

PROPERTIES OF BARIUM MAGNESIUM TANTALATE THIN FILMS PRODUCED BY PULSED LASER DEPOSITION

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Abstract. Barium magnesium tantalate (BMT), a complex perovskite oxide, has garnered interest because of promising dielectric properties, making it a candidate for advanced electronic applications. In this study, BMT thin films were deposited on various substrates by pulsed laser deposition (PLD) using solid-state Nd: YAG laser (266 nm) on substrates of Si, Pt/Si and Al₂O₃ under various substrate temperatures and oxygen partial pressures to assess the impact of these conditions on film properties. The techniques employed for the analysis of the thin films are: Spectroscopic ellipsometry (SE), Scanning electron microscopy (SEM), Energy Dispersive X-Ray (EDX). The optical constants determined using SE exhibited dependence on deposition parameters higher temperatures of substrate during the deposition yielded thin films with higher refractive indices. Moreover, substrate type played a critical role in film-substrate interface quality, influencing film texture and optical response and bandgap. The BMT thin films deposited on Pt/Si have better uniformity and structure, making them more suitable than Si or Al₂O₃ for BMT growth, and have refractive indices (*n*) of 1.82-2.29 at a wavelength of 589nm, direct transition bandgaps of 3.77-4.15 eV and relative permittivity between 25.6 up to 62.5. The findings highlight the tunability of BMT thin film optical properties through careful control of PLD parameters showcasing correlations between growth conditions and functional properties.

Key words: BMT, dielectric, thin films, pulsed laser deposition, material properties

1. INTRODUCTION

The increasing integration of high-performance electronics and optoelectronics in compact formats has driven demand for materials exhibiting stable dielectric constants, low loss tangents, robust optical constants, and high thermal endurance. Complex perovskite oxides with the general formula $A(B'_{1/3}B''_{2/3})O_3$ are particularly promising due to their property tunability via cation substitution and processing control [1]. Among them, Barium Magnesium Tantalate ($Ba(Mg_{1/3}Ta_{2/3})O_3$, BMT) stands out for its high quality factor dielectric material with minimal dielectric loss and chemical and thermal stability—making it an excellent candidate for microwave dielectric resonators and filters [2] [3].

BMT, having the characteristic complex perovskite structure, allows partial substitution and cation ordering between Mg^{2+} and Ta^{5+} to change its properties and behavior [4]. The presence or absence of ordering leads to one of two different phases of its structure: cubic or hexagonal [5]. This ordering, in turn, affects and offers modulation of the optic and dielectric properties, being highly sensitive and dependent to the temperature of processing environment during material synthesis [6]. The high quality factor of BMT, with a constant electric permittivity over a wide range of frequencies, minimal dielectric loss and thermal stability made BMT be used in satellite communications and high precision RF filters [7]. The objective of this research is to give an insight about properties of BMT thin films deposited using PLD under various conditions and their dielectric properties, their topography, their optical constants, bandgaps and element distribution across multiple samples created using different substrates and deposition parameters by analyzing the samples at structural and functional levels using various characterization methods such as dielectric spectroscopy (DS), scanning electron microscopy (SEM), spectroscopic ellipsometry, Tauc plots and Energy Dispersive X-Ray (EDX) Spectroscopy. This way the foundational understanding of BMT's properties changes can be leveraged to help in the future designs for high-performance microwave dielectric devices.

In this study, BMT thin films were synthesized using Pulsed Laser Deposition (PLD), a method known for its stoichiometric transfer, crystallographic fidelity and compatibility with various substrates [8]. The substrates picked for deposition were the following : Si chosen for it's widespread use in microelectronics [9] being a standard for this industry [10] and the particular silicium used for deposition has crystalline structure [11] giving an insight to film growth without lattice matching; Pt/Si which was selected because of its oriented surface which might offer better lattice alignment [12] and Al_2O_3 due to being the most popular ceramic substrate with low processing costs and excellent mechanical and electrical wear properties [17]. The investigation explores how substrate type, deposition temperature, and oxygen pressure influence the optical constants, dielectric behavior, and microstructure of BMT films. These findings contribute to understanding how processing conditions affect performance, enabling the optimization of BMT films for advanced dielectric applications.

2. MATERIALS AND METHODS

2.1 THIN FILM SYNTHESIS BY PULSED LASER DEPOSITION (PLD)

BMT thin films were deposited by PLD using a solid-state Nd:YAG laser (266 nm), pulse rate 10 Hz, spot size 1 mm², and 36,000 pulses. Substrate-target distance was 5 cm. Films were grown on Si, Pt/Si, and Al_2O_3 substrates at substrate

temperatures of 500 °C or 700 °C and oxygen partial pressures of 0.1 mbar or 0.2 mbar with a total of 10 different samples. The chamber base pressure before oxygen introduction was $\sim 10^{-5}$ mbar.

2.2 STRUCTURAL, OPTICAL AND DIELECTRIC CHARACTERIZATION TECHNIQUES

- **Scanning Electron Microscopy (SEM):** Surface morphology and grain structure were examined using high-resolution SEM imaging by using Thermo Fisher Scientific Scios Dual Beam FiB-SEM.
- **Energy Dispersive X-ray Spectroscopy (EDX):** Elemental composition and homogeneity were assessed via EDX mapping and quantification.
- **Spectroscopic Ellipsometry:** Optical constants (n, k) and bandgap were derived over for a wavelength range of 230–830 nm using a HORIBA UVISSEL PLUS ellipsometer. Models employed Tauc-Lorentz and New Amorphous functions.
- **Dielectric Spectroscopy (DS):** Relative permittivity and dissipation factor were determined for films on Pt/Si in the 1 kHz–1 MHz range using Agilent 4294A impedance analyzer.

3. RESULTS AND DISCUSSION

3.1 SURFACE MORPHOLOGY AND ELEMENTAL COMPOSITION

SEM revealed substrate-dependent grain structure.

- **Si substrates:** Films exhibited grain with sizes 30–60 nm and island-like structures evenly distributed across samples with sizes ranging from 100nm and up to 1 μ m.
- **Pt/Si substrates:** Films were uniform, dense, and fine-grained (~ 20 –30 nm), with minimal porosity.
- **Al₂O₃ substrates:** Films showed larger polyhedral grains (300–800 nm) with distinct voids at grain boundaries.

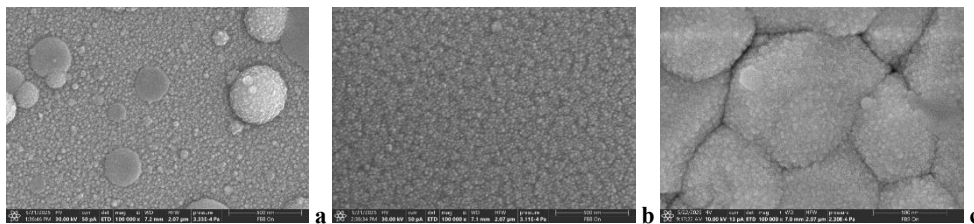


Fig. 1 – Topography of BMT thin films representatives for each substrate at a magnification of 100000: a) Sample 4, Si substrate; b) Sample 7, Pt/Si substrate c) Sample 9, Al₂O₃ substrate.

Films grown at 500 °C and 0.2 mbar showed cracks or enhanced porosity, particularly samples. Pt/Si substrates had the best growth of BMT films with little to no island-like structures which could be advantageous for dielectric applications.

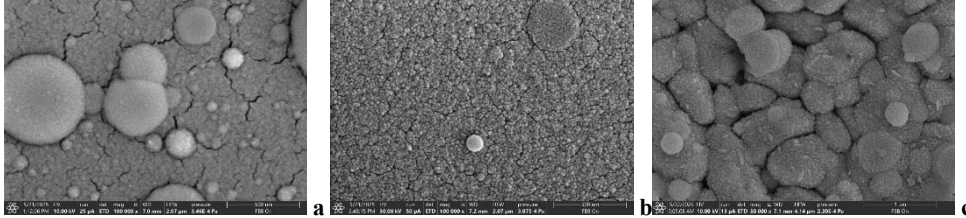


Fig. 2 – Topography of BMT thin films which exhibited cracks and enhanced porosity, all of them were prepared at 500°C temperature and 0.2 mbar pressure for each substrate: a) Sample 3, Si substrate; b) Sample 8, Pt/Si substrate c) Sample 10, Al₂O₃ substrate.

EDX confirmed uniform distribution of Ba, Mg, Ta, and O across films, with no elemental clustering or phase segregation. The Mg:Ta atomic ratios deviated from the ideal 1:2, typically in the range 1:2.4 to 1:4.5, suggesting minor Mg deficiency.

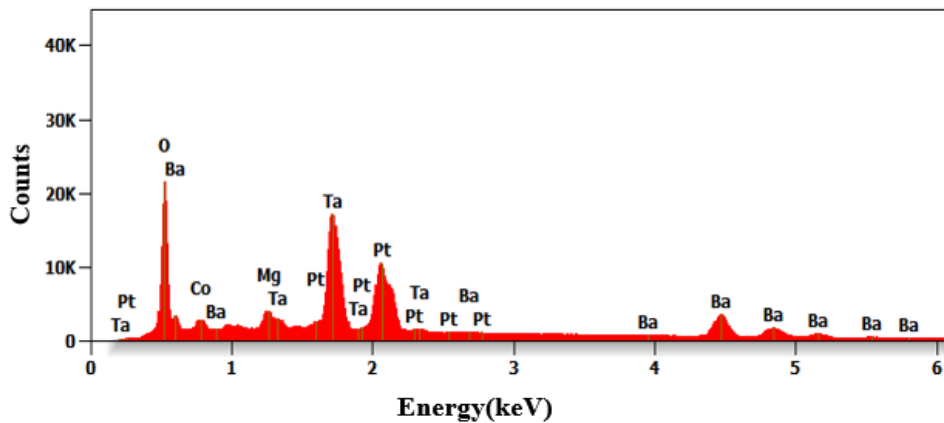


Fig. 3 – EDX spectrum for sample 7, representative of the BMT samples deposited on Pt/Si substrates

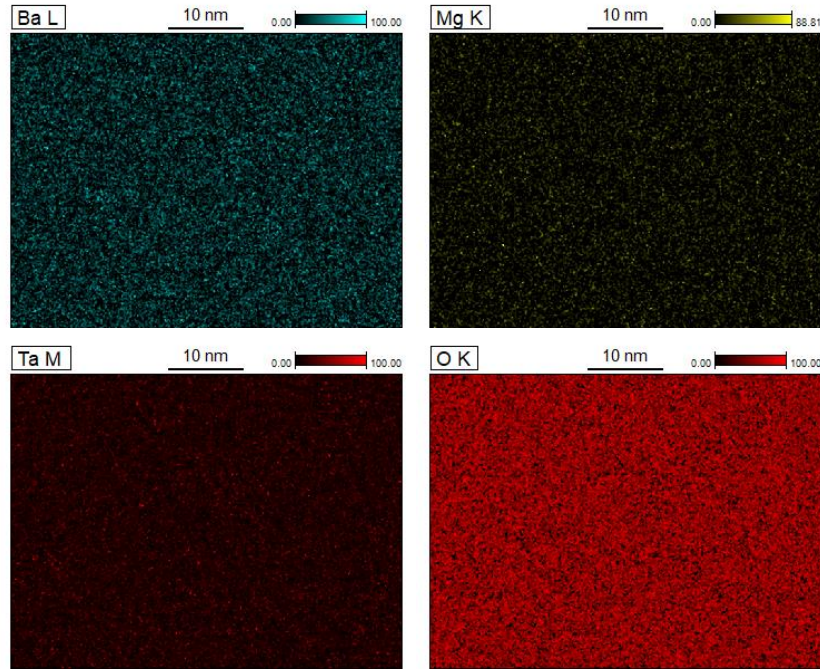


Fig. 4 - Chemical elements distributions of Ba(Mg_{1/3}Ta_{2/3})O₃ (BMT) thin film across sample 7, representative of the samples with Pt/Si substrates

Table 1

Characteristic X-rays of BMT identified in the EDX spectrum (excluding substrate elements which represent the rest to 100%) along with their weight and atom percentages for sample 7, representative of BMT samples deposited on Pt/Si substrates

Element	Extracted Spectrum	Extracted Spectrum	Extracted Spectrum	Extracted Spectrum
	Line Type	Net Counts	Weight %	Atom %
O K	K	101235	12.50	56.55
Mg K	K	16472	1.18	3.53
Ba L	L	63857	29.93	15.78
Ta M	M	185521	29.69	11.88
Total			73.3	87.74

3.2 OPTICAL PROPERTIES

The angles ψ (the amplitude ratio of the s and p polarized light components) and Δ (the phase difference between the p and s polarized light components) were measured and then mathematical models were created for each sample.

After fitting and overlapping each model with the measured data optical constants, thicknesses and bandgaps could be derived for each sample.

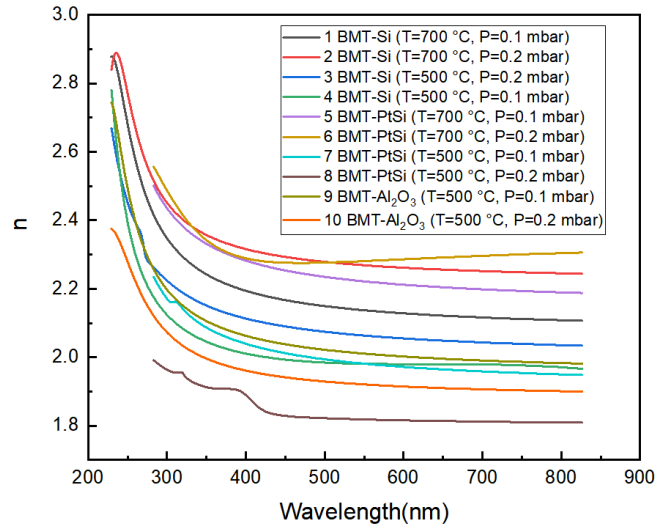


Fig. 5 – Refractive indexes n for the thin film samples grown on Si, Pt/Si, and Al₂O₃ substrates under different temperatures (500 °C and 700 °C) and pressures (0.1 mbar and 0.2 mbar)

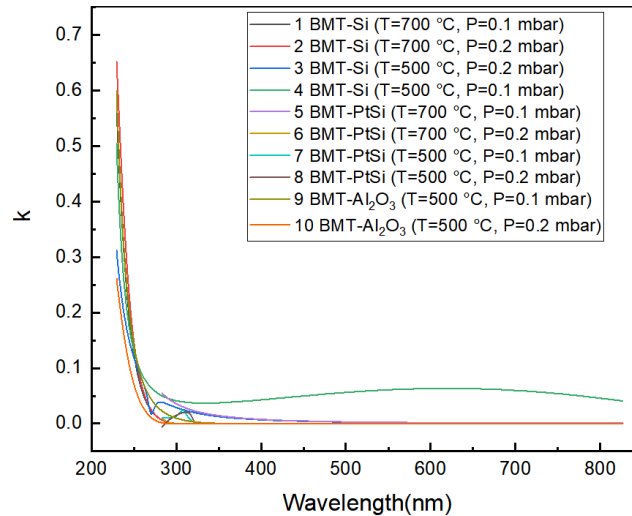


Fig. 6 – Extinction coefficient k for the thin film samples grown on Si, Pt/Si, and Al₂O₃ substrates under different temperatures (500 °C and 700 °C) and pressures (0.1 mbar and 0.2 mbar)

All films displayed refractive indices between 1.8–2.3 at 589 nm, consistent with literature values. Refractive index decreased with increasing wavelength, typical of dielectric materials, and was generally higher for films deposited at 700 °C.

Extinction coefficients were near-zero in the visible and near-IR, with the only exception being sample 4, confirming high transparency, but increased sharply below 275 nm in the UV.

Table 2

Values of the refractive index at the standard yellow doublet 589 nm, Thicknesses and bandgaps determined for each of the BMT thin films grown on Si, Pt/Si, and Al₂O₃ substrates

Sample	1	2	3	4	5	6	7	8	9	10
T (°C)	700	700	500	500	700	700	500	500	500	500
P (mbar)	0.1	0.2	0.2	0.1	0.1	0.2	0.1	0.2	0.1	0.2
Substrate	Si	Si	Si	Si	Pt/Si	Pt/Si	Pt/Si	Pt/Si	Al ₂ O ₃	AlO ₃
Eg (eV)	4.683	4.656	4.4871	4.736	3.77	4.15	3.791	3.814	4.738	4.617
Thickness (nm)	203	91.5	143.7	197.2	147.1	135.8	179.2	194.2	309.8	182.8
n (at 589 nm)	2.122	2.122	2.056	1.973	2.214	2.285	1.974	1.816	2.003	1.916

Bandgaps ranged from 3.77–4.78 eV, determined by Tauc plots assuming direct allowed transitions. Films on Pt/Si had bandgaps (3.77–3.81 eV) closest to the expected value for BMT (~3.55 eV)[13], indicating better crystallinity which could be a cause of the (111) oriented Pt of the Pt/Si substrate which has a lattice constant of 2.71 Å [14][15] while BMT has an interplanar distance of 4.08 Å [16] meaning that on top of 3 Pt crystals could grow 2 BMT crystals. Films on Si and Al₂O₃ often showed higher bandgaps, possibly due to nanoscale disorder or strain. Since the Pt/Si substrates had the best structural growth for BMT along with optical constants and bandgap values closer to the expected ones they were further investigated. The thickness of the films ranged from 91.5 nm up to 309.8 nm.

Zhang et al. (1999) reported very weak absorption below 1200 nm, implying a similar wide bandgap and reported refractive indices between 1.71-1.72 for PLD-grown BMT on fused silica, significantly lower than most values from this research, difference attributed to different substrate and possibly denser films [17].

3.4 DIELECTRIC PROPERTIES

Permittivity (ϵ_r): Values ranged from 25.6 (cracked film) to 62.5 (dense, uniform film) at 1 kHz, decreasing slightly with frequency. Dissipation factor ($\tan \delta$): Films with higher ϵ_r also exhibited higher losses, e.g., sample 5 showed $\epsilon_r \sim 62.5$ at 1 kHz and $\tan \delta \sim 0.09$, while sample 7 had $\epsilon_r \sim 25.6$ and $\tan \delta \sim 0.006$.

These trends highlight the trade-off between permittivity and dielectric loss, and the influence of microstructure on performance.

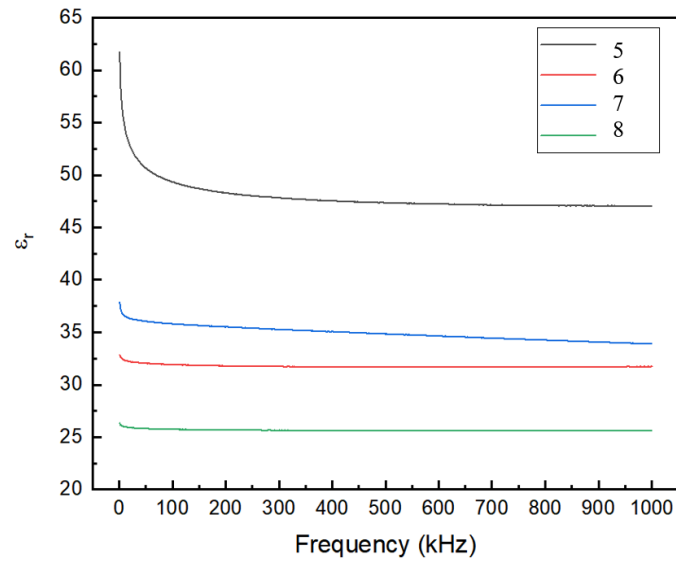


Fig. 7 – Frequency dependency of the relative permittivity for the BMT thin films deposited on Pt/Si substrates, samples 5-8.

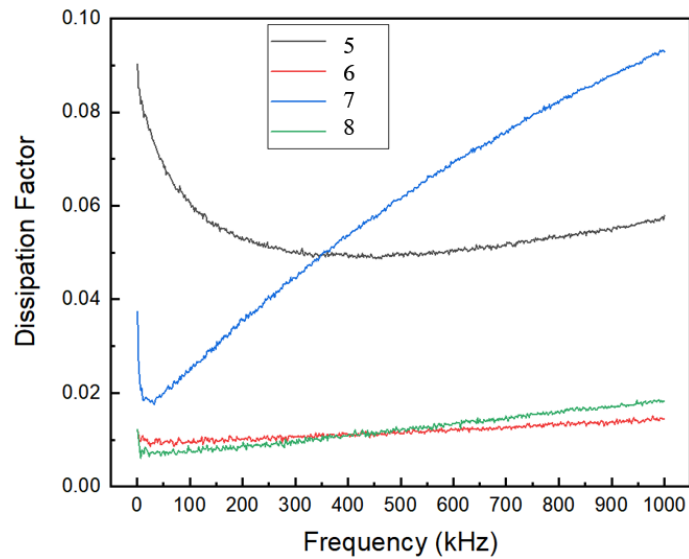


Fig. 8 Frequency dependency of the dissipation factor for the BMT thin films deposited on Pt/Si substrates, samples 5-8.

Between films which were deposited at the same chamber pressure the ones with higher substrate temperature during deposition had the higher dielectric constant values.

By comparison Scarisoreanu et al. reported a bandgap of ~ 4.72 eV for BMT films grown by RF-PLD assisted by plasma, aligning with the higher end of the bandgap range of the samples used for this investigation. This suggests that radiofrequency plasma assistance may induce similar disorder or densification effects as certain PLD conditions. The refractive indices n reported had values ranging from 1.78 up to 2.15, a dielectric constant of $\epsilon_r = 23$ and low transmission in UV which increases in the visible and near IR spectrum consistent with values consistent with determined in this study [19].

Sebastian & Surendran (2006) reported dielectric constants of BMT ceramics doped with glasses prepared by sintering as having values ~ 25 [20] and Busuioc et al. (2012) found dielectric constants around 18–23.5 for BMT thin films on Pt/Si and Al_2O_3 substrates prepared by sol-gel deposition, with some drop attributed to porosity, also showing that higher temperature and shorter times are preferred for annealing due to producing smoother denser surfaces free of cracks [5]. Another report of BMT properties is that of C. Jinga et al. investigating synthesized doped BMT ceramics showing that increasing dopant concentration and sintering temperature led to larger grain sizes and smoother surfaces for Nb^{5+} and V^{5+} doped samples. However, Ga^{3+} doped samples exhibited inhibited grain growth and higher porosity. This also had an effect on dielectric constants where the smoother denser surfaces obtained by doping with Nb^{5+} or V^{5+} had higher dielectric constants going up to 22.4 while the sample doped with Ga^{3+} had lower values and BMT without any dopants showed dielectric constants on the lower end out of the reported values with $\epsilon_r = 13$.

4. CONCLUSIONS

This study demonstrated that Pulsed Laser Deposition enables the fabrication of BMT thin films with tunable optical, dielectric, and structural properties by adjusting substrate type and deposition conditions. Pt/Si substrates produced the most uniform films with refractive indices (1.8–2.3) and bandgap values (3.77–4.15eV) closest to expected standards. Higher deposition temperatures enhanced refractive index and film density, while low temperature / high pressure conditions led to cracks or increased porosity. Dielectric measurements confirmed that films with higher permittivity exhibited greater dielectric loss, with values ranging from 25.6 to 62.5. Overall, the results highlight the critical role of substrate selection and deposition control for optimizing BMT films for microwave dielectric applications.

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