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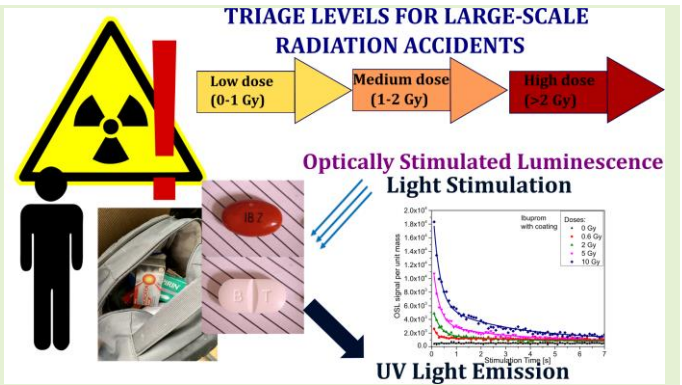
Popular medicines as radiation sensors

Anna Mrozik*, Paweł Bilski

Abstract— During an uncontrolled release of radiation, it is highly unlikely that members of the public will be equipped with personal radiation dose monitors. In preparation for such a situation various personal objects are being investigated as **emergency dosimeters**. The already developed methods are not satisfactory, as they are time-consuming or require destruction of things valuable for victims.

Here we show, that common pharmaceuticals, which frequently may be found e.g. in personal bags, are the perfect candidate as radiation sensors in emergencies. We investigated several over-the-counter medicines for occurrence of the optically stimulated luminescence phenomenon and found that all of them exhibit strong luminescence signal following exposure to ionizing radiation. Its intensity increases linearly with the absorbed dose. The highest sensitivity was shown by the popular painkillers based on ibuprofen and paracetamol. The intensity of their luminescence signal was found to enable measurement of doses well below 1 Gy, what is sufficient for application in emergency dosimetry. Pharmaceuticals are also free of all disadvantages of other emergency dosimeters: their composition is standardized, sampling is immediate, the unit value is usually negligible. We expect our results to be a starting point for broader investigations of various medicines, which should provide a perfect tool for emergency dosimetry.

Index Terms— emergency dosimetry, monitoring radiation, optically stimulated luminescence, radiation sensors



I. INTRODUCTION

In case of a large scale radiation incident (e.g. a major nuclear accident or a terrorist attack with a dirty bomb), thousands of people could be exposed to an unknown amount of ionizing radiation. In such a situation there will be a need for a fast and simple method of assessing the absorbed doses in order to implement the *triage*, i.e. segregation of victims according to the degree of injury to select persons requiring immediate medical treatment [1]. The general public is normally not equipped in dedicated radiation dose monitors. For that reason, in recent years scientist have been investigating effects induced by radiation in various personal objects, that are usually kept close to a human body, with the intention to use them as so-called emergency dosimeters [2-4].

Most of the methods developed for this purpose are based on the luminescence phenomena and especially on optically stimulated luminescence (OSL). Radiation interacting with matter creates free electrons and holes, which in some cases

might be trapped at metastable energy levels related to defects within the substance and remain there for a long period of time [5]. Illumination with the light of a specific wavelength (e.g. blue) may lead to releasing charge carriers, followed by their recombination with the emission of light of a wavelength different than that used for the stimulation (e.g. UV). If the intensity of the emitted light is in proportion to the absorbed dose, a given material may be used as a dosimeter. The OSL phenomenon is nowadays one of the main techniques exploited for personal dosimetry in radiation protection, where high-sensitive detectors made of aluminum oxide [6] or beryllium oxide [7, 8] allow for measuring even very low doses at microgray range. OSL is also used for establishing the age of archaeological objects [9, 10] or geological sediments [11, 12] through measuring natural radiation doses accumulated in them over hundreds and thousands of years.

In the search for the materials suitable for use as emergency dosimeters, the most attention was paid so-far to components of personal electronic devices (mainly smartphones): resistors, capacitors, inductors, integrated circuits [13-16]. Other studied materials included clothing [17], banknotes [18], credit and ID cards [19], plastic from bags and backpacks [20]. Some of the developed methods (especially these based on electronic components) work quite well under laboratory conditions [14, 21], when there is time for laborious preparation and analysis of samples, but their effectiveness is doubtful for real emergencies and field applications, when time factor will be

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of importance. Another very serious disadvantage of using components of electronic devices or other valuable items is that they have to be destroyed to perform a measurement. Especially destroying the victims' mobile phones during a major disaster seems hardly acceptable. The drawback of other proposed materials, like e.g. plastic extracted from various personal items, is the fact that they are produced by a huge number of manufacturers and have non-standardized composition, therefore their luminescent properties are highly variable and not reproducible.

In this paper, we study the possibility of using as emergency dosimeters materials not included so far in the broader research: pharmaceuticals. Although obviously, not everyone carries medicines, it can be assumed that a large part of adults does. Pharmaceuticals are free of all disadvantages listed above: their composition is standardized and repeatable, sampling is immediate and the unit value is usually negligible. Additionally, in most cases, medicines are packaged in a way that protects them from environmental factors, often also from light. Medicines, like practically all non-metallic substances, show some radiation-induced luminescence [22-24]. The basic questions to be answered is whether the intensity of this luminescence is sufficient to measure radiation doses relevant for emergency dosimetry and if so, if this property is prevailing among popular pharmaceuticals. This second condition is equally important as the first one, because even perfect dosimetric properties, but found only in one or two among common drugs or exhibited by drugs rarely carried on the person, would not allow for considering medicines as promising emergency dosimeters.

II. MATERIALS AND METHODS

Within our study, we investigated optically stimulated luminescence of 11 very common medicines, which are likely to be found e.g. in personal bags (Table 1). All of them are over-the-counter analgesics and antipyretics in form of tablets, based on paracetamol (acetaminophen), ibuprofen, acetylsalicylic acid or salicylamide.

Each of the tablets consists of a core and coating. The core contains active substance and other excipients. The features of tablet coatings are very diverse (Fig. 1). Some of them have a thick, glossy coating (e.g. Ibuprom or Nurofen) when the coating of the others (e.g. Panadol or Paracetamol) consists of a very thin, colorless film. All medicines subjected to investigations were purchased randomly at drugstores in the Kraków area in the years 2019-2020. The pure chemical forms of the active ingredients were purchased at Sigma-Aldrich: ibuprofen (PHR1004), paracetamol (Y0001781) and acetylsalicylic acid (A5376).

Nearly all OSL measurements were conducted using the specialized automated Risø DA-20 TL/OSL reader [31,32] (manufactured by DTU Nutech, www.nutech.dtu.dk). For optical stimulation, the reader has built-in two sets of LEDs which emit light at the wavelength of 470 nm (the detection window 300-400 nm) and 870 nm (the detection window 300-700 nm). The reader has also a built-in $^{90}\text{Sr}/^{90}\text{Y}$ beta radiation source for exposure of samples. As a light detector, the EMI

9235QB photomultiplier is used. The maximum thickness of the samples, which can be accommodated in the DA-20 reader is about 1 mm, therefore for measurements of whole medicine tablets another device, with a larger measuring chamber, was applied: the manual Helios reader (manufactured by ZeroRad, <http://zero-rad.com>). Helios reader is equipped with Hamamatsu H8259-01 photomultiplier. The stimulation light wavelength of this reader is 470 nm and the detection window in 300-400 nm, similarly as in DA-20. The sensitivity and the power of stimulating light of Helios reader are however lower than that of DA-20. For measurements with the Helios reader, the samples were irradiated with ^{60}Co gamma-rays.

Most of the OSL measurements were performed on the powder samples. The following standard procedure was adopted: grinding tablets into powder, putting powdered material into a measuring cup, placing the cup in the DA-20 reader, illumination of the sample with blue LEDs to remove the background signal, irradiation with $^{90}\text{Sr}/^{90}\text{Y}$ beta-rays, measurement of the CW-OSL signal. From part of the tablets, the coatings were removed before the grinding. For the measurements, as much powder material was used, as could be accommodated in the volume of the measuring cup (diam. 9 mm, depth 1 mm). Depending on the powder density, the mass of samples varied between 30 mg and 60 mg. All OSL results were normalized to the mass of a sample. In the case of the dose recovery tests (fig. 7B), the tablets were irradiated before grinding them into powder and there was no background removal before the measurement.

TABLE I
LIST OF THE INVESTIGATED MEDICINES

NO	MARKET NAME	ACTIVE SUBSTANCES	MANUFACTURER
1	Ibuprom	ibuprofen	US Pharmacia
2	Ibuprom Max	ibuprofen	US Pharmacia
3	Ibuprom Zatoki	ibuprofen, pseudoephedrine	US Pharmacia
4	Nurofen	ibuprofen	Reckitt Benckiser
5	Nurofen Forte	ibuprofen	Reckitt Benckiser
6	Paracetamol	paracetamol	Accord
7	Panadol	paracetamol	GlaxoSmithKline
8	Panadol Extra	paracetamol, caffeine	GlaxoSmithKline
9	Aspirin	acetylsalicylic acid	Bayer
10	Polopiryna S	acetylsalicylic acid	Polpharma
11	Scorbolamid	salicylamide, ascorbic acid, rutoside	Polpharma

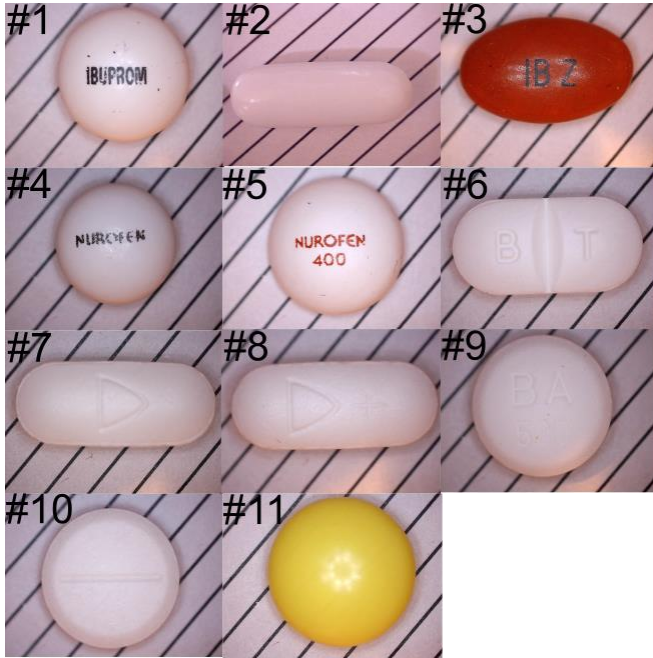


Fig.1. Photos of investigated medicines. Photos showing the investigated medicines in the following order: #1 Ibuprom, #2 Ibuprom Max, #3 Ibuprom Zatoki, #4 Nurofen, #5 Nurofen Forte, #6 Paracetamol, #7 Panadol, #8 Panadol Extra, #9 Aspirin, #10 Polopiryna S, #11 Scorbolamid. The distance between lines of the background is 3 mm.

III. RESULTS AND DISCUSSION

For the blue stimulation a quite considerable OSL signal was measured for all samples. This is illustrated for all investigated pharmaceuticals in Fig. 2 (A, B, C, D), which presents OSL signals (so-called decay-curves) measured under the continuous blue light stimulation. The shape of the OSL decay-curves is quite similar, but the curves show differences in intensity and decay rates.

Additionally, the insets in Fig. 2 present for the selected samples the effects of various radiation dose on OSL intensity. It is also apparent that OSL intensity increases with the increasing dose and the doses well below 1 Gy are measurable. Such sensitivity is adequate for emergency dosimetry. The most commonly accepted triage approach categorizes doses into three ranges: low dose (0-1 Gy), medium dose (1-2 Gy) and high dose (> 2 Gy) [25]. The most critical triage dose level is 2 Gy, above which special medical treatment is required, while persons with lower doses may for the time being remain at home, relieving in this way medical

facilities [2]. The choice of 2 Gy as the critical triage level is based on the data from the Chernobyl victims [26], which indicate that for lower doses no deaths were recorded.

The shape of OSL decay curves can be described by a negative exponential function of time (for constant light stimulation and assuming first-order kinetics). The OSL decay-curve is described by exponential formula: $I_{osl}(t) = I_0 \exp(-t/\tau)$, where, I_0 is the initial OSL intensity at $t = 0$ and τ is the decay constant. Usually, to accurately describe the curves, it is necessary to use a sum of several exponential components, corresponding to the recombination of charge carriers released from different trapping sites.

To show the behavior of the investigated samples three of them were presented (representatives from the groups of pharmaceuticals with different active substance).

The decay curves presented in Fig. 3 A and similarly in 3 B can be described by two exponents and a constant factor to obtain a satisfactory fit. In the case of Panadol (active substance – paracetamol), another mathematical formula was used to describe the OSL decay-curve (Fig. 3 C). It is so-called stretched-exponential function:

$I = I_0 \exp[-(t/\tau)^\beta]$ with $0 < \beta < 1$ [27]. Parameters β and τ depend on excitation conditions. The main reason for the occurrence of the stretched-exponential function is that the electron may return from the conduction band to the electron traps (retrapping). In the case of Panadol sample, there are present two fast components of similar time constants, but one component is a stretched-exponential function with β -parameter equals 0.36.

Fig. 4 compares OSL intensity of all investigated medicines, measured after grinding tablets into powder. Ibuprom data, presented in fig 1A, corresponds to an intermediate level of OSL sensitivity. The difference between the highest (Ibuprom Max and Panadol Extra) and the lowest (Aspirin) signal is about sevenfold, nevertheless, the sensitivity of all investigated medicines is sufficient for establishing triage dose levels. In some cases significant differences between sensitivity of medicines based on the same active substance are observed (e.g. Ibuprom and Nurofen, which both contain 200 mg of ibuprofen). For each type of medicines, two values are presented: with and without coating (i.e. with the coating removed before grinding). The coatings of tablets were found to have a significant influence on the OSL signal. This influence is mixed: for some medicines presence of a coating decreases luminescence (e.g. Ibuprom Max), while for others quite oppositely increases OSL signal (e.g. Panadol Extra). The thick, glossy coating of Ibuprom tablets most probably simply attenuates the light but in the case of samples (e.g. Panadol) - where the coating is very thin, colorless film - it seems that a considerable luminescence signal is generated in the coating itself.

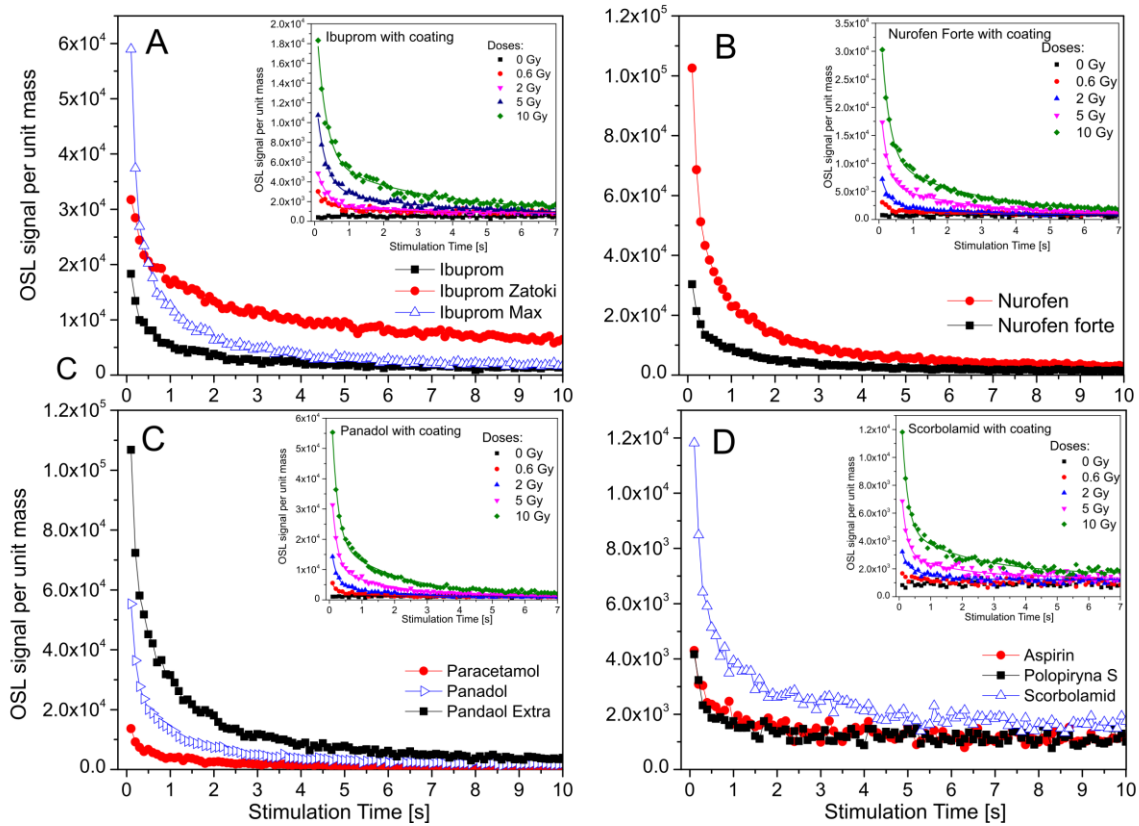


Fig. 2. Examples of OSL decay curves. OSL decay-curves of tablets (powdered together with the coating) with ibuprofen (A and B), paracetamol (C) and acetylsalicylic acid or salicylamide (D), recorded after irradiation with 10 Gy of beta-rays. (Inset - OSL decay-curves of selected tablets recorded after irradiation with different doses (0.6-10 Gy) of beta-rays. The intensity of the OSL signal increases with increasing doses. OSL signal is normalized to the mass of the samples).

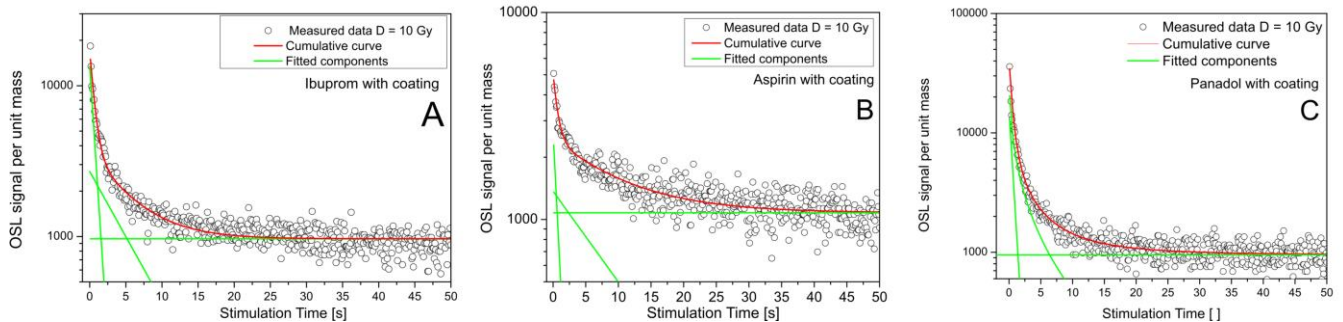


Fig. 3. Deconvolution of the OSL decay-curves. Exemplary deconvolution into single components of the Ibuprofen (A), Aspirin (B) and Panadol (C) OSL decay-curve. Independently from the value of the radiation dose, two exponentially decaying first-order components (and constant background) were needed to fit the decay-curve with R -square parameter of 0.99 (fitted time constants are for Ibuprofen - 0.6; 5.03; Aspirin - 0.69; 9.9 and Paracetamol - 0.14 ($\beta=0.36$); 0.5).

From the practical point of view of emergency dosimetry, removing the coating is an undesirable operation, as it increases the time required for a measurement. Ideally, the measurement would be performed using the whole, not-crushed tablets. Figs. 5 A and B present exemplary results of such measurements performed for two types of tablets with distinctively different coatings, compared with the results for the tablets cut in half and measured with the inside facing the light-detector. The results confirm previous observations. The thick coating of a Nurofen Forte tablet suppressed completely the OSL signal. An opposite effect is observed

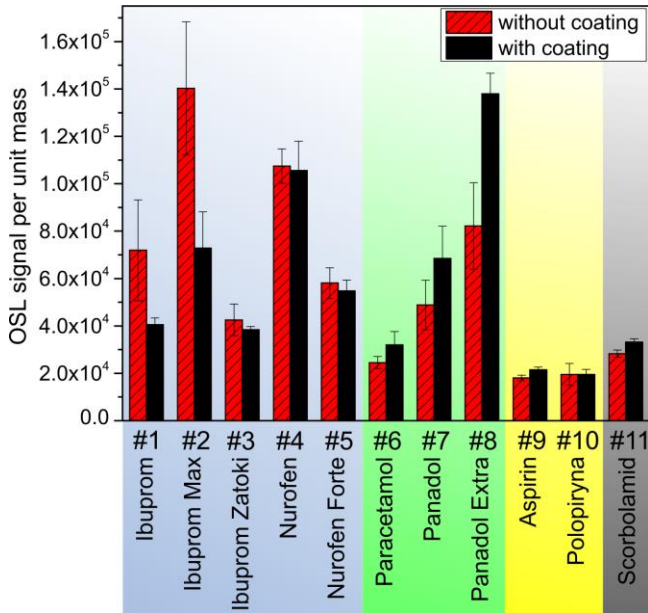


Fig. 4. Sensitivity of investigated pharmaceuticals. Comparison of the OSL intensity of all investigated medicines. Error bars represent 1 SD of 15 measurements.

for Panadol Extra, as the signal of the whole tablet (i.e. measured through the coating) even exceeds that measured from the inner side. These results suggest that each type of drugs may require developing a specific measuring procedure. Each medicine, besides active substances, contains also several other ingredients. A natural question is therefore where the luminescence signal originates from? While the answer to this question is actually not important for the discussed dosimetric application, to get some insight into this issue, we performed a test with chemically pure compounds of ibuprofen, paracetamol and acetylsalicylic acid. The results are presented in Figs. 6 A and B. In the case of ibuprofen, the shape of the OSL decay-curve is nearly identical for both, the pure compound and Ibuprom medicine.

The intensity of luminescence is slightly higher for the pure ibuprofen, which may be easily explained by the presence of inactive ingredients in the Ibuprom. These results, while not conclusive, tend to suggest that in the case of this medicine, most of the OSL signal originates directly from ibuprofen. A similar conclusion may be drawn also from the results of acetylsalicylic acid (not shown). For paracetamol, the situation is less clear. The OSL intensity of the pure compound is almost twofold of that of Panadol medicine. On the other hand, the shape of the decay-curve is significantly different and consists of only a very fast (below one second) decaying component. Taking into account the data from Fig. 5 (effect of coating), one may speculate that the OSL of Panadol is a

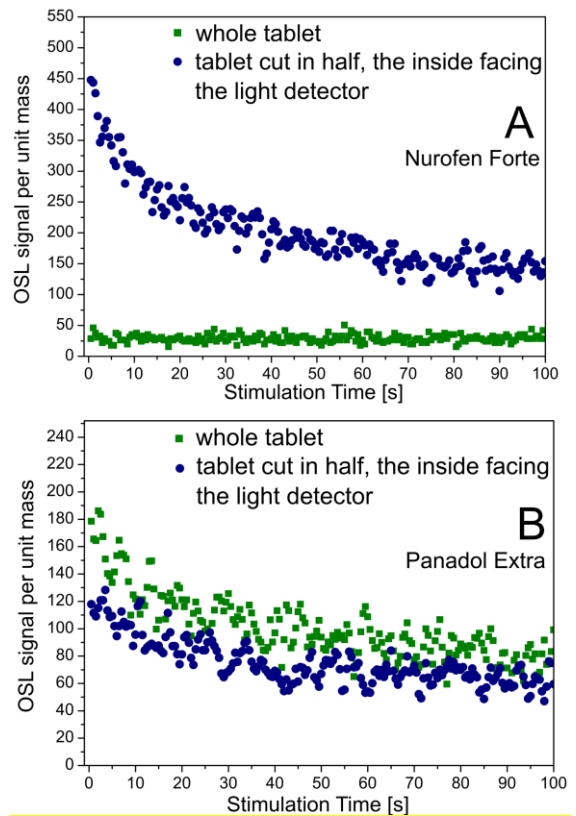


Fig. 5. Influence of coating. OSL decay-curves of Nurofen Forte (A) and Panadol Extra (B), measured for the whole tablets and tablets cut in half and measured with the inside facing the light-detector. Note that to accommodate the large tablets a different measuring OSL device (Helios reader) had to be used, therefore the numbers given in fig. 5 cannot be compared directly with any other.

mixture of the signal from paracetamol, but strongly attenuated by some ingredients of a tablet, and the signal from the film-coating.

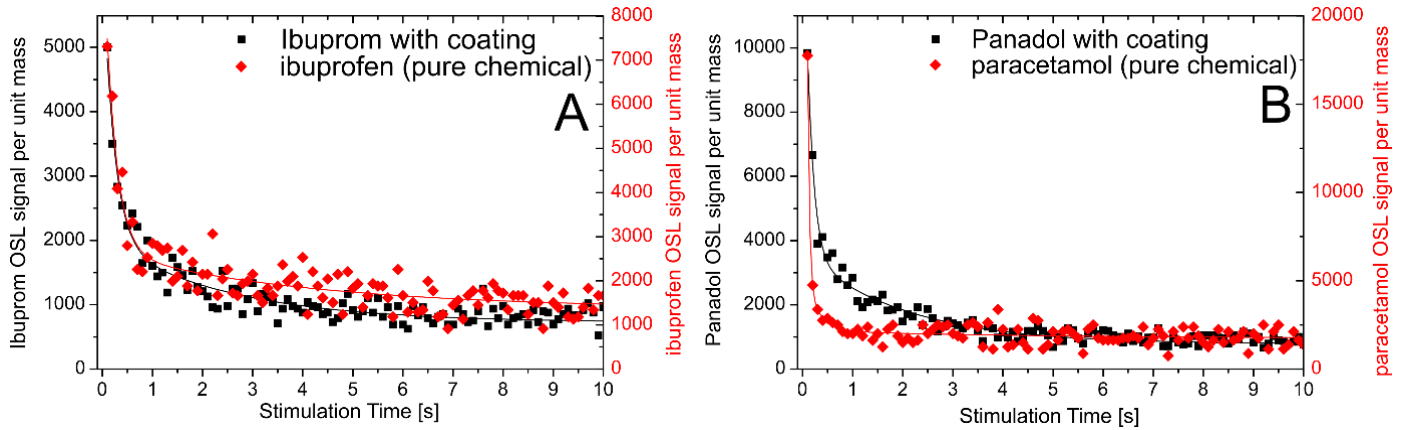


Fig. 6. Comparison of OSL decay curves of investigated pharmaceuticals and pure chemicals. Comparison of pure chemical ibuprofen (A) and paracetamol (B), and complex pharmaceuticals containing these substances (Ibuprom (A) and Panadol (B)).

A very important characteristic of each material to be used as a dosimeter is the dose-response, i.e. the dependence of the signal on the absorbed dose. Fig. 7 presents such data for all investigated samples, measured with the tablet coating. The dose-response in all cases was found to be almost perfectly linear within the tested dose range 0.37 Gy – 10 Gy.

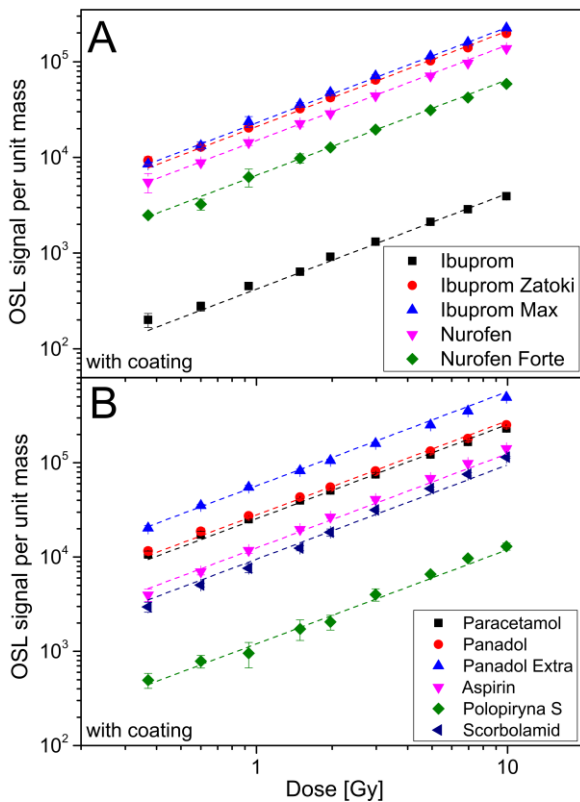


Fig. 7. Dose characteristics of investigated samples (A - pharmaceuticals with ibuprofen and B - pharmaceuticals with paracetamol, acetylsalicylic acid or salicylamide). The dose-response for the different pharmaceuticals was investigated over the dose range from 0.37 Gy to 10 Gy. The dashed lines are the linearity functions of experimental data).

All discussed so far results were obtained with the samples from which any preexisting background signal was removed before irradiation by exposing them to light. These results represent therefore the true intrinsic properties of the studied materials, but in a real emergency situation such approach is obviously not applicable and one has to deal with a possibility of occurrence of a background luminescence signal. It was found that investigated materials demonstrate an initial signal which is not related to radiation dose. A particularly high initial signal was found for medicines based on ibuprofen.

Such background, called usually an ‘initial native signal’, is a well-known problem in emergency dosimetry using e.g. plastic, business cards or banknotes [3,27,18]. The initial signal is an effect of physical or chemical treatment of a given material during its production or utilization. The method of removing the initial native background was proposed by Sholom et al. [28]. It is based on the difference between the shape of the initial signal and the dosimetric signal decay-curves. The decay of the signal induced by radiation is very fast. A great majority of luminescence signal is emitted during the first a few seconds of stimulation, and there is no slow decay-curve components lasting several tens of seconds (see Fig. 2). Oppositely, the initial native signal decays at a much slower rate. This is illustrated in Fig. 8A. The curve marked (3), representing a pure dosimetric (i.e. induced by radiation) signal, after a few seconds of stimulation reaches the background level of an unirradiated sample (2). The curve marked (1), representing a sum of the initial native signal and dosimetric signal, decays slowly and even after 50 s does not reach the background level. The idea of the method is to fit with an exponential function the part of the curve (1) for the range of stimulation time for which the dosimetric signal is known to be close to zero. Then the fitted function is extrapolated to the shortest times and it is subtracted from the measured curve (1) to obtain the dosimetric signal. Fig. 8 B presents the results of the test of this method: comparison between the decay-curve obtained in the described way (‘recovery signal’) and the reference curve (‘dosimetric signal’). The difference between the integrated signals is in the range of a few percents.

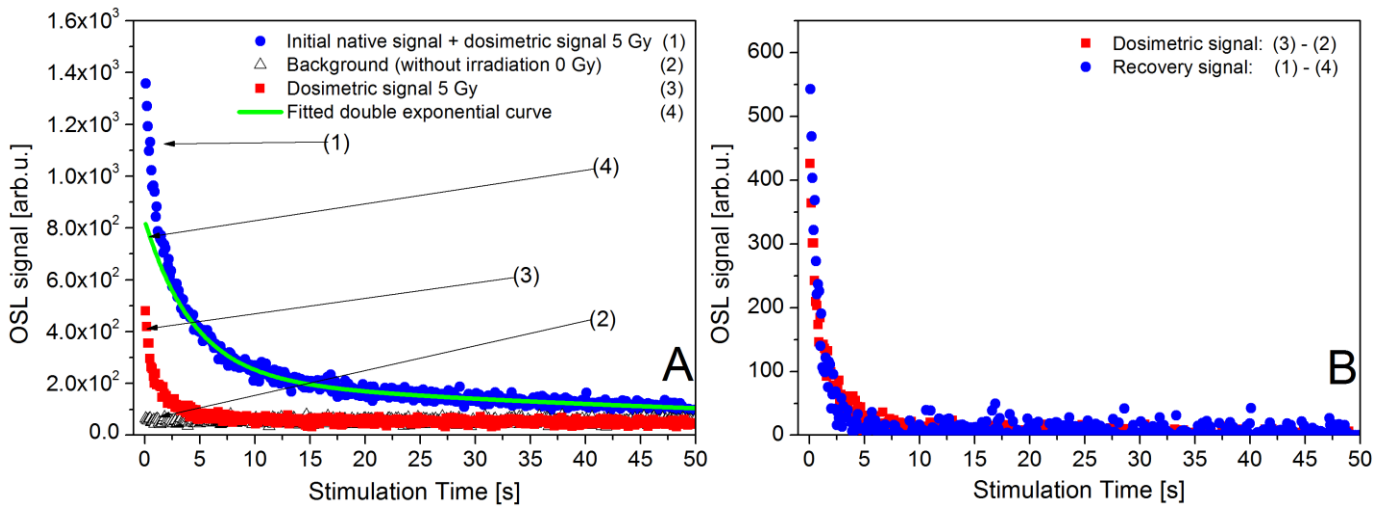


Fig. 8. Subtracting initial native signal. **A.** Three OSL curves, measured with the same sample in a sequence, are shown: (1) - the decay-curve of the sample irradiated with a dose of 5 Gy, containing the initial native signal, (2) - the OSL data from the same sample, but following illumination with blue light, which removed the initial signal (considered as the overall instrumental background), (3) - the OSL decay-curve from the same sample after the second exposure to a beta dose of 5 Gy. The line marked (4) represents the double exponential fit to the data of (1) over the range [6 s – 50 s] ($y=609\exp(-x/4.4)+220\exp(-x/67)$). **B.** Comparison between the result of the initial signal subtraction: curve (1) minus curve (4) compared with the curve (3) minus (2), representing a pure 5 Gy signal.

Due to the presence of the initial native signal, there was a need to perform a more realistic test of capabilities of medicines for measuring emergency doses by including also the presence of this background. To do this, the whole untreated tablets were firstly irradiated with a given dose, then turned into a powder (what also might induce some spurious luminescence) and finally, their OSL signal was measured. The results are presented in Fig. 9 as the ratio of the true doses and measured doses. All measured values are within 20% around the true values, what may be considered as a quite good result for emergency dosimetry.

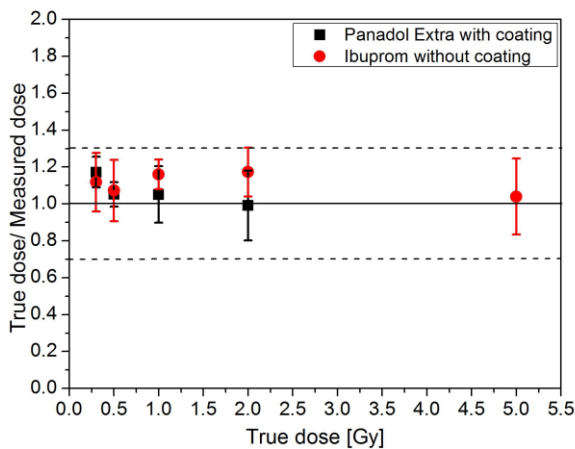


Fig. 9. Recovery tests. The results of dose recovery tests presented as the ratio of the true doses and measured doses. Uncertainty of the final results was assessed by the standard deviation of 5 irradiation trials for different values of dose. In this measurements, differently than in all others, every sample was irradiated firstly and only then powdered. As can be seen, all of the obtained results are below the 20% threshold of the defined ratio.

IV. CONCLUSION

To conclude, the performed investigations confirmed that pharmaceuticals may be excellent candidates to be used as radiation sensors for emergencies. All of the tested medicines exhibited strong luminescence signal following exposure to ionizing radiation. Its intensity was found to enable measurement of doses well below 1 Gy, what is entirely sufficient for application in emergency dosimetry. The highest sensitivity was shown for the medicines based on ibuprofen and paracetamol, which is quite advantageous as they are probably the most commonly used over-the-counter painkillers. Our results suggest also that different types of drugs may require somewhat specific measuring procedures. Because the common medicines are usually produced and marketed in numerous variants (some of them only locally), further broader investigations are certainly necessary and we consider our work to be just a starting point. Such investigations, besides analgesics and antipyretics, should include also other potential candidates, like gastrointestinal medicines or sore throat lozenges. If a sufficiently broad database on dosimetric properties of carry-on medicines is gathered, it will provide a very good tool for emergency dosimetry.

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