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Study of structural properties in polymeric matrix with tungsten particles filler by means of positron annihilation lifetime spectroscopy (PALS)

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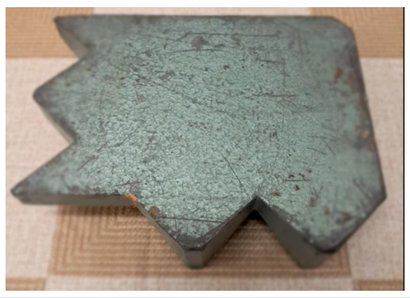
Introduction

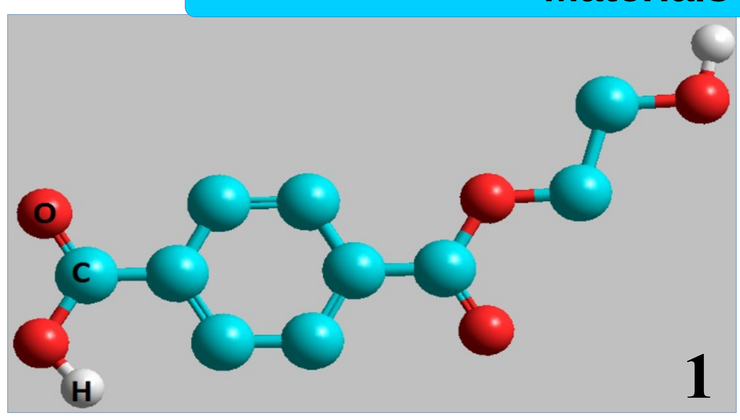
In fields using ionising (gamma, γ) radiation, such as nuclear power and physical sciences one of the most important issues is the safety of people profesionally exposed and the general population. This work deals with one of the three main principles of work with radiation, that is radiological shielding. This work is an introduction to further research.

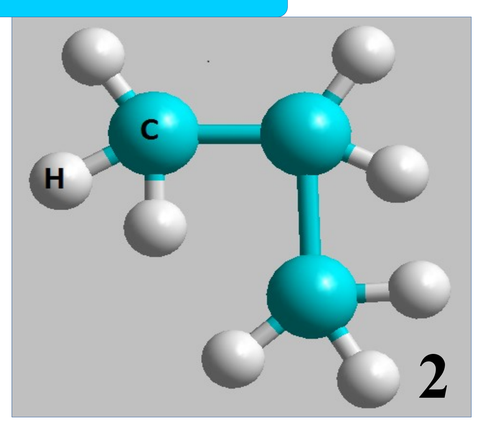
Research problem

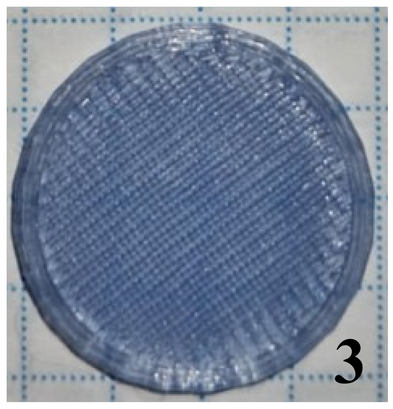
Lead based shields	Composite: polymer + tungsten oxide
Good γ attenuation properties	γ attenuation not examined yet
Lot of research	Initiatory research
Widely used	Accesible; recyclable polymer
Heavy	Light weight
High price	Low price
Toxic	Non-toxic

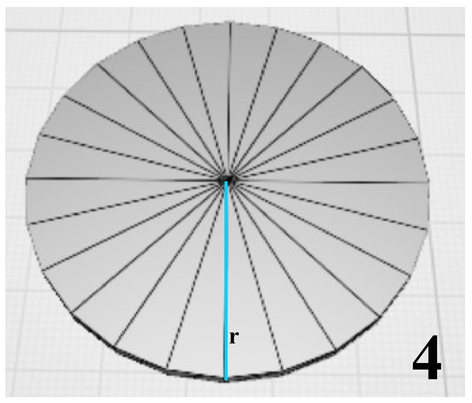


Materials





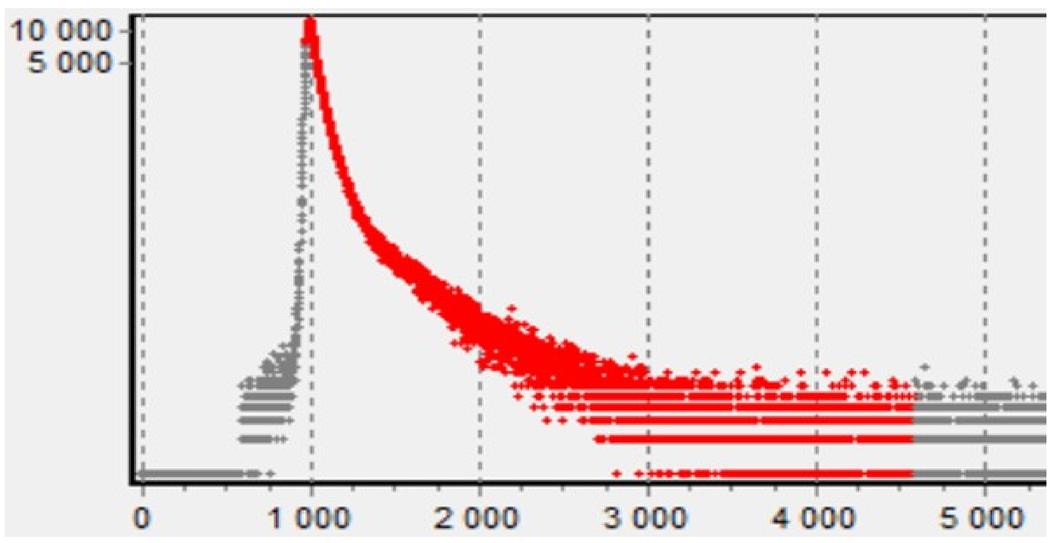




1 – mer structure of PET (polyethylene terephthalate); 2 – mer structure of PP (polypropylene); 3 – 3D printed PET sample; 4 – 3D project for all samples; 5 – 3D printed PP sample; C – carbon; O – oxygen; H – hydrogen.
Diameters of samples: r = 10 mm; h = 2 mm

Methodology

PALS - non-destructive method, based on the interaction between electron and positron. After emission from radiation source, positrons thermalise in matter and are involved in a free state or bound state. Annihilation process is completed and gamma quanta are emitted. We measure the time from emission to annihilation in a material (positron lifetime spectrum). In polymers, there are three components, where the one with the longest lifetime is associated with ortho-positronium annihilation in material's free volumes.



Tao-Eldrup model (T-E) involves equating free volume's shape to a sphere. Knowing the value of the longest lifetime, we can calculate average radius (R) of the free volume's in a material. In the formula, ΔR = 1,66 Å (empirical).

$$\tau_3 = 0,5 \cdot \left[1 - \frac{R}{R+\Delta R} + \frac{1}{2\pi} \cdot sin\left(\frac{2\pi R}{R+\Delta R}\right)\right]^{-1}$$

x3-x2-CDA (coupling decomposition algorithm) - allows for approximating the changes in the structure of the composite, relative to the pure material. This is possible by converting the three components of the positron lifetime into two components that describe the volume with the added tungsten oxide.

As composite matrix, we used recycled PET (polyethylene terephthalate) and PP (polypropylene). In the current ecological situation, it is also necessary to look at the aspect of recycling when choosing the materials to be tested. Polymer samples were created by 3D printing In the next step of research, we plan to mix in tungsten oxide.

Results

PALS measurements were performed using fast-fast coincidence method. Time resolution of the ORTEC spectrometer was about 210 ps. We used Na-22 source sealed in Kapton films. The radius and volume of an average free void in material were calculated using T-E model. In the table below, the samples are as follows: PET-R, PP-R – samples measured previously; PET-U, PP-F – current results; PET-L [1], PP-L [2] – reference results.

Variable	PET-R	PP-R	PET-U	PP-F	PET-L	PP-L
τ_3 [ns]	1,57	2,11	1,62	2,17	1,75	2,22
I_3[%]	9,01	13,27	13,76	12,52	12	25
R [Å]	2,42	2,95	2,47	2,99	2,61	3,04
V [Å^3]	59	107	63	113	74	118

Conclusions

1. We obtained higher I_3 intensities of the τ_3 component, than in previous study where old recycled materials were used. In new, commercial materials (current research and reference articles) the I_3 value is higher.

2. The experimental results indicate that PP has larger free volumes in its structure than PET. This is probably due to differences in the structures of these polymers. Long chains in the PP structure allow for the creation of space for o-Ps, while aromatic rings in PET create areas of greater order, which are not sufficient for o-Ps.

3. 3D printing technology allows for precise production of the desired sample shapes; this does not negatively affect the structural properties of the material. The obtained results are comparable both with the previous experiment (production of samples was carried out by thermal compression of many thin discs) and with reference articles (where sample's thickness was 1,5 mm [1] and 1 mm [2]).

4. Based on the obtained results, we assume that the tested polymers will be able to easily create composites with microparticles of tungsten oxide. In such composites, it will be possible to build WO_3 into the polymer structure. The obtained composite will be tested for gamma radiation attenuation.

5. In future studies the x3-x2-CDA model will be used to compare the free volumes in pure polymers and composites.

Acknowledgements

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Reference articles

[1] https://doi.org/10.1016/0032-3861(94)90042-6 [2] https://doi.org/10.1021/acs.macromol.2c01430