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Paper

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Improvement of neutron sensitivity for lithium formate EPR dosemeters: a Monte Carlo analysis

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Abstract

This work presents the computational analysis of the sensitivity improvements that could be achieved in lithium formate monohydrate (LFM) electron paramagnetic resonance (EPR) dosemeters exposed to neutron beams. Monte Carlo (MC) simulations were performed on LFM pellets exposed to neutron beams with different energy spectra at various depths inside a water phantom. Various computations were carried out by considering different enrichments of ^{6}Li inside the LFM matrix as well as addition of different amounts of gadolinium oxide inside the pellet blend. The energy released per unit mass was calculated with the aim of predicting the increase in dose achievable by the addition of sensitizers inside the pellets. As expected, a larger amount of ^{6}Li induces an increase of energy released because of the charged secondary particles (i.e. ^{3}H ions and α-particles) produced after neutron capture. For small depths in water phantom and low-energy neutron spectra the dose increase due to ^{6}Li enrichment is high (more than three orders of magnitude with respect to the case of with ^{7}Li). In case of epithermal neutron beams the energy released in ^{6}Li-enriched LFM compound is smaller but larger than in the case of fast neutron beams. On the other hand, the computational analysis evidenced that gadolinium is less effective than ^{6}Li in improving neutron sensitivity of the LFM pellets. Discussion based on the features of MC transport code is provided. This result suggests that ^{6}Li enrichment of LFM dosemeters would be more effective for neutron sensitivity improvement and these EPR dosemeters could be tested for dosimetric applications in Neutron Capture Therapy.

Introduction

Neutron Capture Therapy (NCT) is a specific radiation therapy characterised by a biological targeting of cancer at the cellular level. This advanced therapy combines an initial introduction and accumulation of nuclei with high neutron cross section (such as ^{10}B or ^{157}Gd) inside the tumour cells with a following irradiation with thermal neutrons. The ^{10}B or ^{157}Gd nuclei, when hit by thermal neutrons, undergo nuclear reactions where secondary charged particles (such as alpha particles, recoiling ^{7}Li nuclei or Auger electrons) with short subcellular range are released and are able to kill the tumour cells selectively[1–3]. Recently, the development of accelerator-based systems for the neutron beams has given an important impulse to the spread of the experimental research in NCT. One key element for the success of this therapy is the precise and accurate characterisation of the mixed (neutron,γ) field used. Therefore, reliable dosimetric measurements are needed in order to quantify the various (neutron and photon) components of the mixed beam and to improve the therapy. Various dosimetric systems are used for radiation detection such as ionisation chambers[3, 4], semiconductor detectors[5, 6], thermoluminescence (TLD) and optically stimulated luminescence dosemeters (OSL)[7–14], multifoil neutron activation spectrometry[3, 15] and gel dosimetry[16–22]. Each technique has its advantages and disadvantages[23].

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Dosimetry through electron paramagnetic resonance (EPR) has aroused large interest in the last years. This technique can detect the paramagnetic centres (such as free radicals and point defects) produced after exposure to ionising radiations. EPR dosimetry with alanine ($C_3H_7O_2N$) samples has been recognised as reference standard for dose measurements[24–43]. However, recently, new materials were analysed for larger sensitivity than alanine such as ammonium tartrate, ammonium formate, taurine, potassium tartrate hemihydrate, dithionates, phenols, strontium carbonate, sodium tartrate dehydrate and strontium sulphate, clear fused quartz, glasses and some others[31, 44–69].

One compound that is very promising is the lithium formate monohydrate (LFM) which is more sensitive than alanine because of its narrower EPR spectrum, tissue-equivalence, good signal stability and response also for kV photon beams [44, 70–78].

In this work, a Monte Carlo simulation analysis was carried out with the aim of predicting the EPR response of LFM dosemeters with increased ^{6}Li abundance or after addition of gadolinium exposed to neutrons beams for possible dosimetric applications in NCT. In particular, the energy released in these LFM pellets with ^{6} Li or Gd exposed to neutron beams with composite energy spectra and under different irradiation set-ups.

Monte Carlo simulation

Irradiation set-up

Monte Carlo N-Particle–MCNP5[79] radiation transport code was used to estimate the average amount of energy deposition per unit mass into lithium formate dosemeters enriched with ^{6}Li or added with gadolinium oxide. The irradiation set-up adopted was analogous to that used in previous works[42, 52, 54, 80] with the axial system symmetry (see for example Figure 1 of the article[54]). The LFM samples and the dosemeter holder are cylindrical (diameter = 1 cm) and coaxial. The dosemeters are placed inside a holder composed of two water layers (one above the dosemeter and the other below). To simulate measurements at various water depths from surface the measurements of pellets were carried out by setting a water layer (whose thickness was varied from 1.0 up to 10 cm) between the dosemeter and the neutron source. The computational analyses with ^{6}Li-enriched dosemeters were carried out by varying the isotopic abundance of ^{6}Li inside the samples. ^{6}Li/Li ratio ranges from 0% (for samples containing only the ^{7}Li isotope) up to 100% (for samples containing only the ^{6}Li isotope). Lithium with natural isotopic abundance and gadolinium oxide (Gd_2O_3) as additive were considered in the case of dosemeters with gadolinium. Gadolinium oxide, inserted as thin layers 100 μm thick, was placed between layers of lithium formate compound. The amount of gadolinium oxide inside the samples ranged between 0 and 75% of the total sample mass. As the gadolinium content increases the number of gadolinium oxide layers inside the dosemeter increases as well as the content of lithium formate decreases correspondingly.

Neutron source

A disc-shaped neutron source, coaxial with z-axis and placed 20 cm from the bottom of the dosemeter, was considered and three neutron spectra with different energy composition were simulated. More details are provided in Table 1: the first spectrum is mainly composed of thermal neutrons, the second one is a main epithermal neutron and the third one is primarily composed of a fast neutron component.

Results

In Figure 1 the results of the MC simulations related to the energy released inside lithium formate irradiated with a mainly thermal neutron beam (Spectrum A) are reported. The values of the energy released per unit mass (which is an estimate of the absorbed dose) are shown with increasing the isotopic abundance of ^{6}Li inside the samples. The values are normalised to the values without ^{6}Li, i.e. the ratio $\left(\frac{E}{m}\right)_{with\,^6Li}/\left(\frac{E}{m}\right)_{without\,^6Li}$ is shown. Various calculations were performed to simulate irradiations at different depths in a water phantom irradiation.

As expected, the absorbed dose as well as neutron sensitivity increases as the ^{6}Li/Li ratio increases. This was expected because ^{6}Li nuclei have high thermal neutron capture of (i.e. $\sigma \approx 940$ barns) and the secondary charged particles (i.e. ^{3}H and α particle) produced after neutron capture are assumed to deposit energy locally in the dosemeter layer. The absorbed dose with ^{6}Li/Li = 100% is more than 3500 times larger than without ^{6}Li.

Considering the case of Li natural isotopic abundance as reference and dividing the dose values by that absorbed by pellets with a Li natural isotopic abundance, a maximum dose increases larger than a factor 2.5 was observed. This means that large sensitivity improvements are observed for small ^{6}Li contents, with a sensitivity increase of >1000 when ^{6}Li/Li passes from 0 to 7.5% (that is, the ^{6}Li abundance in natural lithium), whereas at higher ^{6}Li contents the energy release enhancement is less evident. It is interesting to note that experimental EPR measurements performed on natural-Li and ^{6}Li-enriched lithium formate dosemeters exposed to a mainly thermal neutron beams have shown that the ^{6}Li-enriched dosemeters

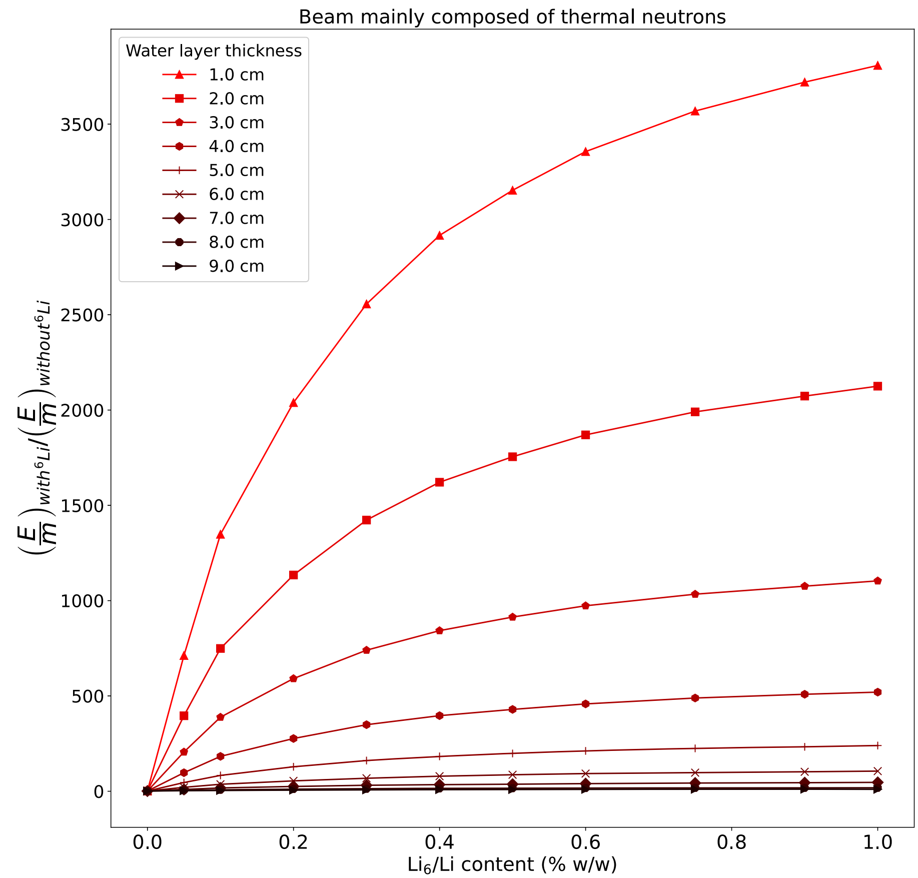

Figure 1. Energy released per unit mass with varying ^{6}Li content inside the sample for various thicknesses of the water layer above the dosemeter. In this case the neutron beam is mainly composed of thermal neutrons. The values are normalised to the values without ^{6}Li.

Table 1. Energy neutron spectra used for Monte Carlo simulation.

	Energy range (eV)			
	0–0.1	0.1–0.4	0.4–10^4	0.4–10^4
Spectrum A	70%	22%	7%	1%
Spectrum B	1%	22%	70%	7%
Spectrum C	1%	7%	22%	70%

have a sensitivity 2.5 times larger than natural-Li pellets[78]. However, differences between the results reported in Lund *et al.*[78] and MC simulations here presented should be underlined. The pellets were experimentally irradiated to a mixed neutron-gamma field, whereas the MC analyses are performed considering only neutrons as incident particles (even though the secondary photons produced during neutron transport are considered). Furthermore, MC simulations assume that energies of secondary ^{3}H and α particles are deposited locally in the dosemeter layer and do not consider effects related to possible local electron–hole recombination and cluster formation. Nevertheless, these results show that, even though MC simulations do not take into account the phenomena of recombination sand saturations of free radicals which influence the EPR response of dosemeters, the computation analysis here performed is able to provide an estimate of response improvement due to ^{6}Li content inside the pellets.

Figure 1 shows that the energy released inside the LFM layer decreases with increase in the thickness of the upper water layer because of the attenuation of the incoming beam inside this layer. The enhancement

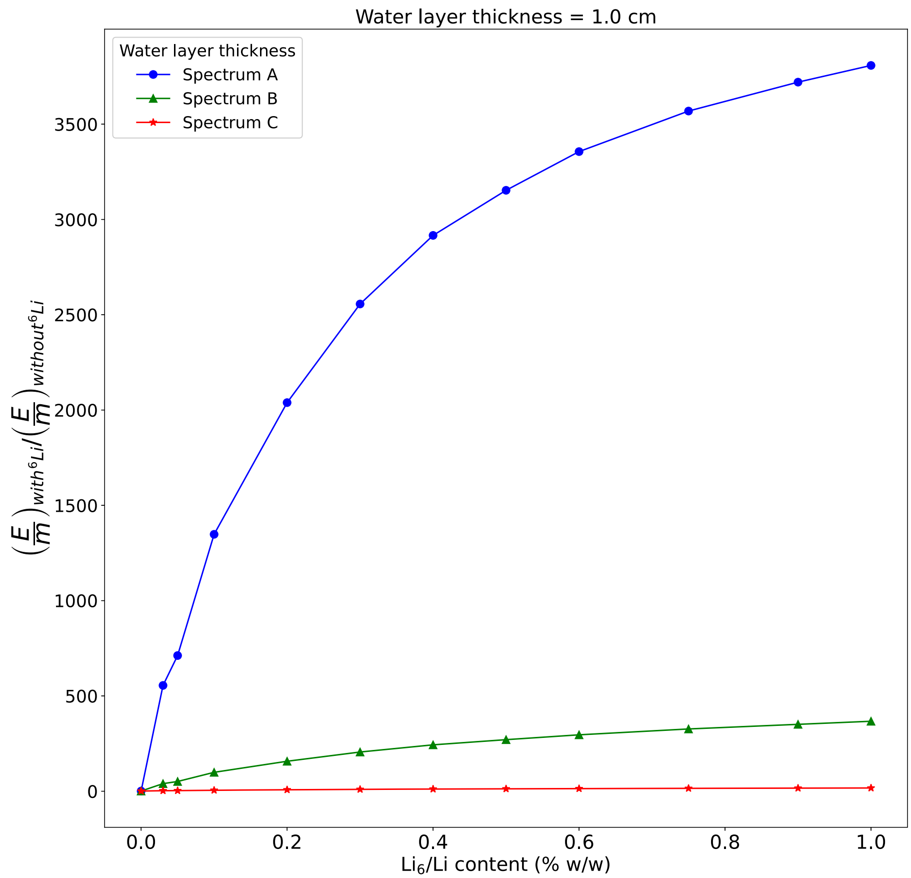

Figure 2. Comparison of energy released per unit mass for lithium formate samples exposed with different neutron spectra ('Spectrum A': mainly thermal neutron beam, 'Spectrum B': mainly epithermal neutron beam, 'Spectrum C': mainly fast neutron beam). The values are normalised to the values without ^{6}Li. The water layer above the pellet is 1 cm thick.

decreases with increase in the thickness of the water layer above the dosemeter (i.e. as the depth at which it is placed increases) and this is due to the fact that the thermal neutron component decreases with the depth. Variations of neutron energy spectra occur with a reduction of the thermal neutron component since the number of epithermal neutrons which are thermalized does not compensate for the thermal neutron losses. This reduces the signal enhancement due to ^{6}Li presence. These trends also provide information about the ability of these neutron beams to penetrate in water and release energy in depth and this could be useful for treatment planning with these neutron beams.

Figure 2 reports the comparison of the Monte Carlo results for lithium formate irradiated with neutron beams characterised by various energy spectra. In this case the thickness of the water layer above the dosemeter is 1 cm. For all neutron spectra considered the energy released increases with the ^{6}Li content. Furthermore, the absorbed dose decreases with increasing the average energy of the neutron beam and this is because ^{6}Li nucleus presents a very high neutron capture cross section for thermal energies, whereas the cross section is smaller for epithermal neutrons and much smaller for fast neutrons. If the energy released for 'Spectrum A' beam is considered as reference, the absorbed dose is about three times smaller for the 'Spectrum B' beam and about eight times smaller for the 'Spectrum C' beam.

The other target of this work is the investigation of the effects on neutron sensitivity related to the addition of gadolinium oxide. Figure 3 shows the energy released per unit mass with increase in the gadolinium oxide concentration inside the dosemeters when the beams is mainly composed of thermal neutrons. The lithium present inside the LFM layers has natural isotopic abundance.

The trend observed decreases with the concentration of Gd_2O_3 inside the pellet. The absorbed dose is decreased to about 30% of its maximum value for

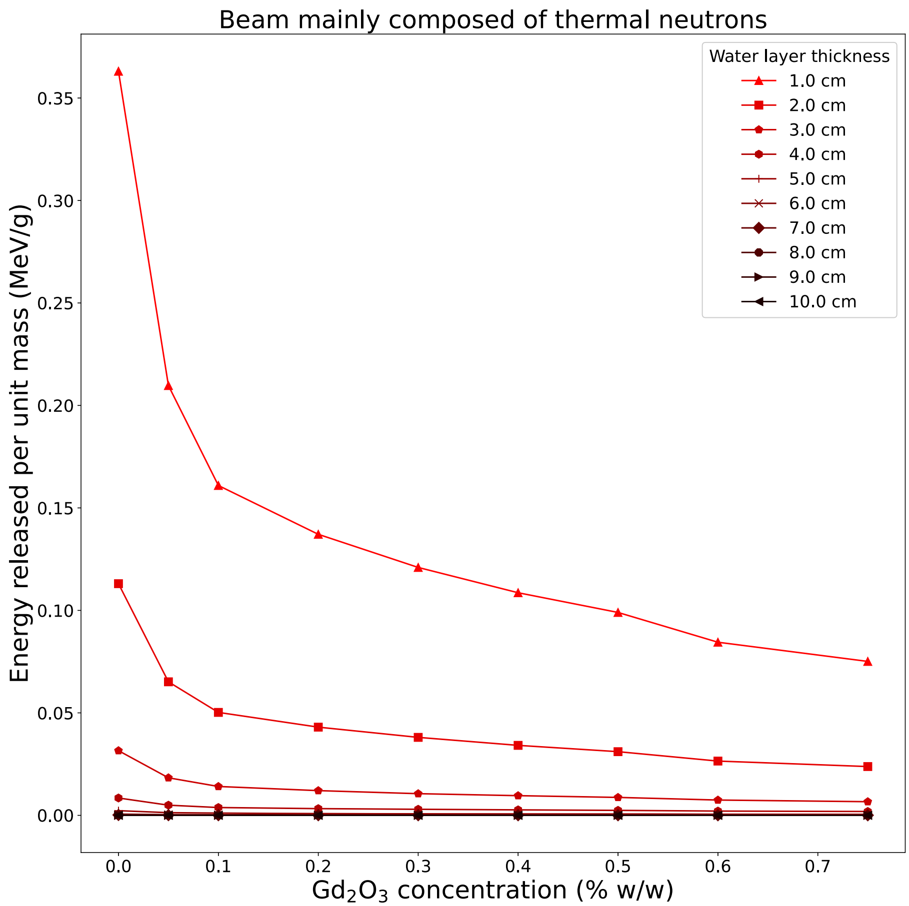

Figure 3. Energy released per unit mass with varying Gd_2O_3 content inside the sample for various thickness of the water layer above the dosemeter. In this case the neutron beam is mainly composed of thermal neutrons.

Gd_2O_3 concentration of 75%. To explain this result some details about the MC computations should be considered. First, MCNP5 does not consider the transport of the secondary charged heavy particles (i.e. ^{3}H and α particle) released after neutron capture by ^{6}Li nuclei but assumes that energies released after the interaction are deposited locally in the place where the neutron capture occurs[79]. Since the Li nuclei are only present inside the dosemeter layer, this implies that the energy released after neutron capture by ^{6}Li is totally absorbed by the LFM mass and guarantees that no energy escapes from the pellets and contributes to pellet dose. In the case of gadolinium-added dosemeters the system is modelled through a sequence of LFM and gadolinium oxide layers. This requires that the neutron interactions happen more frequently inside the gadolinium layers and only some of the secondary particles released can reach the lithium formate layers and contribute to the energy released in them. Therefore, in this last case even though gadolinium has a larger cross section for neutron capture, the total energy released inside the sensitive layers of the dosemeters added with gadolinium oxide is smaller than that due to the presence of ^{6}Li in simple LFM dosemeters. Moreover, an increase of the gadolinium oxide content involves a reduction of the lithium formate inside the pellets, and this justifies the decrease of the absorbed dose with Gd_2O_3 content inside the pellets. Also, in this case of Gd addition the absorbed dose inside the LFM pellet decreases as the water layer becomes thicker and therefore the energy released decreases with increase in the depth the pellets are placed at (see Figure 3). Furthermore, the energy released decreases with increasing the average energy of the neutron beams (data not shown). The absorbed dose is decreased to about 40% of its maximum value for Gd_2O_3 concentration of 75% for 'Spectrum B' neutron beam and to about 50% of its maximum value for Gd_2O_3 concentration of 75% for 'Spectrum C' neutron beam.

These MC results allow to conclude that the addition of gadolinium oxide could not improve significantly the neutron sensitivity of lithium formate pellets, whereas the increment or ^{6}Li content inside the sample could be more effective in improving the EPR response of lithium format to neutron beams. Experimental activity will be performed in future to investigate the actual dependence of EPR response of lithium formate after ^{6}Li or Gd addition.

Conclusions

In this work, analyses of the energy released for unit mass in lithium formate dosemeters exposed to neutron beams were carried out by means of Monte Carlo simulations having the target to predict the sensitivity enhancement achievable through ^{6}Li or Gd addition. As expected, the ^{6}Li addition involves an increase of neutron sensitivity because of its high ^{6}Li cross section for neutron capture and because of the charged secondary heavy particles being able to release energy inside the sensitive material layers. The maximum improvement was obtained for the beam mainly composed of thermal neutrons. The gadolinium addition is less effective than ^{6}Li in increasing the dose release inside the dosemeter layers. All results presented suggest that the ^{6}Li is preferable for improving neutron sensitivity of lithium formate pellets for dosimetric applications in NCT which exploits epithermal neutron beams for treatment of deep tumours.

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