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Research Article

Analysis of dielectric properties of Fe and Co coated thin films on $PbZrTiO_3$

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ABSTRACT

$PbZr_{1-x}Ti_xO_3$ ceramic material, which has ferroelectric properties, attracts great attention from researchers due to its electrical and electromechanical properties. In this study, Fe and Co materials with ferromagnetic properties were coated separately on the ferroelectric $PbZr_{0.5}Ti_{0.5}O_3$ (PZT) ceramic material by vacuum evaporation technique. The variation of dielectric properties such as dielectric constant, dielectric loss, loss tangent, cole-cole diagram of the prepared thin films with frequency was examined. Dielectric constant, dielectric loss, loss tangent, cole-cole diagram of pure PZT, Fe-coated PZT and Co-coated PZT thin films were measured with an impedance analyzer as a function of frequency (in the range of high frequency) at room temperature.

Keywords: *dielectric properties, ferroelectric, piezoelectric, PZT, multiferroic*

PbZrTiO3 üzerine Fe ve Co kaplanmış İnce Filmlerin Dielektrik Özelliklerinin İncelenmesi

ÖZET

Ferroelektrik özelliklere sahip $PbZr_{0.5}Ti_{0.5}O_3$ seramik malzemesi elektriksel ve elekromekaniksel özelliklerinden dolayı araştırmacılar tarafından büyük ilgi görmektedir. Bu çalışmada, Ferroelektrik $PbZr_{0.5}Ti_{0.5}O_3$ (PZT) seramik malzemesi üzerine vakum buharlaştırma tekniği ile ferromanyetik özelliklere sahip Fe ve Co malzemeleri ayrı ayrı kaplanmıştır. Hazırlanmış ince filmlerin dielekrik sabiti, dielektrik kayıp, kayıp tanjantı, cole-cole diyagram gibi dielektrik özelliklerin frekansla değişimi incelenmiştir. Saf PZT, Fe kaplı PZT ve Co kaplı PZT ince filmlerin dielekrik sabiti, dielektrik kayıp, kayıp tanjantı, cole-cole diyagramı oda sıcaklığında frekansın bir fonksiyonu olarak yüksek frekans aralığında empedans analizörü ile ölçülmüştür.

Anahtar Kelimeler: *dielektrik özellik, ferroelektrik, piezoelektrik, PZT, multiferroik*

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I. INTRODUCTION

The chemical and physical properties of materials at the macro scale differ greatly from the physical and chemical properties of the same materials prepared at the nano scale. In fact, a material that is conductive at macro dimensions can become an insulator when reduced to nano dimensions. The fact that materials at the nanometer level show new properties known as the quantum size effect has led to an increase in research on this subject. In this way, it creates potential for technological applications in a wide range of areas such as information storage, magnetic read and write heads in computers, magnetic sensors, magnetic memory, single electron-based quantum information processing devices, microwave electronic devices[1].

The magnetoelectric (ME) effect, that is, the control of magnetic properties by an electric field or the control of electrical polarization by a magnetic field, has been investigated since the 1960s. The ME effect is important for coupling magnetic and electrical properties. In materials with ME effect, control of magnetic domains by electric field and ferroelectric domains by magnetic field emerges. This effect can most clearly be achieved by combining ferroelectric and ferromagnetic materials, and such materials are called multiferroic. Ferroelectric materials are materials that have spontaneous polarization without the application of an electric field, and ferromagnetic materials are materials that have spontaneous magnetization without the application of a magnetic field. The fact that the dielectric and magnetic susceptibilities of these materials are higher than other materials and their non-linear response to external factors make them attractive in terms of application [1-4].

PZT (lead zirconium titanate) is the most commonly used form of ferroelectricity. Piezoelectric materials are materials that can produce electrical signals when mechanical stress is applied or, conversely, mechanical movement when an electric field is applied. Owing to these properties, PZT is used in many different applications such as sensors, transducers and actuators [5]. Iron and cobalt are two important elements known for their magnetic properties - especially ferromagnetic. Iron is naturally magnetic and capable of generating high amounts of magnetic fields. Owing to this feature, it is widely used in the production of magnetic materials. Iron, which is frequently used in devices such as electric motors, generators and transformers based on the principle of electromagnetism, enables strong magnetic interactions. Cobalt, similar to iron, is highly magnetic. Cobalt alloys are used in the medical field in the construction of implants, electronic devices and sensitive magneto-optical systems[6].

In this study, it was planned to produce multiferroic thin films with magnetoelectric properties. For this purpose, ferroelectric PZT material was chosen as the substrate, and two different materials (iron and cobalt) were chosen as the ferromagnetic material. The surface properties and dielectric properties of Pure PZT, Fe-coated PZT and Co-coated PZT thin films were investigated in a high frequency range at room temperature. Cole-Cole plots corresponding to the equivalent electrical circuit of these materials were obtained and the suitability of the samples for electrical circuit applications in technology was discussed.

II. MATERIAL AND METHOD

Fe and Co materials with ferromagnetic properties were coated separately on the $PbZr_{0.5}Ti_{0.5}O_3$ (PZT) ceramic material, which has ferroelectric properties, by vacuum evaporation technique. 100 nm thick Fe and Co films were coated on 0.5 mm thick PZT ceramic material.

Surface investigation of Fe-coated PZT (Fe/PZT) and Co-coated PZT (Co/PZT) thin films was carried out by Scanning Electron Microscope (SEM). Dielectric properties such as real part (ε') and imaginary part (ε'') of the dielectric constant, tangent factor (tanδ) and Cole-Cole plots of the prepared thin films with frequency were examined. The ε', ε'', tanδ, Cole-Cole plots for pure PZT, Fe/PZT and Co/PZT thin films were measured with an impedance analyzer as a function of frequency (in the range of high frequency) at room temperature.

III. EXPERIMENT RESULTS

Surface control of ferromagnetic films coated on PZT by vacuum evaporation method was carried out with Scanning Electron Microscope (SEM). The purpose of SEM imaging is to ensure the quality control of the surfaces of the prepared films. The PZT substrate chosen as a ferroelectric material has a ceramic structure. It was determined that the prepared thin films were largely polycrystalline. At 50 µm magnification, it was observed that Fe/PZT thin films had a smoother surface (Figure 1), while Co/PZT thin films had a rougher structure (Figure 2). At 10 µm magnification, it was observed that Fe/PZT thin films had a tetrahedral structure and had more pores, while the surface structure of Co/PZT thin films changed and had a less porous structure and clusters formed. Additionally, it was observed that the grain size decreased in Co-coated thin films (Figure 1 and Figure 2).

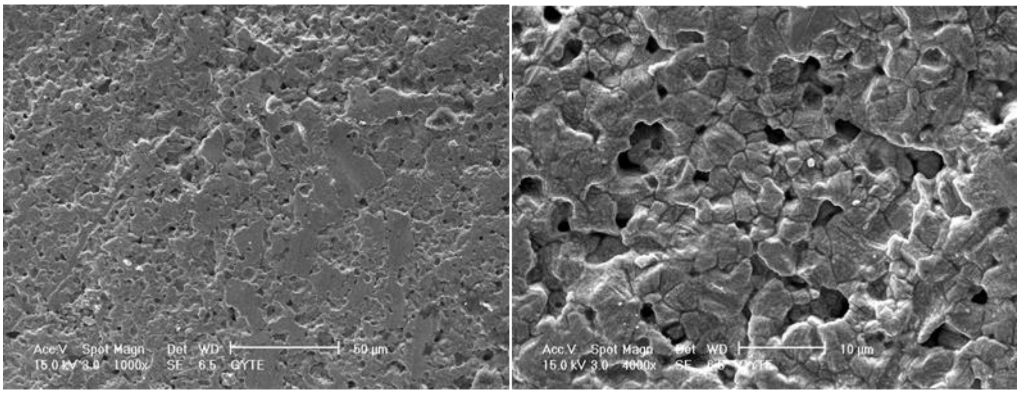
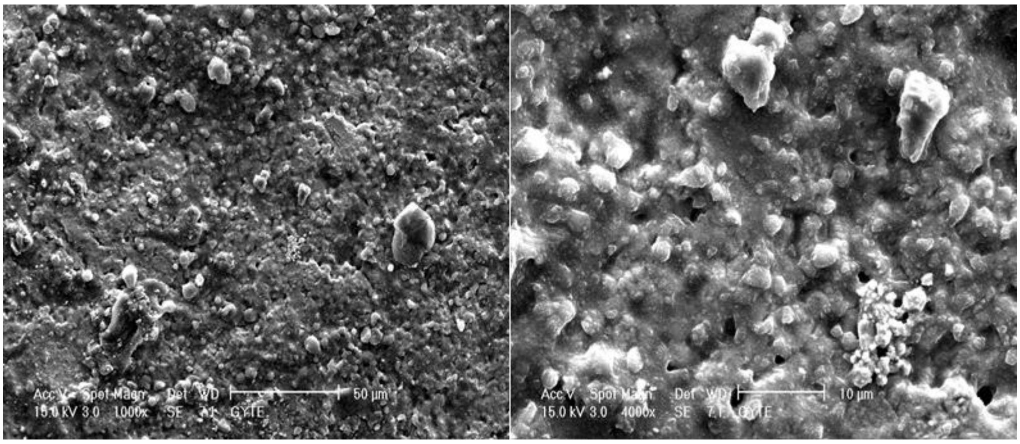


Figure 1. *SEM images of Fe coated PZT thin film*



(b)

Figure 2. *SEM images of Co coated PZT thin film*

The frequency dependence of the dielectric constant, which is the phenomenon that best characterizes dielectric materials, is very important for technological applications of these materials. When an external electric field is applied to dielectric materials, electrically charged electrons, ions and molecules affected by the field change direction. Due to the change of direction of the electrical charges, the electrical charge centers shift and as a result, polarization occurs. It is a good method for examining composites because the response of the materials to alternating current provides important information about the structures of the materials (pores, interfacial polarization, etc.) [7,8,9]. The dielectric permittivity of a material refers to its ability to absorb, transmit and reflect electromagnetic energy. Dielectric Permittivity is a quantity consisting of the ε' and the ε'':

$$\varepsilon^{*} = \varepsilon' - j\varepsilon'' \tag{1}$$

where ε' is the real component and is related to the capacitance of the substance and its ability to store electrical energy. C is the capacitance of the material, C_0 is the capacitance of the vacuum,

$$\varepsilon' = \frac{C}{C_0} \tag{2}$$

ε'' is the imaginary component and is related to various absorption mechanisms of energy distribution. It briefly provides information about losses [8-10]. G is the conductivity of the material, w is the angular frequency.

$$\varepsilon'' = \frac{G}{wC_0} \tag{3}$$

The tangent factor is the ratio of the imaginary part of the complex dielectric constant to the real part. It is used to evaluate the amount of energy released as heat in the dielectric [10, 11].

$$\tan\delta = \frac{\varepsilon''}{\varepsilon'} \tag{4}$$

The reason why the dielectric constant of pure PZT material at room temperature is so high is dipolar polarization[9].

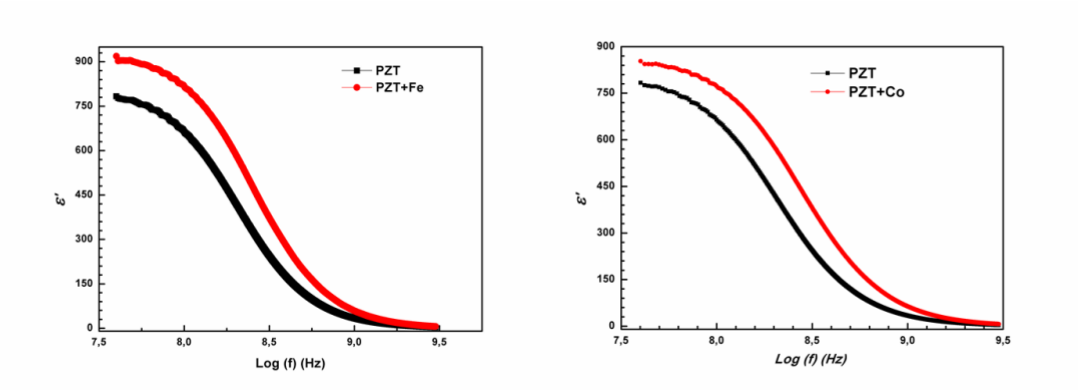


Figure 3. *Real part of dielectric constant (* ε' *)-frequency graph of pure PZT, Co-coated PZT and Fe-coated PZT thin films*

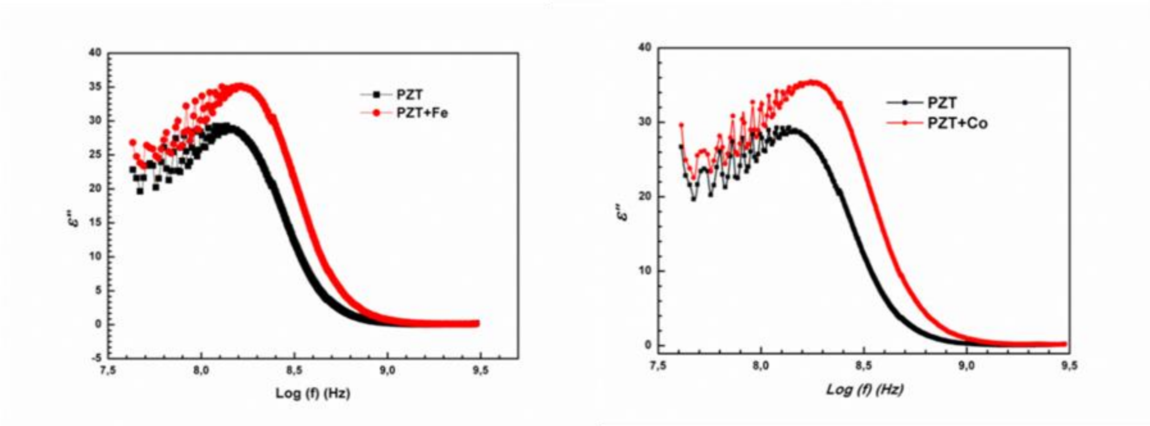


Figure 4. *Imaginary part of dielectric constant (ε'')-frequency graph of pure PZT, Co-coated PZT and Fe-coated PZT thin films*

The frequency-dependent changes of ε' of all thin films are given in Figure 3. It has been observed that ε' of pure PZT, Fe/PZT and Co/PZT thin films decreases with increasing frequency In general, it has been observed that while ε' is higher in the relatively low frequency region, ε' decreases in the high frequency region. In the low frequency region, the sample with the highest ε' was Fe/PZT, while the sample with the lowest ε' was pure PZT. It was observed that the dielectric behavior of all samples was similar. Charged particles moving through the material under the applied external electric field undergo dipolar polarization. This is why dielectric parameters have high values in the low frequency region. The exponential decrease of dielectric parameters with increasing frequency is a result of dielectric relaxation. The main reason the dipolar polarization effect increases in iron and cobalt coated thin films is their contribution to polarization. The fact that these contributions are higher in the iron-coated film is due to less interfacial interaction between Fe and PZT. Frequency-dependent changes in the ε'' of pure PZT, Fe/PZT and Co/PZT thin films are given in Figure 4. It was observed that in all samples, with increasing frequency, the ε'' first increased and then decreased parabolically. While the ε'' of Co/PZT and Fe/PZT samples have similar values in the low frequency region, the pure PZT sample has a lower ε'' value.

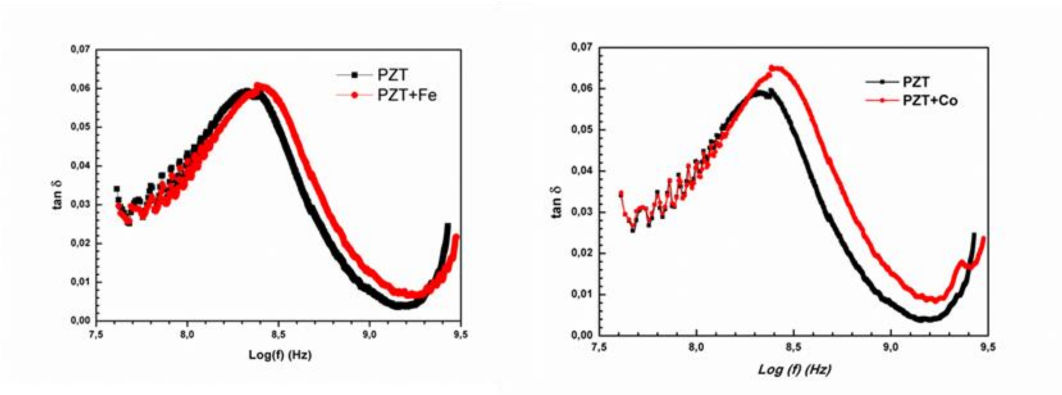


Figure 5. *tangent loss factor (tanδ) -frequency graph of pure PZT, Co-coated PZT and Fe-coated PZT thin films*

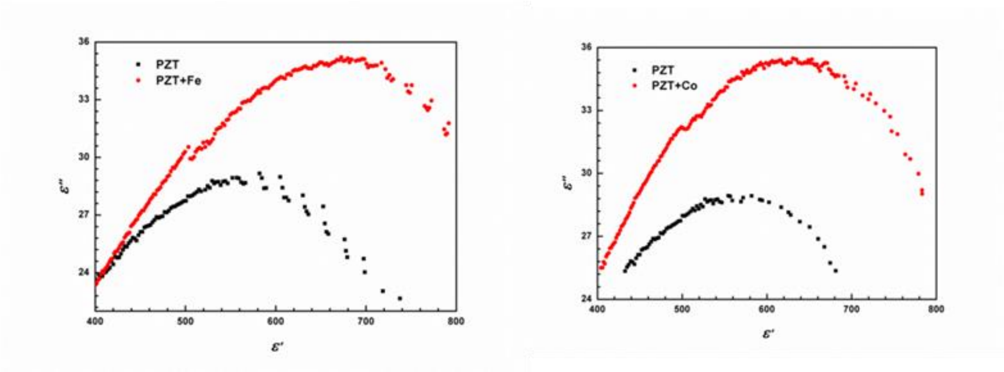


Figure 6. *ε'' - ε' graph of pure PZT, Co-coated PZT and Fe-coated PZT thin films*

Figure 5 shows the frequency-dependent variation of the tangent factor (tanδ) of pure PZT, Fe/PZT and Co/PZT thin films. ε'' and tanδ both give information about dielectric losses but tanδ allows us to examine the material in more detail. As seen in figure 5, a repolarization effect is seen around 9.2 [9]. Figure 6 shows Cole-Cole plots for pure PZT, Fe/PZT and Co/PZT thin films. We obtain information about the energy storage and loss factor in dielectric materials from Cole-Cole relaxation models. Cole-Cole plots are very important for determining whether a dielectric material has a single relaxation time in a dielectric material. For a single relaxation time dielectric material, Cole-Cole plots correspond to a semicircle. As can be seen in the Figure 6, there is a single relaxation time for all samples and this corresponds to an RC circuit. Obtaining this equivalent circuit was found to be very important as it was inspiring in terms of the application of materials as electronic circuit elements.

IV. CONCLUSION

In this study, multiferroic thin films were produced by vacuum evaporation method. Fe and Co materials (100 nm thick) with ferromagnetic properties were separately coated as thin films on ferroelectric $PbZrTiO_3$ (PZT). Surface properties were examined with scanning electron microscopy (SEM), and it was observed that Fe-coated PZT thin films had a smoother structure, while Co-coated PZT thin films had a more porous structure. Real part of dielectric constant (ε'), imaginary part of dielectric constant (ε''), tangent factor (tanδ), cole-cole plots of prepared Pure PZT, Fe/PZT and Co/PZT thin films as a function of frequency (in the range of 100 MHz to 3 GHz) at room temperature with an impedance analyzer measured. It has been observed that the ε' of pure PZT, Fe/PZT and Co/PZT thin films decreases with increasing frequency. Because of the dielectric polarisation, it has been observed that while ε' is higher in the relatively low frequency region, ε' decreases in the high frequency region. In the low frequency region, the sample with the highest ε' was Fe/PZT, while the sample with the lowest ε' was pure PZT. In the figure 3, dipolar polarization was seen and it was observed that Fe had a greater contribution to polarization than cobalt. This is thought to be due to the weaker interfacial interaction between Fe and PZT. It has been observed that the ε'' value of pure PZT, Fe/PZT and Co/PZT thin films first increases with increasing frequency and then decreases parabolically. In the low frequency region, ε'' of the Co/PZT and Fe/PZT samples have similar values, while the pure PZT sample has a lower value. We obtain information about the energy storage and loss factor in dielectric materials from cole-cole relaxation models. Cole-Cole plots are very important for determining whether a dielectric material has a single relaxation time in a dielectric material. The figure 5 shows that the relaxation ends around 9.2 and the re-polarization begins at the same point. The cole-cole plots show that there is a single relaxation in all thin films and this corresponds to an RC circuit. Obtaining this equivalent circuit was found to be very important as it was inspiring in terms of the application of materials as electronic circuit elements. According to these results, it is possible to say that the Fe/PZT thin film shows better dielectric properties in the high frequency region. It is also possible to say that Fe/PZT film is an ideal candidate for application areas such as information storage, magnetic sensor, and magnetic memory[1-4, 12,13].

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