

FABRICATION TECHNIQUES FOR PREPARATION OF LEAD ZIRCONATE TITANATE (PZT) THIN FILMS-A PROMISING SOLID SOLUTION FOR APPLICATIONS AS SENSOR, TRANSDUCERS, MEMS AND ENERGY HARVESTING

Ram Chhavi Sharma

SGT University Gurugram, Haryana, India, rcsharma@sgtuniversity.org

Reetu

SGT University Gurugram, Haryana, India

Vibhawari Srivastava

Babu Banarasi Das University, Lucknow (U.P.), India

Abstract: Lead Zirconate Titanate (PZT) in micrometre thin film attract significant attention due to wide range applications as sensor, transducers, MEMS and energy harvesting. The properties like high magnetoelectric (ME) coupling, breakdown strength, remanent polarization, electric field coupling, ease of miniaturization along with ferroelectric, dielectric and optical properties make it as versatile material for such applications. For effective results, the selection of fabrication technique to prepare a particular solid solution is of significance. Depending on the study undertaken, the different fabrication techniques are used for the preparation of thin film. The prepared PZT thin films were tested using suitable characterization techniques. Some findings and their critical analysis based on research studies undertaken are presented in this article.

Keywords: PZT, Thin film, Fabrication, Sol-gel, Ink jet

1. Introduction

Due to its innate piezoelectric and ferroelectric capabilities, Lead Zirconate Titanate (PZT) has shown promise in a number of scientific applications. [1,2]. The improvement in methods of thin film deposition and characterization and development of new methods, PZT has been explored in various domains to get the desirable outcome. The desired morphotropic phase boundary (MPB) for PZT is provided by the various combinations of Zr/Ti ratio [3]. To get a better shape, PZT has been doped with several minerals, which unintentionally improves its properties even in the non-MPB phase [4]. Doping with Fe^{3+} reduces ferroelectric polarisation but increases magnetization and make PZT multifunctional [5]. The lattice structure is altered as a result of doping with La, Nb, and Fe, which enhances one or more PZT characteristics [6]. In order to give PZT, a proper orientation and multilayer feature, $Bi_{3.25}La_{0.75}Ti_3O_{12}$ (BLT) is employed as a buffer layer in various thicknesses with PZT [7]. This results in superior ferroelectric characteristics. PZT's micro level structure, orientation, and characteristics are all improved by several coatings [8]. The crystal structure and chemical content of PZT can be determined using X-ray photoelectron spectroscopy (XPS) [9]. Utilizing the protective layer, $0.67Pb(Mg_{1/3}Nb_{2/3})O_3-0.33PbTiO_3/Pb$, can safeguard the PZT's inherent features[10]. As the number of contacts rises, O_3 (PMN-PT/PZT) heterostructures are the structure with the highest performance [11]. The PZT thin film develops a capacitive structure during

synthesis, which enables its use as a memory storage device [12, 13]. At both high and low temperatures, PZT has been shown to be a good candidate for the construction of electronic devices [14,15], and working pressure studies have shown that it changes PZT's shape and crystalline structure [16]. When PZT is stressed, its properties remain unchanged at low stress, but as the stress rises, internal fractures develop and the dielectric loss rises [17,18]. Occasionally a thin film surface may be completely damaged by a tiny amount of stress, yet controlled stress methods can repair it [19]. In order to learn more about the PZT lattice structure, this research carefully examines the numerous techniques for creating PZT thin films using a variety of materials. Investigations have also been done into the effects of various material combinations in order to improve the quality of PZT. Analysis is based on a strategy based, on the findings of several research investigations.

2. Fabrication Techniques –Deposition of Thin Film

2.1 Sol-Gel Method

In this method, the metal organic precursor acts as the raw material for the synthesis process. To form the final thin PZT layer, these precursors are spin coated onto the substrate, allowed to dry, and then heated through annealing. To manufacture the best PZT thin film, every stage of the procedure was altered in accordance with the available research. These variations include varied base metal solution ratios, various solvents, spin coating speeds, substrate materials and their temperatures, drying process temperatures, drying process gases, annealing process temperatures, and analysis techniques. We are summarising the major research that has been done in this area based on this.

Sol gel technology was used by A. Wu et al. [19] to create the PZT thin layer on an aluminium substrate. To obtain the stable sol for a longer time, various compounds are used. To obtain a better perovskite structure, the seeding effect, in which pure PZT powder is combined in various ratios with the parent sol, is also examined. To create the stable precursor, these chemicals are combined with various stabilisers in an alcoholic medium and is diluted with ethanol, acetone etc. On an Al substrate, the film is created using the dip coating technique. The film is burnt at approximately 700°C for 1 hour and dried at 120°C for 30 minutes. The temperature at which the perovskite structure is obtained has decreased with the usage of seed at 5% weight, according to XRD and scanning electron microscope (SEM) study. To ascertain the rate at which isotherms crystallised, Oleg Babushkin et al. [20] studied PZT thin film X-ray diffraction. SiO₂, Titanium, and Platinum are measured with silicon substrates at thicknesses of 1µm, 1 nm, and 10 nm, respectively. Spin coating is used to deposit a precursor made using the sol gel process. Its thickness is measured by the repetition of spin coating at a temperature of 350°C. In order to analyse the crystallisation of thin films, high temperature XRD is used. This is what a furnace is made for. After each heat treatment, the thin film's microstructure is also characterised using SEM. Q. Zou et al. [21] employed the sol gel method and the noval polyol metalorganic pathway to produce PZT thin films at low temperatures on Pt/Ti/SiO₂/Si substrates. For analysis purposes, different thermal analyses such as FTIR, XRD etc. are carried out. Glycolate sol is utilised to stop the pyrolysis process in its tracks. Zr and Ti sols are produced by mixing ZrCl₄ and TiCl₄ in pro-pylene glycol (1,2-n-propandiol). To obtain a more complete solution, lead acetate was put in to Ti and Zr. Filtration is used to eliminate PbCl₂ as a by-product. Pb(Zr_{0.52}Ti_{0.48})O₃ was formed by mixing Pb, Zr, and Ti solutions to create Pb/Zr or Pb/Ti.

Pyrolysis took place at 350–400°C, and then annealing took place at 500–600°C. At 250–430°C during pyrolysis, the TGA curve depicts the weight loss of the organic group from the solution. When the PZT precursor is heated to 730°C, DTA analysis shows that PbO is formed and the organic material is broken down. Using spin coating and annealing, a single crystal perovskite structure without crazing is created at 550°C.

For the manufacture of PZT thin films with various Zr/Ti ratios for non-volatile memory applications, Zuleeg R et al., utilized integrated sol gel technique [22]. The substrate for the non-volatile random access memory will be made of Pt, Si, and GaAs. Using the Schlenk process and typical atmospheric conditions, the precursor was created. Precursor was used to create a polymeric solution, which was subsequently spun onto Pt, Si, and GaAs. The XRD, Ellipsometry, Rutherford Backscattering (RBS), and SEM are used to describe the prepared thin film. The stoichiometry of thin films is calculated using RBS. Ellipsometric analysis reveals a shift from 2.0 to 3.0 in the refractive index. PZT thin films were created by Thibault Dufay et al. [23] initially on Al, then transferred to polymer, and finally the Al was removed to leave the polymer. The PZT film is produced on a substrate using the spin coating method, starting with a precursor as the base material. After that, it is heated to a temperature of 650°C to form crystals. Following that, a polyurethane layer is applied on PZT and left exposed for two hours to improve the bonding between them. To have a fresh substrate on the polymer, polyethylene terephthalate (PET) is used. The adhesive is utilised to guard against structural PZT loss during etching. Al is eliminated using a solution of iron chloride. The surface-level PZT peaks are visible by XRD. By initially preparing the precursor solution with the sol gel technique, Xing Wang et al. [24] carried out the deposition. To get phase, surface, and interface information, respectively, XRD, Atomic Force Microscopy (AFM), and SEM are used. PZT layer on substrate is created by combining the spin coating and sol gel method. To produce the desired film, it is warmed, pyrolyzed, and then annealed. PZT film can also be produced using the RF magnetron sputtering technique, with an Ar flow rate of 90 sccm and a chamber pressure of 1.5 Pa. The pyrochlore phase is changed to the perovskite phase by raising the partial pressure of oxygen in O₂/Ar. According to AFM study, the ratio of 10/90 results in a compact and smooth surface. The same ratio produces a dense, homogeneous structure in SEM analysis.

In their study, Ali Shoghi et al. [25], discussed the production of a PZT thin film on FTO glass and determined the ideal heat treatment and sol parameter values. Using suitable ingredients to make PZT, the deep coating process is adopted. The best conditions for producing PZT on FTO glass include a slight pyrochlore phase at 600°C for 5 minutes, followed by surface morphology, roughness and homogeneity, and atomic bands and sol reactions. Jian He et al., [26] produced PZT thin film on a sapphire substrate, and it is then transferred using the spalling method to a flexible polyimide (PI) substrate. Ferroelectric characteristics are also examined, along with XRD and Raman spectroscopy, for the purpose of analysis. At 115°C, a solution of lead acetate trihydrate in acetic acid has been prepared. After waiting and screening for 24 hours, a PZT precursor is created at the end. It is spin coated for 6 seconds at 500 rpm and for 20 seconds at 3000 rpm. 10 minutes of pyrolyzing at 350°C for organic removal, followed by 10 minutes of annealing at 650°C for crystallisation. To get the film, this method is said to have been repeated 20 times. It is annealed once again for 30 minutes at 650°C. Magnetron sputtering is used to create an Au-100 nm/Cr-20 nm layer

on top of it. It has an electroplated Ni stressor layer on it. Next, the spalling process is carried out to get PZT. The PZT diffraction peaks on the Ni layer, the desired perovskite structure, clear visibility of interface with the Ni layer and sapphire has been obtained using XRD, Raman and SEM analysis respectively. Information about the optical characteristics of PZT and the impact of substrate choice was provided by Shagun Monga et al. [27]. The Sol Gel method is used for PZT preparation. The lead acetate trihydrate is taken after being dissolved in acetic acid and heated to 105°C. It is taken in the necessary ratio of zirconium n-propoxide and titanium isopropoxide. For obtaining the necessary precursor, the following options are used. To obtain the necessary viscosity and to lower surface tension, distilled water is utilised in an ultrasonic cleaner for homogeneous solutions. PZT thin films are created using the spin coating technique and are then added to the final prepared solution. On quartz, ITO glass, and corning glass, a thin layer of PZT was deposited. Its optical qualities are examined using XRD, Raman, and UV spectroscopy. When using different substrates, UV-vis spectroscopy reveals varying values of absorbance, reflectance, and transmittance. XRD reveals the perovskite structure at 650°C. Raman spectroscopy demonstrates the presence of the tetragonal phase. Xing Wang et al., [28] analyses the impact of the layer ratio on the thin film after fabricating O_3 on a Pt/Si/SiO₂ substrate. The precursor was prepared and analyzed using XRD and XPS. Figure.1 depicts the improved sol-gel process used to prepare PZT.

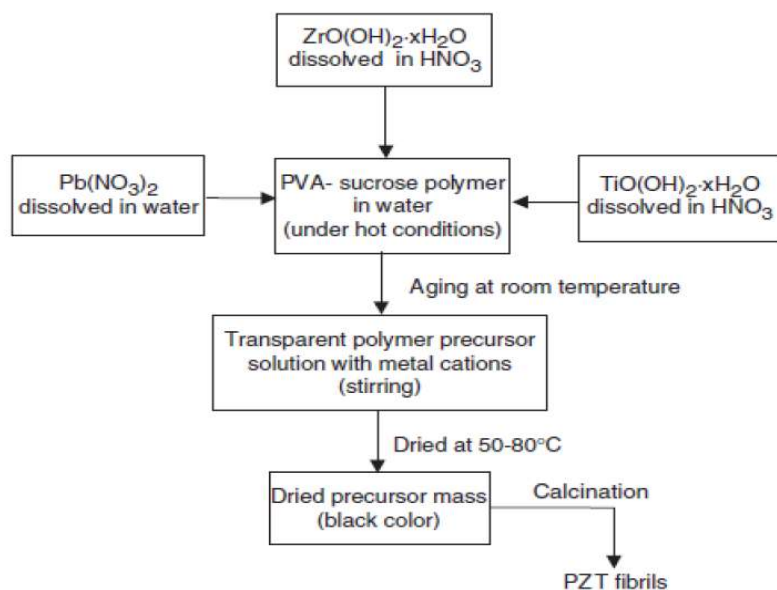


Figure.1 The improved sol-gel process used to prepare PZT.

2.2 Chemical Vapour Deposition (CVD)

Another well-known method for creating PZT thin films is CVD. The equipment utilised to create the film is the method's central component. Modern equipment is produced by many producers in the industry. These devices include bases for the substrate to be placed at various temperatures, chambers for the metalorganic solution to be placed, inert gases and oxygen in particular used at various pressures and temperatures as needed and chambers for sputtering to take place. The research that follows provides information on it.

S.T. Kim et al., [29] used Metal Organic (MO) source in conjunction with the electron cyclotron resonance plasma enhanced chemical vapour deposition technique to create PZT thin layer. Along

with oxygen, precursors such as lead β -diketonate, titanium iso-propoxide, and zirconium t-butoxide are also employed. Radiations at 2.4 GHz are produced by the magnetron. At 500°C, Pt (70 nm), Ti (100 nm), SiO₂(600 nm), and Si substrates are employed. Through XRD and wavelength dispersive X-Ray spectroscopy, the structure and chemical content are analysed. Chemical analysis reveals that the Zr/Ti ratio is 0.35, 0.57, and 0.63, while the resulting film thickness is 1100, 1000, and 900Å. For each of them, a successful single phase perovskite structure can be seen using XRD. The current density likewise rises as the Zr/Ti ratio increases. The maximum dielectric constant is found in Zr/Ti materials with a ratio of 0.57 and a thickness of 1000Å. At 470°C and 500°C, Su Ock Chung et al. [30] created a PZT thin film using ECR PECVD on a Pt/Ti/SiO₂/Si substrate. ECR is produced at 2.4 GHz by matching the microwave frequency to the electron frequency. Zirconium t-butoxide, titanium iso-propoxide, and dipivaloylmethanatolead are all employed as MO sources. The flow rates of Ar, O₂, and MO are all managed. Thermocouple regulates the temperature of the substrate. Only perovskite peaks are visible in XRD at 500°C. TEM also demonstrates that 470°C is insufficient for planer film. PZT thin film was created by Won Gyu Lee et al. [31] using the plasma enhanced chemical vapour deposition (PECVD) technique. Analyses were done on compositional distribution, surface morphologies, phase structures, and electrical characteristics using SEM and auger electron spectroscopy (AES). The fabrication uses Pb (C₂H₅)₄ (TEL, 5N purity), Zr(O-i-C₄H₉)₄ (ZTB, 5N purity), and Ti(O-i-C₃H₇)₄ (TTIP, 5N purity). Source delivery, a vacuum system, and a cold wall reaction chamber make up the device. The substrate is Pt/SiO₂/Si. A few settings are RF power of 20W at 13.56 MHz, substrate temperature of 250C, and chamber pressure of 300mTorr. The concentration of Pb at 650°C showed little variation after annealing under oxygen. According to XRD, the perovskite structure was created between 450 and 550°C. He et al. [32] created a PZT thin film together with the buffer layers of PbTiO₃(PT) and PbZrO₃(PZ), and XRD, SEM, and AFM were used to analyse the surface. As a substrate, Pt(1 1 1)/Ti/SiO₂/Si(1 0 0) is employed. The sources are lead acetate, tetrabutyl titanate, and zirconium nitrate, while the solvent is 2-methoxyethnaol and the catalyst is glacial aceticacid. The membrane filter on a syringe is used to inject the precursor onto the substrate. Sequentially employed in spin coating are the PT (PbTiO₃) solution, PZT-0.6 (PbZr_{0.6}Ti_{0.4}O₃) solution, PZT-0.5 (PbZr_{0.5}Ti_{0.5}O₃) solution, and PZT-0.4 (PbZr_{0.4}Ti_{0.6}O₃) solution. The film is annealed at 700°C after being heated on hot plates at 500°C. According to XRD, PT buffer layers are crucial for obtaining pure perovskite structure. The adhesion between the layers is quite clear and precise, as shown by SEM. According to AFM, as the thickness of the PT buffer layer increases, the PZT's roughness will change. According to Euk Hyun Kim et al. [33], Ph₄Pb, PbCl₂, and PhMgBr were used to create PZT using a novel Pb(IV) precursor. Due to step coverage, deposition rate homogeneity, and throughput (3-5), the liquid delivery metalorganic chemical deposition method is used in this particular work. In this procedure, the precursor is fed into a flash evaporator after being dissolved in the solvent. The film is placed onto the surface in this way. The employment of a precursor is crucial to this method's success. Numerous precursors are accessible, but a specific variety is needed that won't change its physical or chemical features while being prepared. Tetraethyllead (Pb(C₂H₅)₄) and Pb(thd)₂ (thd = 2,2,6,6-tetramethyl-3,5-heptadiketonate) are examples of precursors with improved resistivity at low temperatures and improved solution stability. The initial step was to manufacture tetraphenyl lead (Ph₄Pb) and diphenyl dibromo lead

(Pb_2PbBr_2) using the existing method with minimal modifications. Prior to depositing the PZT layer using MOCVD, the TiO_2 layer on the Si wafer is first prepared. For the creation of PZT, a Pb precursor was dissolved with $\text{Ti}(\text{OiPr})_4$ and $\text{Zr}(\text{OPr})_4$ in butyl ether and tetraglyme. Thus, in Chemical Vapor Deposition method, high quality thin film is formed and the deposition temperature requirement varies 300°C to 800°C .

2.3 Sputtering Method

The fundamental idea behind the technique is to attack the target material with inert gas ions at a predetermined frequency to cause metal atoms to sputter. These atoms were deposited at a close distance on the substrate that was in front of it. The entire procedure was carried out in a sealed chamber with a controlled temperature and pressure. To keep the secondary radiation under control, cooling water is pumped across the target material. This approach underwent development during the ensuing years.

The PZT thin film was produced by P. Muralt et al. [35] by sputtering the sol gel and used in increasing the speed of the micromotor (sensors and actuators). The sol-gel approach makes use of $\text{Pb}(\text{C}_2\text{H}_4\text{O}_2)_2 \cdot 3\text{H}_2\text{O}$, $\text{Zr}(\text{OCH}_2\text{CH}_2\text{CH}_3)_4$, and $\text{Ti}[\text{OCH}(\text{CH}_3)_2]_4$ precursors. The sputtering procedure results in an orientation (100), when three magnetrons are used with a metal target. The results show that the dielectric constants at 100 Hz for the sol gel and sputtering techniques are 950 and 610, respectively. Additionally, the dielectric losses range from 3 to 4.5 percent at 100 Hz. T Hata et al. produced PZT thin film using reactive sputtering. The objective is PbO and ZrTi alloy pellets. It's also recommended to use Zr, Ti, and PbO together. XRD is utilised during analysis. R. F. diode sputtering is a non-magnetron technology used in this work. The target's chemical formula is $(\text{ZrTi} + x\text{PbO})$, where x is the target's PbO's surface area ratio/($\text{SPbO} + \text{SZrTi}$). PbO and ZrTi have total surface areas that are SPbO and SZrTi , respectively. The gases Ar and O_2 are used during the operation. At 190°C , the glass substrate is used. The reliance of the deposition rate on the O_2/Ar ratio and combination of x led to the formation of the solid and clear film. According to XRD, pure perovskite PZT is formed at 450°C and $x = 30\%$. Xin-Shan Li et al. [35] produced a PZT thin film with a single (111) orientation perovskite phase using the sputtering process. We've looked into annealing and deposition temperatures. The FTS-1CB (Osaka Vacuum) instrument is used. Target distance is lowered from 140 mm to 130 mm via the substare. Temperature range: $200\text{-}500^\circ\text{C}$; target: $\text{Pb}_{1.2}\text{Zr}_{0.58}\text{Ti}_{0.42}\text{O}_x$; substrate: Pt/Ti/SiO₂/Si. 120 minutes of RF 700 W at a 4:1 O_2/Ar ratio. At $453\text{-}656^\circ\text{C}$, thin film is annealed. According to XRD, perovskite structure was created by deposition at low temperature (360°C) and annealing at 606°C . According to surface information, the favourable circumstances for PZT thin film are provided by the lower annealing temperature and lower deposition temperature. Others, like Y.C. Lin [36] On a Pt/Ti/SiO₂/Si substrate, PZT thin film was produced utilising the pulsed DC magnetron method. A superior crystalline structure is produced by the examination of the pulse frequency, duty cycle, and gases ratio. When employed as a target, the metallic alloy has a Pb:Zr:Ti ratio of 46:22:22. It uses bipolar pulse power with a duty cycle of between 75 and 95 percent and runs between 10 kHz and 100 kHz. The temperatures for deposition and annealing are 100°C and 750°C , respectively. The target was 60mm away from the substrate when sputtering was done, with a working pressure of 5×10^{-3} mbar and a background pressure of

5×10^{-6} mbar. Annealing occurs at 200°C/s. According to the research, even when the sputtering strength and temperature increase, the surface doesn't become unstable or melt. When pulse rate is lowered, perovskite phase occurs with an O₂/Ar ratio of 1:1 and a duty cycle of 90%. When pulse width is raised, linear deposition happens more quickly. Production of PZT thin layer on Pt/TiO₂/SiO₂/Si was studied using RF magnetron sputtering by K.K. Maurya et al [37]. XRD and cross sectional transmission electron microscopy (XTEM) were employed for the analysis. An RF magnetron made by Inostek Inc. in Korea is used for sputtering. It operates at 2750 C using a substrate made of Pt (150 nm), TiO₂ (20 nm), SiO₂ (300 nm), and Si (675 nm). Pb_{1.1}(Zr_{0.52}Ti_{0.48})O₃ ceramic was employed as the target PZT. 80/20 Ar/O₂ ratio, 20 rpm substrate rotation speed, and 5×10^{-4} Pa base pressure are all acceptable values. The separation between the substrate and the target is 0.110m. The annealing was done PZT at 650°C. At a sputtering pressure of 4.5 Pa for the thickness range of 500 nm, the perovskite structure is produced. XTEM reveals the nanoscale porosity of the interface. Sujin Choi et al. [38] used RF magnetron sputtering to produce PZT. Ar-ions were used to create a depth profile in order to gather surface information. Various O₂ ratios are used in order to acquire the surface profile using XRD. The target is n-type Si, and after being cleaned with acetone, dried with nitrogen, and set on a substrate holder, it is prepared for use. The substrate is heated to 573 K and spun at 5 rpm during the sputtering procedure. 2-inch-thick Pb, Zr, and Ti metals are used for co-sputtering, and Ar and O₂ are mixed throughout the process. The working pressure is 4.5×10^{-6} Torr, while the base pressure is 2.6×10^{-6} Torr. By adjusting the RF power supplied at Pb, Zr, and Ti, the ideal ratio in PZT thin film is preserved. The high conductivity and thickness of the PZT layer are maintained when the O₂ ratio is on the lowering side. As the ratio increases, thickness and conductivity both drop. As the O₂ ratio increases, the PZT formed, according to XRD, becomes more stable. Pb concentration is said to increase as the O₂ ratio increases. Mitra Akhtari Zavareh et al. [39] produced PZT thin films from strontium titanate and strontium ruthenate using powder magnetron sputtering (PMS). XPS, FESEM, and XRD investigation are also conducted. Ar/O₂ is maintained at a 20:1 ratio, at pressure is 0.5 Pa. Sputtering powder is used, and a deposition rate of 0.2–0.3 μm/s is accomplished at 80–100 W. The substrate's distance from the target is 8 cm. The study shows that this method can produce a single crystal with the bulk's characteristics, and offer better surface morphology and composition, respectively.

2.4 Pulsed Laser Deposition (PLD)

In this technique, the laser beam is focussed on the target material taken inside of a vacuum chamber. The target receives enough energy to emit ions, which are then accelerated toward the substrate material resulting the deposition of thin film. Some of the studies undertaken using this technique are as follows.

L'h. Hamedi et al. [40] used the pulsed laser deposition method to produce PZT thin film. The three methods of characterisation are XRD, ECP and RHEED. The substance is SrTiO₃. The following are the PLD process's operational parameters: Target substrate distance is 40 mm, pressure is 0.5 mb, substrate temperature is 500–560 °C, frequency is 2 Hz, and energy is 3.5 J/cm². PbO, ZrO₂, and TiO₂ powdered together to form a target. This results a crystalline structure on a SrTiO₃ substrate. Javad R. Gatabi et al. [41] studied the effects of oxygen pressure and laser fluence on PZT properties.

The FEI Helios Lab 400 scanning electron microscope with EDAX capabilities and the BRUKER D8 ADVANCE XRD apparatus are both used in the technique. $\text{Pb}_{1.1}\text{Zr}_{0.53}\text{Ti}_{0.47}\text{O}_3$, a compound made from plasma components, is used for the PZT target. Under pressure of 1×10^{-10} mbar in a chamber, the deposition occurs on a substrate with a thickness of 2 in. A 248 KrF excimer laser is used in the technique, and oxygen is added using a mass flow controller. PZT would not have enough Pb and Zr because of the divergence of the target. According to EDAX, laser fluence caused the Pb concentration to fluctuate abruptly whereas the Ti content varied slightly. The link between structure and dielectric/pyroelectric properties in up- and down-graded PZT was examined by M. Botea et al. [42]. Pulsed laser deposition is used to produce multilayer PZT (PLD). used XRD and TEM to analyse structures. Hysteresis, current voltage, and capacitor voltage measurements are made to determine the electric and ferroelectric properties. examining the relationship between the pyroelectric properties and frequency.

2.5 Chemical Solution Deposition (CSD)

The development of CSD techniques as a practical, affordable non-vacuum method for PZT fabrication The variety of material systems that can be prepared has rapidly increased as a result of global research efforts, and CSD has now developed into a workable substitute technique for PZT films. The advancement in knowledge of precursor solution chemistry, film-forming principles, and the nucleation and growth of crystalline thin films from amorphous pyrolyzed precursors is at least partially responsible for the advancement in film quality and functional properties.

Information regarding enhancing the thermal stability of PZT thin film using Dysprosium doping was presented by Jongchul Jeon et al. [43]. The change in the MPB at the higher temperature is the primary problem with the PZT structure. To solve the problem, the PZT with Dy doping is created using the CSD process. Lead acetate trihydrate, zirconium n-propoxide, and titanium (IV) isopropoxide are among the precursors for CSD. Titanium (IV) isopropoxide and zirconium n-propoxide were uniformly mixed. To alter it, acetic acid was added. A stable solution was created by adding n-propanol and distilled water. The precursor was then made ready by the addition of dysprosium (III) nitrate hydrate. The substrate is a silicon wafer with a Pt covering. After being spin coated on the substrate at 250°C , the precursor is pyrolyzed at 400°C , then crystallised at 650°C . The desired film thickness may only be obtained through repeated spin coating. The thermal stability was increased by 23%. The $\text{Pb}(\text{Zr}_{0.53}\text{Ti}_{0.47})\text{O}_3$ PZT thin film was produced by Nick Godard and others [44], and the silver electrode was then printed on top using an ink jet printer. The polarisation and piezoelectric response are measured. The data were contrasted using a sputtered Pt electrode. Platinumized silicon ($\text{Si}/\text{SiO}_2/\text{TiO}_x/\text{Pt}$) was used as the substrate to create PZT film.

2.6 Ink Jet Method

The most cutting-edge and promising method for creating expansive and flexible thin films is inkjet printing. The inkjet printing method makes it simple to customise a wide range of complex structures. Itziar Fraile et al. [45] used an inkjet printing technique to create PZT film at room temperature. Organometallic compounds are produced using the metals titanium, lead, and zirconium. It must be combined with water vapour to create sol. Stainless steel substrate is used in PZT nanoparticle ink and stainless steel substrate along with alumina are used in PZT precursor-based ink. 43V piezo-

potential and 10^3 Hz frequency are applied to create drops. The sample was then heated to 40°C for half an hour to eliminate any vehicles prior to sintering. Bathurst et al., [46] utilized a printed PZT thin layer with the ultrasonic sensor. Commercially available solvents such as 1-butanol, 1,2-propanediol, 2-methoxyethanol, and hydrous 2-methoxyethanol were used to create the jettable ink. The created solution maintains the required levels of density, surface tension, and viscosity. The size of the dispersion is held constant. The piezoelectric micro-machined ultrasonic transducer was used to match the PZT's performance. It was discovered that the coupling coefficient ranged from -75 to -95 pC/N. The relative permittivity ranged from 750 to 890. This PZT thin film was used in MEMS applications.

3. Conclusion

The Sol-gel method is simple, based upon the solutions, require low deposition temperature and higher annealing temperature. In Chemical Vapor Deposition method, high quality thin film is formed and the deposition temperature requirement varies 300°C to 800°C . In sputtering, the deposition temperature as well as annealing temperature, both are low. The RF, DC and Microwave sputtering are possible. The perovskite structure is better oriented. Inert gases are utilized in thin film deposition. PLD uses high powered laser pulses at higher substrate temperature. The deposition temperature has wider range and the thin film has good crystalline structure. CSD is a solution based method with better MPB and higher thermal stability while Ink jet produce thin film with uniform solute distribution and better control over the morphology. The cost of thin film deposition is comparatively low.

Acknowledgement

The authors are very grateful to SGT University authorities for providing support and encouragement during the course of this work.

References

1. Y. H. Bing and Z. G. Ye, "Piezo- and ferroelectric $(1-x)\text{Pb}(\text{Sc}_{1/2}\text{Nb}_{1/2})\text{O}_3-x\text{PbTiO}_3$ solid solution system," *Handb. Adv. Dielectr. Piezoelectric Ferroelectr. Mater. Synth. Prop. Appl.*, pp. 173–204, 2008.
2. D. Bochenek, P. Niemiec, R. Skulski, M. Adamczyk, and D. Brzezińska, "Electrophysical properties of the multicomponent PBZT-type ceramics doped by Sn^{4+} ," *J. Electroceramics*, vol. 42, no. 1–2, pp. 17–30, 2019.
3. Z. J. Wang, Y. Aoki, L. J. Yan, H. Kokawa, and R. Maeda, "Crystal structure and microstructure of lead zirconate titanate (PZT) thin films with various Zr/Ti ratios grown by hybrid processing," *J. Cryst. Growth*, vol. 267, no. 1–2, pp. 92–99, 2004.
4. B. Gao *et al.*, "Unexpectedly high piezoelectric response in Sm-doped PZT ceramics beyond the morphotropic phase boundary region," *J. Alloys Compd.*, vol. 836, p. 155474, 2020.
5. N. Kumari *et al.*, "Multifunctional behavior of acceptor-cation substitution at higher doping concentration in PZT ceramics," *Ceram. Int.*, vol. 45, no. 10, pp. 12716–12726, 2019.
6. S. Samanta, V. Sankaranarayanan, and K. Sethupathi, "Effect of Successive Multiple Doping of La, Nb and Fe on Structure and Lattice Vibration of MPB PZT," *Mater. Today Proc.*, vol. 5, no. 14, pp. 27919–27927, 2018.

7. J. Li, P. Li, G. Zhang, J. Yu, Y. Wu, and X. Wen, "The thickness effect of Bi_{3.25}La_{0.75}Ti₃O₁₂ buffer layer in PbZr_{0.58}Ti_{0.42}O₃/Bi_{3.25}La_{0.75}Ti₃O₁₂ (PZT/BLT) multilayered ferroelectric thin films," *Thin Solid Films*, vol. 519, no. 18, pp. 6021–6025, 2011.
8. T. Kobayashi, M. Ichiki, J. Tsauro, and R. Maeda, "Effect of multi-coating process on the orientation and microstructure of lead zirconate titanate (PZT) thin films derived by chemical solution deposition," *Thin Solid Films*, vol. 489, no. 1–2, pp. 74–78, 2005.
9. O. Sugiyama, K. Murakami, and S. Kaneko, "XPS analysis of surface layer of sol-gel-derived PZT thin films," *J. Eur. Ceram. Soc.*, vol. 24, no. 6, pp. 1157–1160, 2004.
10. N. Choudhary, D. K. Kharat, and D. Kaur, "Surface modification of NiTi/PZT heterostructure thin films using various protective layers for potential MEMS applications," *Surf. Coatings Technol.*, vol. 206, no. 7, pp. 1735–1743, 2011.
11. H. Sui, H. Sun, X. Liu, D. Zhou, and R. Xu, "Ferroelectric and dielectric behaviors of sol-gel derived perovskite PMN-PT/PZT heterostructures via compositional development: An interface-dependent study," *J. Eur. Ceram. Soc.*, vol. 38, no. 16, pp. 5382–5387, 2018.
12. M. V. Kamenshchikov, A. V. Solnyshkin, and I. P. Pronin, "Dielectric response of capacitor structures based on PZT annealed at different temperatures," *Phys. Lett. Sect. A Gen. At. Solid State Phys.*, vol. 380, no. 47, pp. 4003–4007, 2016.
13. Z. Jiao, X. Wan, H. Guo, J. Wang, B. Zhao, and M. Wu, "The charge storage characteristics of PZT nanocrystal thin film," *Ultramicroscopy*, vol. 108, no. 10, pp. 1371–1373, 2008.
14. S. Yoshida, H. Hanzawa, K. Wasa, and S. Tanaka, "Enhanced curie temperature and high heat resistivity of PMnN-PZT monocrystalline thin film on Si," *Sensors Actuators, A Phys.*, vol. 251, pp. 100–107, 2016.
15. T. Zhang, W. Li, Y. Yu, M. Wang, J. He, and W. Fei, "Giant electrocaloric effect in compositionally graded PZT multilayer thin films," *J. Alloys Compd.*, vol. 731, pp. 489–495, 2018.
16. A. Bose and M. Sreemany, "Influence of processing conditions on the structure, composition and ferroelectric properties of sputtered PZT thin films on Ti-substrates," *Appl. Surf. Sci.*, vol. 289, pp. 551–559, 2014.
17. R. Wang, E. Tang, G. Yang, and Y. Han, "Experimental research on dynamic response of PZT-5H under impact load," *Ceram. Int.*, vol. 46, no. 3, pp. 2868–2876, 2020.
18. C. Othmani, H. Zhang, and C. Lü, "Effects of initial stresses on guided wave propagation in Preparation, Characterization, Properties and Applications of Lead Zirconate Titanate Thin Films: A Review; multilayered PZT-4/PZT-5A composites: A polynomial expansion approach," *Appl. Math. Model.*, vol. 78, pp. 148–168, 2020.
19. A. Wu, I. M. Miranda Salvado, P. M. Vilarinho, and J. L. Baptista, "Processing and seeding effects on crystallisation of PZT thin films from sol-gel method," *J. Eur. Ceram. Soc.*, vol. 17, no. 12, pp. 1443–1452, 1997.
20. O. Babushkin, T. Lindbäck, K. Brooks, and N. Setter, "PZT phase formation monitored by high-temperature X-ray diffractometry," *J. Eur. Ceram. Soc.*, vol. 17, no. 6, pp. 813–818, 1997.
21. Q. Zou, S. Nourbakhsh, and J. Kim, "Novel polyol-derived sol route for fabrication of PZT thin ferroelectric films," *Mater. Lett.*, vol. 40, no. 5, pp. 240–245, 1999.

22. R. Zuleeg, "Integrated Solgel M Thin-Films on Pt, Si, and Gaas for Non-Volatile Memory Applications," *Ferroelectrics*, vol. 108, no. 1, pp. 37–46, 1990.
23. T. Dufay, B. Guiffard, R. Seveno, S. Ginestar, and J. C. Thomas, "Flexible PZT thin film transferred on polymer substrate," *Surf. Coatings Technol.*, vol. 343, pp. 148–152, 2018.
24. X. Wang *et al.*, "Effect of oxygen partial pressure on crystal quality and electrical properties of RF sputtered PZT thin films under the fixed Ar flow and sputtering pressure," *Vacuum*, vol. 172, p. 109041, 2020.
25. A. Shoghi, H. Abdizadeh, A. Shakeri, and M. R. Golobostanfard, "Sol–gel synthesis of PZT thin films on FTO glass substrates for electro-optic devices," *J. Sol-Gel Sci. Technol.*, vol. 93, no. 3, pp. 623–632, 2020.
26. J. He *et al.*, "Flexible heterogeneous integration of PZT film by controlled spalling technology," *J. Alloys Compd.*, vol. 807, p. 151696, 2019.
27. S. Monga, S. Tomar, P. M. Vilarinho, and A. Singh, "Effect of substrates on optical properties of ferroelectric PZT (52/48) thin films," *Mater. Today Proc.*, no. xxxx, 2020.
28. X. Wang *et al.*, "Enhancements of the electrical properties in $\text{Pb}_{1.25}(\text{Zr}_{0.52}, \text{Ti}_{0.48})\text{O}_3/\text{Pb}_{1.1}(\text{Zr}_{0.52}, \text{Ti}_{0.48})\text{O}_3$ ferroelectric multilayered thin films," *Mater. Chem. Phys.*, vol. 241, no. July 2019, p. 122396, 2020.
29. S. T. Kim, J. W. Kim, S. W. Jung, J. S. Shin, S. T. Ahn, and W. J. Lee, "Electrical properties of PZT thin films deposited by electron cyclotron resonance plasma enhanced chemical vapor deposition," *Mater. Chem. Phys.*, vol. 45, no. 2, pp. 155–158, 1996.
30. S. O. Chung, J. W. Kim, S. T. Kim, G. H. Kim, and W. J. Lee, "Microstructure and electric properties of the PZT thin films fabricated by ECR PECVD: The effects of an interfacial layer and rapid thermal annealing," *Mater. Chem. Phys.*, vol. 53, no. 1, pp. 60–66, 1998.
31. W. G. Lee and Y. J. Kwon, "Preparation of ferroelectric PZT thin films by plasma enhanced chemical vapor deposition using metalorganic precursors," *J. Ind. Eng. Chem.*, vol. 14, no. 1, pp. 89–93, 2008.
32. G. He, Y. Zhang, C. Peng, and X. Li, "Surface morphology and ferroelectric properties of compositional gradient PZT thin films prepared by chemical solution deposition process," *Appl. Surf. Sci.*, vol. 283, pp. 532–536, 2013.
33. E. H. Kim, C. W. Moon, J. G. Lee, M. S. Lah, and S. M. Koo, "Synthesis and characterization of lead (IV) precursors and their conversion to PZT materials through a CVD process," *Polyhedron*, vol. 177, p. 114270, 2020.
34. P. Muralt *et al.*, "Fabrication and characterization of PZT thin-film vibrators for micromotors," *Sensors and Actuators, A: Physical*, vol. 48, no. 2, pp. 157–165, 1995.
35. X. S. Li, K. Yamashita, T. Tanaka, Y. Suzuki, and M. Okuyama, "Structural and electrical properties of highly oriented $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ thin films deposited by facing target sputtering," *Sensors Actuators, A Phys.*, vol. 82, no. 1, pp. 265–269, 2000.
36. Y. C. Lin, H. A. Chuang, and J. H. Shen, "PZT thin film preparation by pulsed DC magnetron sputtering," *Vacuum*, vol. 83, no. 6, pp. 921–926, 2009.

37. K. K. Maurya, S. K. Halder, S. Sen, A. Bose, and S. Bysakh, "High resolution X-ray and electron microscopy characterization of PZT thin films prepared by RF magnetron sputtering," *Appl. Surf. Sci.*, vol. 313, pp. 196–206, 2014.
38. S. Choi, J. Park, J. Kang, A. T. C. Johnson, and Y. C. Kang, "Surface characterization of PZT thin films obtained at various O₂ gas ratios," *Vacuum*, vol. 128, pp. 234–239, 2016.
39. M. Akhtari Zavareh, B. Abd Razak, M. H. Bin Wahab, B. T. Goh, R. Mahmoodian, and K. Wasa, "Fabrication of Pb(Zr,Ti)O₃ thin films utilizing unconventional powder magnetron sputtering (PMS)," *Ceram. Int.*, vol. 46, no. 2, pp. 1281–1296, 2020.
40. L. H. Hamed, M. Guilloux-Viry, A. Perrin, and M. H. Cherkani, "On the epitaxial growth of PZT films by pulsed laser deposition," *Ann. Chim. Sci. des Mater.*, vol. 23, no. 1–2, pp. 377–380, 1998.
41. J. R. Gatabi *et al.*, "Tuning electrical properties of PZT film deposited by Pulsed Laser Deposition," *Ceram. Int.*, vol. 43, no. 8, pp. 6008–6012, 2017.
42. M. Botea *et al.*, "Structural, electric and pyroelectric properties of up and down graded PZT multilayers," *Curr. Appl. Phys.*, vol. 19, no. 7, pp. 804–810, 2019.
43. J. Jeon and K. H. Kim, "Evolution of domain structure in PbZr_{0.52}Ti_{0.48}O₃ thin film by adding dysprosium," *Thin Solid Films*, vol. 701, p. 137940, 2020.
44. N. Godard, S. Glinsek, and E. Defay, "Inkjet-printed silver as alternative top electrode for lead zirconate titanate thin films," *J. Alloys Compd.*, vol. 783, pp. 801–805, 2019.
45. I. Fraile, M. Gabilondo, N. Burgos, M. Azcona, and F. Castro, "Laser sintered ceramic coatings of PZT nanoparticles deposited by Inkjet Printing on metallic and ceramic substrates," *Ceram. Int.*, vol. 44, no. 13, pp. 15603–15610, 2018.
46. S. P. Bathurst *et al.*, "Printing of uniform PZT thin films for MEMS applications," *CIRP Annals* Volume 62, Issue 1, 2013, Pages 227-230