

<https://doi.org/10.47183/mes.2026-478>



SELECTION OF OPTIMAL MATERIALS FOR EPR DOSIMETRY IN CASE OF EMERGENCY RADIATION EXPOSURE

Denis V. Ivanov^{1,2,3}, Elena E. Aladova^{1✉}, Damir R. Baytimirov³, Sergey F. Konev³

¹ Southern Urals Federal Research and Clinical Center for Medical Biophysics, Ozersk, Russia

² M.N. Mikheev Institute of Metal Physics of Ural Branch of Russian Academy of Sciences, Yekaterinburg, Russia

³ Ural Federal University, Yekaterinburg, Russia

Introduction. The electron paramagnetic resonance (EPR) method, based on determining the concentration of free radicals generated in materials surrounding a person under the influence of ionizing radiation, allows for reliable estimates of absorbed doses in individuals exposed to uncontrolled irradiation. However, materials intended for use as objects in EPR dosimetry must meet fairly stringent requirements, including availability, detection limit, temporal stability of the EPR signal, and sufficient radiation sensitivity. Therefore, although ionizing radiation induces free radical formation in virtually all materials, not all of them can be used to obtain information about absorbed radiation doses.

Objective. Selection of optimal materials for assessing absorbed doses of ionizing radiation in victims of accidental exposures using the EPR method.

Materials and methods. A standard Bruker Elexsys E580 X-band spectrometer with a SuperHighQ cylindrical resonator was used to record EPR spectra. A UELR-10-10S2 linear electron accelerator was used for sample irradiation. The study examined samples of biological materials (hair, fingernails), as well as materials commonly associated with humans in everyday life, such as plastic, cotton fabrics, fabric from disposable medical masks, and smartphone glass.

Results. The results showed that each of the studied materials has its own advantages and disadvantages for EPR dosimetry. The radiation sensitivity of hair samples was inversely proportional to their melanin content: the higher the pigment concentration (and, consequently, the darker the hair), the lower the radiation sensitivity and the higher the background melanin signal. To account for the intrinsic melanin signal, an additional measurement is recommended — recording the hair color via a photograph taken under a microscope. Fingernail samples, which, like hair, are keratinous formations but lack melanin, represent an alternative. A drawback of biological materials such as hair and fingernails is their relatively low sensitivity, with a lower limit of detection (LOD) exceeding 10 Gy. High radiation sensitivity was demonstrated by uncolored cotton fabric samples (LOD = 1 Gy). The material of protective masks exhibited several times higher radiation sensitivity. Stable EPR signals were obtained from transparent plastic buttons. Smartphone glass proved to be the most radiation-sensitive material, with a LOD not exceeding 0.5 Gy; however, the variation in sensitivity among such glasses was significant, even among smartphones of the same model. The variability in radiation sensitivity of the investigated samples ranged 20–50%.

Conclusions. When selecting materials for estimating absorbed doses using the EPR method, the following witness materials for radiological events are preferred: smartphone screen glass, uncolored cotton clothing fabrics, materials from protective masks, and transparent plastic. When using hair as a biological dosimeter, the measurement procedure is characterized by increased complexity, and the results may contain a systematic overestimation of the estimated doses, particularly in the case of dark-colored hair. Fingernail samples, in turn, exhibit poor stability of the radiation-induced signal, which is difficult to distinguish from background signals of non-radiation origin. The variability in the sensitivity of the studied sample materials indicates the need for individual calibration using additional irradiation.

Keywords: electron paramagnetic resonance; retrospective dosimetry; radiation sensitivity; radiation-induced signal; accidental exposure; absorbed dose

For citation: Ivanov D.V., Aladova E.E., Baytimirov D.R., Konev S.F. Selection of optimal materials for EPR dosimetry in case of emergency radiation exposure. *Extreme Medicine*. 2026. <https://doi.org/10.47183/mes.2026-478>

Funding: this work was carried out with support from the Ministry of Science and Higher Education of the RF (project No. FEUZ-2023-0013), the state assignment “Spin” (reg. No. 122021000036-3), and the state contract No. 11.310.22.2 under the FMBA’s Federal Target Program on Nuclear and Radiation Safety (2016–2020, extended to 2030).

Compliance with the ethical principles: Ethics Committee approval was not required for this study, as it involved materials science research using anonymized biomaterials without personal identifiers.

Acknowledgments: the authors express their gratitude to A.S. Konev (Research Assistant, IMP UB RAS) for collecting the samples.

Potential conflict of interest: the authors declare no conflict of interest.

✉ Elena E. Aladova aladova@subi.su

Received: 13 Mar. 2026 **Revised:** 22 Apr. 2026 **Accepted:** 4 May 2026 **Online first:** 22 July 2026

УДК 614.876:53.06:623.454.862

ВЫБОР ОПТИМАЛЬНЫХ МАТЕРИАЛОВ ДЛЯ ЭПР-ДОЗИМЕТРИИ ПРИ АВАРИЙНОМ ОБЛУЧЕНИИ

Д.В. Иванов^{1,2,3}, Е.Е. Аладова^{1✉}, Д.Р. Байтимиров³, С.Ф. Конев³

¹ Южно-Уральский федеральный научно-клинический центр медицинской биофизики Федерального медико-биологического агентства, Озерск, Россия

² Институт физики металлов им. М.Н. Михеева Уральского отделения РАН, Екатеринбург, Россия

³ Уральский федеральный университет им. первого Президента России Б.Н. Ельцина, Екатеринбург, Россия

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Введение. Метод электронного парамагнитного резонанса (ЭПР), основанный на определении концентрации свободных радикалов, образующихся в окружающих человека предметах под действием ионизирующего излучения, позволяет получить достоверные оценки поглощенных доз у пострадавших в результате неконтролируемого облучения. Однако к материалам, используемым в качестве объектов для ЭПР-дозиметрии, предъявляются достаточно серьезные требования, такие как доступность, предел детектирования, стабильность ЭПР-сигнала во времени, достаточная радиационная чувствительность и т. д. Поэтому, несмотря на то что под действием ионизирующего излучения свободные радикалы образуются практически во всех материалах, далеко не каждый из них может использоваться для получения информации о поглощенных дозах излучения.

Цель. Выбор оптимальных объектов для оценки поглощенных доз ионизирующего излучения пострадавших в случаях аварийного облучения методом ЭПР.

Материалы и методы. Для регистрации ЭПР-спектров применялся стандартный спектрометр X-диапазона Bruker Elexsys E580 с цилиндрическим резонатором SuperHighQ. Для облучения образцов использовался линейный ускоритель электронов модели УЭЛР-10-10С2. В исследованиях рассматривались образцы биологических материалов (волос, ногтей), а также сопутствующие человеку в быту материалы, такие как пластик, хлопчатобумажные ткани, ткани защитных медицинских масок и стекла смартфонов.

Результаты. Результаты показали, что каждый из исследованных объектов имеет свои преимущества и недостатки для ЭПР-дозиметрии. Радиационная чувствительность образцов волос обратно пропорциональна содержанию в них меланина: чем выше концентрация пигмента (и соответственно темнее волос), тем ниже чувствительность к радиации и тем выше фоновый сигнал меланина. Для учета собственного сигнала меланина целесообразно провести дополнительное измерение — зафиксировать цвет волоса посредством фотографии под микроскопом. Альтернативой могут стать образцы ногтей, которые, как и волосы, являются кератиновыми образованиями, но лишены меланина. Недостатком биологических объектов, таких как волосы и ногти, является высокое значение нижнего предела детектирования — более 10 Гр. Высокую радиационную чувствительность показали образцы неокрашенных хлопчатобумажных тканей (нижний предел детектирования — 1 Гр), материал защитных масок имел чувствительность к облучению в несколько раз выше. Стабильные ЭПР-сигналы получены при облучении прозрачных пластиковых пуговиц. Самым чувствительным к облучению оказался материал стекол смартфонов, предел детектирования по которым не превышал 0,5 Гр, однако разброс чувствительности таких стекол оказался значительным даже при исследовании смартфонов одной модели. Вариабельность радиационной чувствительности исследованных материалов составляла 20–50%.

Выводы. При выборе объектов для оценки поглощенной дозы методом ЭПР предпочтительными являются следующие материалы — свидетели радиологических событий: стекла экранов смартфонов, неокрашенные хлопчатобумажные ткани одежды, материалы защитных масок, прозрачный пластик. При использовании волос в качестве биологического дозиметра процедура измерения характеризуется повышенной трудоемкостью, а результаты могут содержать систематическое завышение оцениваемых доз, особенно в случае волос темного цвета. Образцы ногтей, в свою очередь, демонстрируют низкую стабильность радиационно-индуцированного сигнала, который трудно отличить от фоновых сигналов нерадиационной природы. Разброс чувствительности образцов исследованных материалов указывает на необходимость индивидуальной калибровки с применением дополнительного облучения.

Ключевые слова: электронный парамагнитный резонанс; ретроспективная дозиметрия; радиационная чувствительность; радиационно-индуцированный сигнал; аварийное облучение; поглощенная доза

Для цитирования: Иванов Д.В., Аладова Е.Е., Байтимиров Д.Р., Конев С.Ф. Выбор оптимальных материалов для ЭПР-дозиметрии при аварийном облучении. *Экстремальная биомедицина*. 2026. <https://doi.org/10.47183/mes.2026-478>

Финансирование: работа выполнена при поддержке Министерства науки и высшего образования РФ (тема № FEUZ-2023-0013) и государственного задания «Спин» Гр. № 122021000036-3, а также в рамках реализации государственного контракта № 11.310.22.2, финансируемого ФМБА России по ФЦП «Обеспечение ядерной и радиационной безопасности на 2016–2020 гг. и на период до 2030 г.».

Соответствие принципам этики: исследование не требует одобрения этического комитета, поскольку носит характер материаловедческого, а биоматериалы были обезличены и не содержали персональные данные.

Благодарность: авторы выражают благодарность младшему научному сотруднику Института физики металлов УрО РАН Коневу А.С. за сбор образцов.

Потенциальный конфликт интересов: авторы заявляют об отсутствии конфликта интересов.

✉ Аладова Елена Евгеньевна aladova@subi.su

Получено: 13.03.2026 **После доработки:** 22.04.2026 **Принята к публикации:** 04.05.2026 **Online first:** 22.07.2026

INTRODUCTION

When radiation incidents occur, even in the presence of well-organized dosimetric monitoring, there is a risk of uncontrolled exposure. Furthermore, data from external exposure dosimetric monitoring may require verification, necessitating retrospective dosimetric measurements. In turn, the choice of the correct treatment strategy for patients showing signs of radiation injury depends directly on the assessment of the absorbed dose.

One method that provides reliable estimates of absorbed dose is electron paramagnetic resonance (EPR),

which is based on determining the concentration of free radicals generated in objects and materials surrounding a person under the influence of ionizing radiation. The concentration of free radicals is directly proportional to the accumulated dose. Due to the different nature of substances, their free-radical states differ [1], and the selection of an optimal object for dosimetric measurements is a challenging task. The main parameters to be considered in EPR measurements are the number of radicals generated in the material per unit absorbed dose (radiation sensitivity) and the temporal stability of these radicals. Additionally, the presence of intrinsic paramagnetic centers in the substance that are unrelated

to irradiation and give rise to a non-radiation EPR signal must be taken into account.

The most sensitive object for retrospective EPR dosimetry is tooth enamel [2, 3]; however, its use in dosimetry is limited by the need for tooth extraction and the lengthy sample preparation process for measurements. Nevertheless, research continues actively into alternative approaches that allow measurement of radiation-induced radicals in tooth enamel both *in vivo* [4] and in small chips, making this method less invasive and more convenient for widespread practical application [5, 6]. It should be noted that significant contributions to the development and advancement of technologies for non-extractive tooth enamel sampling have been made by specialists of the FMBA of Russia [7, 8].

In addition to tooth enamel, keratinous fibers — hair and fingernail samples — have been used as objects for EPR studies [9], as have materials commonly associated with humans in everyday life, including plastics, cotton clothing fabrics, and the fabric of disposable medical masks, which are widely used as sanitary-hygiene products [10]. Alongside these, it has been shown that glass from modern smartphones also exhibits high radiation sensitivity [11].

The study purpose was to select optimal materials for assessing absorbed doses of ionizing radiation in victims of accidental exposures using the EPR method. The objective was to perform a comparative analysis of the dosimetric characteristics of various materials (both of biological origin and commonly found in everyday life) when used as objects for retrospective EPR dosimetry and to identify the best candidates.

MATERIALS AND METHODS

EPR was used to study samples of human hair of different colors (dark, brown, light — blond, and gray), fingernail samples, plastic (buttons, pen caps), clothing fabrics (a white cotton laboratory coat), protective medical masks, and samples of smartphone touchscreen glass.

The sample size was determined by the ability to place it in a 5 mm diameter test tube for measurements, with the sample height in the tube not exceeding 10 mm. Due to differences in the density of the materials studied, sample masses ranged 15–200 mg.

A Bruker Elexsys E580 X-band spectrometer with a SuperHighQ cylindrical resonator (Bruker, Germany) was used to record EPR spectra. The spectral recording parameters were optimized to achieve maximum sensitivity without distorting the spectral pattern. The following settings were used: magnetic field modulation — 0.1 G, magnetic field sweep width — 100 G, sweep time — 30 s, microwave power — 2 mW, number of scans — 1–15. The EPR signal amplitude was determined using the peak-to-peak method.

Hair and fingernail samples were obtained from donors aged 20–25 years; gray hair donors were 60 years old. Overall, the study used data obtained from 16 hair

samples, 3 fingernail samples, 10 plastic samples, 20 fabric samples (of which 5 were medical mask fabrics), and 14 smartphone glass samples (of which 11 were Gorilla® Glass and 3 were Tempered Glass).

The pigmentation intensity of the hair samples was determined using an optical microscope with a digital camera, a Nikon Eclipse E200 (Nikon, Japan). The hair was photographed at 400× magnification, and the resulting image was processed in the GIMP graphics editor.

A UELR-10-10S2 linear electron accelerator (NPP KORAD, Russia) located at the Innovation and Implementation Center for Radiation Sterilization of the Physical Technology Institute, Ural Federal University, was used to irradiate the samples. The average electron beam energy was 10 MeV. To rapidly and reliably capture the radiation effect, the samples were exposed to high doses of radiation ranging 10–30 kGy.

The standard deviation of the EPR measurements for the established lower limits of detection of absorbed doses was $\pm 20\%$.

RESULTS AND DISCUSSION

In the case of accidental exposure, hair samples are an accessible and easy-to-collect material; however, their use in EPR dosimetry is complicated by the presence of melanin. This pigment generates its own EPR signal, which is similar in shape to the signal observed after radiation exposure (Fig. 1). The intensity of this signal depends on the melanin concentration, i.e., on the hair color: the darker the hair, the higher the signal (Fig. 2). In this study, the lightness (pigmentation intensity) of the hair was determined by analyzing microphotographs: the brightness of the hair image, expressed as a percentage relative to the brightness of a light background, was measured. Thus, for EPR dosimetry of hair samples, there will always be a systematic error associated with hair color (melanin content). Furthermore, melanin can bind free radicals [12]; therefore, the radiation sensitivity of hair depends strongly on its color (Fig. 3): the lighter the hair, the higher the sensitivity [9, 13].

Additional irradiation of the sample with a known dose (as an alternative to using a calibration curve) reduces the influence of differences in sensitivity but increases the workload. To account for the melanin signal, the hair color (lightness) must be additionally determined (e.g., by microscopic photography), which lengthens the analysis process; even when studying light-colored hair, this procedure is unavoidable. Moreover, chemical treatment of the hair (dyeing, bleaching, etc.) can unpredictably alter the sample's properties, rendering the analysis results unusable.

Fingernails, like hair, are keratinous formations; however, they lack melanin and therefore may serve as a reasonable alternative in dosimetry. Nail material has no background EPR signal, but its radiation sensitivity is

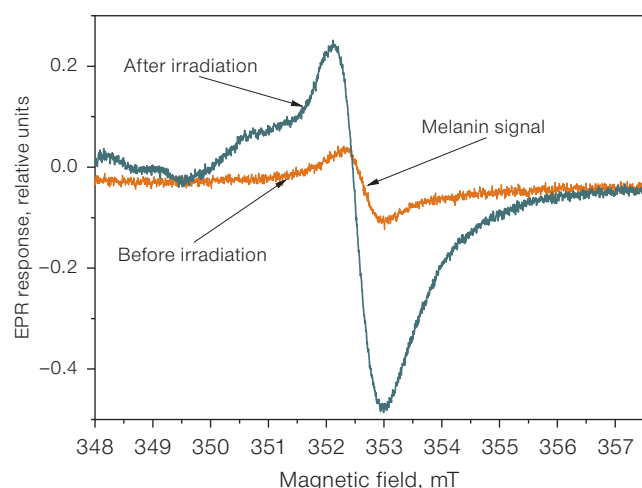


Figure prepared by the authors based on their own data

Fig. 1. Example of an EPR spectrum of black hair before and after irradiation

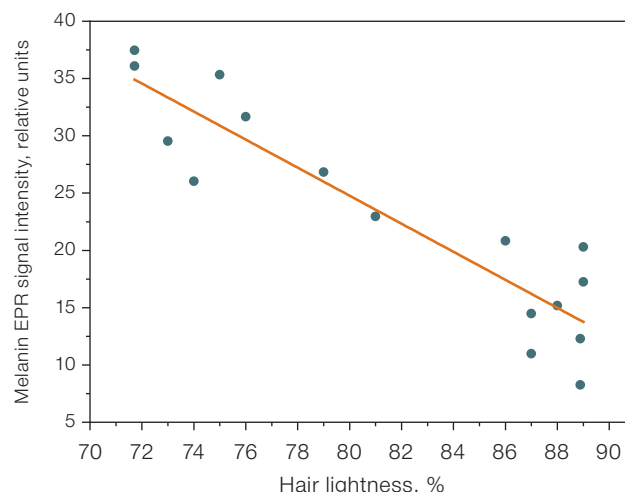


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Fig. 2. Dependence of the melanin EPR signal intensity on hair color

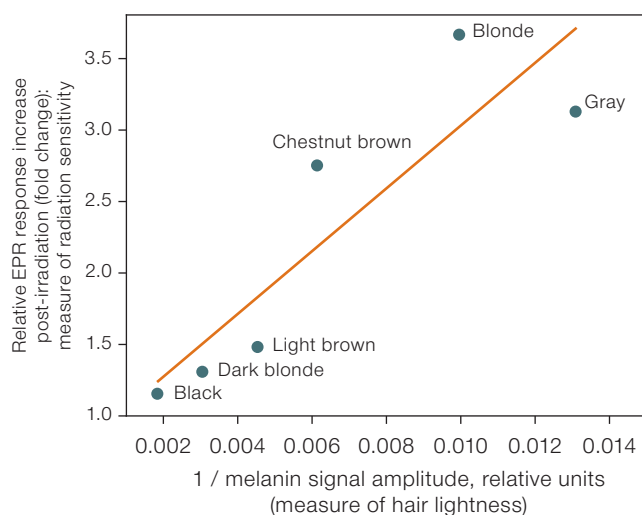


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Fig. 3. Dependence of the relative EPR response to irradiation on hair color

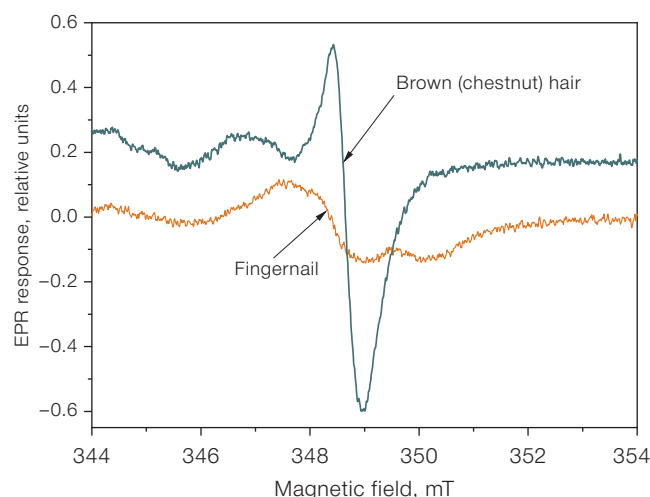


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Fig. 4. Example of EPR spectra of biological materials after exposure to the same radiation dose

significantly lower than that of hair (Fig. 4). Furthermore, the signal intensity decreases rapidly over time (the signal halves within 24 hours), depends on sample humidity [14], and can be overwhelmed by signals from various impurities often present on the nail surface, leading to overestimation of the absorbed dose.

The temporal stability of the EPR signal of the studied materials was assessed by repeated EPR response measurements over a period of 2 months. For all materials except fingernails, the half-life of the EPR signal is 3–4 days or more, which is sufficient for emergency dosimetry purposes [1].

Plastic is a widely used material in the manufacture of many everyday items (mobile phones, buttons, eyeglass frames, etc.). Due to the widespread use of radiation sterilization in the manufacture of medical instruments, detailed studies have previously been

conducted on the effect of irradiation on plastic samples of various compositions [15–17]. It was found that the parameters of the EPR signals and their stability vary significantly depending on the type of plastic [18]. The most stable EPR signals were recorded from irradiated, transparent, uncolored plastic buttons [19]. It should be noted that plastic may have a background non-radiation signal, which is often associated with its coloring (Fig. 5).

Cotton fabrics have fairly high radiation sensitivity and EPR signal stability over several days [20, 21], making them useful as a source of information about levels of radiation exposure. Moreover, in cases of non-uniform exposure, clothing fabric samples from a victim can be used to determine the distribution of absorbed radiation dose over the surface of the body. However, fabric dyes can produce a background EPR

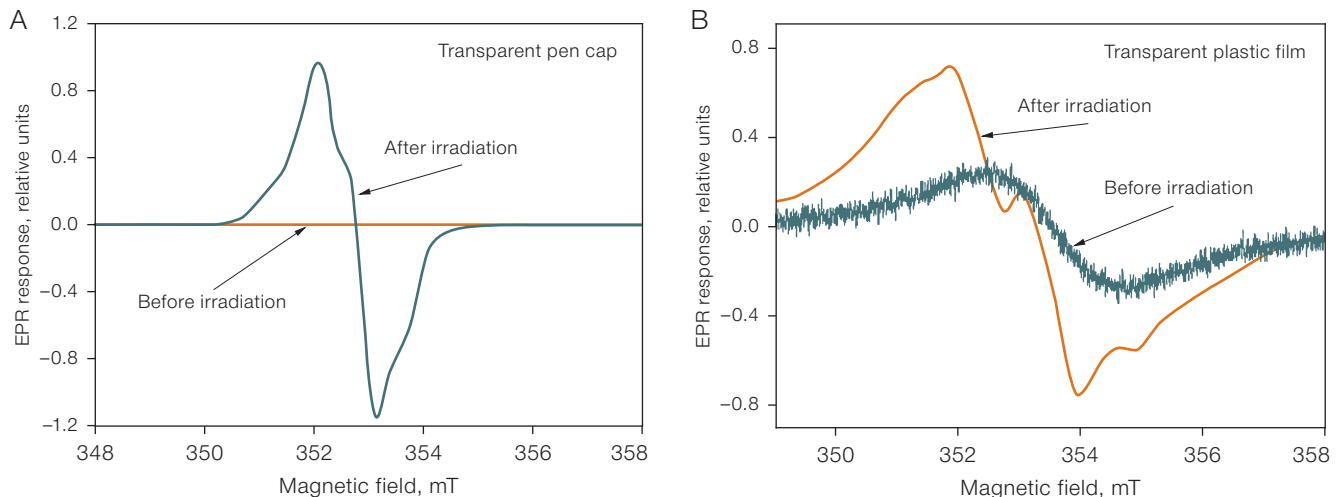


Figure prepared by the authors based on their own data

Fig. 5. Example of EPR spectra of plastic. A — intrinsic background signal absent; B — intrinsic background signal present

signal with high probability. In addition, various factors can complicate the analysis of irradiated cotton: exposure to sunlight, residual content of detergent molecules from laundering, water content, the presence of dust, grease, etc. It should be noted that the fabric of medical masks, which is of a different nature (typically synthetic materials), showed a radiation sensitivity 3.5 times higher than cotton fabrics [10].

The glass of modern smartphones is typically made from a specially chemically strengthened material processed through an ion-exchange process, for example, Gorilla® Glass, developed by Corning Inc¹. Among the candidate materials, smartphone glass is the most sensitive to radiation. However, variations in the chemical processing of such glasses cause a spread in their sensitivity [22], even for smartphones of the same model, which can introduce significant measurement errors [23, 24]. The structure of the EPR spectra can be divided into three types [11]; the two most commonly encountered spectrum types are shown in Fig. 6.

The dosimetric characteristics of the studied biological materials and everyday materials are presented in the table.

The radiation sensitivity of transparent plastic samples varied over a wide dose range: the lower limit of detection (LOD) was 5 Gy for button plastic and reached 15–20 Gy for pen casing plastic. Dyed (colored) plastic may have a background non-radiation signal that leads to an overestimation of the determined absorbed radiation dose.

For hair samples, a pronounced dependence of radiation sensitivity on melanin content was observed: an increase in melanin concentration reduces sensitivity and increases the background signal. Even when studying light-colored hair, color correction is required, performed via optical measurements.

Fingernail material has no background signal but is characterized by poor EPR signal stability and insufficient radiation sensitivity (LOD exceeds 10 Gy).

Cotton fabrics have sensitivity providing a detection limit of 1 Gy and signal stability of several days; however, dyes may produce a background EPR signal. Synthetic fabrics from protective masks showed better radiation sensitivity.

The most radiation-sensitive materials are smartphone glasses: the lower detection limit is 0.5 Gy. However, the variability in sensitivity of the glasses studied is large, even for smartphones of the same model, which can lead to significant measurement error and makes individual calibration of samples using additional irradiation preferable.

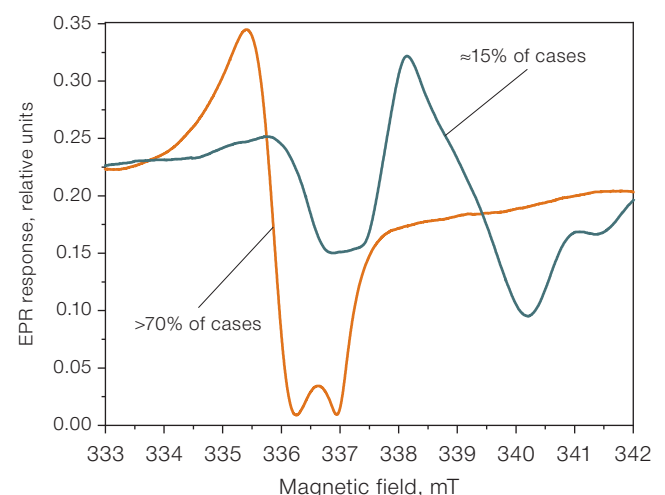


Figure prepared by the authors based on their own data

Fig. 6. Examples of EPR spectra of the two most common types of smartphone glass after irradiation

¹ Corning, Inc. <https://www.corning.com/gorillaglass/worldwide/en.html>

Table. Dosimetric characteristics of the studied materials

Material	Lower limit of detection, Gy	Sensitivity variability, %
Dark hair	20	30
Brown hair	15	30
Blond hair	10	25
Fingernail	10	20
Colorless plastic	5–20	50
Dyed (colored) plastic	25	50
Cotton fabric	1	30
Medical mask fabric	0.7	30
Smartphone glass	0.5	20

The table was compiled by the authors based on their own data

CONCLUSION

- 1. The optimal candidates for EPR dosimetric measurements among the materials considered are smartphone screen glass, as well as uncolored cotton clothing fabrics and medical mask fabrics.
- 2. Transparent plastic items (buttons, ballpoint pen caps) are applicable for dosimetry purposes, but their sensitivity variability is the highest among the materials studied. Samples of dyed (colored) plastic may have a background EPR signal, which may lead to an overestimated dose assessment; therefore, the use of colored plastic is recommended only when no other samples are available.

- 3. The absorbed radiation dose can be determined by analyzing the EPR spectra of hair samples. However, hair analysis is more labor-intensive and may lead to an overestimated result, particularly in the case of dark-colored hair.
- 4. Radiation-induced EPR signals in fingernail samples are difficult to identify and separate from signals not associated with radiation exposure; moreover, they exhibit poor stability. Nevertheless, EPR dosimetry using fingernails is a promising method from the standpoint of biological material availability.
- 5. The high variability in radiation sensitivity that is characteristic of the types of samples considered indicates the need for individual calibration using additional irradiation for each type of sample.

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Authors' contribution. All authors confirm that their authorship meets the ICMJE criteria. Denis V. Ivanov — conduct of the study, data analysis, manuscript drafting; Elena E. Aladova — study planning, manuscript editing; Damir R. Baytimirov — samples irradiation, EPR spectrometry, data analysis; Sergey F. Konev — EPR spectrometry, data analysis.

Authors:

Denis V. Ivanov, Cand. Sci. (Phys.-Math.), ORCID: <https://orcid.org/0000-0001-6757-5587>

Elena E. Aladova, Cand. Sci. (Biol.), ORCID: <https://orcid.org/0000-0003-3277-0512>

Damir R. Baytimirov, Cand. Sci. (Phys.-Math.), ORCID: <https://orcid.org/0000-0003-2004-1988>

Sergey F. Konev, Cand. Sci. (Phys.-Math.), ORCID: <https://orcid.org/0000-0001-9830-4309>