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Synthesis of $MgO:Al^{3+}$ Thin Films for Optoelectronic Applications

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Abstract
$MgO:Al^{3+}$ were deposited onto glass substrates at 40°C using SILAR deposition technique. The Rutherford Backscattering Spectroscopy analysis shows that samples B_1 of 30 cycles and B_2 of 15 cycles and annealed at 250°C and 200°C have thicknesses 346.7 nm and 310.0nm respectively. The elemental compositions of samples B_1 and B_2 are 84.25.22% of oxygen, aluminum of 7.05%, magnesium 8.70% and 80.09% of oxygen, aluminum of 12.79 %, magnesium 7.12%. respectively. The high transmittance (>80%) of Al^{3+} doped MgO thin films suggest its promising material in magnetic memory devices and solar cell applications. The average band gap of the material is 3.05±0.05eV. Its high energy band gap makes it a good candidate for LEDs, FPDs for optoelectronics applications, smart windows and also a possible material applicable in the area of artificial intelligence (AI).

Introduction

MgO thin films are attracting great scientific and technological interest due to their important properties [1]. MgO thin films have important applications in optoelectronic devices, light-emitting diodes, photo-detectors, sensors and solar cells [3]. MgO has remarkable chemical and thermal stability, a wide band gap properties [2]. Doping is a useful technique which modifies the properties of host materials for specific areas of applications. Dopants such as Ag, Zn, Fe, Cr have been used to improve the properties of MgO thin films [5]. Al^{3+} being the doping element here is because of its availability, non-toxic and its cost effectiveness [9]. Al^{3+} being trivalent cations, which provides extra electron by substituting Mg that might improve the opto-electrical properties. The following works have been done on Al^{3+} doped MgO thin films. Payel Maiti *et al* [7] deposited Al^{3+} doped MgO films onto quartz substrate by spin coating technique to investigate the nano-mechanical and optical properties. They reported that their deposited film possesses higher transmittance, higher refractive index value as well as improved elastic modulus. The thin films can be deposited using several techniques such as chemical vapor deposition (CVD) [4], sol-gel [5], sputtering [6], hydrothermal synthesis [7], thermal evaporation [8], spin coating [10], spray pyrolysis [11] etc. In the work, Advanced SILAR method will be used to deposit Al^{3+} doped MgO thin films. The composition, thickness and optical properties of the films will be studied and possible areas of applications will be suggested.

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Materials and Method

The following materials are required to deposit successfully layers of MgO: Al thin films. Magnesium Chloride ($MgCl_3$), Aluminum sulphate ($Al_2(SO_4)_3$ Ammonia NH_3 (as complexing agent), Glass Slides, de-ionized water.

Preparation of Substrates

The substrates were soaked in concentrated solution of hydrochloric acid HCl and Nitric acid HNO_3 in the ratio of 3:1 for 48 hours and were washed with detergents and rinsed in de-ionized water and hanged in slanting order to air-dry. This is done in order to remove unwanted substances such as oil and grease and also to create the nucleation centers for easy and adherent deposition.

The Deposition of Magnesium Oxide Thin Films Doped with Aluminum

The synthesis of MgO thin films doped with 0.06M solution of Al^{3+} using SILAR method constituted: 5ml of 3M solution of ammonia as complexing agent, 3.7g of 0.16M solution of $MgCl_2$ dissolved in $100cm^3$ water and was made to react with H_2O_2 solution forming MgO:Al as depicts in Figure 1, which is the desired result.

The Chemistry and the Process of formation

4ml of 3M solution of ammonia was made to react with the solution of $MgCl_2$, forming magnesium tetra-amine complex ion as given in equations (1).

$$MgCl_2 + 4NH_3 \rightarrow [Mg(NH_3)_4]^{2+} + 2Cl^- \quad (1)$$

De-ionized water was added up to 50ml and the solution was stirred vigorously in order to achieve uniformity in the mixture.

MgO:Al thin films were deposited on substrates in cycles, by dipping the substrates into the beaker containing the cationic precursor of magnesium tetra-amine complex ion and then rinsed in a beaker of de-ionize water, then immersed into the third beaker, containing H_2O_2 solution as anionic precursor, at elevated temperature of 40^0C, the substrates were rinsed in de-ionized water and finally dipped in a beaker containing aluminum complex ion and was rinsed in de-ionized water, after successive immersion and this is repeated based on the number of cycles. The reactions are given in equations (2), (3) and (4). The parameters for SILAR deposition are given in Table 1.

$$[Mg(NH_3)_4]^{2+} + H_2O_2 \rightarrow MgO + H_2O + 4NH_3 \quad (2)$$

$$[Al(NH_3)_4]^{3+} + 2MgO + 2H_2O \longrightarrow Mg_2AlO_3 + 4NH_3 + H_2O \uparrow \quad (3)$$

$$Mg_2AlO_3 + H_2O \xrightarrow{H_2O} Mg_2AlO_3 \quad (4)$$

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Table 1 The deposition of MgO thin films at 40^{0}C

Sample	Dip-time(s) in each reactant	No. of cycle	Dip-time(s) in each Beaker of H_2O
B_1	8	30	4
B_2	8	15	4
B_3	8	20	4
B_4	8	25	4
B_5	8	28	4

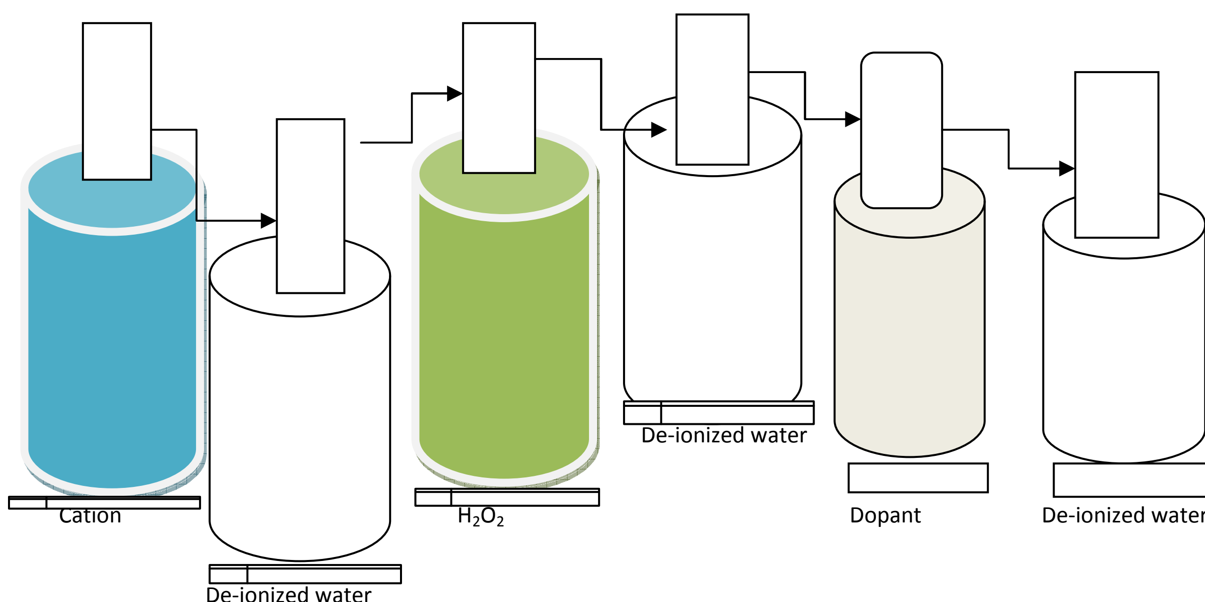

Figure 1 The Setup

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Results and Discussion

4.1 Thickness and Composition measurements

It is often necessary to determine the elements that make up the thin film samples. In this work, atomic compositions were determined, by Rutherford backscattering Spectroscopy (RBS) analysis.

The Rutherford backscattering analysis shows that the sample B_1 has thickness 346.7 nm and sample B_2 has 310.0 nm. Figure 2 and Figure 3 revealed that samples B_1 and B_2 annealed at 250°C and 200°C respectively have 84.25.22% of oxygen, 7.05% of aluminum, 8.70% of magnesium and 80.09% of oxygen, 12.79 % of aluminum, 7.12% of magnesium.

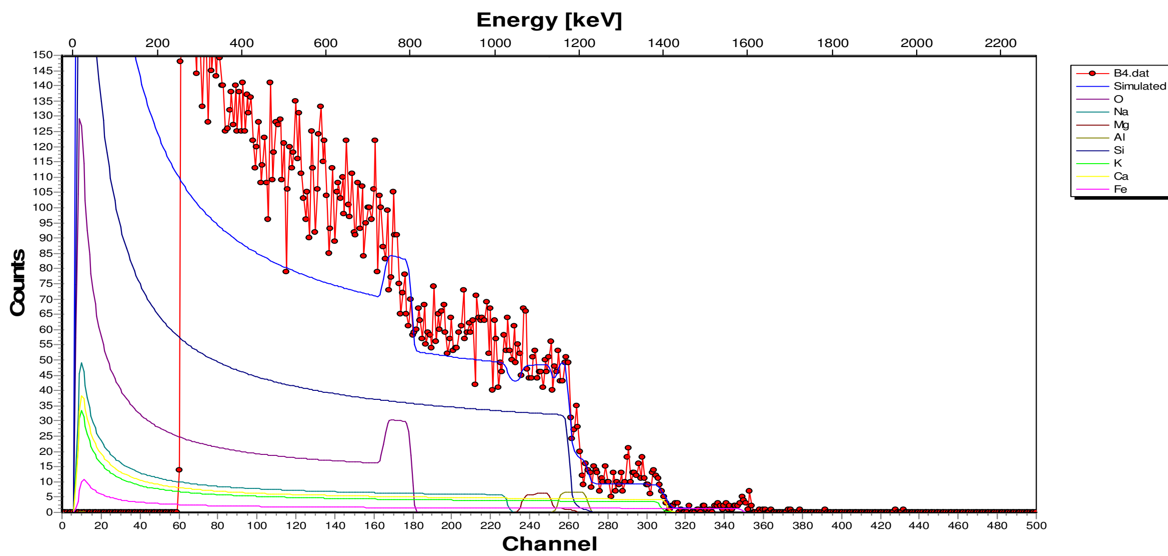

Figure 2 Elemental Composition of Sample B_1

LAYER 1: THICKNESS 346.7 nm

Compo: Mg 8.70% Al 7.05 % O 84.25%

LAYER 2 THICKNESS 5000.0 nm

Compo: Si 35.19% O 51.02% Na 9.81% Ca 2.16% Al 0.02% K 1.41% Fe 0.39%

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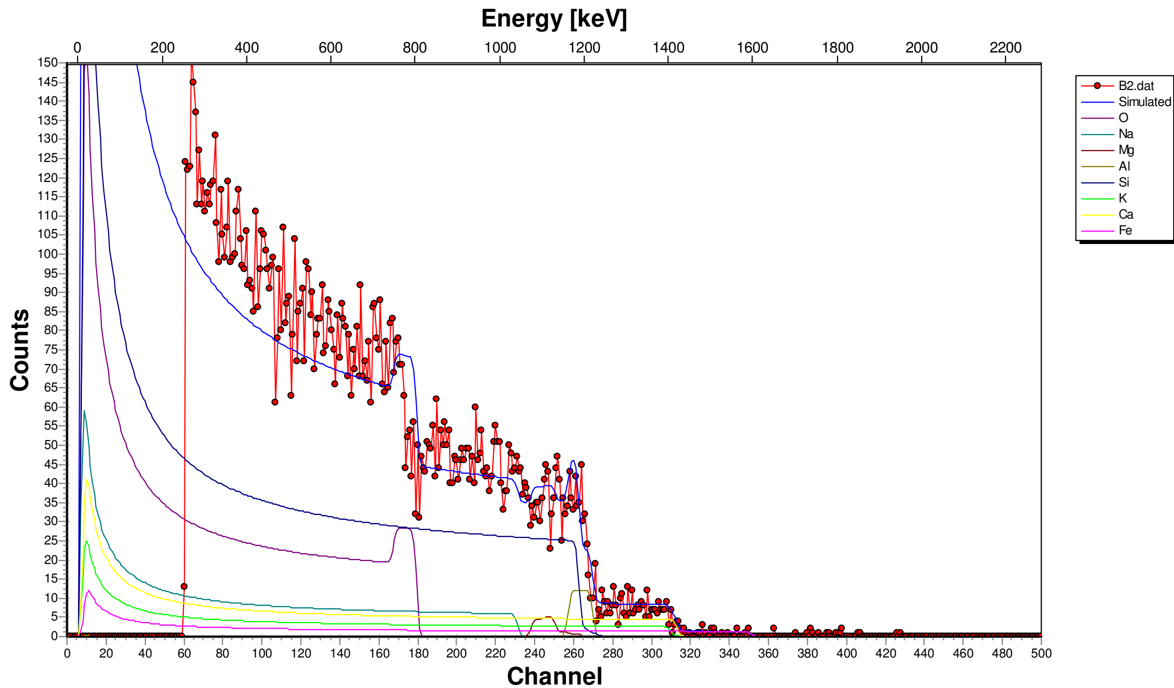

Figure 3 Elemental Composition of Sample B_2

LAYER 1: THICKNESS 310.0 nm

Compo: Mg 7.12% Al 12.79 % O 80.09%

LAYER 2: THICKNESS 5000.0 nm

Compo: Si 35.19% O 51.02% Na 9.81% Ca 2.16% Al 0.02% K 1.41% Fe 0.39%

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Table 2 **Transmittance of Sample B_1 and B_2**

Wave length	Sample B_1	Sample B_2
350	0.8222	0.9117
400	0.91829	0.98692
450	0.92758	0.98273
500	0.94061	0.9873
550	0.93958	0.98253
600	0.93755	0.97858
650	0.93335	0.97284
700	0.92726	0.96683
750	0.91937	0.95889
800	0.91335	0.95308
850	0.90948	0.9494
900	0.90675	0.94704
950	0.90424	0.94489
1000	0.90097	0.94196
1100	0.89493	0.93672

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Transmittance

The optical transmission data in the wavelength range 350nm to 1100nm was measured using UVI double beam Spectrophotometer with serial number 1800. Samples B_1 and B_2 share similar characteristics as indicated on the graph. The samples have high transmittance (>80%) in all regions of electromagnetic spectrum. With the high transparency exhibited by the material, it can be made use as transparent electrode for flat panel displays (FPDs). It also can be used as smart windows in infrared optics, since it has high transmittance in near infrared region.

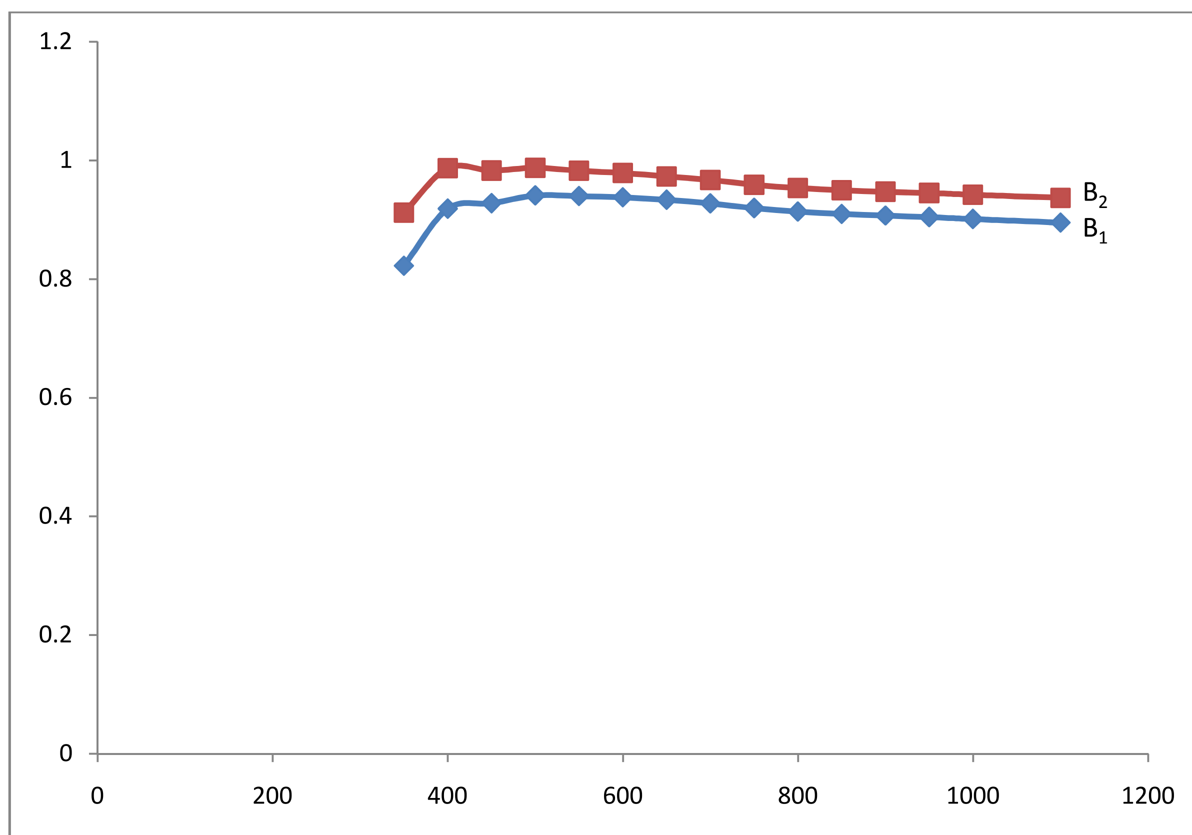

Fig 4 graph of transmittance against wavelength.

Table 3 **shows the band gap values and absorption coefficients**

hv (eV)	**α1^2**	**α2^2**
3.552	3.189E+11	8.893E+10
3.108	6.045E+10	1.804E+09
2.763	4.702E+10	3.158E+09
2.486	3.119E+10	1.700E+09
2.26	3.231E+10	3.232E+09
2.072	3.459E+10	4.879E+09
1.913	3.958E+10	7.890E+09
1.776	4.745E+10	1.184E+10
1.658	5.879E+10	1.834E+10
1.554	6.834E+10	2.403E+10
1.4625	7.490E+10	2.806E+10
1.381	7.972E+10	3.081E+10
1.309	8.430E+10	3.344E+10
1.243	9.047E+10	3.720E+10
1.131	1.025E+11	4.447E+10

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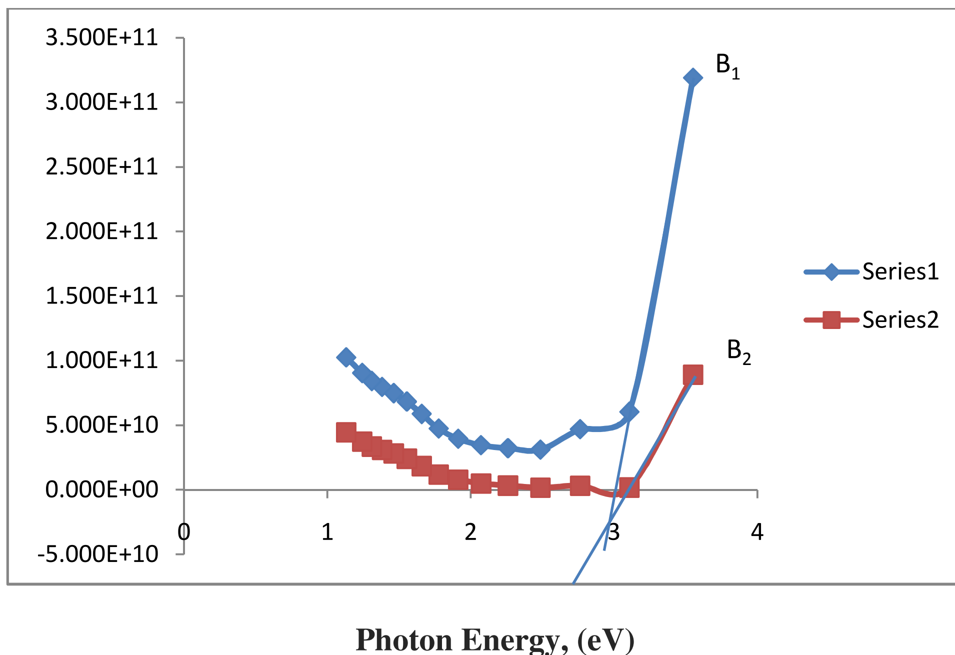
4.2.7 Energy band gap (E_g)

In band structure of solids, the band gap generally refers to the energy difference between the top of the valence band and the bottom of the conduction band in insulators and semiconductors. This is the range where no electron state exists. It is the energy required to free outer shell electron from its orbit about the nucleus to become a mobile charge carrier, able to move freely in solid materials. The band gap is determined in the graph of $(\alpha hv)^2$ against hv, by extrapolating the straight portion of the curve where αhv = 0. B_1 is found to be 3.0±0.05eV and B_2 is found to be 3.1± 0.05eV. It can be used as good material for LEDs, FPDs for optoelectronics applications, smart windows and also a possible material applicable in the area of artificial intelligence (AI).



Photon Energy, (eV)

Fig 5 graph of $(\alpha hv)^2$ $(eV/m)^2$ against hv(eV) for two samples.

Conclusion

The major area of interest is the deposition process in advanced SILAR technique. The synthesis of the samples, shows that the larger the number of cycles, the larger the thickness. Annealing effects may or may not have strong effects on the samples. Rutherford back scattering was used in determining the atomic composition and the thicknesses of the samples. The optical properties were measured using UV1800 series double beam spectrophotometer. Al^{3+} doped MgO thin films were deposited successfully at 40 °C onto glass substrates by SILAR technique. The average optical band gap of $MgO:Al^{3+}$ has been found to be 3.05eV. Thus the obtained results indicate that Al^{3+} doping modifies the optical properties of the MgO thin films significantly with abundant applications ranging from FPDs, LEDs/LCD for electronic/optoelectronic, solar conversions and smart mirror uses.

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