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Experimental Study on The Gamma Ray Absorption Properties of Lanthanum and Cerium Borides

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Abstract: The objective of this study is to investigate the mass attenuation coefficients (μ_m) of lanthanumhexaborides and ceriumhexaborides over a wide photon energy range emitted from the main radioactive sources used in medicine and industry. ^{125}I, ^{99m}Tc, ^{131}I, ^{137}Cs, ^{60}Co and ^{152}Eu gamma ray sources were used in the experiments. The materials synthesized in powder form were first pelletized and then irradiated by photon beams. At the end, it was seen that there is successful consistency between the obtained experimental data and the previous theoretical results. It was also observed that the investigated samples are comparable enough to the known standard gamma shielding materials, especially to lead which is one of the most common one. In conclusion, it is understood that the presently investigated samples have a promising aspect in terms of developing new shielding materials against gamma rays.

Key words: Gamma rays, Hexaborides, Mass attenuation coefficient, Shielding

1. Introduction

Nowadays, with the fast improvements in technology, the use of radiation sources have been significantly highlighted [1-7]. Gamma and x-rays are the types of radiation included in the title of 'ionizing radiation', which is increasingly [8-10] used in the area of nuclear power plants, agriculture, medicine and industry. The ionizing radiation sources have also expanded applications in many areas like nanoscience, biotechnology, semiconductor technology, biology, photochemistry, and geology [11,12].

The main idea of protection from radiation is protecting the employees and the public by ensuring that they get the lowest level of radiation dose as much as possible. The materials having higher densities and atomic numbers are especially accepted as good candidates of the materials which also having higher probability of radiation protection [13]. The protective shielding materials enable workers to expose the radiation not exceeding the juristical dose limits that can be received during the environmental applications.

Developing new protective materials for radiation has also been significant subject of research due to the requirements of getting rid of the harmful effects of ionizing radiation [14,16]. In the radiation shielding studies, the mass attenuation coefficients are the main coefficients to examine and get an idea of measuring the photon interactions with the substance [17,18]. The experimental investigation of μ_m for radiation shielding materials is significantly important besides obtaining theoretical simulation results for them. Thus, there are various theoretical [19,20] and experimental [21,22] studies that have investigated new generation materials for radiation shielding. The materials containing lead are traditional radiation protection materials used for shielding due to their capability of radiation absorption [23].

Some of other radiation shielding materials can be stated as glasses, alloys, thin films and polymer composites [24-29]. In one of the recent study, gamma ray shielding performance and characterization of the graphitic carbon nitride were investigated [30]. There are also some other studies stating that the materials containing Bi_2O_3 nanoparticles have better radiation shielding performance than bulk Bi_2O_3 [31,32]. Several rock samples from different regions of Najran, Kingdom of Saudi Arabia were collected and then evaluated for their shielding performance for γ-rays [33]. R. Arya and his collaborators studied on the next generation gamma ray shielding blocks developed using alumina industry waste [34]. In literature, there are also some theoretical and experimental studies investigating the ionizing radiation absorption properties of materials containing boron compounds [35-37].

Our purpose of this study is to investigate the gamma ray absorption properties of lanthanum and cerium borides experimentally and compare the feature results with the data that have been obtained by Monte Carlo method. So, the μ_m values of experimental results for the radiation absorption are compared with the calculated one obtained by using the developed code. At the end, it was seen that the presented results are very similar and consistent with the calculated ones done by MC along with also those of the previously investigated results for borosilicate, granite, vermiculite, concrete and lead. Thus, it was seen from also experiments that the lanthanum and the cerium borides have high absorption capability of gamma rays when compared to lead which is the standard shielding material.

2. Experimental and Theoretical Details

2.1. Synthesis of lanthanumhexaboride (LaB_6)

For the synthesis of lanthanumhexaboride, the reduction reaction of La_2O_3 with B_2O_3 was used under special conditions. 1 mmol La_2O_3, 6 mmol B_2O_3 and 25 mmol Mg powder were mixed in the porcelain crucible until homogeneous distribution was achieved. The contents of the crucible were heated in the muffle furnace up to 900°C and kept at this temperature for 4 hours and cooled to room temperature. After the cooled sample was transferred into a beaker, it was mixed in 1 M HCl for 24 hours and then kept in concentrated acid for the same time period. After the obtained black precipitate was washed twice with distilled water by filtering, it was dried in an oven at 120 °C, and then annealed at 450 °C for 2 hours.

2.2. Synthesis of ceriumhexaboride (CeB_6)

For the synthesis of ceriumhexaboride; 1 mmol CeO_2, 3 mmol B_2O_3 and 13 mmol Mg powder were thoroughly mixed in a porcelain crucible, and then such contents of the crucible were heated in a muffle furnace up to 800°C. It was kept at this temperature for 2 hours and cooled. The cooled sample was transferred into a beaker and mixed in 1 M

HCl for one day, and then it was kept in concentrated acid for one day too. After the obtained gray precipitate was filtered and washed twice with distilled water, it was dried in an oven at 120 °C and then annealed at 450 °C for 2 hours.

2.3. Preparation of pellet samples

In this part of the study, pelletizing the samples was carried out for the irradiation of LaB_6 and CeB_6 compounds, which were synthesized and obtained in powder form, as seen in Figure 1.

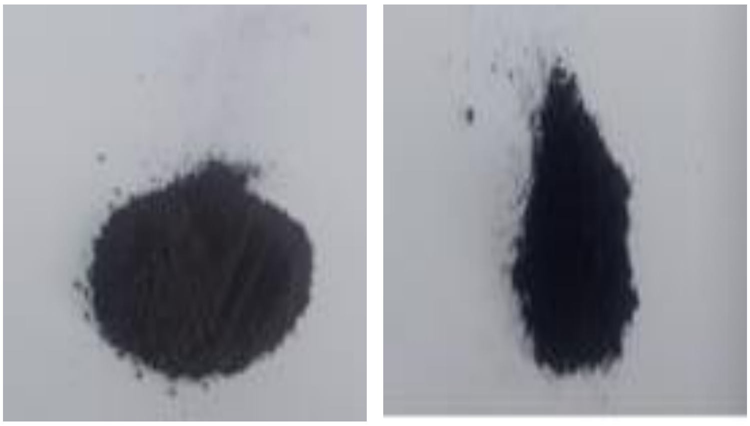

Figure 1. Appearance of LaB_6 and CeB_6 powder samples.

It is necessary to have certain physical properties in order to investigate the radiation absorption properties of materials. For this reason, the 30-ton press machine as seen in Figure 2 was used to make pellets from the synthesized samples. At this stage, cylindrical pellets with a diameter of 3 cm and a thickness of 3-6 mm were produced in a non-dispersible structure under high pressure.

Figure 2. Press machine with 30 tons capacity.

Pellets in desired physical dimensions were prepared, as seen in Figure 3, to be used in gamma spectroscopy system by pressing method under suitable conditions. Thus, the samples were prepared for the irradiation study to determine their radiation shielding properties.

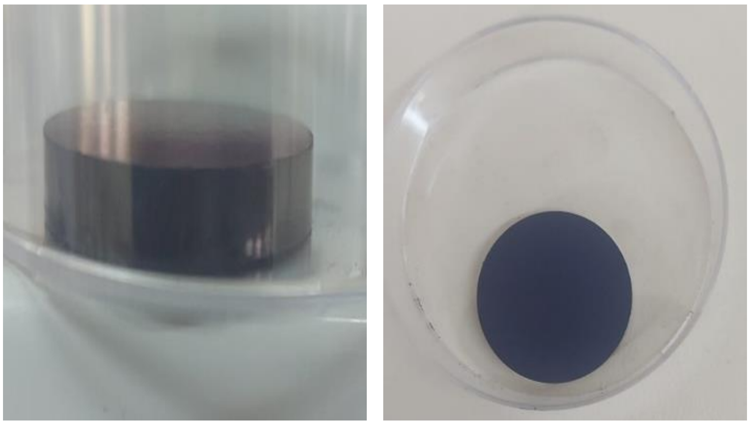

Figure 3. Top and side views of pelletized samples.

2.4. Gamma ray absorption spectroscopy

Gamma ray absorption properties of pellet samples having an appropriate physical size and composing of Lanthanumhexaboride and Ceriumhexaboride compounds were carried out by using two different scintillation detector systems. One of them is an ORTEC/905 Series mark/model gamma spectrometer system which has one block of thallium doped scintillation detector (NaI(Tl)). The experiments using this system were carried out in Nuclear Physics Laboratory located at the Faculty of Science of Ondokuz Mayıs University. The experimental analysis were practised using Scintivision-32 software, which is fully integrated into the calibrated system. The gamma energies of the radioisotopes used in this system are separated in the form of gaussian peaks by multi-channel analyzer. The other detector system is a GE/Discovery NM-630 mark/model gamma camera with multi-block NaI(Tl) detectors. This camera used in the experiments is located in the Nuclear Medicine Laboratory of Ondokuz Mayıs University-Faculty of Medicine. Narrow beam geometry was preferred in both systems used to examine the gamma ray absorption properties of pellet samples. Narrow beam geometry was created by beam collimation using lead cylinder. This cylinder has a diameter of 5 cm and a length of 20 cm, and its hole diameter is 3 mm. In both systems, after narrow beam collimation was achieved, the gamma rays were first dropped in the scintillation crystal. The crystal produces lots of scintillations, and the scintillations create electron avalanches in the photomultiplier tube. These electron avalanches are amplified by the applied voltage, producing an electrical signal proportional to the intensity of the radiation stored in the crystal. The experimental setup and schematic diagram of the both detector systems used are shown in Figure 4. The reason for using two different detector setups in this experimental study is that some radioisotopes and detector setups are located in different laboratories at the Ondokuz Mayıs University. While I-125, I-131 and Tc-99m radioisotopes and the gamma camera are located in the nuclear medicine laboratory, other radioisotopes and the ORTEC brand detector are located in the nuclear physics laboratory. Actually, both detector systems have a thallium-activated scintillation detector NaI(Tl). But the only difference between them is that the ORTEC detector has a single photomultiplyer tube (PMT) while the Gamma camera has multiple PMTs.

The attenuation of beam intensity in the same direction of the incidence due to absorption and scattering of beams is shown in Figure 5 at below. The radiation penetration and absorption properties of the materials as seen in the diagram of such figure are determined according to the Beer-Lambert law, also known as the exponential absorption law, which is also given in Equation 1 [5].

$$I=I_0e^{-\mu\Delta x} \tag{1}$$

Here, Δx is the material thickness and μ is the linear attenuation coefficient. Here, I and I_0 are the radiation intensity in Becquerel (Bq) passing through the material and collected in the detector without the material, respectively. Since $I = I_0$ when there is no material between the source and the detector, I_0 is obtained for all radiation sources.

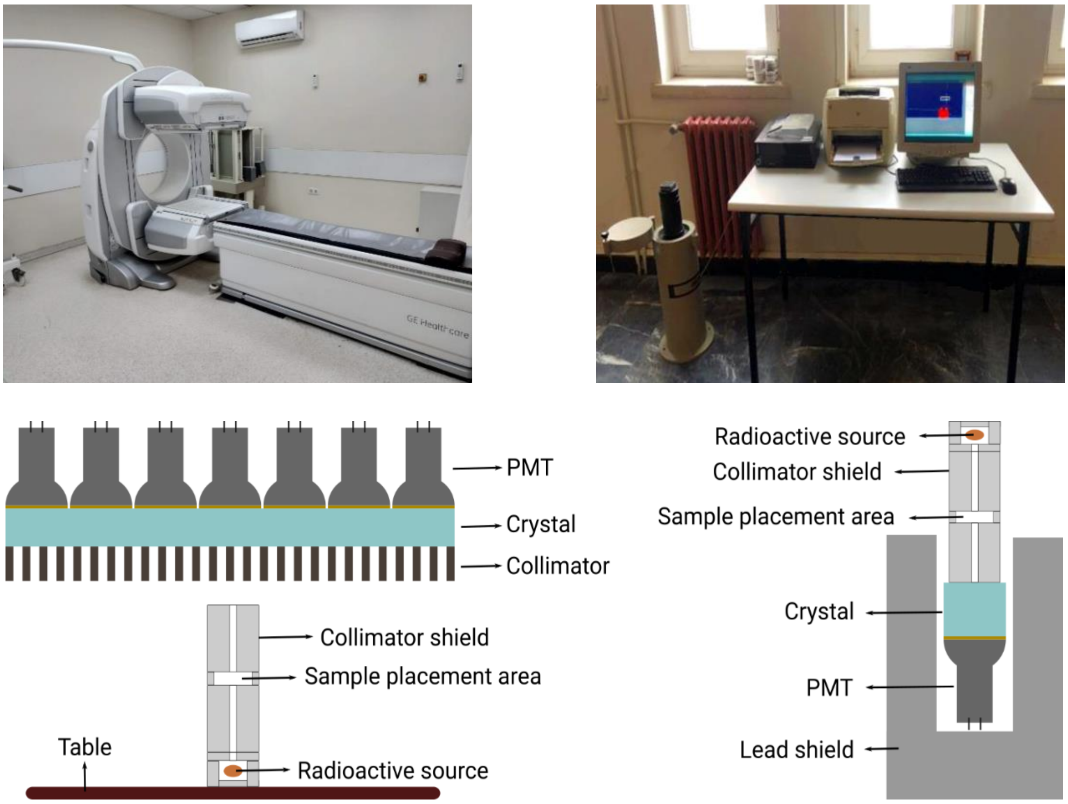

Figure 4. Gamma spectrometer measurement system with scintillation detector; a) GE/Discovery NM-630 Gamma camera and b) ORTEC/905 Series NaI(Tl) dedector.

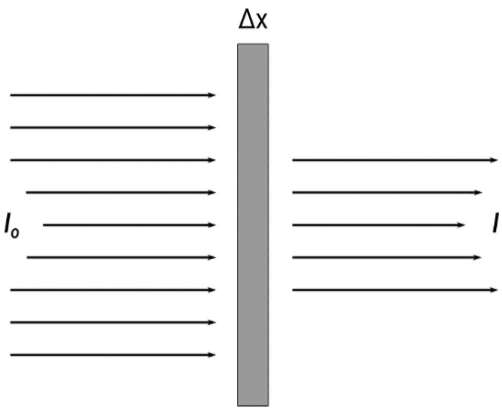

Figure 5. Schematic view of the attenuation in beam intensity.

When a material of Δx thickness is placed between the radiation source and detector, the linear attenuation coefficients (μ) are obtained. Since this coefficient varies with the different densities of the same materials, the μ_m value is much more used instead of μ [37]. This parameter is obtained by dividing the linear attenuation coefficient by the material density (Eq. 2).

$$\mu_m = {}^{\mu}/_{\rho} \tag{2}$$

In the measurements made using the ORTEC detector, the photon intensity was measured with and without the material, taking into account the channels falling within a certain width of the Gaussian peak corresponding to each energy value of the radioactive sources. Photon intensities in the measurements made with the gamma camera were counted by detector channels covering a circular area of 10 mm diameter in the center of the camera head. In this way, the photon absorption coefficients of the materials were obtained experimentally for each photon energy value by using the obtained I and I_0 values.

3. Results and Discussion

In order to calculate the mass attenuation coefficients, the density values of the cylindrical shaped pellet samples were determined experimentally by the classical method. Scales and digital steel calipers were utilized to achieve this precision. The theoretical density values of the synthesized samples and the obtained experimental results were given in Table 1. In order not to create a difference between the theoretical and the experimental data of the gamma ray absorption properties of the pellet samples, no additives were added to the samples during the pelletizing process since not to spoil the original content of the synthesized materials. So, just the cylindrical pellet samples having certain physical dimensions were prepared using a 30-tons press machine. For this reason, the obtained density values of the samples are lower than the theoretical values. It doesn't pose a problem in the comparison of the results since the density does not have an effect on the characteristic absorption parameter which also means the mass attenuation coefficient.

By the present study, the experimental and theoretical results of the mass attenuation coefficients at each energy value emitted from six different radioisotopes for LaB_6 and CeB_6 compound samples are given in Table 2.

Table 1. Theoretical and experimental density values of lanthanumhexaborides and ceriumhexaboride pellet samples.

Sample	**Density (g/cm^3)**	
	Experimental	**Theoretical [38]**
Ceriumhexaboride (CeB_6)	2.51	4.80
Lanthanumhexaboride (LaB_6)	1.92	4.72

Table 2. Theoretical and experimental mass attenuation coefficients of lanthanumhexaborides and ceriumhexaboride pellets for gamma energies emitted from six different radioisotopes.

Radioisotope	Gamma Energy (MeV)	Mass attenuation coefficients (μ/ρ)					
		CeB_6			LaB_6		
		EGSnrc [37]	XCom [37]	Experiment	EGSnrc [37]	XCom [37]	Experiment
^{125}I	0.021	19.7625	19.9400	7.3996	18.5308	18.7200	6.2341
^{99m}Tc	0.140	0.7450	0.7444	0.8082	0.7067	0.7060	0.5753
^{131}I	0.364	0.1334	0.1330	0.1778	0.1298	0.1294	0.1372
^{137}Cs	0.662	0.0776	0.0777	0.0824	0.0765	0.0766	0.0405
^{152}Eu	0.779	0.0693	0.0695	0.0541	0.0684	0.0686	0.0528
^{152}Eu	0.964	0.0606	0.0607	0.0518	0.0600	0.0601	0.0543
^{60}Co	1.173	0.0541	0.0540	0.0423	0.0536	0.0535	0.0801
^{60}Co	1.332	0.0504	0.0503	0.0689	0.0500	0.0499	0.0329
^{152}Eu	1.408	0.0488	0.0488	0.0334	0.0484	0.0484	0.0655

It is clearly understood from the data in Table 2 that there is a good agreement between the theoretical and experimental results. According to the data in the table, the biggest difference between the experimental and theoretical values was obtained for the ^{125}I source with 21 keV gamma energy. It can be said that the reason for this is that the beam energy is low for the 3 mm pellet thickness and the source activity for this radioisotope may also be insufficient. The obtained experimental results were compared to the XCom and the EGSnrc simulation results previously performed by Ozcan et al. [37] for these materials. It was understood that the calculated results in that study were comparable to lead, which is the standard shielding material, and it was strongly supported by the experimental data in this study. The comparisons of experimental data with the previous calculated results [37] for lanthanumhexaborides and ceriumhexaboride are shown in Figure 6.

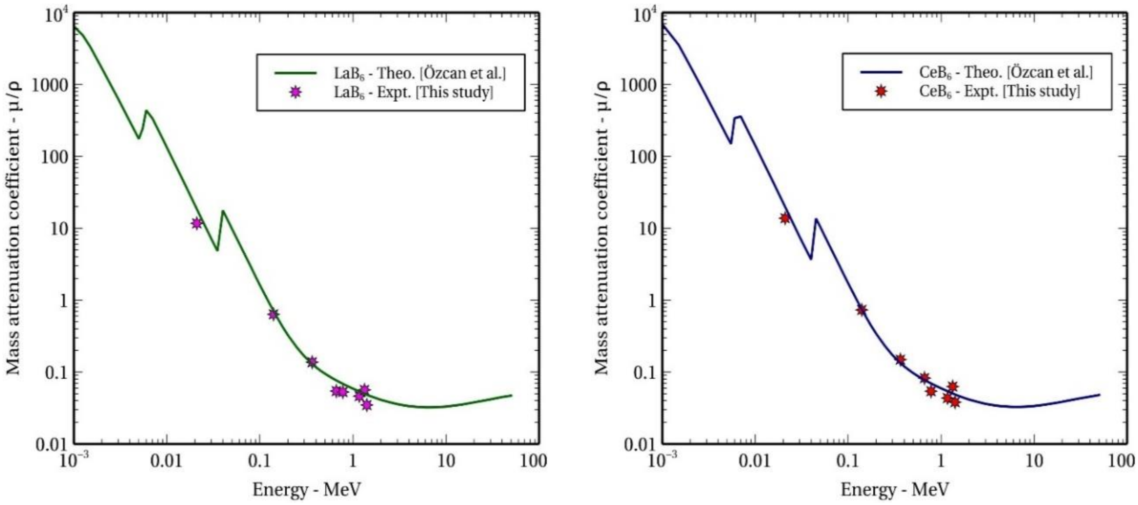

Figure 6. Comparison of the experimental data with the previous theoretical simulation results [37] for lanthanumhexaborides and ceriumhexaboride.

It is understood from the figure that the experimental values decrease exponentially with increasing energy values in consistency with the theoretical values. The small standard deviations appearing in this figure are thought to be the result of unnoticeable defects in the experimental setup like activity values of radioisotopes, or unavoidable statistical errors in the spectroscopy system. From such figure, it is generally seen that the experimental data and theoretical results are quite compatible with each other. The experimentally investigated materials of present study are quite well in terms of absorbing gamma radiation. In Figure 7, the radiation parameter of such materials are also compared to some other known shielding materials such as lead, vermiculite, borosilicate, granite and concrete.

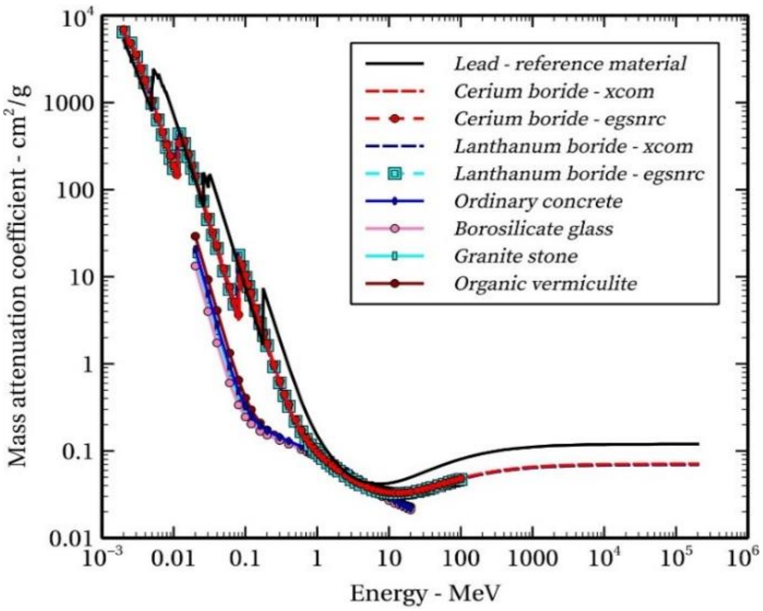

Figure 7. Comparison of the mass attenuation coefficients of lanthanumhexaboride and ceriumhexaboride with some other standard shielding materials (lead, granite, vermiculite, borosilicate and condrete) [36,39,40].

According to this figure, the boride compounds show graphical variations almost similar to lead, especially at gamma energies below 10 MeV. Below the energy of 2 MeV, it provides protectiveness of 92% radiation for 1.5 MeV, 85% radiation for 1 MeV, 63% radiation for 0.5 MeV and 34% radiation for 0.2 MeV, relative to the lead. In addition, according to the mass attenuation coefficients, it is observed that it has exactly the same absorptive property as lead at values corresponding to the K, L and M shell edges at some photon energies below 0.5 MeV. Variations in the mass attenuation coefficients corresponding to these edges can be seen from the peak jumps in the same figure. It should be kept in mind that these boride compounds have a quite low density values (4.80 g/cm^3 and 4.72 g/cm^3) when compared to lead (11.34 g/cm^3).

4. Conclusion

In this study, the mass attenuation coefficients, which are the characteristic parameter for radiation shielding, were experimentally determined for lanthanumhexaboride and ceriumhexaboride using radioactive sources having different gamma energies. The purpose of doing this is to know and understand the gamma absorption properties of such materials. So, it was understood that very appropriate results were obtained when the experimental data were compared to the previous theoretical findings. It is thought that in order to decide a material is a good candidate of radiation shielding material or not, it is a definite requirement to support theoretical ideas by experimental data. Thus, in this study, the expected theoretical results which were calculated previously are also obtained experimentally and it was seen that the results are quite promising ones for future studies that will be held on developing new radiation shielding materials synthesized in powder form. It was also understood from the densities of the hexaborides that being lighter than lead provides a significant advantage in terms of transportation of radiation shielding materials since lead is very heavy. Based on the obtained data, it was concluded that these materials, which are synthesized and produced in powder form, will play an important role in the development of gamma radiation shielding.

Authorship contribution statement

H. Gulbicim: Conceptualization, Methodology, Data Curation, Visualization; **A. Ozcan**: Original Draft Writing, Supervision/Observation/Advice; **N. Turkan**: Methodology, Original Draft Writing, Supervision/Observation/Advice; **M. Aksu**: Data Curation, Original Draft Writing; **E. Kurt**: Data Curation, Visualization.

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.

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