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ANOMALIES IN ELECTRIC CONDUCTIVITY IN γ-IRRADIATED BONE?

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The effect of γ irradiation with doses of 10 to 1000 kGy on the electric conductivity (σ) of dry state bone was studied. Measurements of temperature dependence of electrical conductivity were made for the temperature range of 390–530 K. The obtained T-σ relationship indicated an increase in σ with temperature. An increase in irradiation dose resulted in a decreased σ, excluding the dose of 1000 kGy. For the dose of 1000 kGy the increase in σ was observed. Above the temperature of 510K, σ was dose independent. Activation energy for charge conduction process was calculated. The obtained values for electrical conductivity and activation energy were typical for dielectrics and indicated degradation of the organic component of the bone.

INTRODUCTION

Bone may be considered as a composite material containing a mineral component, hydroxyapatite crystal matter (HAP) and an organic component-collagen fibrils. Interaction between the mineral and organic components influences physical processes in bone such as electric charge transport. The electric properties of bone have been experimentally studied for different physical factors (Behari, 1991; Chakkalakal & Johnson; 1981; Reddy & Saha, 1984; Saha & Wiliams, 1988). Temperature or γ-radiation may modify the spatial structure of the studied material. Irradiation generates free radicals, which can be trapped in the irradiated material for a long period (Alexander & Lett, (1967). Heating of the material liberates the trapped free radicals. In the case of irradiated collagen, the liberated free radicals can take part in cross-linking and degradation of biopolymer (Kubisz & Jaroszyk, 1993; Sintzel, Merkli, Tabatabay & Gurny, 1997). In bone, γ-irradiation influences electrical conductivity (Kubisz, 1999).

In this investigation the effect of the absorbed dose of γ-radiation on the temperature dependence of dc electrical conductivity of bone was studied. The reduction in electrical conductivity with increasing dose of radiation was expected.

MATERIALS AND METHODS

Measurements were performed on irradiated and non-irradiated bones. Samples were obtained from the central part of the diaphyses of adult bovine femurs. The bones were mechanically cleaned and washed in a 0.1 M solution of NaCl soon after sacrificing the 2 years old animals. Samples were withdrawed using a diamond saw with continuous water irrigation to minimise thermal damage during the machining process. Samples, 1 mm thick, and 5 mm in diameter, were cut at 90° to the longitudinal axis of the bone.

Samples were γ-irradiated with the following doses: 1000 kGy, 500 kGy, 100 kGy, 50 kGy and 10 kGy. Irradiation was performed in dry air, at room temperature using a ^{60}Co source, with average energy of γ-quant equal to 1.25 MeV. The dose-rate of irradiation was 2 kGy/hour. After irradiation the samples were covered with silver paste electrodes. Next, they were placed in a measuring device, at room temperature, 24 hours before the experiment.

Temperature dependence of electrical conductivity was measured by the direct current method, within a temperature range of 390–530 K. The voltage was applied to the sample in the direction of the longitudinal axis of the bone. The intensity of the current flowing through the sample was

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measured by a W7-30 electrometer as a function of temperature. All measurements were performed at the electric field strength of $E = 1.00 \pm 0.05$ kV/m. The field strength was in the range of voltage-current linearity where Ohm's law is obeyed (Behari, Guha & Agarwal, 1974).

During the experiment, each sample was continuously heated at a rate of 1 K/min, in the temperature range of 290–383 K under atmospheric pressure. The sample was maintained at the temperature of 383 K until the current intensity reached a constant value. Next the sample was heated in the temperature range of 383–530 K. The temperature of the sample was measured by a constantan-copper thermocouple placed close to the sample. The EMF of the thermocouple was indicated by a digital voltmeter. Current intensity, voltage and temperature were recorded by a computer. Finally, the temperature dependence of dc electrical conductivity was calculated.

RESULTS AND DISCUSSION

On the basis of the recorded current intensity, applied voltage and geometrical dimensions of sample, electrical conductivity σ was calculated. The values of electrical conductivity σ were calculated by using the following equation:

$$\sigma = A^{-1} \times d \times U^{-1} \times I \qquad (1)$$

where A is the cross-sectional area of the measured surface, d is the thickness of the specimen in the direction of measurement, U is the voltage applied to the specimen and I is the measured current intensity (Hill, 1968).

The typical temperature dependency of electrical conductivity obtained in this experiment, for irradiated and non-irradiated samples of bone is shown in Fig. 1. The relative error of the electrical conductivity was less than ± 5%.

The obtained relationships differ in their magnitudes and slopes. In the temperature range of 390–540 K, the increasing temperature led to increasing electrical conductivity. The σ-T relationships for non-irradiated samples showned a local maximum at the temperature of 463 K. This is a typical behaviour of heated bone and dry protein (Kubisz, 1999). Irradiation led to the "smoothing" of the curve with increasing doses. The electrical conductivity of the irradiated bone was significantly lower than that of the controls. In the temperature range of 390–508 K, the increasing doses up to 500 kGy, diminished the electrical conductivity. The dose of 1000 kGy reversed the tendency and an increase in the electrical conductivity was observed. From the temperature of 510 K, for the irradiated bone samples the electrical conductivity was dose-independent, and still lower than for non-irradiated samples.

The range of electrical conductivity in the present study is in a good agreement with other data (Behari *et al.*, 1974; Liboff & Shamos, 1973). The electrode effect could be neglected due to the

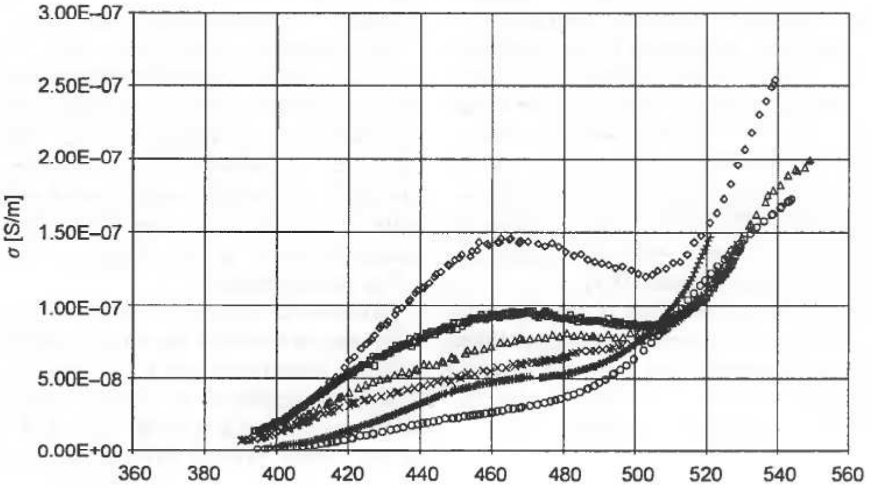

Fig. 1. Temperature dependency of electrical conductivity for non-irradiated bone (◇) and irradiated bone 10 kGy (□), 50 kGy (Δ), 100 kGy (×), 500 kGy (O), 1000 kGy (+)

calculated values of conductivity, less than 10^{-6} $\Omega^{-1}m^{-1}$, the solid state of samples and low level of hydration during measurements (annealing at the temperature of 383 K) (Davies, 1969). The contribution of the electrode effect will only be significant for materials having a low sample dielectric constant (Reddy & Saha, 1984). As bone has a very high dielectric constant (Reddy & Saha, 1984; Kosterich, Foster & Pollock, 1983) the polarisation problem should not be significant.

On the basis of these studies the activation energy E_A of the charge conduction process for irradiated and non irradiated bone could be determined according to the following formula (Liboff & Shamos, 1973):

$$\sigma(T) = \sigma_0 \exp\left(\frac{E_A}{2kT}\right) \quad (2)$$

where σ stands for electrical conductivity, T for temperature, and k for the Boltzmann's constant.

Fig. 2 is an Arrhenius plot for the electrical conductivity for non-irradiated and irradiated samples. Calculations were performed in the intervals of linearity of the following relationship (Liboff & Shamos, 1973):

$$\ln\sigma = f\left(\frac{1}{T}\right) \quad (3)$$

The activation energies of the charge conducting process was calculated for two temperature ranges: 400–420 K and 510–530 K. The averaged activation energies of the charge conduction process, embarked with the standard deviation less than 3%, are summarised in Table 1. At the lower temperature range, up to dose 100 kGy, activation energy remained constant and the increase in the activation energy was observed for the dose 500 kGy and 1000 kGy. The higher activation energy, could indicate generation of the dose of 500 kGy traps hosting free charge carriers deeper than for other doses.

Table 1. Activation energy (kJ/mole)

Dose kGy	Temperature range 400–420 K	Temperature range 510–530 K
0	175	142
10	170	127
50	162	133
100	162	128
500	192	135
1000	230	140

In the temperature range of 510–530 K, the activation energy was dose independent. Most probably, the thermal decomposition of the organic component of the material was responsible for this phenomenon because this temperature range was above the melting point of collagen in dry bone (Marzec, Kubisz & Jaroszyk, 1996; Nguyen, Vu & Wilkes, 1974).

All the activation energies, being less than 260 kJ/mole indicates that dc electrical conductance is

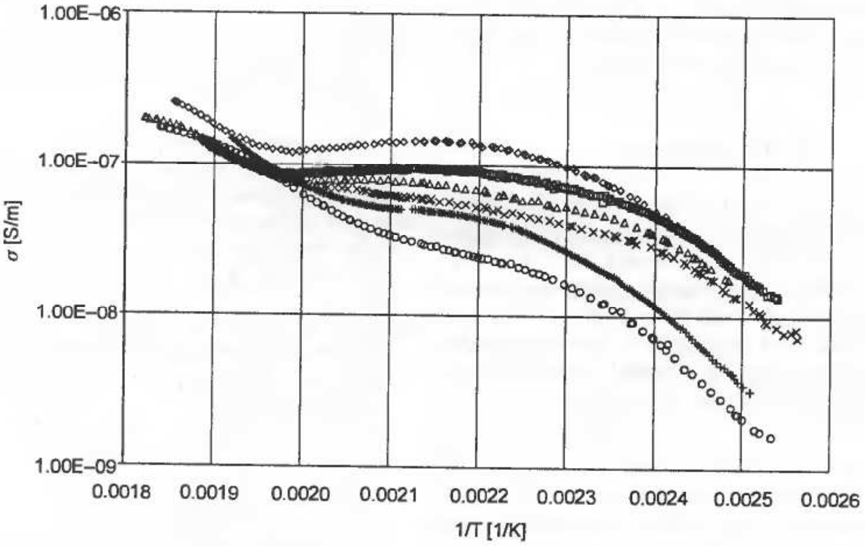

Fig. 2. Arrhenius plot for electrical conductivity, 0 kGy(◇), 10 kGy (□), 50 kGy (Δ), 100 kGy (×), 500 kGy (○), 1000 kGy (+)

not connected with charge carriers injected from the electrodes used but instead and belong to excited electrons from the valence band (Eley, 1968).

In this study the samples were heated to 530 K, the temperature of the beginning of the amino acids pyrolysis (Jakubke & Jeschkeit, 1989). Moreover, up to the temperature of 673 K HAP does not change its physical form (Holden, Clement & Phakey, 1995). Most probably the organic component of the material is responsible for the observed behaviour of the collagen-HAP system.

During the exposition to γ-rays of ^{60}Co, Compton electrons irradiate a sample. The electron beam interacts with bone, leading to electron excitations and structural defects trapping free radicals generated during irradiation. The increase in the electrical conductivity with temperature could be caused by an increase in the number of charge carriers liberated from the traps or/and by the increase in the mobility of charge carriers caused by the thermal energy supplied to the samples. Simultaneously, the charge transport phenomenon is disturbed by the collagen chain scission generated during irradiation, decreasing the free path of charge carriers and decreasing the electrical conductivity of bone, up to the dose of 500 kGy. The increase in the electrical conductivity for the dose of 1000 kGy could be caused by the increase in the number of free radicals generated, even if the deeper traps were generated (higher activation energy).

Considering the results obtained, we could not neglect the action of γ-radiation on HAP but it was not discussed in this paper. The observed increase in the electrical conductivity for the does of 1000 kGy needs further studies and especially the dose reversing the tendency must be found.

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