

EPR study of gamma - irradiated homeopathic medicines

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The EPR studies of homeopathic medicines from plant (aconite, arnica, belladonna, bryony, dulcamara, symphytum and eupatorium) and animal (cuttle) origin before and after gamma-irradiation are reported. Before irradiation all samples are EPR silent. After gamma-sterilization however they all exhibit a specific “sugar-like” EPR spectrum due most probably to sucrose used as an excipient. These stable free radicals can be used to identify radiation processing a long time after irradiation.

Keywords: homeopathic pills; gamma – irradiation; EPR.

INTRODUCTION

Gamma rays have been intensively used to sterilize drugs, medical devices and foodstuffs [1]. One of the main advantages of the process is the opportunity for sterilization of the products even in their final packages [2]. This process however must be under control. Electron Paramagnetic Resonance (EPR) spectroscopy is an unambiguous technique for determining the origin and stability of the radiation induced free radicals in the solid state. Thus, it has been accepted by the European Union as a standard procedure for identification of irradiated foods [3; 4; 5]. As for foodstuffs, it could be interesting to be able to prove whether or not a drug has been irradiated [6]. In view of this a wide variety of gamma-irradiated antibiotics were studied by EPR [7]. The character and stability of radicals in drugs following ionizing radiation varies depending on their structure and the propensity of the solid matrix to free radical trapping. The decay rates of gamma-induced free radicals in drugs that have a wide application in anticancer [8] and anti-asthmatic therapy [9] have been investigated. The EPR spectroscopic properties were determined and suggestions concerning the possible structure of gamma induced radicals for anti-emetic [10] and antipyretic drugs [11] have been made. In these samples however there is only one substance – an active pharmaceutical ingredient (API). Most of the other pills are a mixture of an API which varies from 2 -10% of the weight of the pill and the remaining 98 – 90% are excipients [12]. This case is similar to the homeopathic pill case in which the

API is extremely low and thus the identification of the radiation sterilization is possible only from the EPR spectra of the excipients. Therefore, previous research [12, 13] on the stability of the induced radicals by irradiation and mechanical treatment in some excipients will be very useful.

In the present communication we report the EPR spectra obtained before and after gamma irradiation of some homeopathic medicines in order to prove the samples have been irradiated or not. Also, the stability of the radiation induced free radicals is determined.

EXPERIMENTAL

Materials

Homeopathic pills from different plant (aconite, arnica, belladonna, bryony, dulcamara, symphytum and eupatorium) and animal origin (cuttle) were purchased from a local pharmacy and were divided in two portions. The first batch was passed for irradiation, the second was separated as control samples. After gamma-irradiation the samples were kept under normal environmental conditions (temperature 19–22 °C, humidity 70–80%), available in the laboratory.

Irradiation

Homeopathic pills were simultaneously gamma-irradiated by the “Gamma 1300” irradiator with a single dose of 10 kGy. The irradiation was performed at room temperature and in the air. All further manipulations of the irradiated samples were performed at least 72 h after irradiation in order to avoid any interference by the radiation induced short living paramagnetic species.

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Instrumentation

EPR measurements were performed at room temperature on a JEOL JES-FA 100 spectrometer operated in the X-band. Standard cylindrical cavity operating in the TE₀₁₁ mode was used. All samples were accommodated in quartz EPR sample tubes (i.d./o.d. 4/5 mm). The parameters used to record a single scan EPR spectrum of each sample were: microwave power 1 mW; modulation frequency 100 kHz and amplitude 0,25 mT; sweep width 10 mT, sweep time 2 min. and time constant 1 sec.

RESULTS AND DISCUSSION

EPR spectra

Before irradiation all studied pills do not show EPR spectra.

After irradiation all homeopathic medicines exhibit a typical EPR spectrum (Figure 1). It is worth to note that the same EPR spectrum was recorded from sucrose, white and brown sugar [14]. This feature, as well as its independence of the origin of the sugar [15] is very important from a dosimetric point of view because EPR can be used also for emergency radiation monitoring. The previous ENDOR [16] and high-frequency EPR studies [17] of irradiated sucrose show that the spectrum shown in Figure 1 is complex. At least three separate EPR spectra due to different paramagnetic species are superimposed on it [17]. In homeopathic pills sucrose is most probably used as an excipient. This is in accordance with the well-known fact that to reach the homeopathic concentrations, a preparation selected starting substance or compound is serially diluted in water or ethyl alcohol. Thus we cannot record any EPR spectrum from an active ingredient.



Fig. 1. EPR spectrum of homeopathic medicines after gamma-irradiation.

Study of the stability of radiation induced free radicals

The fading kinetics of the radiation induced EPR signal is an important characteristic of the materials because after irradiation it limits the time interval in which the identification of radiation processing is unambiguous. Since typically, for the given excipient this period strongly depends on the structure of the material under study, its storage conditions before and after irradiation, the humidity, exposure to air, light and temperature. In order to find the stability of radiation-induced EPR signals in homeopathic pills their kinetics were studied for a period of 200 days after irradiation. The studies show that radiation induced radicals in sucrose remain stable (Figure 2). Each point of Figure 2 is an average of at least three measurements. This result is imported in direction of emergency purpose because it is known that sucrose is found to be a very promising radiation sensitive material for gamma-irradiation with respect to both retrospective and reference solid state EPR dosimetry [18].

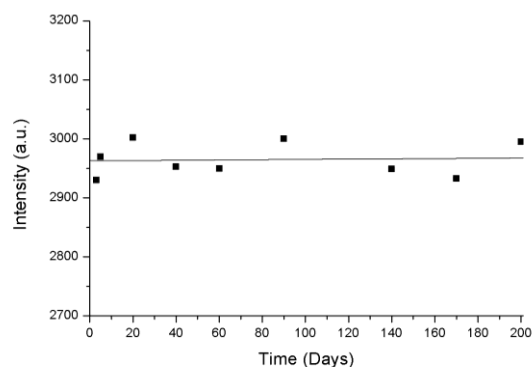


Fig. 2. Kinetics of the stability of a radiation induced signal in homeopathic medicines.

CONCLUSION

The results of EPR studies on homeopathic pills from: aconite, arnica, belladonna, bryony, dulcamara, symphytum, eupatorium and cuttle show that gamma-irradiation induce free radicals of sucrose, which is used as an excipient. No EPR spectra from active substances were record most probably due to their low concentration. Thus, the gamma-induced free radicals in sucrose present as excipient can be used for identification of radiation processing a long time after irradiation.

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ЕПР ИЗСЛЕДВАНЕ НА ГАМА-ОБЛЪЧЕНИ ХОМЕОПАТИЧНИ ЛЕКАРСТВА

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(Резюме)

Представени са резултати от ЕПР изследване на хомеопатични лекарства от растителен (самакитка, арника, беладона, дива тиква, червено кучешко грозде, черен оман и еупаториум) и животински произход (сепия) преди и след гама-облъчване. Преди облъчване не се регистрира ЕПР спектър. След гама-стерилизация при всички проби се детектира специфичният т. нар. „захароподобен“ ЕПР спектър, който най-вероятно се дължи на захароза, използвана като пълнител. Тези стабилни радикали може да се използват за идентифициране на радиационната обработка дълго време след облъчването.