Contribution ID: **908** Type: **Poster**

Poly(hydroxyethyl methacrylate) as a host matrix for the radiolytic synthesis of hydrogel –Au(0) nanocomposites

Thursday, 19 May 2022 17:15 (8 minutes)

As a part of a project aiming at the post-fabrication modification of 3D-printed objects by radiation processing, we are currently investigating the formation of gold nanoparticles by irradiating poly(hydroxyethyl methacrylate) (PHEMA)-based hydrogels, swollen with aqueous Au(III) solutions.

The radiolytic behavior of PHEMA has apparently not been deeply investigated nor reported in the literature, in spite of its quite common uses in many biomedical applications. We have therefore examined in some detail the radiolytic behavior of commercial linear PHEMA samples or of covalent networks (PHEMA-networks) prepared by photopolymerization of HEMA in presence of (ethyleneglycol)dimethacrylate (EGDMA).

Various spectroscopic and analytic methods were used to monitor the effects of electron beam radiation to assess PHEMA's reactivity during irradiation in the solid amorphous state for doses ranging from 5 to 100 kGy. Intrinsic viscosity measurements and molecular weight (MW) determination using size exclusion chromatography (SEC) confirmed the propensity to undergo chain scission, a common trend observed with poly(methacrylates) submitted to irradiation. The radiolytic yields for cross-linking and scission were assessed. Spectroscopic data allow to propose a reasonable mechanism accounting for these observations.

EB-irradiation (10 –100 kGy) of poly(HEMA) networks with various initial cross-link densities was shown to have a moderate impact on the swelling characteristics and thermo-mechanical properties of the materials treated in the dry or swollen state. However, gravimetric measurements revealed that irradiation induces the formation of extractables.

Different approaches were explored to synthesize the nanocomposites with inclusions of gold nanoparticles: (i) direct Au(III) reduction within the matrix at various doses and (ii) pre-irradiation of dry or water-swollen disks followed by soaking in Au(III) solutions. The efficiency of Au(III) reduction was evaluated by UV-vis spectroscopy.

These results confirmed that in spite of trend for main chain scission, direct Au(III) reduction by exposing the swollen PHEMA-based matrices could be achieved at doses where network degradation is kept at a very low level. The efficiency of Au(III) reduction was confirmed and quantified by UV-vis spectroscopy.

The effect of network mesh size which is dependent on the content in cross-linking agent (0.5, 1, 2 and 5 wt-% of EGDMA) was shown to modify the UV-vis. spectra of the nanocomposites, presumably by controlling the nanoparticle size via the diffusion rate of Au(III) ions, of reducing species and/or of nanoparticles formed upon radiolytic synthesis.

Sub-millimeter 2D PHEMA-based patterns were successfully converted into nanocomposites using the direct reduction method within the hydrogel matrix.

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Session Classification: Radiation Chemistry

Track Classification: Radiation Chemistry