

Influence of the Annealing Temperature on the Thickness and Roughness of $\text{La}_2\text{Ti}_2\text{O}_7$ Thin Films

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Abstract

In this work, the impact of the substrate annealing temperature on the thickness and roughness of $\text{La}_2\text{Ti}_2\text{O}_7$ thin films was verified. A group of LTO Thin films was grown on Si (100) substrates successfully via pulsed laser deposition technique (PLD) at various annealing temperatures with a constant numbers of pulses and energy per pulse. Scanning Electron Microscope (SEM) and Atomic Force Microscope (AFM) were used to investigate the thickness and roughness of the deposited $\text{La}_2\text{Ti}_2\text{O}_7$ thin films. The average thickness of the thin films was decreased due to the increasing in the annealing temperature linearly; the maximum thickness was found (231 nm) when LTO thin film deposited at 500°C. The root mean square roughness was increased linearly with increasing the substrate Temperatures. The minimum roughness was found (0.254 nm) when LTO deposited at (500°C). From the obtained results, its clear evidence that the annealing temperature has an influence on the thickness and roughness of the LTO thin films.

Keywords

$\text{La}_2\text{Ti}_2\text{O}_7$ Thin Films, PLD, Perovskites, Annealing Temperature

1. Introduction

During the last few years, many experimental and theoretical studies were blessed to the study of perovskite materials because of their fascinating properties in-

cluding, ferroelectricity, superconductivity [1] [2] [3], pyroelectric and optical properties [4] [5]. Depending on these unique properties perovskite ceramics have distinct remarkable applications such as tunable microwave devices and piezoelectric devices [6] [7], sensors and wireless communications [8] [9], and some environmental applications [10]. Perovskites have the basic formula ABX_3 , where A and B are cations with A larger than that of B and X is often times oxygen but also other large ions are possible such as halides, sulfides and nitrides [11]. The perovskite structures exist in Layered perovskite, double perovskite, and Triple perovskite [12]. Lanthanum titanium oxide is a member of the layered perovskite family [13], which has high Curie temperature and excellent piezoelectric and electro-optic properties. This makes $La_2Ti_2O_7$ thin films a powerful candidate for a variety of applications in electrical and optical devices [14] [15]. The physicochemical properties of these materials are dependent on the crystal structure, lattice defect, exposed lattice plane, surface morphology, particle size, and specific surface area as well as the pore structure [16].

Many researchers were interested in examining the properties of perovskites thin films with different deposition techniques. In 2003, D. Todorovsky *et al.* introduced some properties of $La_2Ti_2O_7$ single-phase films using spray pyrolysis deposition technique, while in 2014 C. Paven *et al.* used RF magnetron sputtering to deposit $La_2Ti_2O_7$ in different substrates [17] [18]. Particularly in 2006 Ji Won and Sergei Orlov conducted an extensive parametric studies and defined the optimal condition for achieving the better surface quality of the $LiNbO_3$ thin films deposited by PLD. Followed by two years in 2008 Havelia and Balasubramanian used a pulsed laser deposition and offered a study on the influence of the substrate type on the growth morphology, phase selection of $La_2Ti_2O_7$ and $LaTiO_3$ thin films on $SrTiO_3$ (100) and $SrTiO_3$ (110) substrates respectively [19] [20].

Lately, many researchers investigated the effect of deposition temperature on thin films properties such as Yuli Xiong and Hai Zheng in 2011 reviewed the influences of annealing temperature against the structure, morphology, optical and electric properties of Lithium Lanthanum Titanate (LLTO) thin films prepared by RF magnetron sputtering [21]. While in 2018 Jianchao and Wanmin summarized the impact of deposition temperature on the refractive index and the extinction coefficient of $LaTiO_3$ thin films deposited by electron beam evaporation on Si and fused quartz substrates accompanied with other study of $La_2Ti_2O_7$ thin films as well in the same year [22] [23]. Obviously, increasing the deposition temperature and the variation of the thermal stability during the deposition is highly effective for the thin film properties. The properties of $La_2Ti_2O_7$ films deposited by Pulsed Laser Deposition under different temperature fields have not been summarized in the published literatures yet, so the impact of annealing temperature on the structure and roughness of LTO thin film were considered in this paper.

2. Experimental Methodology

Lanthanum titanium oxide powder was prepared using traditional solid-state

method, by mixing a stoichiometric amount 2:1 of (Aldrich 99.99%) TiO_2 and La_2O_3 (Aldrich 99.99%) powder. After grindings using a mortar and pestle, the mixture was successively calcined at 1400°C for 4 hours using High-Temperature Furnace (Delta Power Controls). In order to improve the synthesis efficiency, the calcined product was pelleted using uni-axial pressing (Kimaya Engineers). The formed pellet was then heat-treated in air at 1100°C for 8 hours with immediately grinding. For the deposition process, a 45.8% density target was obtained by uni-axial pressing of the LTO powder under 20 MPa followed by a sintering at (1350°C) for 10 hours in air. Then the sintered pellet was characterized by a (Rigaku) X-ray diffractometer and ultimately the LTO pellet was placed as a target in the film deposition process. Before the deposition, Si substrates were cleaned with RCA to eliminate all irrelevant objects from the surface and finally the substrate diced into (4 cm^2) pieces [24].

LTO thin film deposited on (100) Si substrates using PLD system as shown in **Figure 1** via a KrF excimer laser (COHERENT LMC; $\lambda = 248\text{ nm}$; $t \sim 20\text{ ns}$) with constant laser frequency (3 Hz) and fluency (200 mJ), together with a target substrate distance of 5 cm for 85 minutes under vacuum pressure $\sim 10^{-6}$ mbar at different annealing Temperatures 500°C , 600°C and 700°C , Separately.

The structural analysis taken using the Rigaku X-ray diffractometer technique (Cu $K\alpha$ radiation), while the thickness and the roughness of the thin films were obtained using a Ultra 55 scanning electron microscope SEM (ZEISS) and BRUKER Atomic Force Microscope (AFM using contact tip with a Rectangular with a length of 1 micrometer), respectively.

3. Results and Discussion

3.1. Sample Characterization

The XRD pattern of the LTO target at room temperature was analyzed and determined using X'Pert High Score Plus software which was classified into a monoclinic structure the lattice a : 13.0150 (\AA) , $b = 5.5456\text{ (\AA)}$ and $c = 7.8170\text{ (\AA)}$ which are fairly consistent with those stated in the literature as exhibited in **Figure 2** [26].

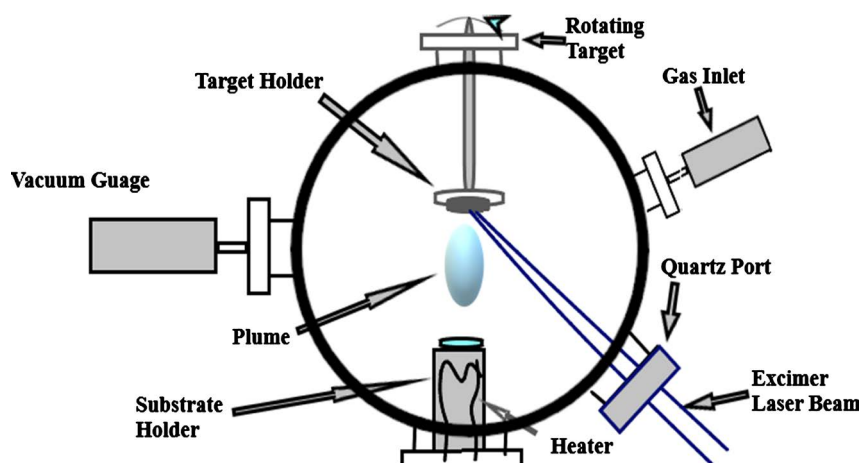


Figure 1. Schematic Diagram of PLD System [25].

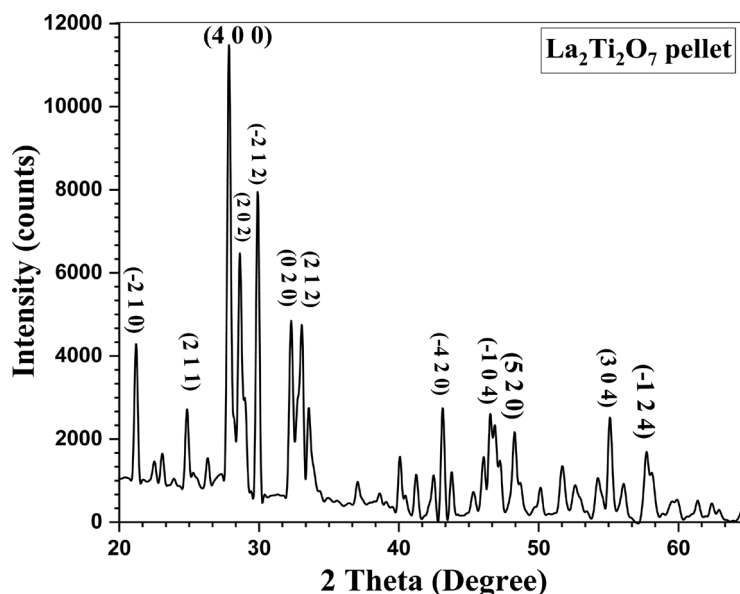


Figure 2. The XRD pattern of the LTO target at room temperature [26].

Table 1. The average thickness of the LTO thin films deposited at different annealing Temperatures.

Annealing Temperature (°C)	500	600	700
Average Thickness (nm)	231	228.3	178.6

3.2. Thickness and Roughness Measurement

Table 1 displays the average thickness of the thin films measured from SEM images (see **Figure 3**) at different annealing temperatures, As it's shown in **Figure 4** the thickness of the film decreased with the increasing of the annealing temperature for all the three samples. because the deposited molecules become more active due to energy gained from the substrate holder as resulting in increasing their motion with increasing the temperature as well as reducing their potential force to the substrate which is appearing in fewer micro-porosity defects, higher compactness and thinner thickness of these films [23].

The surface seems very smooth with a small presence of monotonic grains (see **Figures 5-7**) that leads to a slightly increased in the roughness of the thin films with the increasing of the annealing temperature as shown in **Figure 8**. The value of the root means square (RMS) of LTO films that is the average of height deviations taken from the mean image data plane calculated by the Nano scope Analysis software using the Equation (1) [27], and presented in **Table 2**.

$$\sqrt{\frac{\sum Z_i^2}{N}} \quad (1)$$

The (RMS) value of the minimum roughness equal to 0.256 nm for the thicker film with a thickness of 231 nm and highest roughness found 0.672 for the thinner thin film at 178.6 nm, which were deposited at 500°C and 700°C, respectively.

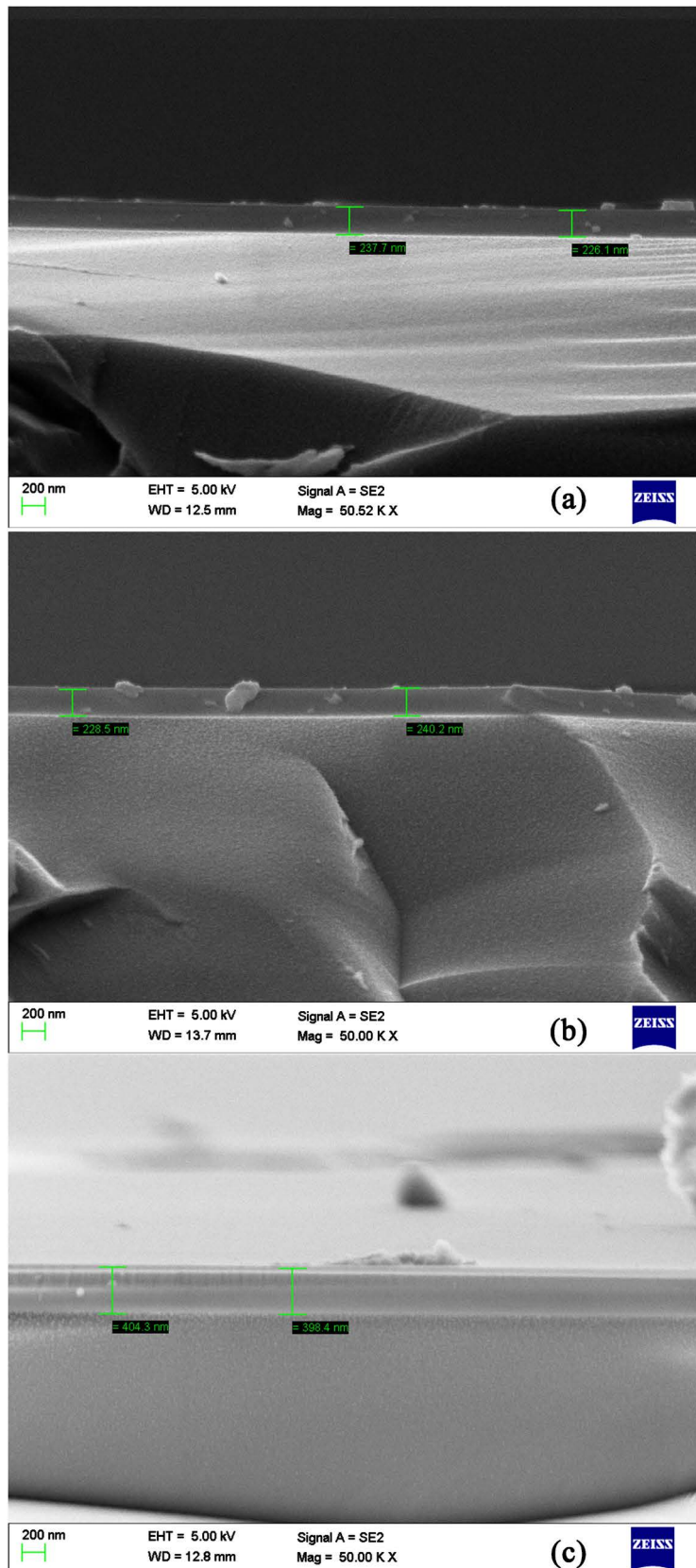


Figure 3. The SEM cross section image of the LTO thin films deposited in different temperatures (500°C, 600°C and 700°C), respectively.

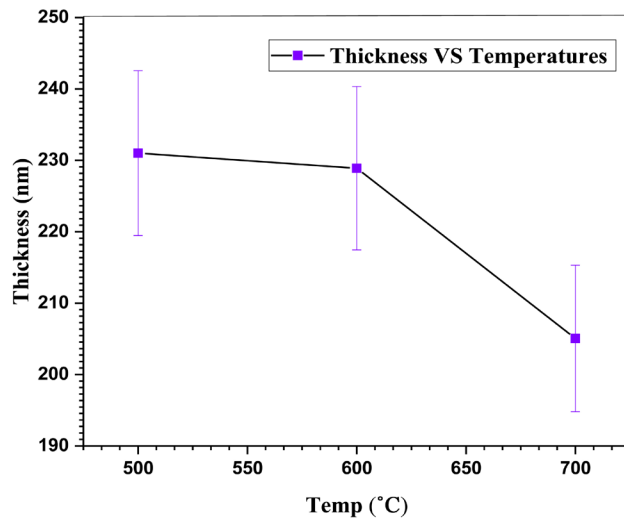


Figure 4. The Relationship between thickness and annealing temperature for the LTO thin films.

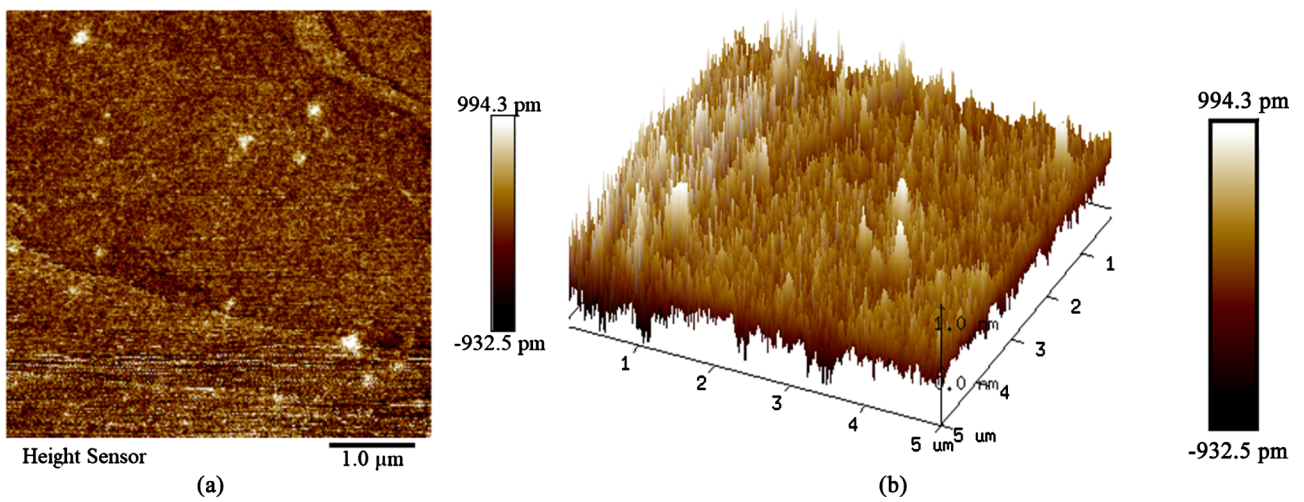


Figure 5. Three dimensional and Top view AFM images of the LTO thin films deposited at 500°C.

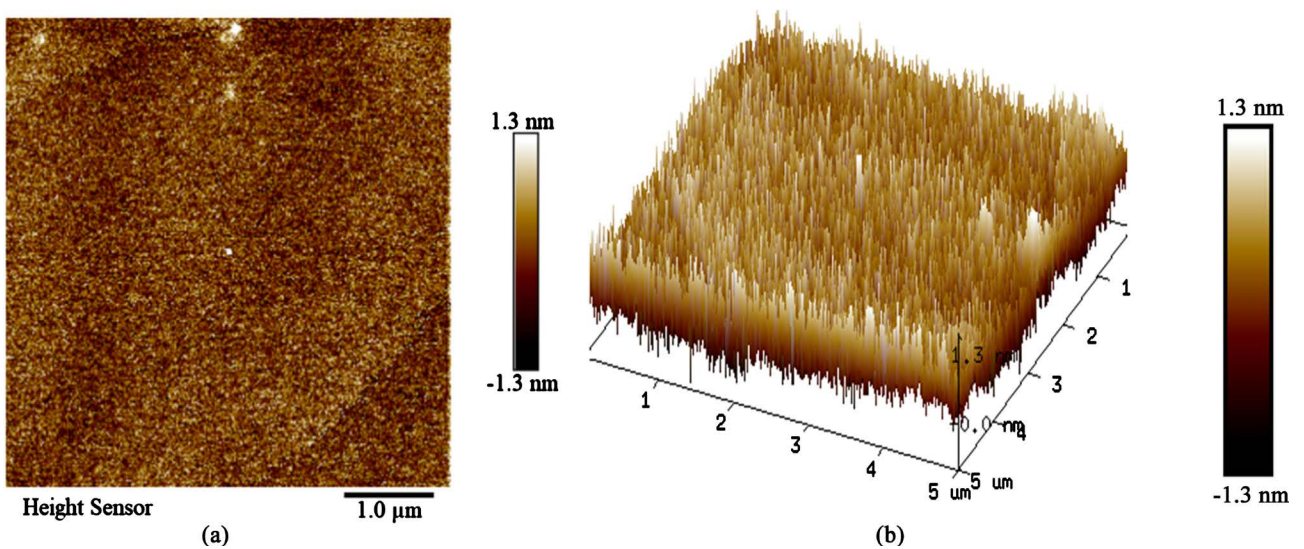


Figure 6. Three dimensional and Top view AFM images of the LTO thin films deposited at 600°C.

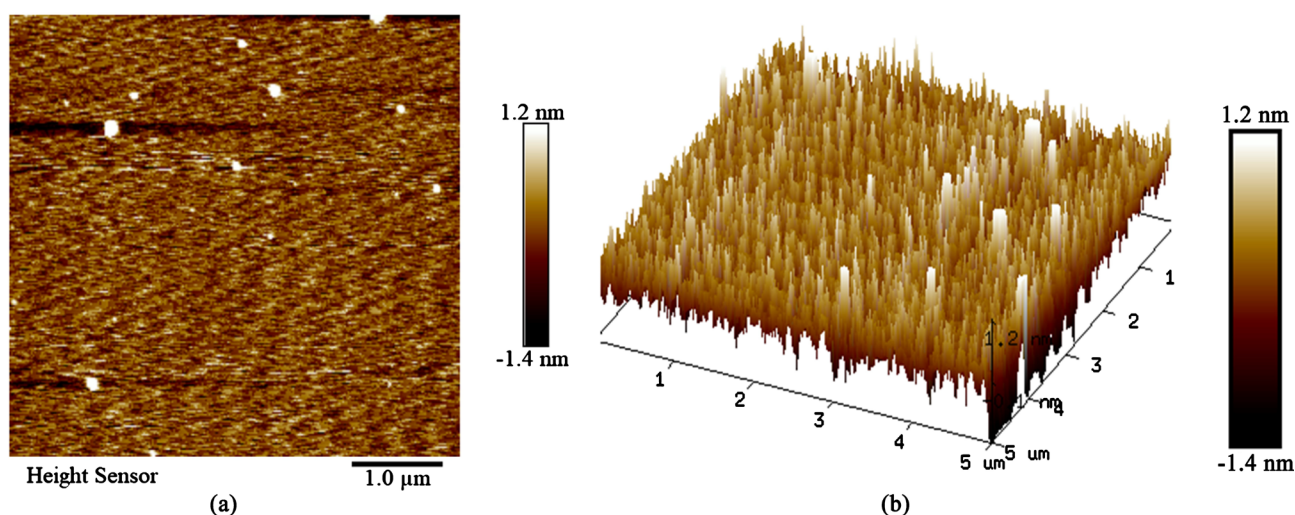


Figure 7. Three dimensional and Top view AFM images of the LTO thin films deposited at 700 °C.

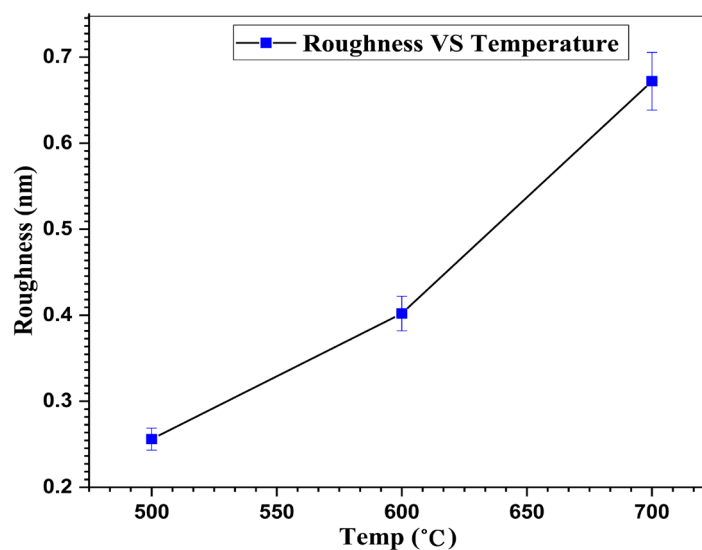


Figure 8. The Relationship between the roughness and annealing temperatures.

Table 2. The roughness value (RMS) of the LTO thin films deposited at 3 Hz in different annealing Temperatures.

Annealing Temperature (°C)	500	600	700
Roughness (RMS) nm	0.256	0.402	0.672

4. Conclusions

In this work, a monoclinic $\text{La}_2\text{Ti}_2\text{O}_7$ target synthesized by a solid-state method successfully and confirmed by XRD using X'Pert High Score Plus software. Pulsed laser deposition system was capable of synthesizing LTO thin films on Si (100) substrate at different annealing temperatures. During the deposition, the thermal annealing of LTO films in vacuum had a noticeable effect on thickness and the roughness of the films, the average thickness was found to be decreased while roughness (RMS) were linearly increased with the increasing of annealing

temperatures. Based on SEM and AFM results, it can be concluded that the annealing temperature influences the thickness and the roughness of the LTO thin films.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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