

## PLD DEPOSITED $\text{Al}_2\text{O}_3$ THIN FILMS FOR TRANSPARENT ELECTRONICS

M. ION<sup>a,b</sup>, C. BERBECARU<sup>a</sup>, S. IFTIMIE<sup>a</sup>, M. FILIPESCU<sup>c</sup>,  
M. DINESCU<sup>c</sup>, S. ANTOHE<sup>a,d,\*</sup>

<sup>a</sup>*Faculty of Physics, University of Bucharest, Atomistilor 405, Magurele-Ilfov, 077125, Romania,*

<sup>b</sup>*National Institute for Research and Development in Microtechnologies 126A, Erou Iancu Nicolae Street, 077190, Bucharest, ROMANIA*

<sup>c</sup>*National Institute for Laser, Plasma and Radiation Physics, Str. Atomistilor, Nr. 409 PO Box MG-36, 077125 Magurele, Bucharest Romania*

<sup>d</sup>*Horia Hulubei Foundation, Atomistilor 407, Magurele-Ilfov, 077125, Romania*

Alumina ( $\text{Al}_2\text{O}_3$ ) films of 800 nm thicknesses were obtained by pulsed laser deposition method. The chemical composition and the morphology of as grown films were investigated by Energy Dispersive X-ray spectroscopy (EDX), Scanning Electron Microscopy, Atomic Force Microscopy and optical transmittance spectroscopy (OTS) techniques. EDX results show a stoichiometric transfer between target and substrates and the bandgap energy and refractive index obtained by OTS were found to be in good agreement with the bulk values. Dielectric characterizations of  $\text{Al}_2\text{O}_3$  films were performed, in a sandwich structure using titanium (Ti) as back electrodes (100 nm) and aluminum (Al) as top electrodes (100 nm). The temperature dependence of permittivity and dielectric losses of Ti/ $\text{Al}_2\text{O}_3$ /Al capacitors have been measured between 123 - 423 K at selected frequencies in the 42 Hz - 5 MHz ranges. The dielectric constant of  $\text{Al}_2\text{O}_3$  determined from capacitance measurements was found to be around 8.3 at 273 K. The temperature and frequency dependence of both the permittivity and dielectric loss values are weak in the investigated temperature and frequencies ranges.

(Received September 13, 2012; Accepted October 16, 2012)

*Keywords:* Alumina thin films, Pulsed Laser Deposition, Dielectric properties.

### 1. Introduction

Dielectric films with a large relative permittivity are of great importance in a variety of electronic applications, based on metal-oxide-semiconductor (MOS) technology. In the race to find a better replacement for silicon dioxide in MOS devices, oxides of hafnium and aluminium have emerged as leading candidates [1]. Robertson and Peacock have shown that alumina ( $\text{Al}_2\text{O}_3$ ) has excellent conduction and valence band offsets on contact with Si [1, 2]. This makes  $\text{Al}_2\text{O}_3$  ideally suited for use with *n*- and *p*-type Si over a range of doping levels. There is however a disadvantage in using  $\text{Al}_2\text{O}_3$  in MOS structures, as compared to other high-*k* dielectrics: its intermediate dielectric constant limits the capacitance density [3].

In spite of that,  $\text{Al}_2\text{O}_3$  has many interesting physical properties that make it suitable for thin film applications. Because of their good transmittance in the visible region and chemical stability,  $\text{Al}_2\text{O}_3$  films are widely used as antireflective and protective coatings [4] and also for transparent electronic device applications [5]. Different techniques were used to produce alumina thin films: atomic layer deposition [6-8], chemical vapour deposition [9], magnetron sputtering, and pulsed laser deposition (PLD).

---

\* Corresponding author: [santohe@solid.fizica.unibuc.ro](mailto:santohe@solid.fizica.unibuc.ro)

Here we report our results on producing alumina thin films with good chemical homogeneity and surface morphology by PLD in a reactive atmosphere. As the physical properties of the  $\text{Al}_2\text{O}_3$  films depend strongly on their microstructure, scanning electron microscopy (SEM) and atomic force microscopy (AFM) data of the surface are presented. Dielectric characterizations (permittivity and dielectric losses) of the films were performed in a temperature range of 123 K – 423 K. The results of optical characterizations are also presented and discussed.

## 2. Experimental details

800-nm films of alumina were deposited by PLD either directly on optical glass substrates (for optical characterizations) or on a 100 nm evaporated Ti layer deposited on glass substrates. The optical glass substrates were previously cleaned with acetone, isopropyl acid and distilled water. In the case of those used for dielectric characterization, an argon plasma treatment was performed in order to obtain a good adhesion of the metallic layer.

PLD is a relatively simple technique, widely used in obtaining various materials, including complex oxides. It allows for the possibility to produce ultrathin films from materials that are difficult to be processed by other methods. This technique is currently used for deposition of a large class of materials, including superconductors, piezoelectric, ferroelectric or biocompatible materials.

The films were deposited in a PLD system containing an ArF laser operating at the wavelength of 193 nm. The laser radiation is focused on the mounted target inside the reaction chamber at  $45^\circ$  angle of incidence. The substrate holder is mounted in a parallel plane to the target surface and can be electrically heated up to temperatures of  $800^\circ\text{C}$ . The deposition chamber, shown schematically in Fig.1, was initially evacuated to a pressure less than  $10^{-5}$  mbar. The deposition conditions were collected in Table I.

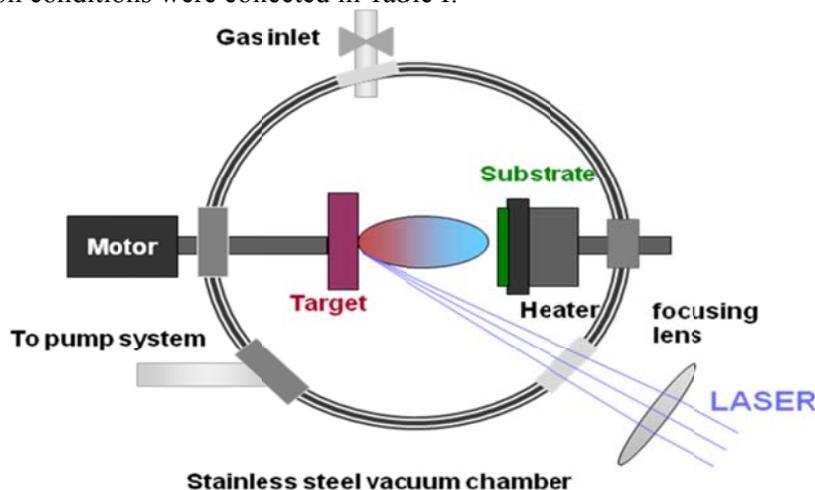


Fig.1 Schematic figure of the laser ablation chamber.

Table 1 Alumina PLD deposition parameters

Laser	Used wavelen gth(nm)	Angle of incidence	Distance (cm)	Fluence (J/cm <sup>2</sup> )	Energy (mJ)	Pulses	Preliminary pressure (mbar)	Oxygen pressure (mbar)	Deposition rate (Å/pulse)
ArF	$\lambda=193$	$45^\circ$	4	4	22	384.000	$2 \times 10^{-6}$	$5 \times 10^{-5}$	0.02

To perform the characterization of prepared dielectrics, a second aluminium electrode was deposited on top of some of the PLD deposited  $\text{Al}_2\text{O}_3$  films. Prior the deposition of the top electrode, the films were thermally treated at  $300^\circ\text{C}$  for 30 min. Dielectric measurements were carried out on Ti/ $\text{Al}_2\text{O}_3$ /Al capacitors, by using a RLC automatic bridge (Hioki-3532).

The morphology of the film surface was analyzed by SEM and AFM (in non-contact mode). Optical transmission spectra were measured at room temperature, by using a Perkin-Elmer Lambda 35 spectrometer.

### 3. Results and discussions

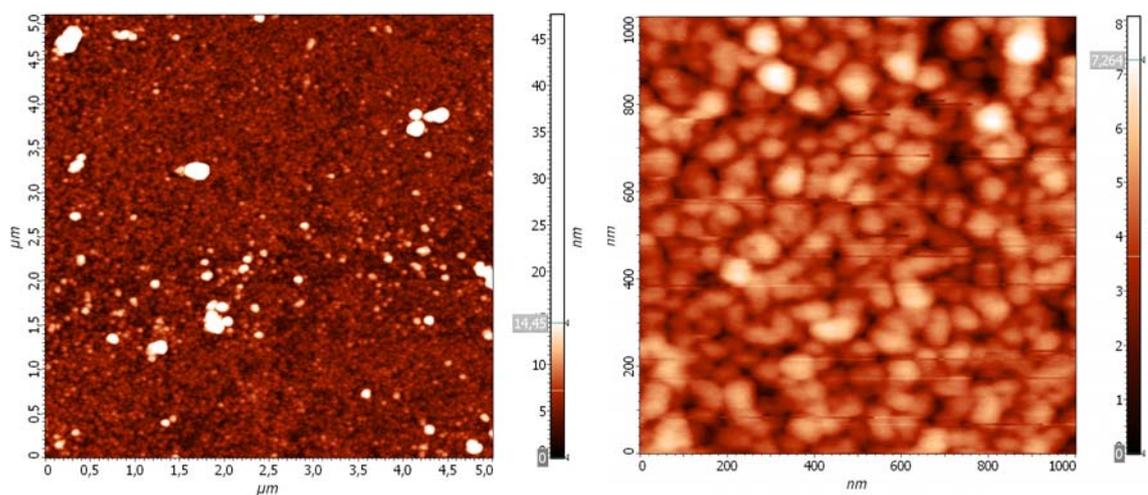


Fig.2 AFM images obtained for PLD alumina (800 nm).

The AFM images (fig. 2) of the alumina thin films deposited at room temperature by PLD, and the SEM images (fig.3) of the same films show an average diameter of grains of about 50 nm.

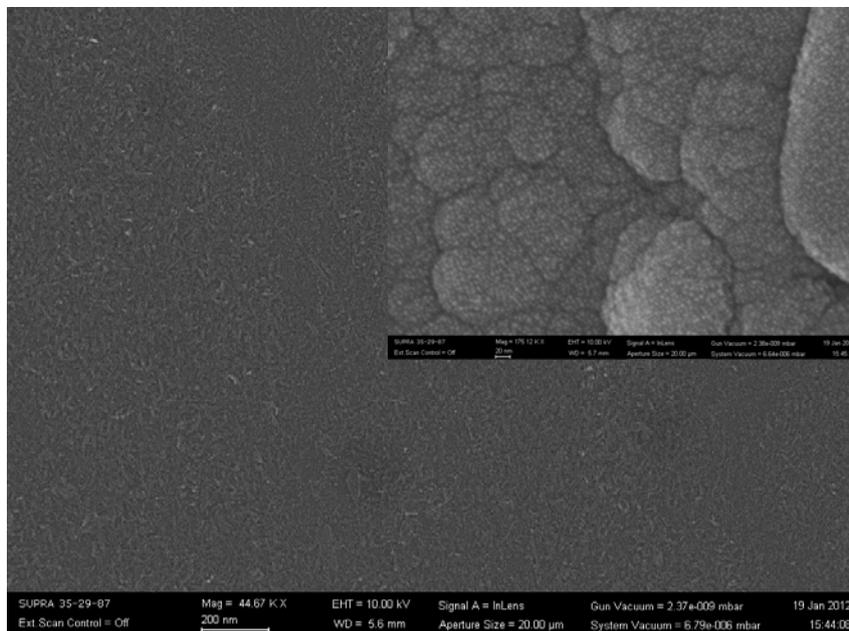


Fig.3 SEM images obtained for PLD alumina (800 nm)

The parameters describing the surface morphology, as extracted from AFM scans, were collected in Table 2. The surface has a typical roughness for PLD, and is relatively flat, with extreme peaks (as indicated by the value greater than one of  $S_{sk}$ ), characterized by a sharp distribution, as revealed by the large value of  $S_{ku}$ .

Table 2 Alumina PLD AFM parameters

Root Mean Square, Sq	2.7 nm
Surface skewness, Ssk	6.0
Coefficient of kurtosis, Sku	55.8

Energy Dispersive X-ray spectroscopy (EDX) results (Fig.4) show a stoichiometric transfer from target to film.

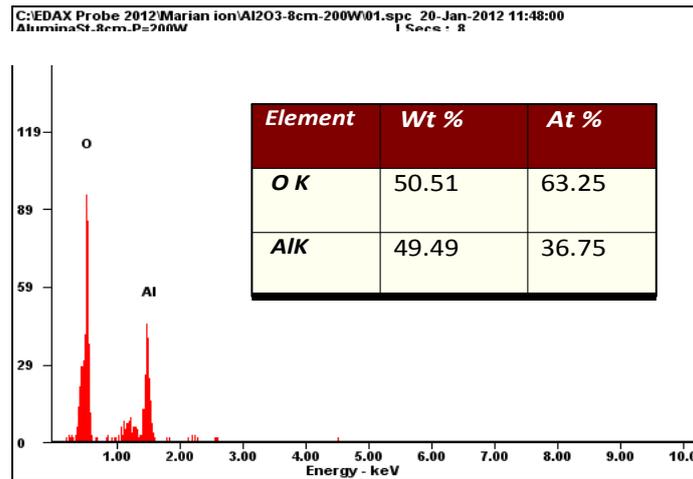


Fig.4 Results of EDX investigation of a PLD alumina film (800 nm thick).

Optical properties of the films were characterized by recording their transmittance spectra, shown in Fig. 5. In the visible region, the average transmittance of the  $\text{Al}_2\text{O}_3$  films on glass varies from about 84% to 90%. The sharp absorption edge in the wavelength range 280-300 nm is due to the glass substrate.

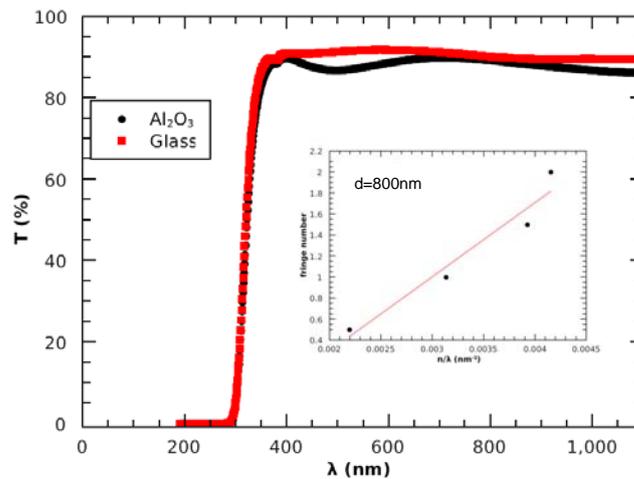


Fig.5 Transmittance spectra of  $\text{Al}_2\text{O}_3$  films and glass (inset- thickness for  $\text{Al}_2\text{O}_3$  film)

The dispersion of refractive index  $n$  was extracted from fringes data in the transmitting region using Swanepoel's theory [10,11], and is shown in Fig.6.

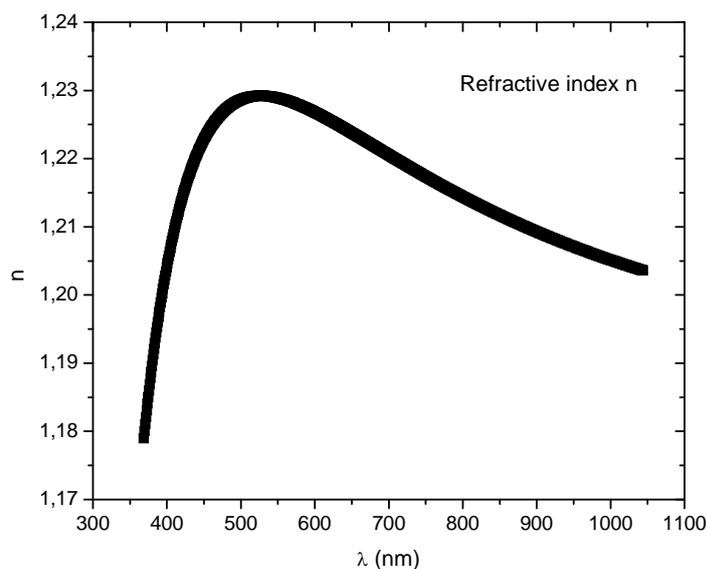


Fig 6 Refractive index dispersion of the alumina thin films.

For dielectric characterizations,  $\text{Al}_2\text{O}_3$  films were deposited on Ti electrodes (100 nm). After thermal treatments at 300 °C for half hour, a top Al electrode was deposited and dielectric measurements were carried out on Ti- $\text{Al}_2\text{O}_3$ -Al capacitors, by using a RLC automatic bridge (Hioki-3532). The temperature dependence of permittivity and dielectric losses of capacitors were measured in the range 123 - 423 K, at selected frequencies in the 42Hz - 5MHz domain. The dielectric constant of  $\text{Al}_2\text{O}_3$  determined from capacitance measurements was found to be around 7.2 at 273 K. Lower dielectric losses values, fewer than 1 %, were found at 10 kHz. A good stability of the permittivity and loss values was found in the investigated temperature and frequency ranges.

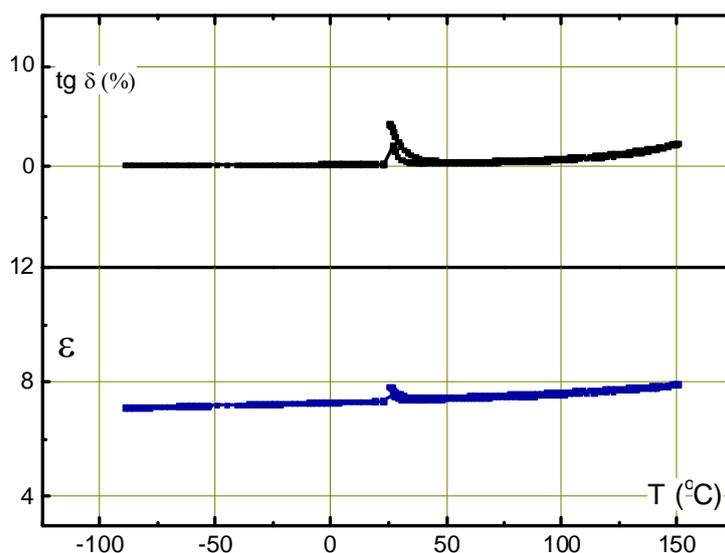


Fig 7 Temperature dependence of permittivity and dielectric losses of capacitors in the range 123K – 423 K (10 kHz).

#### 4. Conclusions

Al<sub>2</sub>O<sub>3</sub> films, 800 nm thick, were successfully obtained by pulsed laser deposition (PLD) method. The films show a good chemical composition as demonstrated by EDX data, and a relatively smooth surface, characterized by the presence of some extreme grains with sharp distributions.

The dielectric constant of alumina films determined from capacitance measurements, was found to be around 7.2 at 273 K and increases monotonically with the temperature in the range 123 - 423 K. Lower dielectric losses values under 1 % were found at 10 kHz.

#### References

- [1] J. Robertson, *J. Vac. Sci. Technol. B* **18**, 1785 (2000).
- [2] P. W. Peacock and J. Robertson, *J. Appl. Phys.* **92**, 4712 (2002).
- [3] R. Ravindran, K. Gangopadhyay, S. Gangopadhyay, N. Mehta, N. Biswas, *Appl. Phys. Lett.* **89**, 263511 (2006).
- [4] J. Tauc, in: *Amorphous and liquid semiconductors*, edited by J. Tauc. Plenum, New-York, 1974;
- [5] F. Di Fonzo, D. Tonini, A. Li Bassi, C.S. Casari, M.G. Beghi, C.E. Bottani, D. Gastaldi, P. Vena, R. Contro, *Appl. Phys. A* **93**, 765 (2008).
- [6] S.M. George, *Chem. Rev.*, **110**, 111 (2010).
- [7] R.L. Puurunen, *J. Appl. Phys.* **97**, 121301 (2005)
- [8] G.S. Higashi, C.G. Fleming, *Appl. Phys. Lett.*, **55**, 1963 (1989).
- [9] F. S. Ohuchi and R. H. French, *J. Vac. Sci. Technol. A* **6**, 1695 (1988).
- [10] R. Swanepoel, *J. Phys. E: Sci. Instrum.* **16**, 1214 (1983).
- [11] R. Bazavan, L. Ion, G. Socol, I. Enculescu, D. Bazavan, C. Tazlaoanu, A. Lorinczi, I.N. Mihailescu, M. Popescu, S. Antohe, *J. Optoelectron. Adv. Mater.* **11**, 425 (2009).