and above–atmospheric pressured reactors with or without carrier gasses at temperatures in a wide range such as 200–1600 °C (Creighton and Ho, 2001). Even though there are different process configurations for the chemical vapor deposition method, they all have some common points:

1. Reactant gases are injected into the system and the reaction takes place on the substrate.
2. The temperature of the substrate is always under control.
3. The reactions of gaseous reactants on the substrate surface form the desired solid thin film.
4. By–products should be removed from the reactor environment as they will create impurities (Dopkin and Zuraw, 2003).

Physical vapor deposition

Physical vapor deposition (PVD) technique is applied by evaporating or sputtering materials under vacuum. The process is based on the principle of separating atoms from the surface of a material that will be coated and adsorbing to the surface of the substrate to be coated.

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Physical vapor deposition processes are applied with two ways including evaporation and sputtering methods. The evaporation method is to deposit the coating material, which is evaporated by a heater in a vacuum environment, as a thin film layer on the material to be coated. The vapor phase required for PVD coatings is obtained by resistance, induction, electron bombardment and cathodic arc evaporation methods (Türküz, 2006). The evaporation process is generally carried out under 10^{-5}–10^{-6} torr vacuum (Bunshah, 1980).

In the sputtering method, the solid raw material is ionized with aid of high energy and reactive gases and transformed into plasma to be adhered on the material to be coated in a controlled manner. The disadvantages of the system are the low deposition rate and low ionization effect in the plasma and the increase of the substrate temperature in the sputtering technique. In recent years, the effect of these disadvantages has been reduced by applying magnetic fields in sputtering technology (Smallman et al., 2007). The most important advantage of the sputtering method is that alloys with different evaporation rates at different vapor pressures can be successfully deposited without changing their composition. The high consumption of electricity causes the cost of the method to be higher than the evaporation method (Sönmezoğlu et al., 2012).

Recently, Mundra et al. (2021) developed an integrated deposition system comprising Physical Vapour Deposition (PVD) and Chemical Vapour Deposition (CVD) as sub systems to achieve a wide range of thin film depositions. The developed system can find applications to develop thin films of a range of material on various substrate materials.

Liquid Phase

Liquid phase deposition techniques are classified as sol–gel coating, chemical bath coating, electrochemical deposition method and Langmuir–Blodgett coating method. Liquid phase coating methods are low cost methods and appropriate for large scale coatings with any geometry.

Sol–gel deposition

Sol–gel coating process (soft chemistry) is based on the principle of adsorbing the solution of the substance to be coated on a surface and converting it into a solid phase by using dip coating, spin coating and spray coating methods (Toygun et al., 2013). In sol–gel methods hydrolysis and condensation of molecular precursors is used to prepare organic and inorganic materials or thin films. The most important factors affecting the rate of hydrolysis and condensation reactions are pH, water ratio, temperature, catalyst type and concentration (Türhan, 2000). The applicability of the sol–gel process at room temperature and the ability to obtain products in various shapes, sizes and compositions have led to the increasing use of this technology in various scientific and engineering fields. Beside its good application under laboratory conditions, its use is gradually increasing for large–scale productions (Pierre, 1998).

Dip coating method is based on the principle of immersion and extraction of the substrate at the same speed. The dip coating method takes place in five stages: dipping, drawing up, coating, percolation, and evaporation. In the dipping stage, the sustrate is dipped into the solution at a constant speed. It is kept in the solution for a short time. Subsequently, the substrate is pulled up at the same speed. In the coating stage, the parts of the substrate that come out of the solution are covered with the solution (Özer, 2007). In the flow–through stage, the solution coated on the substrate flows down dropwise. In the evaporation stage; the excess solution that cannot drip evaporates away from the surface and a dry gel is obtained on the substrate. The dry gel formed on the substrate becomes a film after oven drying.

Spin coating method is based on the principle that the solution is dropped on the substrate while rotating it around an axis horizontally and the solution spreads on the substrate surface by the effect of the centrifugal force. The spin coating method consists of 5 steps. In the drip step, the solution is dropped onto the substrate fixed on the rotating surface. The initially standing substrate is rotated at a certain speed and for a certain time. The substrate should reach the desired rotational speed in the shortest possible time. Because the rotational speed affects the uniform thickness of the film. During rotation, the solution dropped onto the substrate is spread over the entire surface of the substrate by centrifugal force. If the solution is dripped more than necessary, the excess solution is blown over the substrate. During the rotation, the thickness of the film decreases. At the end of the rotation, the thickness of the film becomes uniform over the entire surface of the substrate. After

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evaporation and oven drying, a thin film is obtained (Brinker and Sherer, 1990).

In the spray coating method, the solution prepared to be coated is spread on the substrate surface with the help of a pressurized inert gas or air. After this process, thin film is obtained by applying evaporation and oven drying processes.

Chemical bath deposition

The chemical bath deposition (CBD) method consists of a heated magnetic stirrer, a water bath, substrate and substrate holder, a thermometer and a pH meter.

The chemical bath deposition method is based on slowing down the reaction of the ions that will form the film in solution. The cleaned glass substrate is immersed in the prepared solution for a certain time to form thin films on the surface of the glass. Among the techniques used to prepare the thin film, CBD is a relatively inexpensive and simple method (Canci, 2009). In this system, we can list the parameters affecting the film quality and film thickness: pH value of the solution, temperature and reaction time of the solution, solvent concentration, structure and concentration of the catalysts used, annealing temperature and time, drying and drying atmosphere.

The most important advantages of the CBD method are that it can be applied at low temperature and atmospheric pressure, does not require expensive equipment, and is suitable for coating film on large surfaces (Sönmezoğlu et al., 2012).

Electrochemical deposition

Metal coating or conductive polymer coating studies can be performed in the electrochemical coating technique (Aslan et al., 2016). By applying electrochemical polymerization process for the conductive polymer coating, the monomers in the solution can be polymerized on the electrode surface and coated on the surface. In electrochemical polymerization, the monomer is scanned in the anodic region by cyclic voltammetry or coated with a constant potential/current. The main factor in not performing polymerization in the cathodic region is polymerization. It is generally not achieved as a result of the instability of the radical anion consisting of monomers in this region. Electrochemical polymerization is carried out through a 3–electrode system. These are the working electrode, reference electrode and auxiliary electrode. In the selection of the working electrode and auxiliary electrode, factors such as the type of monomer to be studied and the purpose of the study should be taken into account. If the spectroelectrochemical behavior of the polymer film in the visible region is to be examined, a transparent indium tin oxide (ITO) coated glass electrode should be used as the working electrode. Platinum (Pt) electrodes are generally used as auxiliary electrodes due to their inertness and high conductivity. Silver wire, Ag/AgCl and standard calomel electrode (SCE) are the most commonly used reference electrodes.

Langmuir–Blodgett coating

This method was invented by Irving Langmuir and Katharine Blodgett for creating highly ordered monolayer films at the air/water interface and transferring these films to substrate surfaces. The Langmuir–Blodgett (LB) method entails the use of amphiphilic molecules with a polar "head group" and a hydrophobic "tail". When such amphiphilic molecules are dispersed onto the surface of water, the head groups point down, because they are strongly solvated by water, and the hydrophobic tails point up. The LB method has recently been used to coat substrate surface with monolayer and, after multiple transfers, multilayer films. Although setup of the coating process is difficult and there is a need of amphiphilic material for coating, excellent film thickness control, homogenous film formation and multilayer structures with varying layer compositions are possible with LB coating method (İpek, 2015).

While two surfaces are covered with dip coating and Langmuir–Blodgett technique, a single surface can be covered in spin coating. In spray coating and electrochemical coating techniques, a single surface or both surfaces can be coated optionally. Plating with dip coating technique and Langmuir–Blodgett method is generally similar. However, with the Langmuir–Blodgett method, a more precise, controlled and nano–metric thickness or molecular size coating can be obtained. It is more difficult to make a molecular size sensitive coating in the dip coating technique. Both techniques involve the immersion of the substrate in the solution, and the speed of dipping and removal in both techniques is an important parameter for the smoothness of the coating.

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ZnO THIN FILMS

In recent years, wide band gap semiconductor materials have attracted great attention from researchers and industry. The structural, electronic and optical properties of semiconductor materials used in many areas can be changed depending on the control of doped elements (Üzar, 2018). Doping process is carried out depending on the dopant concentration of the host material (Gottardi et al., 2013). ZnO, a semiconductor material, has an optical band gap of 3.37 eV and a large exciton binding energy of 60 meV. ZnO has ideal properties such as low processing cost, being abundant in nature and non–toxic elements, resistance to radiation, high mechanical and thermal stability (Gezgin and Gündoğdu, 2021). ZnO is often called II–VI semiconductor because zinc and oxygen belong to groups 2 and 6 of the periodic table, respectively. It is known that II and VI group double compound semiconductors crystallize either in cubic zinc sulfide or in hexagonal wurtzite structure (Figure 2) where each anion is surrounded by four cations at the corner of a tetrahedron (Şener, 2019).

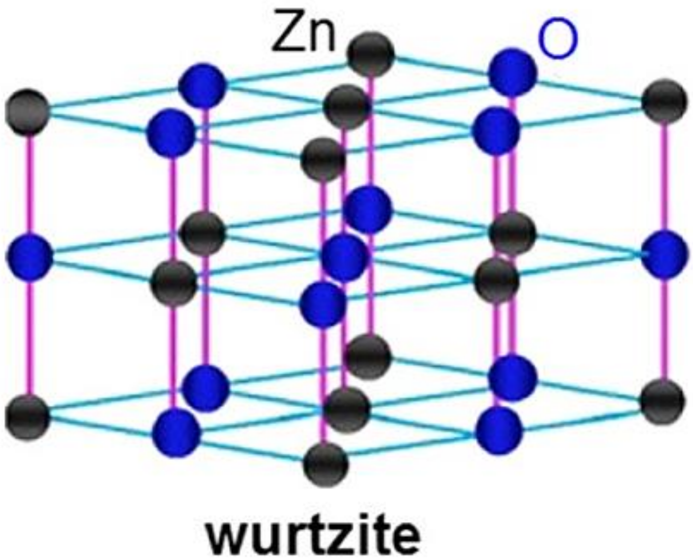

Figure 2. ZnO wurtzite structure (Kung and Sreenivas, 2016).

ZnO is preferred for use in many technological areas such as solar cells, light emitting diodes, gas sensors and flat panel displays due to its good optic and electrical properties, as well as its non–toxic, low cost, easy production (Anand et al., 2018a; Frieiro et al., 2020; Gottardi et al., 2013; Üzar, 2018). In addition, it is used in different applications such as blue luminescent devices, solar cell windows, laser diodes, optical sensors (Kayani et al., 2020). It is known that transition metal and rare earth element doped ZnO are called diluted magnetic semiconductors and these semiconductors are used in different areas due to their structural, electrical, magnetic and optical properties (Asikuzun et al., 2017). Rare earth ions are particularly interesting for areas such as photovoltaic photocatalysis, optoelectronic devices, and flat panel displays (Gottardi et al., 2013) and it is also known that the optical activity of the material doped with rare earth ions is increased (Petersen et al., 2010).

REE DOPED ZnO THIN FILMS

Gd and Al co–doped ZnO thin films for optoelectronic applications were obtained by Anand et al., (2018), and these films were prepared using nebulizer spray method. The films were determined to have an optical transmittance above 86% and good crstallinity. It has been stated that Gd co–doped thin film can produce high optical and good electrical conductivity for optoelectronic applications (Anand et al., 2018b). In a study carried out by Siraj et al., (2015), it has been reported that Y–doped ZnO thin films obtained by pulsed laser deposition have high transparency in visible region and can also be used in optoelectronic devices like solar cells, thin film transistors. Fenwich et al.,

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(2007) investigated optical, magnetic, and structural behavior of transition metal and rare earth–doped (Gd, Eu, Tb) ZnO thin films. It was stated that rare earth–doped samples could be useful for optoelectonic applications. Minami et al., (2000) investigated the transparency and conductivity properties of Sc and Y doped thin ZnO films prepared using magnetron sputtering. It was determined that ZnO: Sc thin films had lower resistance than ZnO: Y thin films, and also an average transmittance of over 85% in the visible range were obtained for both of the thin films. Dielectric and magnetic properties of Ce–doped ZnO thin films prepared using sol–gel dip coating method were investigated by Kayani et al., (2020). It was found that the magnetic properties of thin films obtained with different Ce concentrations were better at 1% addition ratio of Ce, but the dielectric properties of these films were better at 5% of Ce. In a study, undoped ZnO and rare earth–doped ZnO thin films were prepared by sol–gel spin coating method (Üzar, 2018). The structural, optical, electrical properties and solar cell performances of undoped ZnO, Yb–doped ZnO, Eu–doped ZnO, Eu/Yb co–doped ZnO thin films were investigated. The average optical transmittance of the rare earth doped ZnO samples was found to be 98% at different regions in the visible light. The resistivity of the rare earth–doped ZnO samples was increased, and the solar cell performance of the ZnO nanostructures was improved with doping Yb and Eu elements. Er–doped ZnO thin films were obtained by spin coating method, and the structucal and optical properties of these films depending on Er concentration and annealing temperature were investigated by Vettumperumal et al. (2015). It was reported that the average crystal size of 27.44 nm at 500 °C and 29.28 nm at 600 °C were obtained and the average transmission was to 80% with the calculated value of optical band gap being 3.26–3.32 eV. In the study reported by Llusca et al., (2014), Er doped ZnO and Er, Yb co–doped ZnO thin films were prepared with RF magnetron sputtering under different O_2–rich atmospheres. It has been determined that the atmosphere during deposition affects the optical activity and the optical activity of erbium ions increases under the flow of oxygen. Also, visible up–conversion emission increased with the addition of Yb. Sm amd Al co–doped ZnO thin films for opto–electronic applications were investigated by Anand et al., (2018a). These films were deposited on the glass substrate using nebulizer spray pyrolysis method. Sm and Al co–doped ZnO thin films were showed high transparency (around 90%). Thin films have been found to be higly suitable for opto–electronic device applications (Anand et al., 2018a). Ce, Tb and Eu doped ZnO thin films were prepared by Frieiro et al., (2020) using RF magnetron sputtering and they developed monochromatic light emitting devices in blue for Ce, green for Tb and red for Eu (Frieiro et al., 2020). Luo et al. (2012) investigated photoluminescence properties of undoped and Ce–doped ZnO films prepared by DC magnetron sputtering. They copared the structural, optical, and photoluminescence properties of Ce doped ZnO thin films and undoped ZnO thin films. In addition, Ce–doped ZnO thin film showed a wide blue luminescence, and it was determined that these films could be used for potential applications in optoelectronic devices. Petersen et al., (2010) studied the structural and optical properties of Eu–doped ZnO thin films. These films were prepared using sol–gel method and magnetron reactive sputtering. When the structural studies of the films were evaluated, it was determined that the films prepared with magnetron reactive sputtering had better structural quality than the films prepared with sol–gel technique. In the study reported by Soumahoro et al., (2011), Yb–doped ZnO thin films were prepared by spray pyrolysis technique. It was determined that the thin films, which were evaluated for structural, optical, and electrical properties, had a visible transmittance of around 75–90% at about 375 nm for the main absorption edge at 3.3 eV. Er–doped ZnO nano thin films were prepared by sol–gel method and microstructure and electrical properties of the films prepared with different Er concentrations were examined by Asikuzun et al., (2017). It was observed that the microstructural, electrical and optical properties of ZnO films were improved with the contribution of rare earth element. Microstructure and chemical structure of undoped and Nd–doped ZnO thin films prepared by RF co–sputtering were studied by Gottardi et al. (2013). As a result of the changes in the film composition, structural changes were determined especially at low Nd atomic concentration (Nd $\leq$ 3 at.%). Transparent, antibacterial and conductive thin films can be achieved with REE doped ZnO thin film coating (Minami et al., 2000).

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CONCLUSION

In summary, especially in the 2000s, it has been observed that many studies have been carried out on rare earth element doped zinc oxide thin film coatings. Sol–gel technique and mangetron sputtering method were preferred as thin film coating methods. The sol–gel technique has been preferred because it is economical and does not require special conditions. The magnetron sputtering method has been applied by many researchers because it produces smoother and higher quality thin films. However, it is very difficult to cover very large surface areas with the magnteron sputtering method as in the sol–gel method. Magnteron sputtering method is more suitable for coating small surfaces. As a result of the studies, an improvement in the desired direction has been observed in the electro–optical properties of ZnO thin films doped with rare earth element. On the other hand, doping the ZnO thin film with rare earth ions increased the optical activity of the material. ZnO thin film having high optical and good electrical conductivity for optoelectronic applications were achieved and the solar cell performance of the ZnO nanostructures was improved. Also, additive ratio of the REE ions affected the physical properties of ZnO films.

CONFLICT OF INTEREST

The Author report no conflict of interest relevant to this article.

RESEARCH AND PUBLICATION ETHICS STATEMENT

The author declares that this study complies with research and publication ethics.

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