



Research Papers

Effect of annealing atmosphere on MgO thin film for tunneling magnetoresistance sensor

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ABSTRACT

Magnesium oxide (MgO) thin films are the most currently used as magnetic barrier shields in tunneling Magnetoresistance (TMR) sensors. In this work, MgO (50 nm) film on silicon substrates was annealed at different temperatures (250, 350, and 450 °C) and atmospheres (Argon, Nitrogen, and air). The structure, morphology, chemical composition, and thickness were investigated by using a focused-ion beam scanning electron microscope, atomic force microscopy, and X-ray photoelectron spectroscopy techniques. Increasing annealing temperature in a normal air atmosphere increases the film's thickness and roughness due to grain growth induced by Magnesium hydroxide (Mg(OH)₂) formation. On the contrary, annealing in Argon does not significantly affect the thickness and roughness of the film. Furthermore, the dielectric breakdown and hardness of the film increase. The mechanism of the change of physical properties of the film due to annealing was elucidated by a qualitative model.

1. Introductions

Magnesium oxide (MgO)-based magnetic tunnelling junctions (MTJs) are extensively used in magnetic sensors in high areal density hard disk drives (HDDs) [1] (Fig. 1(a)). This is because MgO single crystal has a very high tunnelling magnetic resistance (TMR) ratio compared to amorphous barriers [2,3]. Many researchers reported that using crystalline MgO as a tunnel barrier material could eventually increase the TMR ratios by as high as 6000% [4–6]. Practically, an annealing temperature above 350 °C is an essential procedure to attain a high TMR value in the CoFeB/MgO/CoFeB based junctions [7,8]. Some works suggest that thermal annealing can improve dielectric strength, a lifetime, reduced porosity defects, and reduced leakage current of oxide films [9–12]. However, the annealing process may cause undesirable oxide degradation, changes in chemical composition, microstructure, and interlayer mixing (i.e., migration of material from another layer) of the MgO layer in MTJs [13,14].

In the case of MgO, thermal annealing can remove surface absorbents

such as water, hydroxyl (OH⁻) [15,16], and carbonyl (CO⁻) groups [17]. The annealing process may also change the interfacial structures of the MTJs [13,14]. It can increase the crystallinity of the interface layer between MgO and CoFeB; this yields a significant increase in magnetoresistance [18,19]. However, the annealing process may decrease the breakdown voltage of the MTJs [20]. It also found that increased annealing temperature elevates the MgO surface roughness [21–24]. In addition, annealing in an inert gas environment enhances the dielectric constant [25,26]. On the other hand, when the magnesium oxide film is annealed at a high temperature, the oxygen environment yields more oxygen vacancies, resulting in a decrease in the electrical resistance [27–29]. In the last decade, although many researchers have studied the annealing effect on MgO-based MTJs, very few works report the effect of annealing on MgO-based magnetic barrier shields. While the magnetic sensor detects the change in a magnetic field, a magnetic field from another area may interfere with the signal. Therefore, it is required to use a magnetic shield and electronic insulator formed of soft magnetic material (NiFe) and MgO, respectively, to minimize possible

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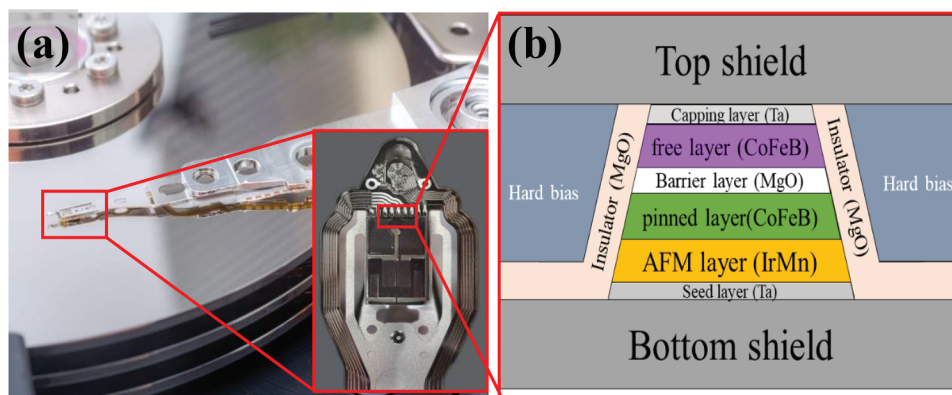


Fig. 1. (a) Schematic illustration of the HDD (b) schematic of typical thin-film multilayer stack cross-section for a magnetic sensor in hard disk drive reader head.

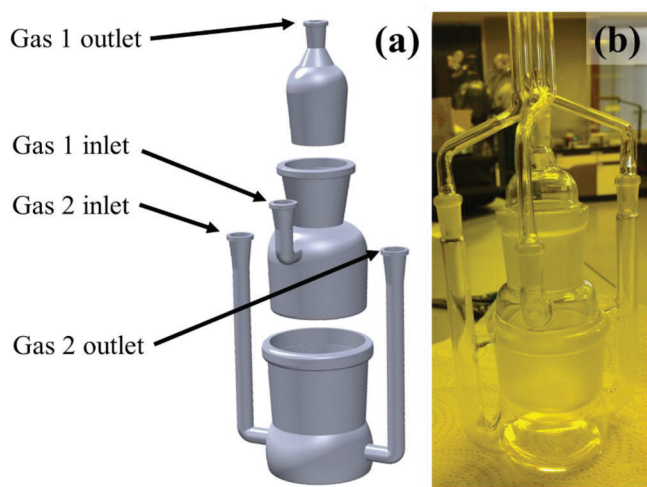


Fig. 2. (a) schematic representation of custom-built in-house annealing setup and (b) actual image of the annealing gas chamber.

interference. It is often positioned on the magnetic sensor's left and right sides to improve signal quality [30]. Theoretically, the magnetic shield is installed at the lateral of a magnetic sensor (Fig. 1(b)). The main function of the magnetic shield is to reduce the electronic noises of MTJ sensors. The noises in MTJs dramatically decrease TMR sensitivity and reduce the signal-to-noise ratio (SNR). In order to improve the SNR, the noise in MTJs must be minimized [31,32]. Therefore, the magnetic shield must be an excellent electrical insulator and have a smooth surface [33].

This work reports the systematic study of the effect of annealing conditions of MgO thin films which were fabricated as magnetic barrier shields. The change of surface morphology, physical, chemical, and electrical properties after the film was annealed at different gas atmospheric temperatures was investigated. The results were analyzed using atomic force microscopy (AFM), X-ray photoelectron spectroscopy (XPS), focused ion beam scanning electron microscopy (FIB-SEM), and electrical properties.

2. Material and methods

In this work, 50 nm thick MgO films on 10 nm Ruthenium (Ru) on silicon (Si) substrates (provided by Western Digital, LLC, Fremont, CA, US) were used to study the annealing in different environments. These MgO films were annealed at 250, 350, and 450 °C with Argon (Ar), Nitrogen (N₂) and normal air atmospheres using a custom-built in-house gas chamber (as shown in Fig. 2(a)). The samples were kept at the

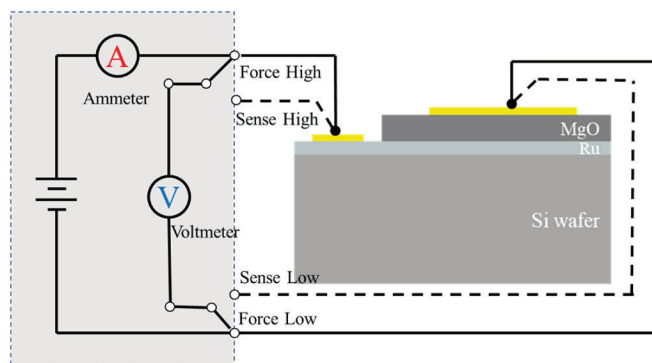


Fig. 3. a schematic diagram of the MgO thin film prepared for dielectric breakdown measurement.

bottom of two cups and allowed the gases to enter the respective gas chamber. The custom-built chamber was kept in the oven and annealed at 250, 350 and 450 °C for 2 h. The ramp-up and ramp-down rates of an oven were 5 °C/min. The gas inlets of the chamber were connected to the Ar and N₂ gases with a flow rate of 100 cm³/min. In the case of conventional air annealing, samples were annealed with a flow rate of 100 cm³/min by a conventional air pump. The actual image of the custom-built in-house gas chamber is shown in Fig. 2(b).

Tapping mode atomic force microscopy (AFM, XE-120 Park Systems, scan frequency 0.2 Hz, 512 lines) was used to investigate the surface roughness of the films, and the measured scan area was 5 × 5 μm². AFM images were analyzed using XEI software (Park Systems) to extract the root mean square (RMS) roughness values. Focused ion-beam scanning electron microscopy (FIB-SEM, dual-beam, Auriga, SE detector, 5 kV, Zeiss) was used to determine the topographical information and cross-sectional images (Gallium Ion source, 5 kV). Additional research was carried out using X-ray photoelectron spectroscopy (XPS) at Beamline 3.2 of the Synchrotron Light Research Institute (SLRI) in Thailand to investigate MgO films' chemical composition and depth profile. The XPS measurement parameters are as follows: the survey spectrum was collected from 0 to 1400 eV with 1 eV energy step and 117.5 eV pass energy. High-resolution Mg 2p, O 1s, and Ru 3d spectra were acquired with a 0.1 eV energy step, a 23.5 eV pass energy, and five cycles. The depth profile analysis of the MgO thin films was sputtered using an *in-situ* argon ion (Ar⁺, 1 keV) source with 1 mA beam current, 2 × 2 mm² beam size, and a 10 min per step size. The XPS spectra analysis was performed using the commercial software CasaXPS (Casa Software Ltd). In-house prepared circuits were used to measure the dielectric breakdown of MgO films. The schematic diagram of the dielectric breakdown measurement is shown in Fig. 3. A 10 nm Gold (Au) electrode was deposited on both MgO film (as a top electrode of 10 × 10 mm²) and on

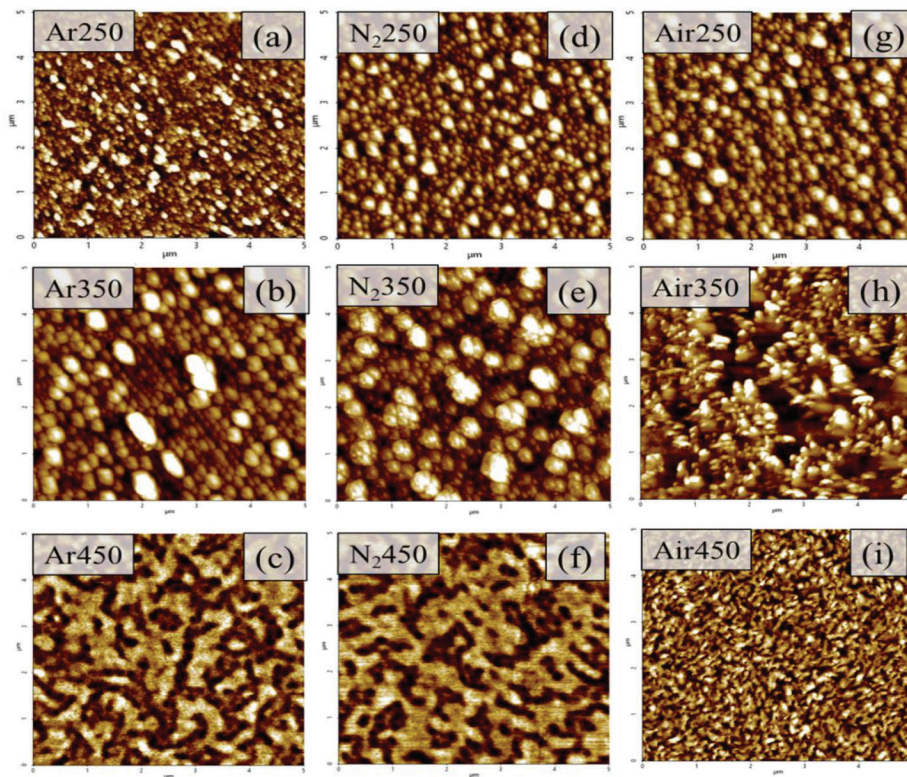


Fig. 4. surface roughness of MgO films at different gas atmospheric temperatures. Figures (a-c) depict the Ar gas annealing, figures (d-f) represent the N₂ gas annealing, and figures (g-i) represent the air annealing. Here annealing temperatures are 250, 350, and 450 °C.

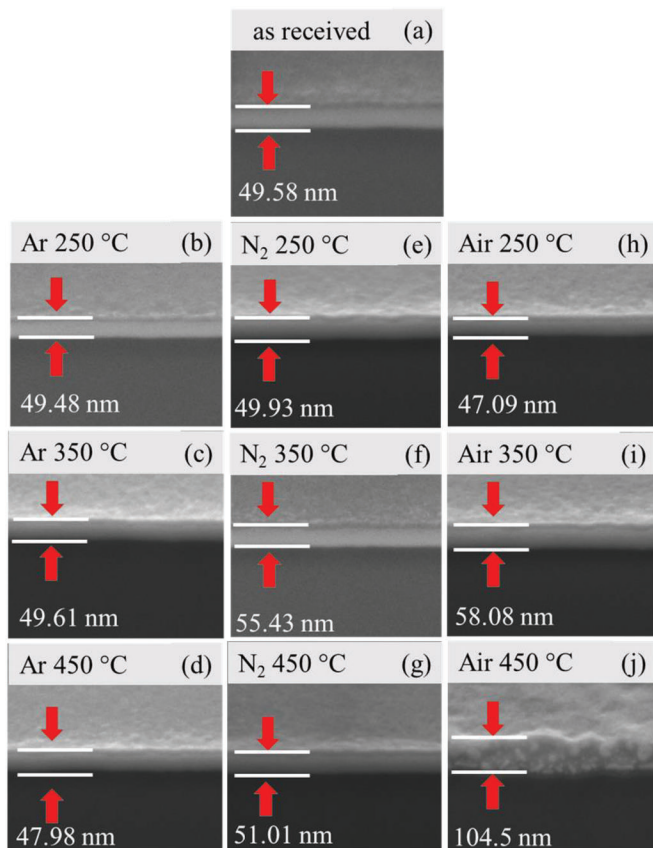


Fig. 5. SEM-FIB cross-section of (a) as-received, (b-d) argon, (e-g) nitrogen and 295 (h-j) normal air 250–450 °C samples, respectively.

the Ru electrode (as a bottom electrode of $2.5 \times 10 \text{ mm}^2$). The electrodes were connected to a voltage source unit (Keysight B2901A) to measure the current-voltage (I-V) curves.

3. Results and discussion

3.1. AFM roughness

Fig. 4 shows the topographic AFM images recorded over $5 \times 5 \mu\text{m}^2$ of MgO thin films. The Ar gas annealing showed an increased RMS roughness value from 1.74 to 2.53 nm as the annealing temperature increased from 250 to 450 °C, as shown in Fig. 4(a-c). Similarly, the N₂ gas annealed MgO film RMS values are 2.47 and 4.10 and 3.61 nm, respectively, for 250, 350 and 450 °C, as shown in Fig. 4(d-f). The increased film roughness with an increase in annealing temperature is related to grain growth and aggregation [34]. However, in the case of air annealed samples, the RMS values are higher, and the values are 2.82, 3.53, and 5.12 nm, respectively, for 250, 350 and 450 °C, as shown in Fig. 4(g-i). It is found that the surface roughness is increased with increasing temperature in all the gas environments. The maximum surface roughness is observed in the case of air-annealed films at a higher temperature. This might be due to the grain growth effect in the MgO films, and this grain growth is maximum for air-annealed samples. High annealing temperatures increase film thickness from grain growth, which can be the reason for the increase in surface roughness when the annealing temperature increases [35,36].

3.2. FIB-SEM analysis

MgO film thicknesses were estimated from FIB-SEM cross-sectional images, as shown in Fig. 5. Pristine MgO film showed a smooth surface, homogeneous, with an average film thickness of 49.58 nm, as shown in Fig. 5(a). The Ar gas annealed MgO film thickness values are 49.48, 49.61, and 47.98 nm, respectively, for 250, 350, and 450 °C, as shown in

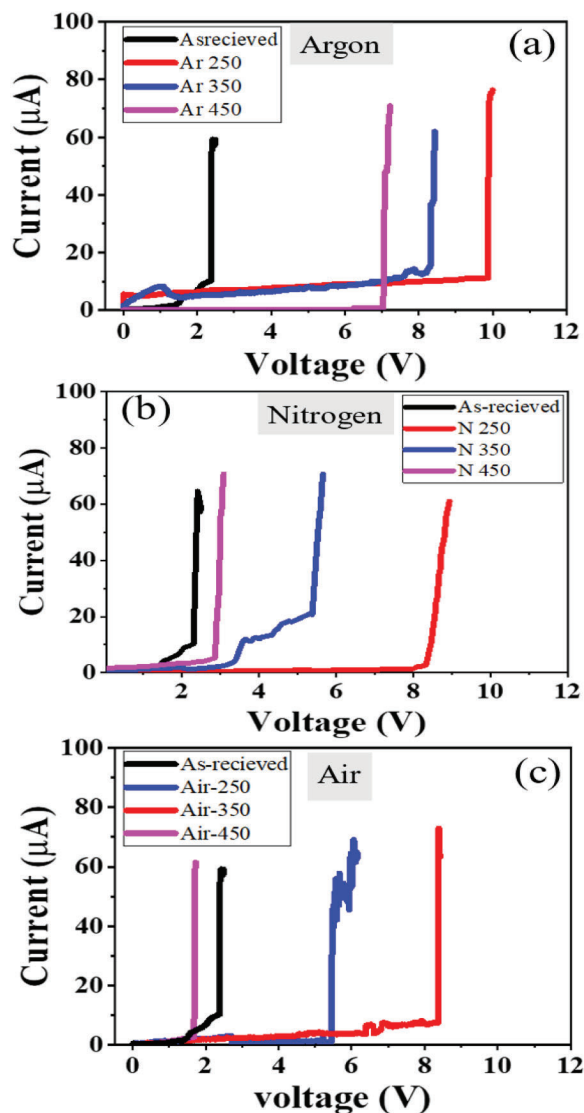


Fig. 6. (a-c) I-V curve of the Ar, N₂ and normal air annealed MgO films. Current jump indicates the dielectric breakdown voltage.

Fig. 5(b-d). Similarly, the N₂ gas annealed MgO film thickness values are 49.93, 55.43, and 51.01 nm, respectively, for 250, 350, and 450 °C, as shown in Fig. 5(e-g). In the case of N₂ and Ar annealed films, the film

thickness did not change significantly with increasing annealing temperature. However, when the MgO thin film was annealed under an air atmosphere, the thickness of the film increased with the annealing temperature. The thin film thicknesses of air annealed films are 47.09, 58.08, and 104.5 nm, respectively, for 250, 350, and 450 °C, as shown in Fig. 5(h-i). These experimental results are consistent with Lin et al. [37], which showed that high temperatures could promote grain boundary movement and agglomerate other grains during annealing. The increased film thickness is observed in air annealed films, and the increase of film thickness is very high for air annealed films compared to other gases. The creator's surface is visible throughout the film, possibly due to the high surface roughness induced by grain growth or hydroxide formation. Grain growth occurs because of grain surface energy reduction. It is observed that temperature variation stimulates and migrates to the grain boundaries, increasing the grain size of the material during the annealing process. The result is consistent with the previous study by Aswal et al. [22].

3.3. Dielectric breakdown analysis

Typically, the isotropic MgO has a critical field strength between 100 and 350 kV/cm, which means that 50 nm MgO film has the 0.25–1.75 V critical field strength [1–3]. As seen in Fig. 6, the current-voltage (I-V) curves are measured for Ar, N₂ and air-annealed MgO films. When the voltage is low, the current is of few micro amperes, and when the voltage reaches a certain threshold, a rapid rise in current indicates the onset of dielectric breakdown. This irreversible change in conductivity correlates to the thin film's hard dielectric breakdown [4]. Fig. 6 shows the as-received film dielectric breakdown voltage (DBV) at around 2.2 V and is consistent with the predicted value.

The Argon annealed sample breakdown voltages are 10.2, 8.4 and 7.2 V, respectively, for 250, 350, and 450 °C, as shown in Fig. 6(a). While nitrogen gas annealed film's dielectric breakdown voltages are 8.3, 7.2 and 5.5 V, respectively, for 250, 350, and 450 °C, as shown in Fig. 6(b). Similarly, the air annealed sample breakdown voltages are 8.2, 5.6 and 1.7 V, respectively, for 250, 350, and 450 °C, as shown in Fig. 6(c). The result showed that the DBV decreases with the increase of annealing temperature due to multiple factors. One factor is the formation of oxygen vacancies, which can act as electron traps. Eventually, increase the leakage current and decreases DBV. At higher annealing temperatures, the mobility of Mg ions in the film increases, making it easier for oxygen vacancies to form [38–40]. However, the effect of oxygen vacancies alone cannot entirely explain the decrease in DBV. In principle, DBV value should be the same for films annealed in Argon and Nitrogen since both are inert gases. However, normal air annealed DBV value is less than those annealed in Argon and Nitrogen atmospheres. This suggests that factors other than oxygen vacancies are also involved

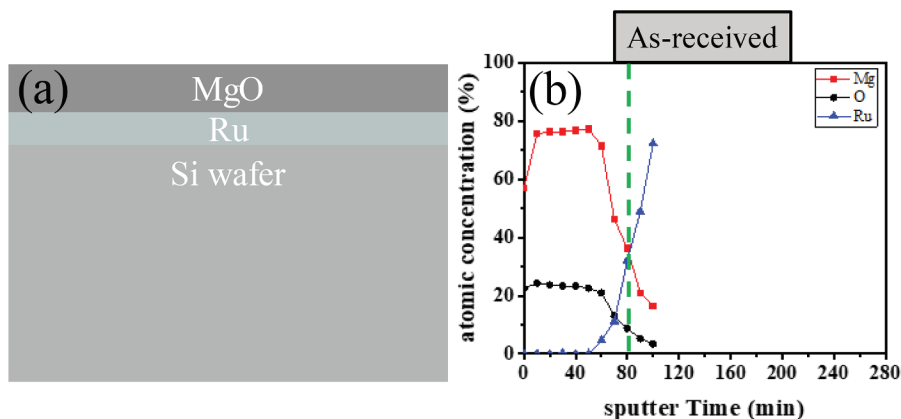


Fig. 7. (a) Schematic drawing of MgO layer structure (b) XPS depth profile of pristine MgO/Ru/Si film to determine the atomic concentration of Mg as a function of film depth.

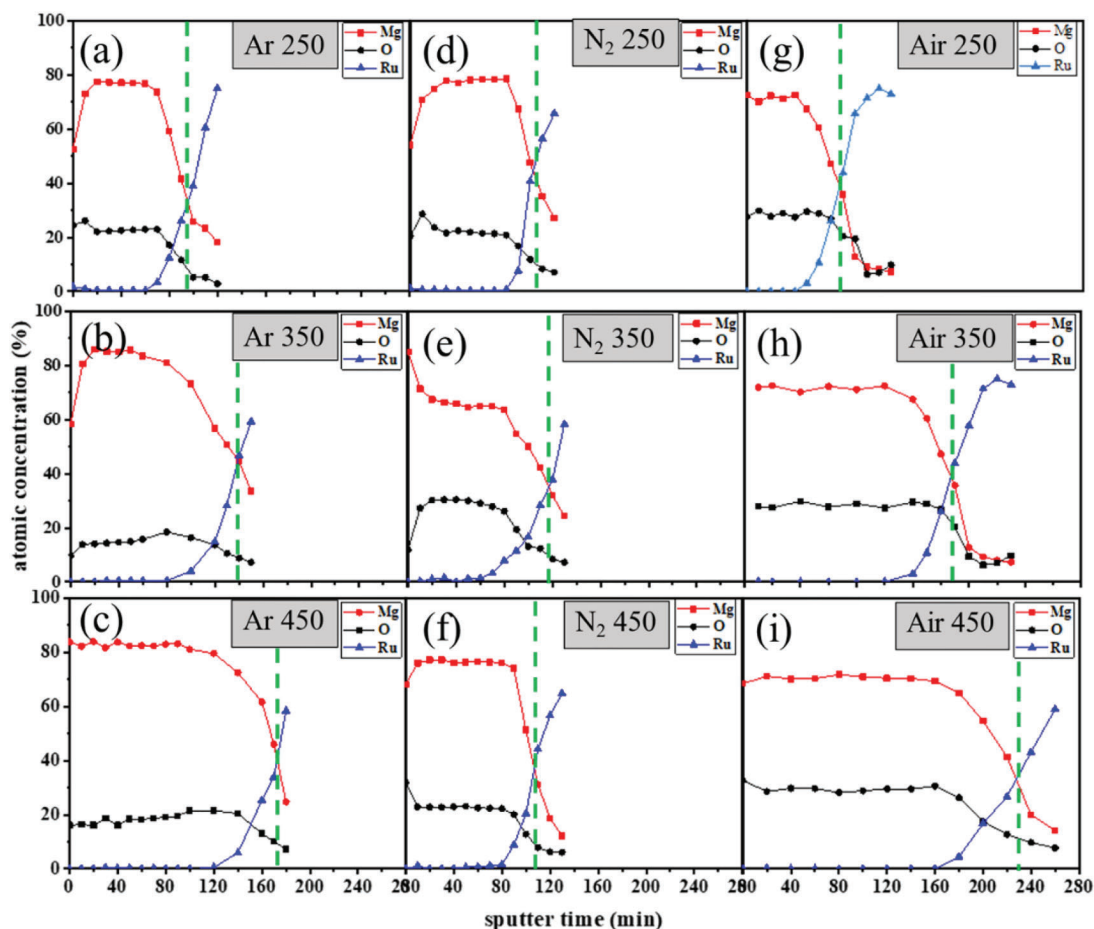


Fig. 8. Ion depth profile of MgO thin film with different annealing environments. High-resolution peak intensity of Mg2p, O1s, and Ru3d as a function of etching time for the fresh sample, N₂ and Ar. The rising of the Ru peak intensity is used to calculate the etch rate.

in the BVD transformation of MgO films. One of the factors is a change in MgO film density, which is caused by the reaction with oxygen and air humidity in the annealing atmosphere. Oxygen is highly reactive and can react with MgO to form Mg(OH)₂ [41]. This compound has a lower dielectric breakdown voltage than MgO, resulting in the reduced dielectric strength of the thin film. This phenomenon explains why MgO films annealed in an Ar atmosphere prevent MgO oxidation and have the highest BVD. In contrast, those annealed in the air showed the lowest BVD due to the formation of Mg(OH)₂. Another factor affecting the BVD in MgO films is grain formation. Annealing at higher temperatures causes grain expansion, which leads to an increase in the film thickness. This increase in thickness reduces the film's electric field strength, resulting in a lowering of the BVD. The mutual combined effects of these phenomena are playing together and yield a complex outcome in DBV.

3.4. XPS analysis of MgO films

Fig. 7(a) shows the schematic drawing of the pristine film's MgO layer structure and XPS depth profile. Pre-sputtering of MgO films was done for 10 min to remove the surface contamination. High-resolution XPS spectra were recorded, and the procedure continued until the Ru film layer reached. The Mg, O and Ru atomic concentrations significantly varied as the sputtering time increased (as shown in Fig. 7(b)). The area under the O1s, Mg2p, and Ru3d peaks were plotted as a function of etching time. Initially, both Mg and O concentrations increased slightly (this might be due to surface impurities) and were further saturated for a certain period (this is due to the core MgO film content). As the sputtering time progressed further, both Mg and O concentrations dropped, and Ru (bottom electrode film) concentration

rose sharply. This is due to the end of MgO film and starting of the bottom electrode layer. On average, the rate of MgO etching rate is 0.63 atoms/min. The sputtering rate was calculated by dividing the sputtering time by film thickness measured from FIB-SEM images. The detailed XPS analysis of MgO films was discussed in our previous publication [42].

The depth profile results obtained from different gas annealed MgO films are shown in Fig. 8. Ar gas annealed film depth profiles are shown in Fig. 8(a-c). As the annealing temperature increases, the sputter rate decreases in Ar gas annealed films. The sputtering rates of the annealed films are 0.53, 0.35, and 0.28 nm/min, respectively, for 250, 350, and 450 °C. The decrease in the sputter rate indicates that the film hardness becomes greater as the annealing temperature increases. This might be due to an increase in film density and crystallinity, which is consistent with the Oswald et al. (2020) [43] result. The N₂ annealed film depth profiles are shown in Fig. 8(d-f). The sputtering rates of N₂ annealed films are 0.54, 0.47, and 0.47 nm/min with increasing annealing temperatures from 250 to 350 and 450 °C, respectively. The sputtering rate slightly decreased and did not change much in N₂ annealed films. This indicates that the annealing temperature has little effect on the degree of film hardness.

However, the sputtering rates of the air annealed films are 0.56, 0.44, and 0.46 nm/min, respectively, for 250, 350, and 450 °C, as shown in Fig. 8(g-i). The results showed that the sputter rate was similar to that of the annealed film in a nitrogen atmosphere. On the other hand, increasing air annealing temperature leads to an increase in thin film thickness, and variation in sputtering time is due to varying film thickness. Moreover, the sputtering yield variation results from the varying film densities [44]. As film thickness increases, film density drops; this

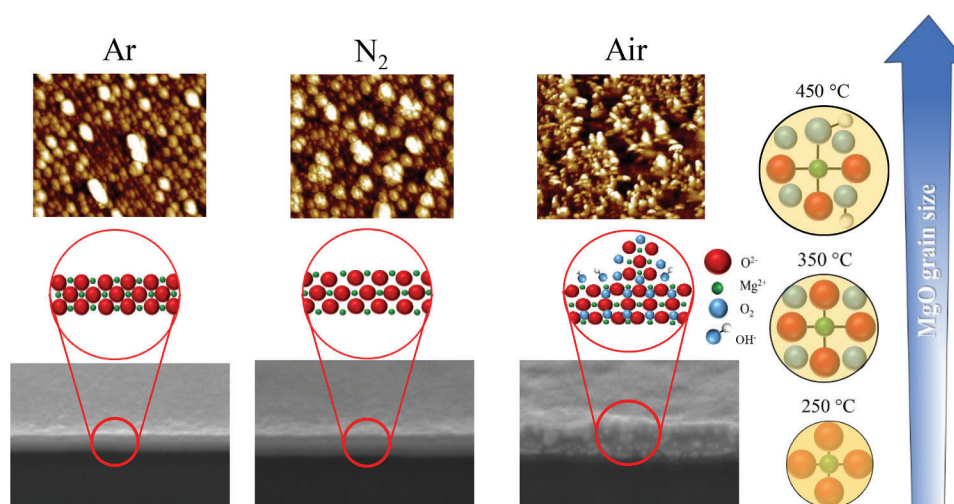


Fig. 9. Mechanism of annealed MgO thin film grain expansion and hydration reaction mechanism.

leads to the increase of the sputtering rate.

3.5. Grain expansion mechanism

This study indicates that gas environments and annealing temperatures influence MgO films' surface morphology, composition, and dielectric properties breakdown voltage. Fig. 9 shows the basic understanding and a schematic representation of the hydration reaction mechanism. MgO film easily reacts with the oxygen content in the atmosphere and initiates the hydration reaction within the film. Annealing temperature influences the oxygen content in the atmosphere and the hydration reaction rate. The oxygen content in Ar and N₂ gases is significantly less; hence the observed less hydration reaction (in all the temperatures). However, oxygen content in the air is adequate to initiate the hydration reaction for all temperatures. The larger MgO grains create more voids between grain boundaries and allow the water molecules in the air (in the form of moisture) into the gaps. The primary reason for this phenomenon is the change in film thickness caused by the oxygen concentration in the air flowing through the annealing chamber, which results in oxygen gas diffusion and adsorption on the thin film's surface [22,45].

Additionally, increasing annealing temperature increases the lattice constant of MgO, resulting in grain growth of MgO thin films [46–48]. As a result, films annealed in the air have a lower free carrier concentration than those annealed in inert gas. When annealing in an oxidant reduction environment, the grain boundaries act as carrier traps, creating a decrease/increase in carrier concentration [49].

4. Conclusions

In conclusion, the physical and chemical properties of the MgO films annealed at different temperatures in different gas environments were examined. The MgO film thickness slightly increases when annealed in Ar and N₂ gases. However, the film thickness is drastically increased after air annealing. The reason behind the increase in film thickness is hydration reaction-assisted grain growth. Oxygen content in the different annealing environments is the main contender for the hydration reaction of MgO films. The maximum surface roughness of air annealed films is due to the surface scatters that evolved due to drastic grain growth. The smooth surface of films is observed when annealed below 350 °C, and the roughness of the films increases after 450 °C. The FIB-SEM results revealed that film thickness increased twice of a pristine film when annealed air atmosphere at 450 °C. Although the film density decreases with the annealing temperature, the dielectric breakdown voltage is decreased as the temperature increases. The argon-annealed

film demonstrated the lowest reduction in dielectric breakdown voltage. In contrast, the air annealed thin film showed the most significant decrease in dielectric breakdown voltage, affecting the film shielding performance and signal output quality of the TMR sensors. In addition, the Ar annealing can reduce residual stress and humidity in MgO thin films rather than N₂ gas do. Therefore, the Ar annealing process could prevent the film thickness expansion, film surface roughness, and reduction of dielectric strength of MgO thin films.

CRediT authorship contribution statement

Mongkol Kongtungmon: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Visualization. **Laddawan Supadee:** Conceptualization, Resources. **Worasom Kundhikanjana:** Conceptualization, Methodology, Writing – review & editing, Data curation. **Pattanaphong Janphuang:** Investigation. **Ratchadaporn Supruangnet:** Investigation. **Suparat Tongpeng:** Investigation, Data curation. **Watcharin Jongpinit:** Visualization. **Dhanunjaya Munthala:** Conceptualization, Writing – review & editing. **Soodkhet Pojprapai:** Conceptualization, Methodology, Resources, Data curation, Writing – review & editing, Visualization, Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

The data that has been used is confidential.

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