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Absorbed Dose measurement using nanomaterial

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ABSTRACT: Introduction: The basic features of nanophosphors specified by their structural state are propounded. New era in determination of absorbed dose is emphasized by using nanophosphor Lithium Tetraborate dosimeters. Objective: They can be used for measuring absorbed dose of ionizing radiations more efficient than known micro-scaled materials in medical, personal and environmental dosimetry. Methodology: Lithium carbonate reagent and boric acid were used as precursors. Poly vinyl pyrrolidone (PVP) was used as a capping agent to reduce the agglomeration of the particles. Besides, nitrate of copper (II) were used as dopants. Deionized water was used as the solvent. All materials which, utilized without any further purification, were purchased from Sigma Aldrich Company (United Kingdom) with purities more than 99.8% and used without further purification. Results and Discussion: The effect of a high concentration of surface trapping centers and grain size of particles on the nanophosphor luminescence characteristics is noted. These features determine some effective Thermoluminescence properties, which are essential for radiation detection. The luminescence and dosimetric properties of nanophosphors of different compositions are described. It is noted that the consequence nanophosphors show promise results in linearity as an advanced material for detecting efficiently ionized radiation doses.

Keywords: *Absorbed dose; Detection; Dosimetry; Lithium tetraborate; Nanophosphor; Thermoluminescence; Thermal treatment*

INTRODUCTION

Ever since (Daniels, *et al.*, 1953) reported for the first time on the Thermoluminescence (TL) as a technique in radiation dosimetry the TL has attracted tremendous attention in science, industry and medicine, where ionizing radiation is used for radiation dose monitoring. They showed that the irradiated material contains stored energy, which could be released thermally. Daniels firstly introduced LiF as a good dosimeter, due to its high sensitivity, and small pellets. LiF was applied to measure internal doses at the Hospital of Oak Ridge Institute of Nuclear studies by cancer patients (Daniels, *et al.*, 1953). Later, it was applied to determine radiation following an atomic weapon test by (Cameron, Suntharalingam, and Kenney, 1968). Cameron and his colleagues (1960s) introduced that the presence of impurities could improve the efficiency of LiF. The intensity of LiF was improved in the presence of impurities. Due to enormous intensification of attempts and a massive literature that now exists on radiation dosimetry (more

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than 50% of publication on Thermoluminescence are focused on dosimetry), much researchers are fulfilled on extending other dosimeters (McKeever, 1988). Table 1 depicts some popular TLD materials. Moreover, some advantages of TL dosimeters (Olko, 2010) are ; High sensitivity of new materials like LiF: Mg, Cu, P, Easy handling (no light sensitivity), Simple TLD readout – Effective and flexible gas heating, Neutron dosimetry possible with Li-6 and Li-7 , TL glow curve used as a quality control, Individual calibration of dosimeters possible, and Dose re-estimation possible Ultrafine structures having an average phase or grain size the order of 10^{-9} m are classified as nanostructured materials (NSMs). Currently, in a wider meaning of the term, any material that contains grains or clusters below 100 nm, or layers or filaments of that dimension, can be considered as "nanostructured". The interest in these materials has been stimulated by the fact that, owing to the small size of the building blocks (particle, grain, or phase) and the high surface-to-volume ratio, these materials are expected to demonstrate unique mechanical, optical, electronic, and magnetic properties. Nanoparticles demonstrate exotic catalytic, optical and electronic properties. Their properties vary with the manufacturing technique used for controlling their size and shape, thus making them interesting building blocks for nanoscale structures, assemblies and devices. Miniaturization of structures by electron-beam lithography and mechanical techniques is approaching the theoretical limits of about 50 nm. Hence, it is necessary to develop alternative methods to enable further miniaturization of chemical objects (Azonano, 2015).

Nanoparticles have potential applications in many different fields. Engineered nanoparticles are specifically designed and formed with customized physical properties in order to fulfill the requirements of specific applications. They can serve as the end product, like sensor for special purposes, pharmaceutical drugs and quantum dots, or they can serve as components in end products, as in the case of carbon black in rubber products. For both purposes, the physical properties of the nanomaterials play a key role in their performance. The novel physical and chemical properties of nanomaterials pave way to interface electronic signal transduction with biological recognition events and to design advanced bioelectronic devices with innovative functions (Kortov, *et al.*, 2014).

Main properties of nanophosphors

Nanophosphors present a type of phosphors consisting, which include nano powders, pressed compacts, nano ceramic materials, films, glasses with photoactive nano clusters, and nano powder coatings of different thickness. Manufacture and storage of powder nanophosphors with particles1-10 nm in size present difficulties. Such powders are unstable and tend to aggregation. Nanophosphors with particles 20–100 nm in size are convenient in use. A high concentration of surface atoms and defects at multiple nano grain boundaries may be regarded as one of the fundamental properties of nanophosphors. They create surface charge-carrier trapping centers, which are analogous to bulk centers, but have a different energy depth. The translational symmetry breakdown and limitation of the free path of electrons by the nanoparticle size alter the selection rules, bring about new optical transitions, increase the oscillator power, and change the luminescence decay time (Suzdalev, 2005). If the nanoparticle size becomes comparable with the de Broglie wavelength or the Bohr exciton size, quantum confinement effects come into play, changing the forbidden gap width and leading to appearance of new energy levels (Kortov, 2010).

Lithium Tetraborate Dosimeter

One of the most important missions in TL dosimetry investigation is expansion of new detector materials for radiation monitoring. Lithium tetraborate (LTB) is a popular and significant starting material in this extension for a long time. The TLD material needs to be tissue equivalent and highly sensitive. Moreover, it is needed for measurements to be done in laboratory conditions that require the possible smallest size of TLD material. Other than these properties, the TLD should not be toxic. In order to the importance of tissue equivalent factor, the ICRP published recommended diagnostic reference levels for medical imaging modalities. Luminescence detectors of ionizing radiation are now comprehensively used in individual dosimetry services, due to their excellent dosimetric properties. The most frequently used personal dosimeters are based on Optically Stimulated Luminescence (OSL), radio photoluminescence (RPL) or Thermoluminescence. Luminescence detectors have also applied in clinical dosimetry, especially around new radiation modalities in radiotherapy, such as Intensity Modulated Radiotherapy (IMRT) or ion beam radiotherapy (Olko, 2010). LTB with its effective atomic number $Z_{eff} = 7.39$ has the property being nearly tissue equivalent that makes it a very promising material in the field of personal and clinical dosimetry. Although, un-doped LTB crystal shows TL emission and its energy dependence is low. Its sensitivity is much lower than those of the commonly used detector materials. The idea of adding an activator material came very early. In the 1960s Schulman *et al.*, synthesized LTB: Mn for dosimetric purposes. The doped LTB with Mn shows a bit higher sensitivity, but the emission of the material is in the red region activator to enhanced key factors of TL dosimeters. In order to the superior results of synthesis detectors with Cu, it is preferable dopant element.

LTB: Cu is not a wanted aspect for routine application in dosimetry. It should be emphasized that almost all of the commercially prepared lithium which is not favorable for the light detecting systems of the commonly used TL readers. Thus, a lot of other efforts were made for finding dopants and possibly co dopants giving better TL properties. Some of transition metals improved named drawback such as, Cu, Ag, Ni, Co. even though In and Ce was applied as borates doped by Cu, change the color (become brownish) after repeated annealing steps.

LTB synthesis techniques

There are two methods to prepare doped and un-doped LTB phosphors; “melting technique” and “sintering technique” (Bjarnard and Kase, 1987). The precursors of the melting method are lithium carbonate and boric acid and for doping sufficient aqueous solution of the desired dopant is needed whereas, the precursor of the sintering method is raw LTB with the solution of a dopant in acetone or alcohol. So, doped and un-doped LTBs can be ideally prepared by controlling some important factors such as, temperature, concentration of precursors, ratio of dopant substance, and retention time of the master mixture before undergoing to the thermal treatment stage in the furnace. The mentioned factors have a high impact on TL proper-

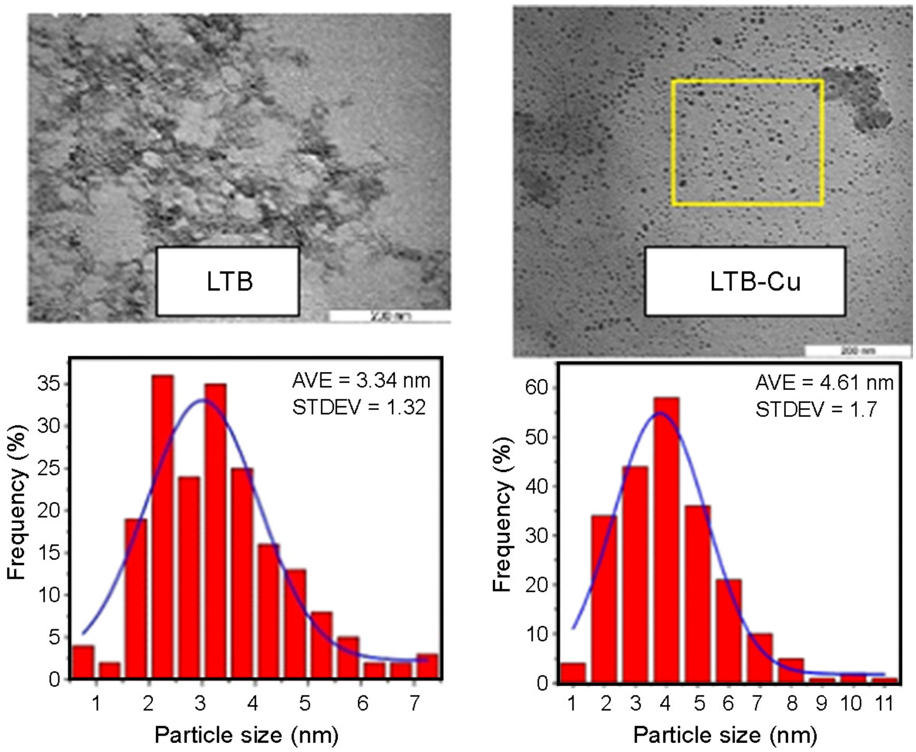

Fig. 1. The TEM results of LTB and LTB-Cu nanophosphors

Table 1. Characteristics of popular TLD materials (McKeever, 1988)

Phosphor	Z_{eff}	Linear Range (rad)	Saturation level (rad)	Thermal fading	Pre-annealing procedures
LiF: Mg, Ti	8.14	5_*10^{-3}-10^2	10^5	~5-10%	400C, 1 h 800C, 24 h
$Li_2B_4O_7$: Mn	7.4	10^{-2}-300	3_*10^6	5% in 60 days	300C, 15 min
$CaSO_4$: Dy	15.3	10^{-4}-3_*10^3	10^5	7-30 % in 6 months	400C, 1 h
BO	7.13	10^{-2}-50	5_*10^5	7% in 2 months	600C, 15 min

ties of dosimeters. Very fine particles and narrow size distributions, which can be reached by forming doped and un-doped LTBs in wet stage prior to thermal treatment stage, were formed. Furthermore, the synthesis and characterization of the Cu doped LTB (LTB: Cu) nanoparticles by thermal treatment method were reported previously as well (Khalilzadeh, *et al.*, 2015, Khalilzadeh, *et al.*, 2016). Table 2 is shown variety methods for preparing LTB. Figs. 1 and 2 are illustrated some results of synthesized Nanophosphors.

RESULTS AND DISCUSSION

Nanophosphor LTB and Cu doped LTB Results:

I. TEM

TEM images (Fig. 1) of LTB & LTB-Cu showing particles with spherical morphologies and almost mono-dispersed distribution. Moreover, the linearity of both materials through 10-3 to 10 3 Gy is highly acceptable as comparison to other researches results. Table 1 shows comparison of some characteristic factors between different TLD materials.

II. Linearity

The dose vs. TL response of LTB & LTB-Cu nanophosphors can be seen in (Fig. 2). The linearity is investigated in the range of medical and personal dosimetry from 0.1 by 0.15×10^3 Gy. It was found the nanoparticles show very good linearity from the named dose intervals without any superlinearity up to 0.15×10^3 Gy. The LTB illustrates a very good trend as well as the LTB-Cu. Moreover, the particle sizes of current research are compared with different researches' results, which displays in (Table 2).

CONCLUSIONS

The doped and undoped nanophosphor lithium tetraborate borate has been synthesized by innovative single step thermal treatment method. The specific objective of the present study is the named methods can synthesis remarkable nanophosphor LTB &LTB-Cu dosim-

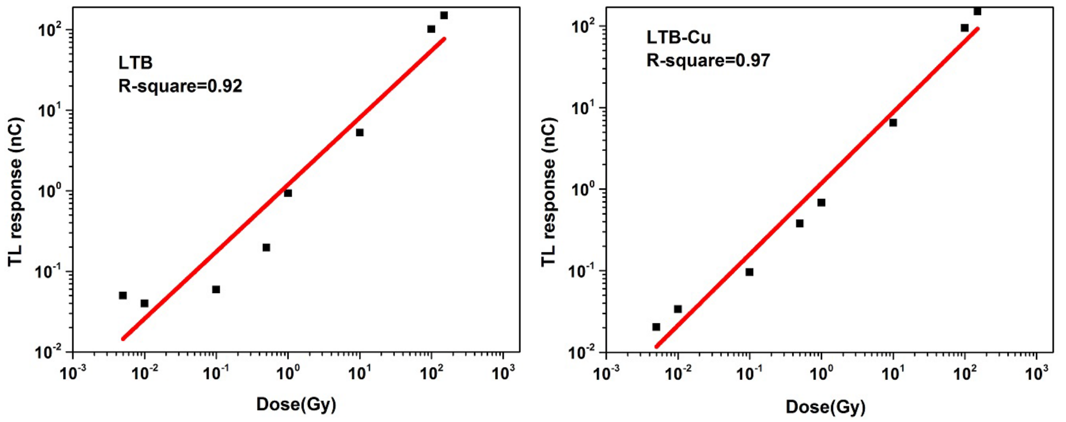

Fig. 2. The Linearity results of LTB and LTB-Cu nanophosphors

Table 2. Variety methods for preparing LTB

Synthesis method	Sample	surfactant	Dopant	Application	Size	Reference
Bridgman	LTB: Cu single crystal	-	Cu	Scintillator for neutron detection	-	Bui The Huy et al. 2009
Conventional meltquenching method	LTB Glassy sample	-	Ag	Dosimeter	-	El-Adawy, A. et al., 2010
Combustion method	LTB Nanocrystal	-	Cu	Dosimeter	25-30 nm	Singh, K. et al., 2011
Solution combustion	LTB Crystal	-	Cu, Ag	Dosimeter	-	Doull, B, et al., 2013
Single step thermal treatment method	LTB chip Nanophosphore	PVP	-	Dosimeter	-3.5 nm	Khalizadeh et al., 2013
			Cu		3.92 nm	
			Ag		3.5 nm	

eters. The most unique feature of current research is a linearity of the nanophosphor over a wide range of doses without any supralinearity and sublinearity. It should be emphasized that interesting properties can be seen in non-doped lithium tetraborate nanoparticles. It is worth noting that nanophosphor LTBs can be suggested as a Thermoluminescence dosimeter material for evaluating the absorbed dose for personnel and medical dosimetry purposes.

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