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Radiation-induced degradation of galactomannan polysaccharides

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Abstract

In this study, guar gum, tara gum and locust bean gum were irradiated in a gamma cell in the solid state. The change in their molecular weights were determined by size exclusion chromatography analysis and the change in their viscosity values with change of temperature and irradiation dose were determined. Chain scission yield, $G(s)$, and degradation rate values were calculated. The calculated $G(s)$ values is 1.09 ± 0.16, 1.07 ± 0.06, 0.85 ± 0.10 for GG, TG and LBG, respectively. The effect of mannose–galactose ratio and initial molecular weight of these gums on the degradation behavior were discussed.
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1. Introduction

Galactomannans are neutral polysaccharides that occur in substantial amounts in the endosperm of the seeds of some leguminous plants. Structurally they consist of a β(1–4)-D-mannose backbone to which galactose units are attached α(1–6). Of the number of galactomannans known, guar gum (GG), locust bean gum (LBG), and tara gum (TG) are the most used in applications in, for example, the food, pharmaceutical, and chemical industries as thickening agents or stabilizers due mainly to the high viscosity they give at low w/w concentrations. While guar gum commonly has a mannose-to-galactose ratio (M/G) of approximately 2, M/G ratio is 3 and 4 for TG and LBG, respectively [1].

Their functional and physical properties (including solubility, gelling behavior, and viscosity) are related to the molecular structure, sugar composition, degree and distribution of branching, and polymerization [2].

In many industrial applications, the depolymerization of linear polysaccharides is essential. For example, guar solutions, which are used as hydraulic fracturing fluids in oil and gas recovery, need to be degraded to facilitate the outflow of oil. In addition, to understand the solution properties of guar as well as other water-soluble biopolymers, it is often necessary to degrade the native polymer to prepare samples with various molecular weights (MW) [3]. Depolymerization of polysaccharides has been widely studied. Though acid and enzymatic hydrolysis [4–6] are most common, other methods such as thermal [1], γ-irradiation [7], extrusion, ultrasonication [6,8] and free radical degradation are also reported [9].

However, mostly enzymatic hydrolysis is used to degrade galactomannans. There is only one work about degradation of guar gum with γ-rays in solution state. Jumel et al. investigated the change of absolute molecular weight and viscosity properties of guar gum samples irradiated in solution state with gamma rays in a 0–10 kGy dose range. They observed that the molecular weight and viscosity of the guar gum decreased with irradiation and this decrease was relatively steep at low irradiation doses (0.1–0.8 kGy). The decrease was much slower at high doses in both molecular weight and viscosity. They also investigated the change in the number of chain breaks per molecule G(s) and concluded that G(s) values at lower irradiation doses were higher than those at higher irradiation doses, i.e. more molecules were affected by low irradiation doses [10].

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In this study, GG, TG and LBG samples were irradiated in a ^{60}Co-gamma cell in the solid state. The change in their molecular weights were determined by SEC analysis and the change in their viscosity values with change of temperature and irradiation dose were determined. G(s) and degradation rate values were calculated.

2. Experimental

Commercially available locust bean gum (LBG), guar gum (GG) were obtained from Sigma–Aldrich and used as received, tara gum (TG) was obtained from domestic TGS Company.

The polymer samples were placed in tightly closed containers and irradiated to required doses (2.5, 5, 10, 20, 50, 100 and 150 kGy) in a Gammacell 220 type ^{60}Co-gamma irradiator at room temperature in air.

Irradiated and unirradiated samples were analyzed using a Waters Breeze model Gel Permeation Chromatograph. $NaNO_3$ (0.1 M) was used as the eluting solvent. Water 2000-1000 hydrogel columns were used for molecular weight analyses and universal calibration was constructed by using narrow molecular weight poly(ethylene oxide) standards. The Mark–Houwink parameters used for PEO were $K = 6.4 \times 10^{-5}$ dL/g and $a = 0.82$. The K and a constants for GG, TG and LBG were calculated by using the equation derived by Sittikijyothin et al. [11]. Corresponding K constants for GG, TG and LBG were taken as $K = 3.85 \times 10^{-6}$, $K = 2.89 \times 10^{-6}$, and $K = 2.31 \times 10^{-6}$ dL/g, respectively, and a constant is 0.98 for all gums.

The change of their viscosity with temperature and irradiation dose was measured by using a Brookfield DVII + Pro viscometer equipped with a SC4-21 type spindle at different rates.

3. Results and discussion

3.1. Controlling of molecular weight of polysaccharides

It is very well known that polysaccharides in dry form or in solution degrade when exposed to ionizing radiation. For the investigation of the effect of gamma rays on the molecular weight of guar gum (GG), tara gum (TG) and locust bean gum (LBG), $\overline{M}_w$ and $\overline{M}_n$ values of gums were evaluated by using size exclusion chromatography (SEC) analysis. The SEC chromatograms of LBG irradiated in the dose range of 2.5–75 kGy are given in Fig. 1 as examples. Similar unimodal chromatograms are obtained also for GG and TG samples. As the irradiation dose increased, the SEC chromatogram of the LBG sample is shifted to higher retention volumes indicating the molecular weight of the sample is decreased with irradiation.

By using universal calibration curve, average molecular weight of galactomannan samples were evaluated. The change of weight ($\overline{M}_w$) and number average ($\overline{M}_n$ molecular weights with irradiation dose is given in Figs. 2 and 3, respectively. As can be seen from these two figures, both average molecular weights decrease rapidly until 20 kGy and then this decrease was slowed down at higher doses and almost the same final molecular weight value was reached by all galactomannan samples.

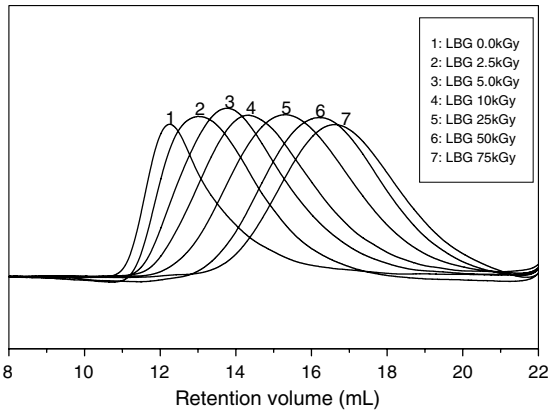

Fig. 1. Normalized GPC chromatograms of LBG irradiated to indicated doses.

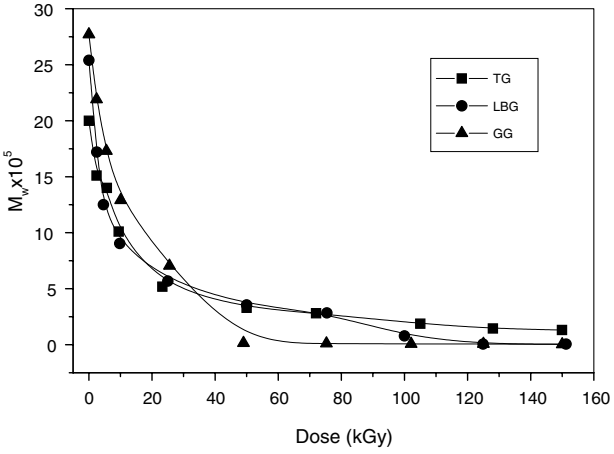

Fig. 2. The change in weight average molecular weight ($\overline{M}_w$) of galactomannans with dose.

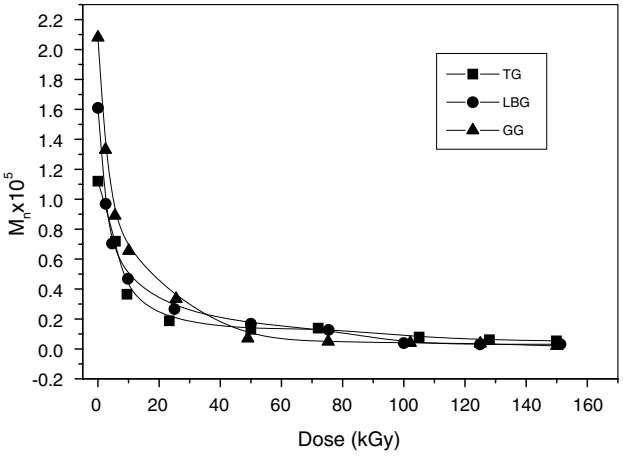

Fig. 3. The change in number average molecular weight ($\overline{M}_n$) of galactomannans with dose.

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