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Radiation-induced oxidation in gamma-irradiated UHMWPE modified with hydroxyapatite

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INTRODUCTION

The medical use of ultra-high molecular weight polyethylene (UHMWPE) as the bone replacement as well as the manufacture of sterile sealed cases and envelopes require detailed investigations on the chemical stability in different conditions, including radiation sterilization. To impart higher similarity to substituted hard parts of osseous system, the addition of hydroxyapatite (HAP) to the polymeric material is an attractive option. The present study investigates the radiation behavior of three polymer composites consisting of UHMWPE and LDPE to which HAP was added. Some of samples were stabilized with rosemary extract powder (RM). The irradiation doses are in the range of radiation-sterilisation ones.

EXPERIMENTAL

Materials

The UHMWPE/LDPE/HAP composites were obtained by melting in a BRABENDER Plastograph, under a mixing temperature of 180°C for 10 min and screws rotation rate of 40 rpm followed by the thermal processing in a laboratory press type POLYSTAT 200 at the following conditions: temperature: 165°C, pressing time 3-6 minutes, pressure of 125-150 atm and cooling time of 30 minutes.

Table 1 - Composites based on UHMWPE/LDPE

Sample code	UHMWPE, wt%	LDPE, wt%	nHAP, wt%	RM, wt%
S1	20	80	-	-
S2	18	72	10	-
S3	17.91	71.64	9.95	0.5

Instruments and methods

Gamma exposure (Ob-Servo Sanguis, Hungary) was carried-out in air at room temperatures, with a ^{60}Co sources at a dose rate of 1.1 kGy h^{-1}. Irradiation doses: 0, 25 and 50 kGy;

Chemiluminescence spectroscopy (LUMIPOL 3, Slovakia): both isothermal and nonisothermal procedures (25-250 ^{o}C) were applied;

Differential Scanning Calorimetry (Setaram DSC 131 EVO, France): Non-isothermal mode (β = 10 °C min^{-1}), inert atmosphere (N_2, 50 ml min^{-1}). The crystallinity degree was obtained using the ratio of the measured melting enthalpy of LDPE and the enthalpy of 100% crystalline PE (293.6 $j \cdot g^{-1}$);

FTIR spectroscopy (Jasco IR-4200, Japan): spectral range:4000-400 cm^{-1}, 50 scans/spectrum. The carbonyl (CI) index values were obtained as the ratio of the absorbance at 1720 cm^{-1} and the absorbance of 1461 cm^{-1} (used as internal standard).

RESULTS

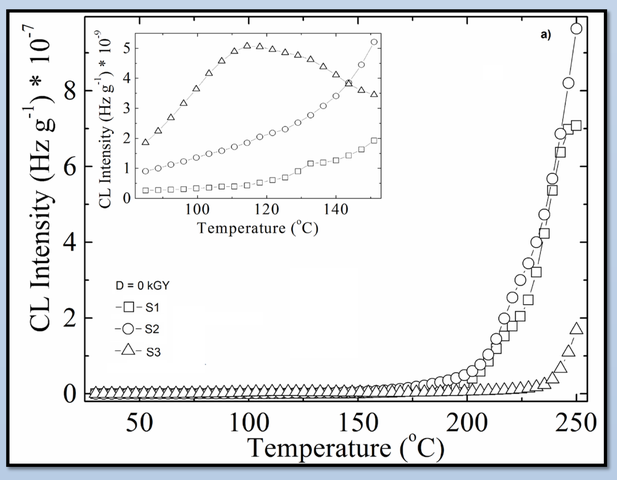

Fig. 1 - Nonisothermal chemiluminescence spectra recorded on S1, S2 and S3 samples: a) 0 kGy; b) 50 kGy

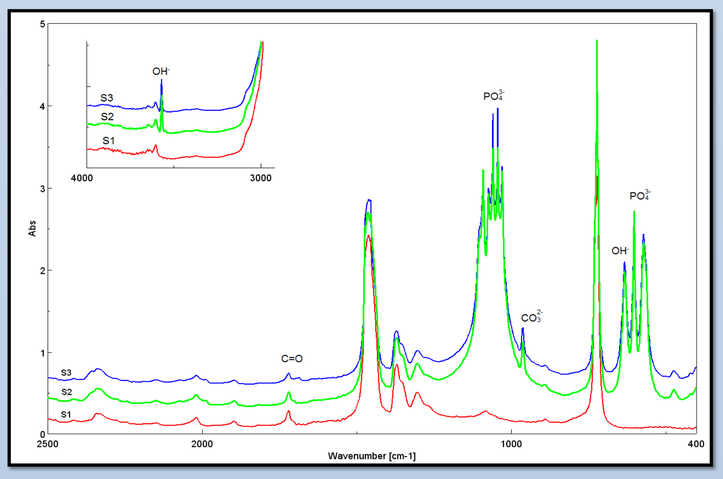

Fig. 2 – FTIR spectra recorded on unirradiated samples

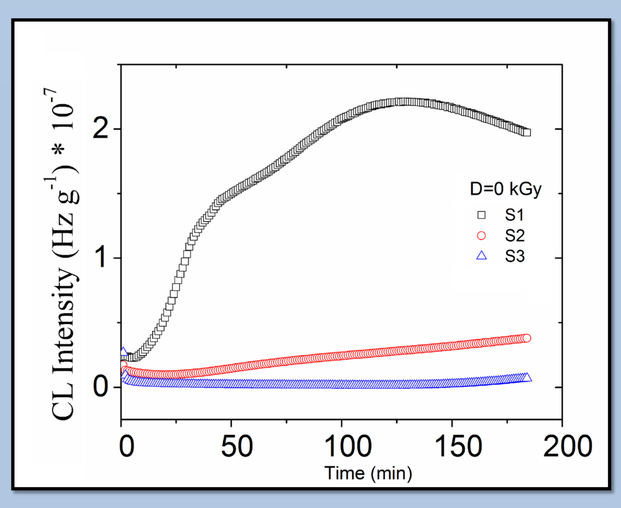

Fig. 3 - Isothermal chemiluminescence spectra (180°C) recorded on S1, S2 and S3 samples.

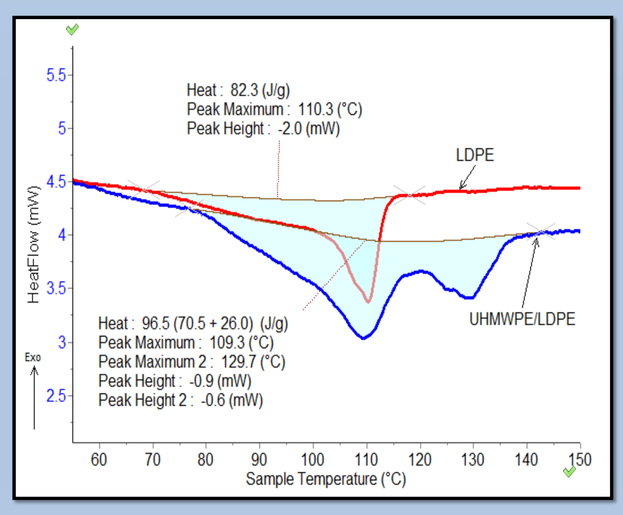

Fig. 4 - DSC curve recorded on S1. Comparison with LDPE.

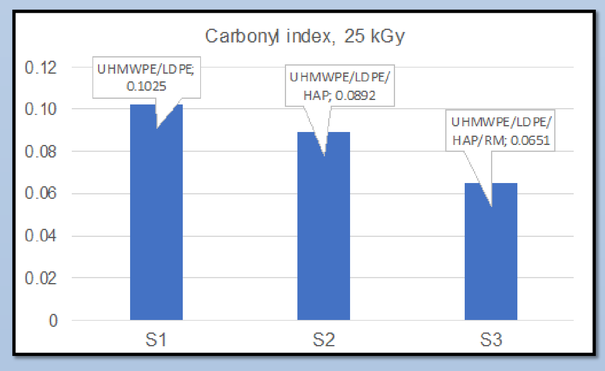

Fig.5 - Carbonyl index variation. Sample composition influence.

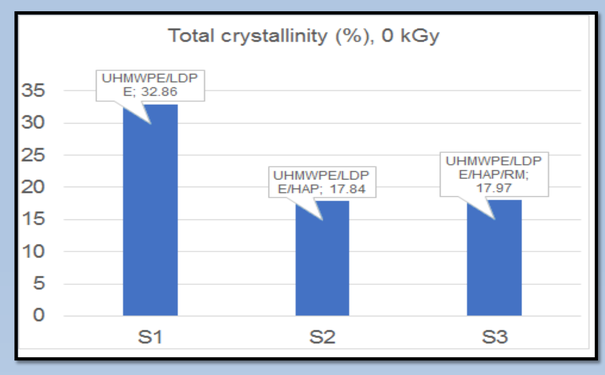

Fig. 6 – Total crystallinity degree variation. The γ-irradiation (in 0-50 kGy range) did not modify significantly the total crystallinity.

CONCLUSIONS

The influence of hydroxyapatite (HAP) and rosemary (RM) powder is noticed by the improvement of the kinetic parameters: the decrease of oxidation rates and the extension of oxidation periods.

The carbonyl index variation can be explained in terms of inhibition promoted by the large surface HAP particles (playing the role of free radicals adsorbent) and by the free radicals scavenging activity of rosemary components, where the predominant active principle is carnosic acid. Both HAP and RM were effective in materials processing: thermo-formation and radiation stabilization.

The use of HAP and RM with UHMWPE/LDPE blends makes possible the manufacture of products for long term applications, requiring high stability against various stress factors.