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Challenges in personal and clinical dosimetry using $Li_2B_4O_7$ and MgB_4O_7 as TLD and OSLD

Oliveira[a], L.S.S., Souza[b], L.F., Donald[a], G.G.,
Emidio[a], M.F.S., Novais[c], A. L. F. Souza[a], D.N.

[a]*Departamento de Física, Universidade Federal de Sergipe (UFS), 49100-00 São Cristóvão, SE, Brazil*
[b]*Centro de Desenvolvimento da Tecnologia Nuclear, CDTN, 31270-901 Belo Horizonte, MG, Brazil*
[c]*Faculdade de Engeharia Mecânica, Universidade Federal do Sul e Sudeste do Pará, Marabá, Pará, Brazil*
divanizia@gmail.com

ABSTRACT

Thermoluminescent dosimeters (TLD) and optically stimulated luminescent dosimeters (OSLD) are essential in radiation dosimetry. Such dosimeters can be easily transported due to their small size and can be used in *in vivo* dosimetry and anthropomorphic simulations. In this work, the dosimetric properties of $Li_2B_4O_7$ and MgB_4O_7 compounds were evaluated based on their response to the applied stimulus, either thermal or optical. The linear dose response range of the luminescent signal, its fading, the lowest detectable dose, and reproducibility are important parameters in determining a good dosimeter for clinical and personal dosimetry. Therefore, the objective of this work was, based on studies performed by other authors on dosimetric characterizations of doped and codoped Li_2B4O_7 and MgB_4O_7, to highlight those compounds with the greatest potential for applications in personal and clinical dosimetry using TL and OSL techniques. Considering the results described in other works, the materials that stood out for use in personal and clinical dosimetry were $Li_2B_4O_7$:Cu and MgB_4O_7:Dy,Na. In several of the reported studies, no data related to the lowest detectable dose, fading, and reproducibility of the luminescent signal of the investigated compounds were found. There are many possibilities for investigating these two types of compounds for the purpose of using them in personal and clinical dosimetry. Further studies will provide a broader scientific basis for choosing appropriate dosimetric materials for these applications.

Keywords: **clinical dosimetry, personal dosimetry, borate materials.**

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1. INTRODUCTION

Thermoluminescent dosimeters (TLD) and optically stimulated luminescent dosimeters (OSLD) are essential in radiation dosimetry, particularly in clinical and personal applications. Such dosimeters can be easily transported due to their small size and can potentially be used in in vivo dosimetry and in anthropomorphic simulations.

The characteristics that must be considered in determining whether a material is a potential dosimeter depend on the application for which it is intended. The linear dose-response range of the luminescent signal, the fading of this signal, and its reproducibility are important parameters in determining a good dosimeter. Also, the smaller the lowest detectable dose (LDD), the more likely the dosimeter will be suitable for clinical and personal dosimetry. Currently, approximately 90% of TLDs are used in personal dosimetry [1]. Although the use of TLD and OSLD is less frequent in clinical dosimetry than in personnel dosimetry, these dosimeters are advantageous in the clinic because they can be used for dose measurements in anthropomorphic phantoms and for *in vivo* dosimetry [2]. OSLD also have the possibility of immediate or near-immediate response. The dose of interest for these areas is up to 20 Gy [3].

In thermoluminescence, the measurement of the absorbed dose of low linear energy transfer (LET) radiation is quite simple and reproducible [4]. On the other hand, for the optically stimulated luminescence (OSL) technique, materials with capture centers with a larger cross section for photoionization are employed [4]. The advantages of the OSL technique compared to thermoluminescence (TL) are the relative simplicity of the instruments required for the measurement of the luminescent signal, the high dose sensitivity of the materials, and the possibility of multiple readings from the same dosimeter to check a certain dose [5].

Personal dosimetry evaluates absorbed doses in human tissue superficially, which is the case for the lens of the eye (3 mm) and skin (10 mg/cm^2), and deeply (1000 mg/cm^2 - 1.0 cm). When considering doses from low-energy beams, such as low-energy beta radiation and X-rays with energy below 15 keV, the concern is with the superficial part of the skin. For more energetic radiation, such as gamma rays, high-energy beta particles, X-rays above 15 keV, and neutrons, the concern is with the doses deposited inside the body of the person who has been exposed. The equivalent dose range of interest in personal dosimetry is from 10 mSv to 1 Sv [3]. Equivalent dose is calculated by multiplying the absorbed dose to the organ or tissue with the radiation weighting factor. Both units, sievert (Sv) and gray (Gy), are special names for J/kg [3].

For personal and clinical dosimetry, one always looks for materials with an effective atomic number (Zeff) similar to that of soft human tissue, i.e., tissue equivalent materials [3]. The use of tissue equivalent dosimetric materials (Zeff = 7.42) is necessary to reduce the variation in the response of a dosimeter with radiation quality; in other words, to reduce its energy dependence [6,7].

Boron-based TLD and OSLD have been prominent in studies on dosimetric materials for clinical and personal dosimetry, with $Li_2B_4O_7$ (Zeff = 7.25) and MgB_4O_7 (Zeff = 8.2) being the most renowned [8-10]. An important characteristic of borate materials is the possibility of applying them for thermal neutron dosimetry. The intrinsic sensitivity of borates to thermal neutrons can be enhanced by incorporating B-10 in their compositions due to an increase in the cross section of the material to this type of radiation. In a comparative study on the use of MgB_4O_7:Ce,Li, Al_2O_3:C and a neutron sensitive detector prepared using a mixture of Al_2O_3:C and $^6Li_2CO_3$, it was observed that MgB_4O_7:Ce,Li samples enriched with B-10 showed an OSL signal comparable to that of the other two dosimeters [11]. Souza *et al*. [12] studied the use of MgB_4O_7:Ce,Li for OSL dosimetry of neutrons produced in the interaction of megavoltage photon beams. Recently, using a Monte Carlo simulation, Moreira *et al*. [13] evaluated the sensitivity of the vinyl polychloride (PVC) films loaded with MgB_4O_7 for the detection of thermal neutrons and observed significant differences regarding the type of boron isotope present in the luminescent sensitivity of the films. According to the simulations, the incorporation of B-10 resulted in a higher OSL sensitivity of the films than with the other boron isotopes.

Although doped magnesium tetraborate (MgB_4O_7, abbreviated as MBO) was initially studied for TL dosimetry [14], more recently, its dosimetric properties have been evaluated with the OSL technique. Studies on MBO doped with Sm, Gd, Tb, Dy, Tm, Ce, and Li have already been performed [1,15], and it has been observed that MBO doped with Dy and Tm shows a luminescent sensitivity fifteen times higher than that of the LiF:Mg,Ti dosimeter (TLD-100). Currently, there is increasing interest in the study of the OSL properties of Ce and Li doped MBO [8]. Recently, Souza *et al*. [16] studied the compounds MBO:Ce,Li and $80MgB_2O_4$–20MBO:Ce (glass) as alternative dosimeters for testing the quality of 6 MV photon beams. These authors observed that $80MgB_2O_4$–

20MBO:Ce (glass) shows a more intense OSL response and a lower standard deviation in the measurements when compared to MBO:Ce,Li. Regarding MBO:Ce,Li, a challenge in using this material for dosimetry is in minimizing the initial fading of its luminescent signal [17].

Lithium tetraborate ($Li_2B_4O_7$, abbreviated to LBO) is a matrix widely applied in acoustics, electronics, linear and nonlinear optics, and piezoelectric and pyroelectric devices [18,19]. LBO, due to its 9 eV bandgap, has high UV transmittance at wavelengths up to 155 nm [20]. For dosimetry, dopants such as Ce, Ni, Cu, Ag, P, Mn, and Ag are incorporated into the LBO matrix, with Cu and Ag resulting in a higher luminescent efficiency [8,10,19,21-25].

Other borate matrices, such as SrB_4O_7, BaB_4O_7, and CaB_4O_7, have also been explored for use as luminescent dosimeters [26-30]. Pure SrB_4O_7 has a TL efficiency similar to the commercial dosimeter TLD-700 (^{7}LiF:Mg,Ti) [31]. Mishra *et al.* [30] investigated the TL and mechanoluminescence properties of SrB_4O_7:Dy and observed that this material has a linear dose response to gamma rays in the range of 0.0 to 1.4 kGy. SrB_4O_7 doped with Eu shows a linear TL response between 0.1 and 2.0 Gy and an OSL response between 0.1 and 20 Gy, with a TL sensitivity 200 times higher than that of TLD 500 (α-Al_2O_3:C) [26]. BaB_4O_7 doped with Ce has a TL sensitivity approximately 1.8 times higher than that of TLD-100 (LiF:Mg,Ti) and a linear response over a dose range of 1.5 mGy to 1.3 kGy, with a fading of 5% after 30 days [27]. CaB_4O_7, although having a high Zeff of 12.5, shows a sensitivity 8 times higher than that of TLD-100 when doped with Dy [28,29,32,33].

Considering that both LBO and MBO compounds are very promising for luminescent dosimetry, we should question which of them has the potential for use in clinical and personal dosimetry. To answer this, in this paper, we conducted a literature review that aims to compare studies on dosimetric characterizations of these materials and identify those that are applicable for personal and clinical dosimetry using the TL or OSL technique. Our expectation is that this study will help identify the challenges to be faced for the routine use of these materials in dosimetry.

2. MATERIALS AND METHODS

The search for the papers discussed in this study was performed via the Google Scholar site, which is a site for searching academic literature. The results obtained were filtered using the following keywords: magnesium tetraborate in radiation dosimetry; MBO dosimetry; lithium tetraborate in radiation dosimetry; LBO dosimetry; borates in radiation dosimetry. The focus of this research was articles on clinical and personal dosimetry, so documents such as patents, dissertations, theses, and conference abstracts were not considered. Only papers published between 2000 and 2020 were of interest for this study. From the papers, information was extracted on the type of beam used in the irradiation of the materials, beam energy, LDD, fading, reproducibility, and energy dependence of the luminescent response.

The parameters chosen to define the materials with the most suitable luminescent characteristics for use in personal and clinical dosimetry were the linear dose response between 10^{-4} Gy and 20 Gy, with little or no energy dependence, LDD lower than 20 μGy, fading less than 10% in 90 days, and a reproducibility with a maximum standard variation of 5% in 10 consecutive cycles of irradiation, readout, and annealing.

3. RESULTS AND DISCUSSION

3.1. Dosimetric characteristics of MBO and LBO

The phenomenon of luminescence can be observed in various materials from known forms of stimulation; however, to take advantage of this phenomenon in radiation dosimetry, it is necessary to consider some requirements, what is the most appropriate reading method and a suitable dose response range, among other relevant factors depending on the area of interest. Although it is hard work to find a material that meets all the characteristics required for a good dosimeter, it is interesting to identify those that have a greater number of ideal characteristics.

Because the expected doses in personal or clinical dosimetry are usually very low, the LDD is an important parameter in determining how close the material is to an ideal dosimeter; a lower LDD value indicates that the material is more likely to be suitable for these types of dosimetry [11, 19]. LBO and MBO compounds have stood out in dosimetry for their applicability in thermal neutron dosimetry. In general, magnesium tetraborate shows high sensitivity in thermoluminescent dosimetry and lithium tetraborate has high UV transmittance for certain wavelengths [11,12,15,34].

A good dosimeter also depends on other parameters such as linear dose range, fading, and reproducibility. To aid in the search for TL and OSL based MBO materials that are best suited for individual and clinical monitoring, Table 1 reports characteristics of materials studied in the last two decades. Among the works, three addressed MBO:Dy,Na [14,35,36], another three studied MBO:Dy [37-39], three analyzed MBO:Ce,Li [11,34,40], and others studied MBO doped with Mn and codoped with Tb [41], Dy, Ca, Na [42], Dy, Li [34], Mn [43], Cu [43]; MBO-B_2O_3 as glass [44], Ce [45]; Tb [46]; Gd, Li [47]. Concerning LBO, in five works, this material was doped with Cu [7,9,21,48,49], undoped LBO was studied by [9, 25], and doped with Ag by Ozdemir *et al.* [25] (Table 2).

Regarding the analyses, among the papers on MBO (Table 1), in eleven of them, the thermoluminescent properties of the investigated materials were studied, and another four investigated the OSL response to dose. In only two papers, analyses of the energy dependence of the luminescent signal of MBO:Dy,Na were performed [14,35]. Furetta *et al*. [14], Cano *et al*. [35], and Bahl *et al*. [36] characterized MBO:Dy,Na and found promising dosimetric results from MBO, such as a variation in the signal reproducibility less than 3%, fading less than 15% over three months, and LDD lower than 2 mGy. Different from other works, Furetta *et al*. [14] and Prokić [42] analyzed data obtained from more than one reader to determine the LDD of MBO. In this work, we chose to include in Table 1 the data obtained from the equipment that registered the lowest LDD in these two studies.

Among the six works that investigated LBO (Table 2), only one used the OSL technique and the other five investigated the dosimetric properties of materials using the thermoluminescence technique. Uniquely, Furetta *et al*. [49] studied the energy dependence of the LBO luminescent response. This dependence was less pronounced for the thermoluminescent response of the LBO:Cu samples than that of the LBO:Cu,In samples irradiated with energies in the range from 33 to 161 keV. No energy dependence was observed in the thermoluminescent response of the samples irradiated with ^{137}Cs and ^{60}Co.These same materials also showed good dosimetric properties, luminescent signal reproducibility with a standard deviation less than 2% in ten cycles of irradiation, readout, and annealing, fading less than 10% over three months, and LDD lower than 2×10^{-2} mGy.

The relevant factors for a dosimeter to show an adequate LDD are its high dose sensitivity and a luminescent emission coherent with the spectral response of the TLD reader used in the measurement [11]. The procedure for obtaining the LDD is to separate and stimulate (heat or light) a batch of non-irradiated samples and then to read them, obtaining the background adjusted to the individual sensitivity factor of the samples. The process of calculating the LDD is to apply a linear function interpolation, that is, to construct new data points from already known points, taking into account the sum of the background with three times the standard deviation of the non-irradiated samples and then multiplying this sum by the system calibration factor [11,46].

As observed in Tables 1 and 2, MBO samples doped with Ce, Li [11,34], Dy, Ca, Na [42], Dy, Li [34], Dy [50], Mn [43], Cu [43], Ce [45], Gd, Li [47], and LBO doped with Cu [7,49] were exposed to beta radiation. Among these, the one that was best for personal and clinical dosimetry is LBO:Cu [49], as it showed a linear dose response between 10^{-3} Gy and 10^{3} Gy, LDD of 20 μGy, and fading around 10% over 90 days.

In the analyzed studies, only MBO materials doped with Dy, Na [14,35], Mn, Tb [41], Dy, Ca, Na [42], MgO-B_2O_3 as a glass [44], Tb [46], Gd, Li [47], and LBO doped with Cu [48,49], Cu, In, Ag [21], and Cu, In [21,49] were irradiated with gamma radiation (^{60}Co, ^{137}Cs). For this type of radiation, the material that stood out for dosimetry was MBO:Dy,Na [14], which is useful for monitoring doses from 9×10^{-5} Gy up to 100 Gy, with LDD of only 2 μGy and a luminescent signal decay of 8% over a storage time of 90 days.

The respective objectives of each of the works that have been cited in our discussion have been met. However, it should be considered that for the use of the materials investigated in these papers more dosimetric analyses would certainly need to be performed. Of the fourteen studies reported in Table 1, only three evaluated the luminescence of materials irradiated with doses on the order of micrograys. Table 2 also reports three papers with dose response analyses of this order of magnitude. Only half of the studies investigated fading. It should also be considered that some of the studies may have employed laboratory structures more suitable for dosimetric investigations focusing on personal and clinical dosimetry. Therefore, it can be said that the systematization of this type of dosimetric study is a challenge in itself.

Table 1: Dosimetric characteristics of MBO in the papers analyzed.

Material	Work (Reference)	Source	Linear dose range (Gy)	LDD (mGy)	Fading (% - Days)	Reproducibility (±% - cycles)	Technique
MBO:Dy,Na	[14]	^{60}Co, ^{137}Cs, X-rays	$10^{-4} – 2 \times 10^{1}$ *	2×10^{-3}	8% - 90	± 2% - 10	TL
MBO:Dy,Na	[35]	^{60}Co	$2 \times 10^{-4} – 5 \times 10^{1}$ *	2.33×10^{-2} 2.43×10^{-2}	10% - 60	< ± 7.5% - 10	TL
MBO:Dy,Na	[36]	Proton, ^{137}Cs	$1 – 4.5 \times 10^{2}$ **** $10^{-1} – 10^{3}$ *	10^{-1} 10^{-2}	< 7% - 30 < 15% - 30	< ± 3% - No Info	TL
MBO:Ce,Li	[11]	^{90}Sr/^{90}Y, Pu(Be)	$\sim 5 \times 10^{-1} – \sim 10^{1}$ (OSL)***	< 1	< 4% - 6 (OSL)	~10% - 3	OSL
MBO:Ce,Li	[40]	^{90}Sr/^{90}Y, X-rays	$10^{-1} – 10^{1}$ ***	1	OSL ~20% - 3	No Info	OSL
MBO:Mn,Tb	[41]	^{137}Cs	$10^{-1} – \sim 5 \times 10^{3}$ *	No Info	~10% - 30	No Info	TL
MBO:Dy,Ca,Na	[42]	^{90}Sr/^{90}Y, ^{60}Co, X-rays	$10^{-5} – 5 \times 10^{2}$ *	2×10^{-3}	0% - 180	± 2% - 10	TL
MBO:Dy,Li MBO:Ce,Li	[34]	^{90}Sr/^{90}Y	$2 \times 10^{-1} – 4 \times 10^{1}$ *** $2 \times 10^{-1} – 10^{2}$ ***	No Info	100% - 40 < 1% - 40	No Info	OSL
MBO:Dy	[50]	^{90}Sr/^{90}Y	$5 \times 10^{-3} – 2$ ***	No Info	No Info	No Info	TL
MBO, MBO:Mn MBO:Cu	[43]	^{90}Sr/^{90}Y	$1 – 6 \times 10^{1}$ ***	No Info	13.8% - 48	No Info	TL
MBO	[44]	^{137}Cs	$2 \times 10^{1} – 10^{5}$ * $10^{1} – 5 \times 10^{3}$ *	5	TL - 20% - 15 OSL - 1 h	~20% - 6	TL and OSL
MBO:Ce	[45]	^{90}Sr/^{90}Y	2.25 – 36 ***	No Info	5% - No Info	< ± 10% - 10	TL
MBO:Tb	[46]	^{60}Co	$1 – 10^{1}$ *	5×10^{-2}	No Info	No Info	TL
MBO:Gd,Li	[47]	^{90}Sr/^{90}Y ^{60}Co, ^{137}Cs	$10^{-3} – 10^{3}$ *	No Info	< 10% - 63	< ± 10% - 10	TL

* Gamma; ** X-rays; *** Beta; **** Proton beam

Table 2: Dosimetric characteristics of LBO in the papers analyzed.

Material	Work (Reference)	Source	Linear dose range (Gy)	LDD (mGy)	Fading (% - Days)	Reproducibility (±% - cycles)	Technique
LBO:Cu	[7]	No Info (Beta rays)	$10^{-3} - 10^{3}$ ***	10^{2}	No Info	No Info	TL
LBO LBO:Cu	[9]	^{60}Co and ^{137}Cs	$10^{1} - 4.5 \times 10^{2}$ * $10^{2} - 5 \times 10^{3}$ *	No Info	18% - 30 7% - 30	50% - 10 No Info	TL
LBO:Cu,In,Ag LBO:Cu,In	[21]	^{60}Co	$10^{-4} - 3 \times 10^{3}$ *	10^{-2}	< 10% - 90 < 3% - 90	± 2% - 10	TL
LBO:Ag	[25]	X-rays	$10^{-1} - 10^{2}$ **	3	No Info	No Info	OSL
LBO:Cu	[48]	^{137}Cs, ^{60}Co and ^{90}Sr/^{90}Y	$10^{-3} - 10^{3}$ *	2×10^{-2}	11% - 90	No Info	TL
LBO:Cu,In LBO:Cu	[49]	^{60}Co, X-rays	$\sim10^{-4} - 10^{3}$ * $\sim10^{-4} - 10^{3}$ *	2×10^{-2}	< 10% - 90	< 2% - 10	TL

* Gamma; ** X-rays; *** Beta; **** Proton beam

3.2 Mechanisms involved in the luminescence of MBO and LBO

There is no systematic study on the luminescent properties of pure MBO, so it is important that a thorough study be performed. There is still a lack of detailed studies on MBO, which makes it difficult to choose appropriate dopants for its use in dosimetry [15]. Oliveira *et al*. [51] performed a theoretical study on MBO using density functional theory calculations and found information about the valence band, which is dominated by the excited O 2p states; in the conduction band bottom, a predominance of Boron (trigonal) 2p states was found. The same study reinforced the lack of experimental data in the literature for comparison with the theoretical data obtained.

Porwal *et al*. [52] investigated Tm-doped MBO by electron paramagnetic resonance (EPR) and observed that upon heating the material to 200 °C, the BO_3^{2-} related lines were destroyed. With this information, the authors were able to propose that upon irradiation of the sample, the anionic borate groups BO_3^{3-} act as hole capture centers, subsequently converting to BO_3^{2-} (BO_3^{3-} + h+ to BO_3^{2-}), and the dopant Tm^{3+} acts as an electron capture center, becoming Tm^{2+}. When the material was heated, the holes previously trapped in the BO_3^{2-} charge-capture centers recombined with Tm^{2+} electrons to become Tm^{3+}.

In the study by Torres-Cortés *et al*. [53], the composites MBO:Dy and MBO:Dy,Na were investigated, and it was observed that MBO doped only with Dy exhibits a low TL signal with a peak around 250 ºC. The co-doping with Na (MBO:Dy,Na) intensifies the TL signal and increases the temperature of its peak, which is located at 222 ºC. So far, no studies explaining the role of Dy and Na in the MBO matrix have been found. Nevertheless, the EPR studies by Gundu Rao *et al*. [54] suggest that Dy acts as hole traps in the irradiation process of the $CaSO_4$ material and the co-dopant Na acts as a compensator for excess negative charges of the SO_4^{-1} centers. As experimental and theoretical studies investigating the structural and electronic properties of MBO:Dy,Na have not yet been found, it is suggested that the Dy and Na dopants in the MBO matrix have a similar behavior to that of $CaSO_4$, acting as hole traps and charge compensators.

Although some of the studies in Table 1 showed that MBO samples doped with Mn, Tb [41], Ce, Li [34], Mn [43], and Cu [43] exhibited luminescence with a linear dose response and fading less than 10% for up to three months storage, these same studies did not investigate the LDD and reproducibility of the TL/OSL emission. Similarly, MBO doped with Dy [50] exhibited a luminescence with a good linear dose response, but the study did not investigate the LDD, fading, or reproducibility of the TL signal.

LBO crystals contain large concentrations of oxygen and lithium holes. As these holes have opposite effective charges, they act as charge compensators, with two lithium vacancies compensating one oxygen vacancy [55]. Although LBO exhibits luminescent properties when pure, the temperature at which this phenomenon occurs is impractical for dosimetry. During irradiation at -196 ºC, the oxygen vacancies play the role of electron traps that remain stable at room temperature. However, the lithium vacancy together with the adjacent oxygen ion play the role of a hole trap, which has a strong electron decay at temperatures above -173 ºC [55]. Pure LBO crystals are not suitable for radiation dosimetry, as it is impractical to maintain their proper luminescent properties outside of stringent laboratory conditions.

Copper doping can act as electron and hole trapping centers in the LBO. These traps represent defects in the LBO:Cu crystal that can change their charge states. One of the defects results from the replacement of a lithium vacancy by Cu without causing nearby disturbances because the Cu^{1+}

ions act by trapping holes. A second type of defect is caused by the replacement of Cu^{2+} near a lithium vacancy. The other defects are interstitial Cu^{1+} ions in different environments performing the function of charge compensators for lithium vacancies [55].

4. CONCLUSIONS

Considering the results of the works conducted by other authors, it is not yet possible to state which dopants are ideal for using MBO and LBO for TL or OSL dosimetry. Especially for the use of these materials as OSLD, of the twenty papers reviewed, only four described results obtained using OSL. The materials that stood out for use in personal and clinical dosimetry were LBO:Cu and MBO:Dy,Na. In several of the reported studies, no data related to the LDD, fading, and reproducibility of the luminescent signal of the investigated compounds were presented. Therefore, there are many possible investigations on these two types of compounds for the purpose of using them in personal and clinical dosimetry. One challenge is to systematize the dosimetric characterizations so that these materials can have routine use in dosimetry. Further studies will provide a broader scientific basis for choosing appropriate dosimetric materials for personal and clinical dosimetric applications.

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