

# We are IntechOpen, the world's leading publisher of Open Access books Built by scientists, for scientists

6,900

Open access books available

186,000

International authors and editors

200M

Downloads

Our authors are among the

154

Countries delivered to

TOP 1%

most cited scientists

12.2%

Contributors from top 500 universities



WEB OF SCIENCE™

Selection of our books indexed in the Book Citation Index  
in Web of Science™ Core Collection (BKCI)

Interested in publishing with us?  
Contact [book.department@intechopen.com](mailto:book.department@intechopen.com)

Numbers displayed above are based on latest data collected.  
For more information visit [www.intechopen.com](http://www.intechopen.com)



# Fabrication of Dielectric Thick Films by Electrophoretic Deposition and Their Characterization

*Vineetha P and Venkata Saravanan K*

## Abstract

In this chapter, we have discussed about the fabrication of  $\text{MgTiO}_3$  and  $(\text{K}_{0.5}\text{Na}_{0.5})(\text{Nb}_{0.7}\text{Ta}_{0.3})\text{O}_3$  (KNNT) thick films via EPD, including the preparation of stable suspension, which is the essential requirement for EPD. Acetone and isopropyl alcohol are used as solvents for  $\text{MgTiO}_3$  and  $(\text{K}_{0.5}\text{Na}_{0.5})(\text{Nb}_{0.7}\text{Ta}_{0.3})\text{O}_3$ , respectively. In both the cases triethanolamine (TEA) is used as the surfactant. It was observed that with increase in applied voltage and deposition time, the deposition of particle on the substrate has increased. For KNNT particle the optimum voltage and time to get quality film was 100 V and 10 min respectively. The surface morphology and film thickness was analyzed from SEM images. The thickness of KNNT and  $\text{MgTiO}_3$  films was found to be ~ 32 and 18–20  $\mu\text{m}$  (for film sintered at  $900^\circ\text{C}$ ) respectively. In the case of  $\text{MgTiO}_3$  thick films, the sintering temperature plays an important role in the quality of films since it affects the density and grain growth. An increase in the sintering temperature, from  $800$ – $900^\circ\text{C}$ , resulted in increase in the dielectric constant and reduced loss. The dielectric values of the KNNT and  $\text{MgTiO}_3$  thick film at 1 MHz was 320 and 18.3 respectively.

**Keywords:** electrophoretic deposition, dielectric thick film,  $\text{MgTiO}_3$ ,  $\text{K}_{0.5}\text{Na}_{0.5}\text{Nb}_{0.7}\text{Ta}_{0.3}\text{O}_3$ , dielectric constant

## 1. Introduction

Miniaturization of electrical components have inherent advantages like less space usage, low cost, high speed and easy transportation owing to its size and light weight. In most industrial applications of piezoelectric materials, crack-free thick or thin films with a uniform microstructure are required. Lapping down a bulk material to the desired thickness is a tedious and time consuming process and thus corresponding thick films are apparently a promising alternative. Thus better understanding of the fabrication methods is necessary to make future generations of miniaturized components/materials. There are many coating techniques available to develop the materials in nano or micro sizes, which are either expensive or follow a tedious processing deposition methodology. On the other hand, preparation of thick film by the electrophoretic deposition has attained much attention owing to its high versatility, cost effectiveness, simple equipment design and process requirements [1]. Generally, thick films of any solid can be deposited by EPD provided the solid assumes its form as powder or a colloidal suspension [2]. In the following sections

we have included in details the preparation of stable suspensions of KNNT and  $\text{MgTiO}_3$ . EPD comprises of two steps; the first step includes the preparation of a stable suspension of the material to be deposited and movement of the particle towards the oppositely charged electrode and the second step involves the deposition of the particle on the substrate surface to form a film. After the first two deposition steps, a heat-treatment step is normally needed to further densify the deposited films and to eliminate porosity in the film [3, 4]. Compared to other advanced shaping techniques, EPD offers easy control of the thickness and morphology of a deposited film through simple adjustment of the deposition time and applied potential [5].

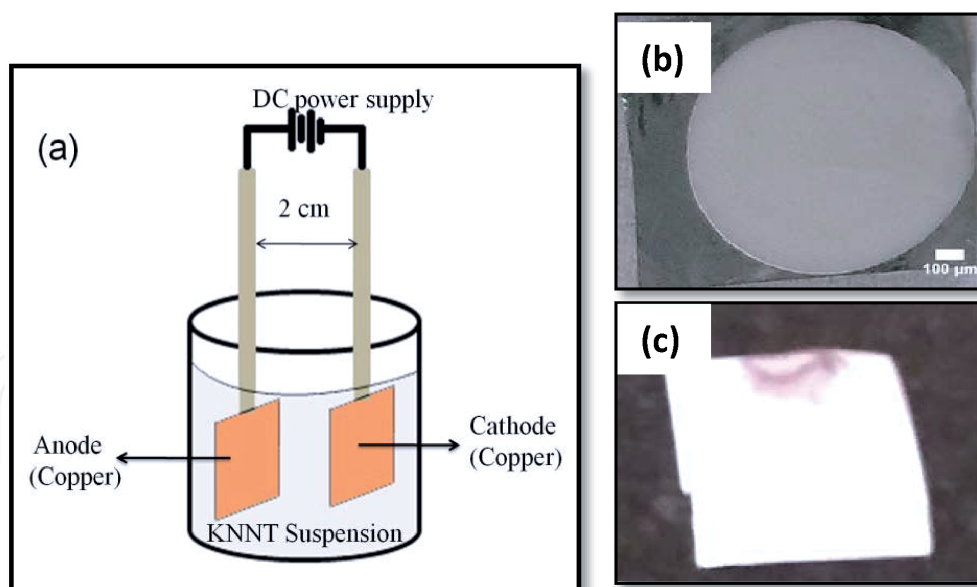
Considering the environmental and health risks of lead-based materials, lead-based materials in the electronics industry are now being replaced with lead-free materials. In lead-free ceramic materials, potassium sodium niobate-based materials have received much attention due to its superior piezoelectric properties and high Curie temperature [6]. A piezoelectric value of 80 pC/N has been reported for pure KNN ceramics prepared by solid state reaction route method [7]. However compared to the pure KNN material, doped materials have got high piezoelectric as well as ferroelectric properties. So far, studies have been carried out to improve the piezoelectric properties of KNN ceramics by partial substitutions of Li at A-site and/or Ta at B-site, [8, 9]. Tantalum doping in the B site has a major effect on the materials properties and a piezoelectric value of 205 pC/N has been reported for  $(\text{K}_{0.5}\text{Na}_{0.5})(\text{Nb}_{0.7}\text{Ta}_{0.3})\text{O}_3$  (KNNT) ceramics [10]. Even though the bulk ceramics such as KNNT and  $\text{MgTiO}_3$  based materials have good electronic properties [10], miniaturizations of electronic devices demand smaller and lighter components with properties comparable to that of bulk material. Potassium Sodium Niobate (KNN) based layers are being employed in applications such as high frequency transducer and in ultra sound imaging [11, 12] and magnesium titanate ( $\text{MgTiO}_3$ ) based materials are used in many applications such as high density capacitors, resonators and filters, in wireless communication systems, global positioning system, antennas for communications, radar and broadcasting satellite, operating at micro-wave and millimeter-wave frequencies [13]. The versatile use  $\text{MgTiO}_3$  is due to its high quality factor ( $Q_f = 160,000$  GHz,  $Q_f = 1/\tan\delta$ ), low dielectric constant ( $\epsilon_r \sim 17$ ) and negative temperature coefficient of resonant frequency ( $\tau_f = -50$  ppm/ $^\circ\text{C}$ ) [14].

## 2. Experimental

### 2.1 Stable suspension

In order to prepare a stable suspension of any ceramic, it is desirable to have nano grain sized powders, hence the precursor for  $\text{MgTiO}_3$ , which contains excess of MgO ( $\text{Mg}_2\text{TiO}_4$ ), prepared by Solar Physical Vapor Deposition (SPVD) was used. The details of the SPVD process could be found elsewhere [15]. The main advantage of SPVD is direct synthesis of nanopowders without any contamination. Moreover, the process is eco-friendly since it uses solar energy which is available abundantly in nature. The KNNT powder required for the EPD was prepared by conventional solid state reaction technique [16, 17]. The KNNT and  $\text{Mg}_2\text{TiO}_4$  powders were thoroughly mixed in isopropyl alcohol and acetone, respectively, followed by ultra-sonication and rigorous magnetic stirring. Ultra sonication and magnetic stirring was performed to get homogeneous and well dispersed suspensions.

In both the cases, Triethanolamine (TEA), which is an organic base, was added as the dispersant to increase the zeta potential and colloidal stability of particles in the suspension. It is known that  $\text{H}^+\text{TEA}$  species generated by proton capturing from the isopropyl alcohol/acetone are chemically adsorbed on the ceramic nanoparticles via



**Figure 1.**  
 (a) Schematic representation of customized EPD cell (b) optical micrograph of  $\text{MgTiO}_3$  thick film [17], and  
 (c) KNNT thick film.

strong hydrogen bonding thereby enhancing their zeta potential and hence the colloidal stability. Also, the addition of TEA is resulted in increase of pH (7 to 9 for KNNT and 6 to 8.2 for  $\text{Mg}_2\text{TiO}_4$ ). The charging mechanism of KNNT in TEA added isopropyl alcohol will be discussed in the coming section. Care is taken while adding TEA since the excess amounts of TEA may lead to the sedimentation of the particle [18].

## 2.2 Electrophoretic deposition

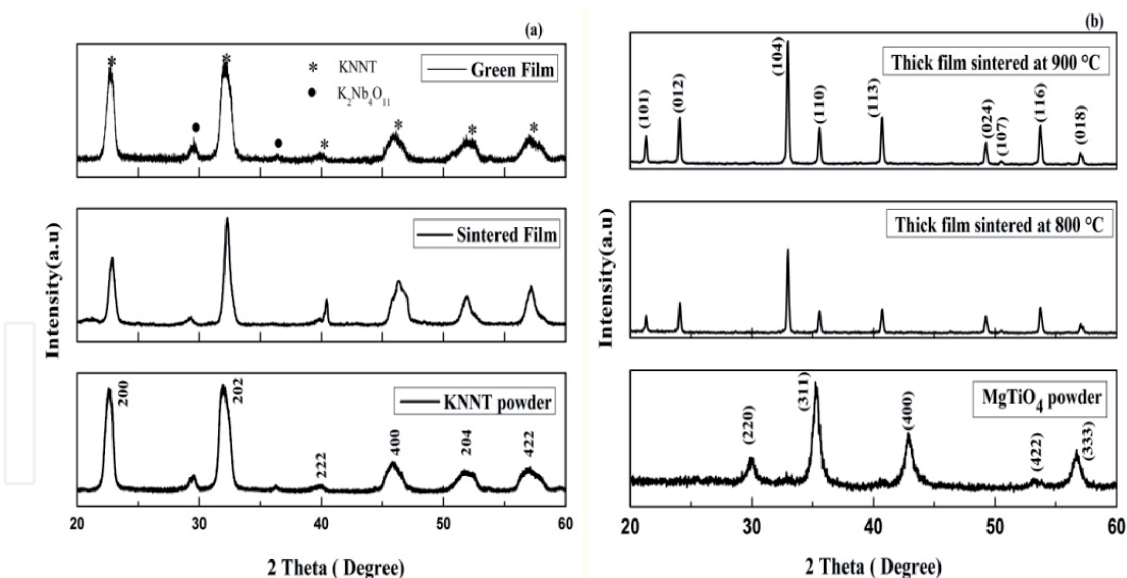
The EPD was performed at room temperature using a customized EPD setup/cell (**Figure 1a**) and a voltage source (Keithely 2636 B). Pre-cleaned platinum (Pt) plates and foils are used as the electrodes. One of the peculiarities of EPD is that electrodes with various shapes and dimension can be used for deposition. The distance between the two working electrodes was kept constant at 2 cm. The deposition of KNNT was carried out at 100 V for 10 min and  $\text{Mg}_2\text{TiO}_4$  at 60 V for 1 min. The picture of thick films obtained is shown in **Figure 1**. The deposition of KNNT as well as  $\text{MgTiO}_3$  particles happened in the cathode, indicating the negative charge of particles in the suspension. The deposition of particle at cathode is known as cathodic deposition. After deposition the KNNT films were annealed in an oven at 200°C for 1 h and  $\text{Mg}_2\text{TiO}_4$  films were dried in desiccators to evaporate the solvent. Then the KNNT films are sintered at 1000°C for 2 hr. For better compaction the dried  $\text{Mg}_2\text{TiO}_4$  films were isostatically pressed at 150 MPa for 5 minutes before carrying out the sintering process in air at 800, 900 and 1000°C for 15 minutes in a box furnace.

X-Ray diffraction technique was used for the phase analysis of the films and the microstructures were characterized using a scanning electron microscope (SEM). Later dielectric measurements were carried-out on all the thick films for which metallic top electrodes were sputter deposited through shadow mask.

## 3. XRD analysis

XRD patterns of KNNT powder, green and sintered KNNT thick films are shown in **Figure 2a** and the XRD patterns of the SPVD obtained  $\text{MgTiO}_3$





**Figure 2.**

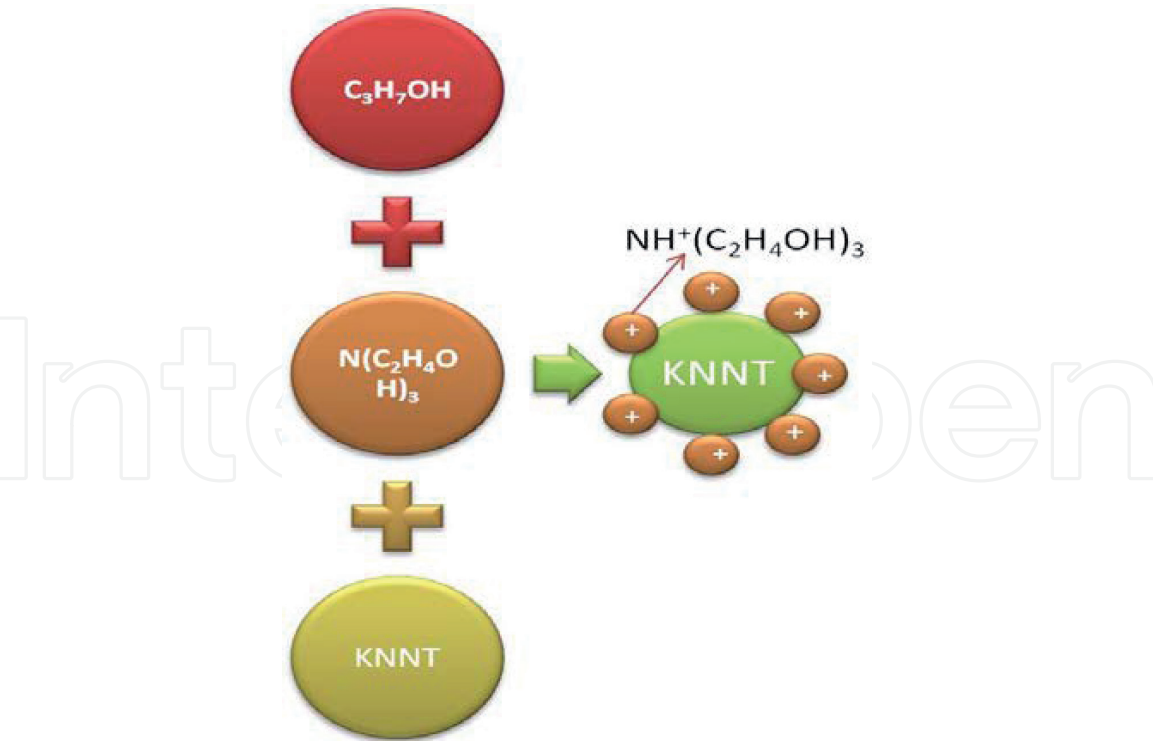
(a) XRD patterns of KNNT powder, green film and sintered thick films (b) XRD patterns of  $Mg_2TiO_4$  nanopowder, EPD films sintered at 800 (15 min) and 900°C (15 min) [17].

nanoparticle and the thick films sintered at different temperatures is shown in **Figure 2b**. XRD analysis of KNNT revealed that the powder as well as the films has perovskite monoclinic structure (JCPDS 770038) along with some secondary phase. On comparing the patterns of green and sintered films, no additional peaks were observed, indicating that the quality of the film merely depend on the quality of the powder. However, in the case of  $Mg_2TiO_4$ , a phase change between the SPVD nanopowders and the sintered films was observed. An apparent decrease in the peak width between the nanopowders and the sintered films as also observed in  $Mg_2TiO_4$  samples, which indicate an increase in the crystallite size of the sintered films [19]. Even though the starting powder was  $Mg_2TiO_4$ , a pure  $MgTiO_3$  phase without secondary phases was obtained after sintering because nanopowders were very fine and very reactive. Moreover, the XRD patterns were matched to crystallographic data on synthetic geikielite crystalline phase and the peaks were indexed to a rhombohedral unit cell with  $a = b = 0.5057(2)$  nm and  $c = 1.3890(4)$  nm [17].

#### 4. Charging mechanism and deposition

Depending on the surface charge on the particle in a suspension, the deposition can be anodic or cathodic deposition. The deposition of a negatively charged particle on anode is called anodic electrophoretic deposition and the deposition of a positively charged particle on cathode is called cathodic electrophoretic deposition. The surface charge of the particle can be easily modified by the appropriate choice of solvent or/and surfactant [18, 20]. Moreover, different surface charge can be introduced on KNN particle using a single solvent with different surfactants [21].

Here we have observed that the KNNT particles were deposited on cathode yielding a smooth deposit. Cathodic deposition of KNNT particle indicates that KNNT particles have acquired positive charge in the medium. Since TEA is a weak base, it increases the pH of the suspension and when it is added to isopropyl alcohol it takes the proton from isopropyl alcohol and acquires positive charge. The overall reaction can be written as [18].



**Figure 3.**  
 Schematic representation of charge acquisition of KNNT particle in TEA added isopropyl alcohol medium.



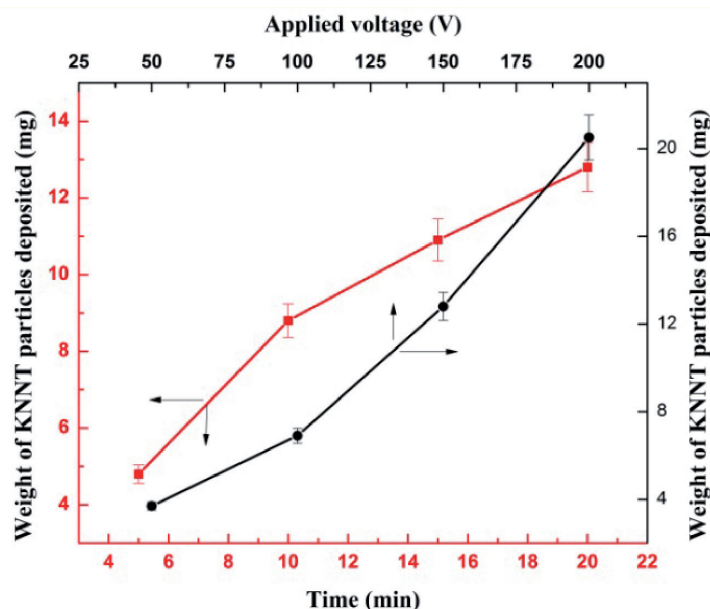
This charged species  $NH^+(C_2H_4OH)_3$  get adsorbed on the surface of the KNNT particles confer positive charge to them and the electrostatic repulsion force between TEA adsorbed KNNT particles prevent the agglomeration of the particles in the medium. Upon applying an electric field of proper strength, the charged particles get attracted towards oppositely charged plate and there it get neutralized and the deposition of KNNT particles occurs on the plate leaving  $NH^+(C_2H_4OH)_3$  back to the solution. Even though the stabilization of suspension is via electrostatic repulsion force, the high molecular weight TEA may also contribute to some steric stabilization mechanism [18]. The green and sintered film showed good adhesion with the Pt-electrode and showed no micro cracks, peelings, or pores (**Figure 3**).

### 5. Effect of EPD voltage and time on the deposition weight

According to Hamaker’s equation the yield of electrophoretic deposition ( $w$ ) is related to electrophoretic mobility ( $\mu$ ), surface area of the electrode ( $A$ ), electric field strength ( $E$ ) particle mass concentration in the suspension ( $C$ ) and deposition time ( $t$ ) through the following relation [22].

$$w = \int_{t_1}^{t_2} \mu . E . A . C dt \tag{2}$$

**Figure 4** shows the variation of weight of KNNT particles deposited on substrate at various dc voltages and the variation in deposit weight with deposition times at a particular voltage (100 V). From **Figure 4**, it is evident that the rate of deposit increases with the increase in applied voltage and deposition time.



**Figure 4.**

*Amount of KNNT particles deposited on substrate at various dc voltages and various deposition time (at constant voltage of 100 V).*

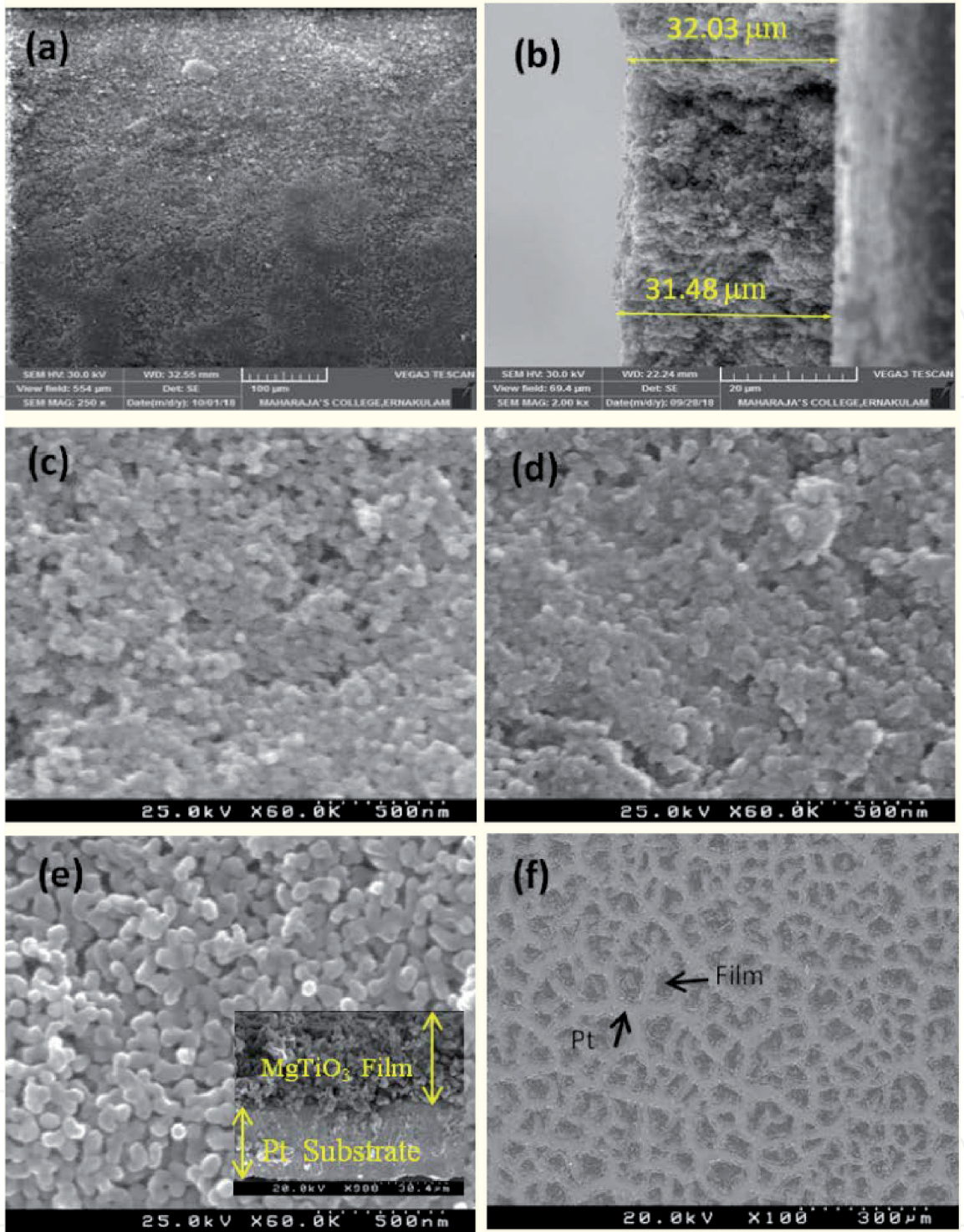
At high voltage and longer deposition durations, though the rate of deposition was high, it does not yield smooth film surface. The surface morphology was very rough and the films tend to peel off from the substrate due to the low adhesion between the film and the Pt substrate. At low voltages and short deposition time, the amount of deposit was too small to form a uniformly thick film. Since we obtained films with good surface morphology at 100 V, this film was used for further studies.

## 6. SEM analysis

When there is dominance of gravity and/or adhesive force of suspension over the inter particle van der Waals force, dislodgement of particle may occur during the lifting of the substrate out of suspension. This may lead to crack formation or micro cavities on the film surface. Hence, to study the quality of the deposited film, SEM images were taken and analyzed. The SEM images of KNNT thick film sintered at 1000°C are shown in **Figure 5a**. From **Figure 5a**, it is evident that KNNT particles are well deposited on the substrate with good compaction. In addition, no cracks were observed on the surface indicating that the processing conditions are favorable to get crack free thick films. Thickness as well the uniformity in thickness was analyzed from SEM images using Image J software and it was found that the film had uniform thickness of 32  $\mu\text{m}$  over the entire deposited region.

SEM micrographs MgTiO<sub>3</sub> thick film is shown in **Figure 5c–f**. A regular shaped grain with an average size of 50 nm can be observed in as deposited green films (**Figure 5c**). Whereas a porous microstructure with an average grain size of 60–70 nm was observed in MgTiO<sub>3</sub> thick films sintered at 800°C (**Figure 5d**). It is clear from **Figure 5e** that the films sintered at 900°C have more homogeneous and denser microstructure with higher grain size (100 nm). However, at higher sintering temperature (1000°C) inhomogeneous grain growth and delaminating effects were observed. The cross-sectional (CS) image of film sintered at 900°C was taken to find the thickness (inset of **Figure 5c**) and the thickness was about 18–20  $\mu\text{m}$ .



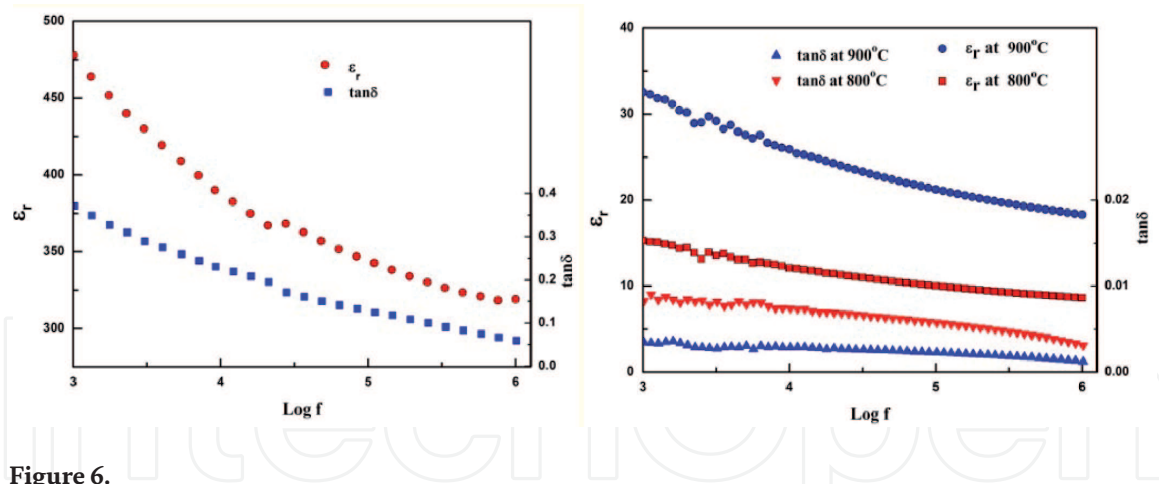


**Figure 5.** SEM images (a) surface morphology of the sintered KNNT thick film (b) SEM showing the thickness of the KNNT film sintered at 1000°C. SEM micrograph of (c) as-deposited green film. (d–f) SEM images of MgTiO<sub>3</sub> thick films sintered at (b) 800°C (15 min), (c) 900°C (15 min); the inset shows the cross sectional SEM image of the film sintered at 900°C and (d) 1000°C (15 min) sintered film showing delamination (the dark island like regions are the film and the lighter area is the Pt substrate).

### 7. Dielectric constant ( $\epsilon_r$ )

Variation of dielectric constant and dielectric loss with frequency at room temperature for sintered KNNT thick film is obtained using a metal- insulator-metal structure and the data is shown in **Figure 6**. Dielectric constant value of 320 (at 1 MHz) was obtained for sintered KNNT thick film. The dielectric value of the KNNT film (320) at 1 MHz was lower than the value reported for KNN thick films fabricated via electrophoretic deposition in acetone (393) [21] and aqueous





**Figure 6.** Dielectric constant ( $\epsilon_r$ ) and  $\tan \delta$  as function of frequency for (left) KNNT thick film sintered at 1000°C (right) MgTiO<sub>3</sub> thick films sintered at 800 and 900°C.

media (495) [20] and the higher values of later two might be the result of dense film obtained by isostatic pressing and higher sintering temperature (1100°C). Dielectric permittivity and loss tangent ( $\tan \delta$ ) of MgTiO<sub>3</sub> films were evaluated over a frequency range of 1 kHz to 1 MHz, using an impedance analyzer (HP 4284A precision LCR Meter). Since the samples sintered at 1000°C were not of good enough quality for electrical studies, the films sintered at 800 and 900°C were used for the studies. Among these two samples, those sintered at 900°C exhibited a good dielectric constant  $\sim 18.3$  ( $\tan \delta \sim 0.0012$ ) at 1 MHz compared to films sintered at 800°C ( $\epsilon_r \sim 8.6$  and  $\tan \delta \sim 0.0031$ ). The low density and porous microstructure can be the reason for low  $\epsilon_r$  values for the films at sintered at 800°C.  $\epsilon_r$  value obtained for samples sintered at 900°C was comparable with that of other literature reports [23, 24].

## 8. Conclusions

(K<sub>0.5</sub>Na<sub>0.5</sub>)(Nb<sub>0.7</sub>Ta<sub>0.3</sub>)O<sub>3</sub> and MgTiO<sub>3</sub> thick films were fabricated successfully by electrophoretic deposition method from TEA added isopropyl alcohol medium and acetone medium, respectively. We suggest that the stability of the KNNT suspension is mainly due to electrostatic stabilization mechanism and the NH<sup>+</sup>(C<sub>2</sub>H<sub>4</sub>OH)<sub>3</sub> ions being adsorbed on the KNNT particle is responsible for the observed positive surface charge in KNNT particles. From SEM images we confirmed that KNNT particles as well as MgTiO<sub>3</sub> particles were well deposited on the substrate and the thickness was found to be 32  $\mu\text{m}$  and 18–20  $\mu\text{m}$  (for film sintered at 900°C) respectively. In the case of MgTiO<sub>3</sub> thick films, the sintering temperature plays an important role in the quality of films since it affects the density and grain growth. An increase in the sintering temperature, from 800–900°C, resulted in increase in the dielectric constant and reduced loss. Moreover, no cracks were observed on the surface of the thick films, indicating that the processing conditions are favorable to get crack free thick films. The dielectric values of the KNNT and MgTiO<sub>3</sub> thick film at 1 MHz was 320 and 18.3 respectively, which is in agreement with the earlier reported values. For these reasons we can say that compared to other solution based techniques EPD is a cost effective and simple technique to fabricate high quality thick films.

## Acknowledgements

The authors acknowledge Department of Physics, Central University of Kerala for providing XRD characterization facilities and Maharajas College,

Ernakulam for SEM facility. VSK. acknowledges the facilities provided by CUTN, UGC for the start-up grant and DST-SERB for the Early Career Research grant (ECR/2015/000273). VP acknowledge the facilities and financial support offered by CUTN.

## **Conflict of Interest**

The authors declare no conflict of interest.

## **Author details**

Vineetha P and Venkata Saravanan K\*  
Department of Physics, School of Basic and Applied Sciences, Central University of Tamil Nadu, Thiruvarur, India

\*Address all correspondence to: [venketvs@cutn.ac.in](mailto:venketvs@cutn.ac.in)

## **IntechOpen**

© 2019 The Author(s). Licensee IntechOpen. This chapter is distributed under the terms of the Creative Commons Attribution License (<http://creativecommons.org/licenses/by/3.0>), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited. 

## References

- [1] Corni I, Ryan MP, Boccaccini AR. Electrophoretic deposition: From traditional ceramics to nanotechnology. *Journal of the European Ceramic Society*. 2008;**28**:1353-1367. DOI: 10.1016/j.jeurceramsoc.2007.12.011
- [2] De Riccardis MF. *Ceramic Coatings-Applications in Engineering*. China: InTech; 2012. 286p
- [3] Sarkar P, Nicholson PS. Electrophoretic deposition (EPD): Mechanisms, kinetics and applications to ceramics. *Journal of the American Ceramic Society*. 1996;**79**:1987-1202. DOI: 10.1111/j.1151-2916.1996.tb08929.x
- [4] Besra L, Liu M. A review on fundamentals and applications of electrophoretic deposition (EPD). *Progress in Materials Science*. 2007; **52**:1-61. DOI: 10.1016/j.pmatsci.2006.07.001
- [5] Heavens N. Electrophoretic deposition as a processing route for ceramics. In: Binner GP, editor. *Advanced Ceramic Processing and Technology*. Vol. 1. Park Ridge, NJ, USA: Noyes Publications; 1990. pp. 255-283
- [6] Wu J, Xiao D, Zhu J. Potassium–sodium niobate lead-free piezoelectric materials: Past, present, and future of phase boundaries. *Chemical Reviews*. 2015;**115**:2559-2595. DOI: 10.1021/cr5006809
- [7] Egerton L, Dillon DM. Piezoelectric and dielectric properties of ceramics in the system potassium-sodium niobate. *Journal of the American Ceramic Society*. 1959;**42**:438. DOI: 10.1063/1.2436923.
- [8] Ming et al. Piezoelectric properties of (Li, Sb, Ta) modified (Na, K) NbO<sub>3</sub> lead-free ceramics. *Journal of Applied Physics*. 2007;**101**:054103. DOI: 10.1063/1.2206554
- [9] Zang et al. Perovskite lead-free (Na<sub>0.5</sub>K<sub>0.5</sub>)<sub>1-x</sub>(LiSb)<sub>x</sub>Nb<sub>1-x</sub>O<sub>3</sub> piezoceramics. *Applied Physics Letters*. 2006;**88**:212908
- [10] Lv YG et al. Tantalum influence on physical properties of (K<sub>0.5</sub>Na<sub>0.5</sub>)(Nb<sub>1-x</sub>Ta<sub>x</sub>)O<sub>3</sub> ceramics. *Materials Research Bulletin*. 2009;**44**:284-287. DOI: 10.1016/j.materresbull.2008.06.019
- [11] Lau ST, Li X, Zhou QF, Shung KK, Ryu J, Park D-S. Aerosol-deposited KNN–LSO lead-free piezoelectric thick film for high frequency transducer applications. *Sensors and Actuators A: Physical*. 2010;**163**:226-230. DOI: 10.1016/j.sna.2010.08.020
- [12] Zhu et al. (100)-textured KNN-based thick film with enhanced piezoelectric property for intravascular ultrasound imaging. *Applied Physics Letters*. 2015;**106**:173504. DOI: 10.1063/1.4919387
- [13] Ferri EAV, Sczancovski JC, Cavalcante LS, Paris EC, Espinosa JWM, de Figueiredo T, et al. Photoluminescence behavior in MgTiO<sub>3</sub> powders with vacancy/distorted clusters and octahedral tilting. *Materials Chemistry and Physics*. 2009;**117**:192-198. DOI: 10.1016/j.matchemphys.2009.05.042
- [14] Istianah, Lestari R, Baqiya MA, Pratapa S. In: *Proceedings of International Conference on X-Ray Microscopy and Smart Matter (ICXSM 2011)*, June 13, 2011; Solo, Indonesia. 2011. pp. 1-4
- [15] Apostol I, Saravanan KV, Monty CJA, Vilarinho PM. Solar physical vapor deposition: A new approach for preparing magnesium titanate nanopowders. *Applied Surface Science*. 2013;**285**:49-55. DOI: 10.1016/j.apsusc.2013.07.155



- [16] Vineetha P, venkata Saravanan K. Fabrication of  $(K_{0.5}Na_{0.5})(Nb_{0.7}Ta_{0.3})O_3$  thick films by electrophoretic deposition. AIP Conference Proceedings. 2018;**1953**:090041
- [17] Apostol I, Mahajan A, Monty CJA, Venkata Saravanan K. Nanostructured  $MgTiO_3$  thick films obtained by electrophoretic deposition from nanopowders prepared by solar PVD. Applied Surface Science. 2015;**358**:641-646. DOI: 10.1016/j.apsusc.2015.09.060
- [18] Farrokhi-rad M, Ghorbani M. Stability of titania nano-particles in different alcohols. Ceramics International. 2012;**38**:3893-3900. DOI: 10.1016/j.ceramint.2012.01.041
- [19] Monty CJA. Characterization and properties of nanophases prepared by solar physical vapor deposition (SPVD) in the solar reactor heliotron. Arabian Journal for Science and Engineering. 2010;**35**(1C):93-116
- [20] Mahajan A et al. Unleashing the full sustainable potential of thick films of lead-free potassium sodium niobate  $(K_{0.5}Na_{0.5}NbO_3)$  by aqueous electrophoretic deposition. Langmuir. 2016;**32**:5241-5249. DOI: 10.1021/acs.langmuir.6b00669
- [21] Dolhen M et al. Sodium potassium niobate  $(K_{0.5}Na_{0.5}NbO_3)$ , KNN) thick films by electrophoretic deposition. RSC Advances. 2015;**5**:4698-4706. DOI: 10.1039/c4ra11058g
- [22] Hamaker HC. Formation of a deposit by electrophoresis. Transactions of the Faraday Society. 1940;**36**:279-283. DOI: 10.1039/TF9403500279
- [23] Kim ES, Seo SN. Evaluation of microwave dielectric properties of  $MgO-TiO_2$  system by dielectric mixing rules. Journal of the Korean Ceramic Society. 2010;**47**:163-168. DOI: 10.4191/KCERS.2010.47.2.163
- [24] Wu HT, Jiang YS, Cui YJ, Zhang XH, Jia X, Yue YL. Improvements in the sintering behavior and microwave dielectric properties of geikielite-type  $MgTiO_3$  ceramics. Journal of Electronic Materials. 2013;**42**:445-451. DOI: 10.1007/s11664-012-2349-2