Mechanical Properties and Effect of the MgO Content on the Dielectric Breakdown in the MgO/Binder Mixtures

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Abstract: In this study, the mechanical and electrical properties of the MgO powder and MgO solved in binder are investigated. For this purpose, the X-ray diffraction, scanning electron microscopy, energy dispersive X-ray analysis and electric field (E) -current (I) characteristics techniques are employed. Namely, the lattice parameters, the strain and the dislocation density of pure MgO powders and MgO solved in the binder are determined. The effect of MgO content on the electrical conduction is also investigated. The studies revealed that the binder is mostly composed of Na2O2: SiO2: CO3. The X-ray diffraction patterns which were recorded for a 50% MgO-50% binder mixture were related to the cubic structured MgO as major phase, the hexagonal structured Na2O2 and SiO2 as minor phases. While both of the MgO and Na2O2 caused compressing strains in grains of sizes of 50 and 27 nm, respectively, the SiO2 phase exhibit stretching strain in grains of a size of 21 nm. The solving of the MgO powder in the binder enlarged the grains of MgO from 9 to 50 nm. On the other hand, the E – I variations have shown that the amount of the MgO content in the binder highly shifts the dielectric breakdown limit toward lower values of applied electric field. The critical electric field at which the breakdown takes place reduces from 286 to 14 V/cm as the MgO content is increased from 20% to 99%, respectively.

Key words: MgO, binder, X-ray, dielectric properties.

1. Introduction

Magnesium oxide (MgO) is an attractive material for use in many technological applications [1-5]. Since 1996 developments on the AC PDP (Plasma display panels) got advantage from MgO binder as a protective layer [1], I. Koiwa et al. reported the ability of lowering the drive voltage by using MgO powder and binder. A screen-printed protective MgO layers containing no MgO liquid binder showed a short half-life of 800 hours while the MgO binder extended the half-life of luminance to 5,000 hours. Nowadays, MgO is being used as substrate to establish magnetic anisotropy [2] and as substrate for ZnO piezotronic pressure sensor [3] in which the MgO nano-layer is used as the electron-tunneling modulator of the sensors. In addition, the MgO is implanted for the design of magnetic tunnel junctions based random access memory cells [4].

Magnesium oxide layers are reported to exhibit some novel features that nominate it to be used in optoelectronics [5-7]. A 100 μm thick metal semiconductor metal device made of MgO displayed photovoltaic effect with an open circuit voltage of 0.12-0.47 V and short circuit current density of 3.9-10.5 μA/cm2. On the other hand, p-MgO/n-Ga4Se3S heterojunctions exhibited tunneling device characteristics with a wide range of negative resistance [6]. The tunneling device displayed excellent capacitance voltage tunability at 100 MHz, which make it attractive to be used as a varactor device that is able to amplify, store and resonate electromagnetic signals.

In the scope of these important applications, here in this article we aim to explore some of the properties of the MgO nano-powders and investigate its properties when solved in a binder. The study is of importance as
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it provides an easy way for producing the MgO thin films from liquid phases with effective costs. Particularly, in this article we will discuss the effect of binder addition on the mechanical properties of MgO including the structural phases, grain size and strain and dislocation densities. In addition, the effect of MgO content on the dielectric breakdown limit will be explored.

2. Experimental Setup

High purity MgO (99.99%) powders and silicate binders which were prepared by the Alfa Aesar firm were mixed to convert the MgO into its liquid phase. Different MgO content ratios were used. The mixtures were painted onto chemically cleaned glass substrates and left to dry for 24 hours in an isolated medium. The resulting films were not subjected to any heat treatment. The liquid binder lose 40-50% of its weight when it dries. The SEM (Scanning electron microscopy) images were recorded using JOEL JSM-7600F. The compositional analysis was carried out using the energy dispersion X-ray spectroscopy, which is attached to the SEM system. The X-ray diffraction was recorded using Rigaku Ultima IV. The current-voltage characteristics were recorded using Keithley 230 voltage source and Keithley 6485 picoammeter.

3. Results and discussion

Due to the high melting temperature of MgO, the MgO powders are solved in a binder that allows its existence in liquid phases. The solved MgO powders (MgO paste) are used in the design of optoelectronic devices [5-7] as optical windows. For this reason, there is a need to investigate the changes in the structure and in the mechanical properties of this material upon changing its phase from solid to liquid. The X-ray diffraction patterns of the MgO powder are displayed in Fig. 1a. The diffraction patterns of the powder are of cubic nature of structure with maximum peak reflection in the (200) direction. The analysis of the reflected lines which appear in the Fig. 1 was carried out with the assistance of “Trear 92” software. The calculated lattice parameter for this powder is found to be 4.203 Å. It is consistent with the PDF card no.: 00-001-1235. To explore the mechanical properties of the powders, the reflection peaks are analyzed using Scherrer equation:

$$\beta \cos(\theta) = \frac{0.94 \lambda}{D} + 4 \varepsilon \sin(\theta)$$

with $\beta$ being the peak broadening at full wave half maximum positions. In this equation, $\varepsilon$ is the strain, which arises from displacement of the unit cells about their normal positions, $D$ is the crystallite size and $\lambda$ is 1.54178 Å. The plot of $\beta \cos(\theta) - \sin(\theta)$ which is displayed in Fig. 1b for the powder reveal a straight line which allows determining the strain and grain size as $2.05 \times 10^{-3}$ and 9 nm, respectively. The value of the stain indicates that for a lattice constant ($a$) of 4.203 Å, the compression displacement in the unit cell is $\Delta a = 8.6 \times 10^{-3}$ Å. In other words, for each thousand cells, 8.6 cells are missed as a result of compressing stress. The dislocation density in the grains ($\delta$) which is calculated from the relation, $\delta = 15 \varepsilon / (aD)$[8], is found to be $8.1 \times 10^{10}$ line/cm$^2$.

A recent study that discusses the strain effects on the properties of MgO has shown that the strain can be an advantage for technological applications such as data storage, transistors, solar cells and strain gauges [9]. Loong et. al. have shown that the quantum transport across magnetic tunnel junctions can be significantly affected by the introduction of controllable mechanical strain. In addition, the strain at the atomic level is suggested as a new way to enhance and tune the quantum properties in nanoscale materials and devices.

Fig. 1a also displays the X-ray reflection spectra of the silicate binder that is used to solve the MgO and convert it to its liquid phase. The X-ray is recorded for the silicate binder film after it dries 24 hours (loses 40% of its weight upon drying). The silicate displayed only one reflection peak at 32.4°. The results of the
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EDXA (Dispersive energy X-ray analysis) which are illustrated in Fig. 2a and that relates to the scanning electron microscope image shown in Fig. 2b, indicated that the binder is composed of carbon, oxygen, silicon and sodium. The average atomic ratios of the composition are Na$_2$O : SiO : CO$_3$. The SEM images for a 120,000 magnifications which are displayed in Fig. 2b reflects an amorphous nature of the film. Some of the existing islands may relate to the formation of some randomly distributed grains.

On the other hand, Fig. 2c illustrates the SEM image for a 120,000 magnification of the mixture of MgO and binder. The liquid mixture are composed of 50% MgO to 50% binders. The image clearly reflects a polycrystalline nature of the MgO/binder film with uniformly distributed grains. The average grain size of grains appearing in the mixture image is around 90 nm. Consistently, the X-ray diffraction patterns which are
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presented in Fig. 1a showed a polycrystalline nature of the films. The X-ray patterns which were indexed using the same software revealed the cubic MgO peaks (shown by dashed lines in Fig. 1a with the maximum peak being located at 42.9° and oriented in the (002) direction. The other peaks which are not related to the cubic MgO structure are carefully analyzed assuming all the possible compositions of the structure. In the scope of software analysis and typical X-ray card comparisons, two apparent minor phases appeared in the X-ray spectra. The phases are Na2O2 and SiO2. The indexed peaks which appear in the Fig. 1 relate to the hexagonal Na2O2 with lattice parameters of $a = 6.22 \, \text{Å}$ and $c = 4.47 \, \text{Å}$ [10] and to trydimite hexagonal SiO2 with lattice parameters of $a = 5.00 \, \text{Å}$ and $c = 8.16 \, \text{Å}$ consistent with the PDF card no: 00-001-0378. The carbon dioxide was not apparent in the X-ray patterns. The not appearing of the CO2 phase may be due to its non-homogeneous distribution through the structure.

The analysis of the peak position and peak broadening of the X-ray spectra of the mixture (MgO + binder) with the help of Scherrer equation allowed determining the strain and grain size from the slope and intercept of the solid line of the $eta\cos(\theta)-\sin(\theta)$ plot which are presented in Fig. 1b. The strain and grain size are found to be $1.79 \times 10^{-4}$ and 80 nm, respectively. The dislocation density of the cells in the mixed structure is $7.98 \times 10^9 \, \text{line/cm}^2$. When the minor phase peaks are excluded and only the peaks that related to MgO are included in the strain analysis, the compressing strain, the average grain size and the dislocation density are found to be $6.25 \times 10^{-4}$, 50 nm and $4.46 \times 10^{10} \, \text{line/cm}^2$, respectively. On the other hand, when the major phase (MgO) peaks are excluded from the analysis and only those of Na2O2 are considered the relative $\varepsilon$, $D$ and $\delta$ along the $a$-axis are found to be $1.75 \times 10^{-4}$, 27 nm and $1.56 \times 10^{10} \, \text{line/cm}^2$. For the minor phase of SiO2 the stretching strain, the grain size and the dislocation density (along $a$-axis) are found to be $4.78 \times 10^{-3}$, 21 nm and $6.82 \times 10^{11} \, \text{line/cm}^2$, respectively.

The above mention numerical data suggest that the strain which is associated with the crystallization of SiO2 which is one order of magnitude greater than those of MgO and Na2O2 is a reason for the occurrence of compressing strain in the MgO and Na2O2 grains. The values of the dislocation density and grain size reported for the MgO in powder and thin film form is consistent with those reported for MgO prepared by the sol-gel technique [11] in which the Magnesium nitrate (MgNO3 : 6H2O) and sodium hydroxide (NaOH) are used as starting materials to participate the MgO nano-size powders.

As one possible application of these types of mixtures we study the electric field ($E$) –current ($I$) relationship between two parallel electrodes for samples which contains different concentrations of MgO. The $I$ – $E$ dependence which is displayed in Fig. 3 exhibited interesting features that depend on the MgO concentration in the samples. When no MgO was added to the binder, the current –electric field variation follows an ohmic nature of contact for all applied fields less than 183 V/cm. In the region of 184-324 V/cm, the current is slowly increasing with increasing electric field. For values larger than 324 of applied electric fields, the $I$ – $E$ relation is again ohmic. When a 20% of MgO powder is solved in the binder (20% of MgO added to the binder in liquid phase before drying) and the curve is registered again, a sharp jump in the current is observed at an electric field of 286 V/cm (see Fig. 3) the high to low current ratio at this critical field is $\sim 7.2$. Increasing the MgO contents to 50, 67 and 99% decreased the critical electric field to 79, 29 and 14 V/cm, respectively. The high to low electric current ratios are not affected by the MgO content. The abnormal increase in the current values at these critical electric fields is usually regarded as dielectric breakdown. The dielectric breakdown in MgO are previously observed to be strongly dependent on the impurity type doped into the
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structure. MgO crystals doped with Cu, Co, or H is reported to suppress the breakdowns, while the doping of Fe, Ni, Cr, or V enhanced it [12, 13]. The sharp decrease in the value of the electric field with increasing MgO content can be assigned to the increase in the grain size that is associated with increasing MgO content. Previous studies on dielectric breakdown on MgO have shown that the dielectric breakdown is correlated with the grain size in MgO. The breakdown is initiated at the grain boundaries most often at triple junctions of grain boundaries [14]. On the other hand, studies on high-quality MgO tunnel junctions with a range of barrier thickness revealed two types of dielectric breakdown in MgO layers. One is soft breakdown and is attributed to tunnel junction imperfections. And another is hard and assigned to reaching in the critical electric field across the tunnel barrier [15].

4. Conclusions

In this article we have discussed the structural properties of the pure and binder solved MgO nano-powders. When solved in the binder, the cubic MgO structure contained some minor Na$_2$O$_2$ and SiO$_2$ hexagonal phases. While the MgO and Na$_2$O$_2$ exhibited compressing strains the hexagonal SiO$_2$ exhibited stretching strain. In addition, upon mixing, the MgO fine grains enlarged from 10 to 50 nm when mixed in binder. On the other hand, when the concentration of the MgO which is solved in the binder is increased the critical electric field where the dielectric breakdown take place shifts toward lower values reaching a minimum critical field of 14 V/cm when the MgO content reaches 99%. These variations provide wide tunability in the electrical properties of the MgO.

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