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Effect of Irradiated Gum Tragacanth on Rheological Properties of Oil in Water Emulsion

N. Mollakhalili Meybodi[1], M.A. Mohammadifar[2*]

1. *Department of National Nutrition and Food Technology Research Institute, Faculty of Nutrition Sciences and Food Technology, Shahid Beheshti University of Medical Sciences, Tehran, Iran*
2. *Department of Food Science and Technology, National Nutrition and Food Technology Research Institute, Faculty of Nutrition Sciences and Technology, Shahid Beheshti University of Medical Sciences, Tehran, Iran*

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Abstract

Background: Emulsions are thermodynamically unstable systems that can be kinetically stable by adding substances known as emulsifiers and/or thickening agents. By respect to the functional properties of gum tragacanth (GT), and its breakage probability during irradiation treatment, the major aim of this study was to investigate the stability of oil in water emulsion in the presence of different doses irradiated GT by respect to its viscosity and viscoelastic properties at constant temperature.

Methods: The effect of irradiation treatment was studied on the rheological properties of 10% w/w oil in water emulsions systems containing 0.5% w/w GT that is irradiated at doses of 0, 3, 5 and 10 kGy that are called NIGT, GT 3, GT 5, GT 5 and GT 10, respectively. In order to monitor the effect of irradiation treatment on rheological properties, rheometer has been used. All treatments were performed three times and the data were analyzed using SPSS 16.0. Values were considered to be significantly different if $p<0.05$.

Results: Irradiation treatment affects on rheological properties of GT containing oil in water emulsion both in steady state and oscillatory one. Increasing the irradiation dose up to 10 kGy decreases the consistency index from 1.88 to 0.076 and increases the flow behavior index from 0.42 to 0.764. Irradiation treatment also makes the systems behaves more sol like. It should be noted that these differents are dose dependant.

Conclusion: Rheological properties of irradiated GT containing emulsion are dose dependant. These changes may be due to the gums structural rearrangement due to irradiation. It is necessary to apply appropriate dose by considering its desirable function.

Introduction

Emulsions are thermodynamically unstable systems that are usually composed of two immiscible liquids, oil and water. Oil in water emulsions are composed of oil dispersions as small spherical droplets in the aqueous phase (Charoen et al., 2012). Emulsion instability is due to the increase in interfacial area following emulsification. So, they will be collapsed as two phases attempt to minimize the contact area. Despite the fact that emulsions are thermodynamically unstable systems, it can be kinetically stable (metastable) for a reasonable period of time, if their destabilization velocity is sufficiently low compared with the expended lifespan (Mirhosseini et al., 2009). With this fact in mind adding substances known as emulsifiers and/or thickening agents prior to homogenization will enhance the activation energy of system to form metastable emulsions (Charoen et al., 2012; McClements, 2005; Dombrowski et al., 2007). Food products such as mayonnaise and other emulsified products if not properly formulated, will

*Corresponding author
E-mail: mohamdif@ut.ac.ir

have poor storage stability and so the quality and safety of the products will be decreased.

Gum tragacanth (GT) is a complex, heterogeneous anionic hydrocolloid that consists of two fractions: water-swellable (Bassorin) and water-soluble (Tragacanthin). These ratios are different among different species (Mohammadifar et al., 2006; Karimi and Mohammadifar, 2014). As a result of these two fractions, it can act either as a thickening agent and/or emulsifier, so it is called bifunctional emulsifier (Farzi et al., 2013).

The main commercially used GT producers comprise Iran following by Turkey and Syria. GT has been used since 1961 in different industries due to its perfect properties such as high resistance to heat and acid (Gorji et al., 2011), high viscosity at low concentration (Pangborn et al., 1973), good suspending action and emulsifying properties.

Based on Food Chemical Codex, GT is a dried gummy exudation obtained from *Astragalus gummifer Labillardiere* or other Asiatic species of *Astragalus* (Anderson and Bridgeman, 1985) that is allowed as a food additive (code number E413) at the level of 0.2-1.3% by weight of their product that are used (Balaghi et al., 2011).

By regard to the botanical origin of GT and its contamination probability, it is necessary to make it safe for food consumption (Blake et al., 1988).

Food irradiation that is also called minimal processing technology is a treatment to improve food safety and quality, in which food products are exposed to electron or electromagnetic rays directly. Irradiation process has been approved by IAEA, FAO and WHO in doses up to 10 kGy however; it is applicable in doses as high as 75 kGy for some specific products (Farag Zaied et al., 2007; Aliste et al., 2000).

More than 55 countries use irradiation treatment on a commercial scale (Alijani et al., 2011). Since commercial application of food irradiation become more accepted (Blake et al., 1988; Dogan et al., 2007; Farag Zaied et al., 2007) today, it is used for food ingredients and ready to eat meals, too (Farkas and Mohacsi-Farkas, 2011). The effect of irradiation treatment on functional properties of natural polysaccharides that are used as rheological modifiers has been investigated in different studies. However, some studies indicated an inverse change in desirable characteristic of gums such as guar, agar, carageenan and alginate (Aliste et al., 2000; Gupta et al., 2009; Dogan et al., 2007), others show that pectin and salep gums didn't affected by irradiation treatment (Dogan et al., 2007).

Since, this using ordinary empirical shelf-life testing is time consuming and not practical, dynamic mechanical rheological testing can provide a rapid solution. In emulsions, the underlying structure and interaction of the fluid droplets hold the stability of the product against settling or separation. By respect to the functional properties of GT, either as an emulsifier or thickening agent that is directly connected to its structure, and its breakage probability during irradiation treatment, the major aim of this study was to investigate the stability of oil in water emulsion in the presence of different doses irradiated GT. Also, the effect of irradiation treatment on the viscosity and viscoelastic parameters at constant temperature was evaluated.

Materials and methods

Materials

Iranian gum tragacanth (*Astragalus gossypinus*) was collected from plants growing in the central mountainous area of Isfahan province, Iran. The raw gum was grounded and sieved. Powdered gum with mesh size between 200 and 500 was used in this study. Commercial sunflower oil from the same lot was prepared from a local market**.**

Irradiation

The powdered gum samples were kept in sealed polyethylene bags and irradiated at 0, 3, 5, 10 kGy at ambient temperature and at a fixed dose rate of 3.41 Gy/s from a Co^{60} gamma irradiator (Gammacell 220, AECL) at Nuclear Science and Technology Research Centre (Tehran, Iran). The gamma irradiator was calibrated using the Fricke dosimeter. Co^{60}, manufactured in Nordion International Co. Ltd., Ottawa, ON, Canada, was used for gamma ray source. The samples were kept at room temperature.

Preparation of gum dispersions

Irradiation treated GT dispersions (0.5% w/w) were prepared by adding 0.5 g of gum powder (200–500 µm) to 89.5 ml of distilled water, the mixture were kept on a magnetic stirrer at room temperature. Sodium azide (0.05% w/w) was added to prevent microbial growth. The solution was stored at 4 ^{o}C for an overnight to ensure that the hydration of the gum was complete. These amounts were chosen based on previous studies.

Production of Oil/Water emulsions

For the preparation of Oil/Water (O/W) emulsions, 10% (w/w) sunflower oil was added gradually to the gum dispersions and homogenized for 15 min at 13500 xg (based on previous studies) by Ultraturax (IKA T 25, Deutschland, Germany). Emulsions were ice-coated in order to prevent temperature fluctuations.

Rheological properties

Steady shear viscosity, strain and frequency sweep oscillatory shear tests were performed with a Physica MCR 301 rheometer (Anton Paar GmbH, Graz, Austria) equipped with a concentric cylinder measurement system with a radius

ratio of 1.0846. The temperature was adjusted to 25 °C with a peltier system equipped with fluid circulator with an accuracy of 10^{-2}. Rheological data were collected using Rheoplus software version 3.21 (Anton-Paar).

Flow curves were obtained at shear rates of 0.05–1000 s^{-1} at 25 °C, and then a power law model was used to describe the rheological properties of emulsions at middle shear rates. The flow behavior index (n) and consistency coefficient (m) values were obtained by fitting the shear rate versus apparent viscosity to the power law model (Eq. (1)):

$$\mu_a = m.\gamma^{(n-1)} \quad (1)$$

Where μ_a is the apparent viscosity (Pa.s), m is the consistency coefficient ($Pa.s^n$), γ is the shear rate (s^{-1}) and n is the flow behavior index (dimensionless).

Strain sweep tests were performed at strain of 0.1–600% and fixed frequency of 1 Hz to determine the linear region of viscoelasticity. Frequency sweep tests were carried out at frequency of 0.05- 50 Hz and constant strain of 1% to evaluate the dynamic rheological properties such as G' and G''.

Statistical analysis

Analytical values are based on the mean and standard deviation of three replicates. For all rheological measurements, the reported values are based on the mean of three replicates. Analysis of variance (ANOVA) was used for the data analysis (SPSS 16.0). When the F-values were significant ($p<0.05$) in ANOVA, Duncan's multiple-range test was used to compare treatment means. Statistical differences between samples were also calculated using student's t-test for selecting data. Values were considered to be significantly different if $p<0.05$.

Results

Flow curves of 10% w/w oil in water emulsions containing 0.5% w/w different doses irradiated GT are presented in Fig. 1. The apparent viscosities of emulsions containing 0.5% w/w irradiated GT were compared as a function of shear rate. As shown, all samples showed shear thinning behavior or pseudoplastisity. This means that there is a decrease in their apparent viscosity with increasing the shear rate for all systems.

Table 1 indicates the power law parameters for all systems. The high determination coefficients showed that the model was well fitted to the data. As shown, increasing the irradiation dose decrease the consistency coefficient (m) significantly ($p<0.05$), in a way that this index of systems containing 10 kGy irradiated GT is 24 fold lower than Non-Irradiated GT (NIGT) containing systems. Flow behavior index is an indicator of shear rate dependency of systems. According to Table 1, increasing the irradiation dose up to 10 kGy increased flow behavior index from 0.42 in systems containing NIGT to 0.764.

The strain sweep tests have been done to determine the limits of the linear viscoelastic domains for 10% w/w oil in water emulsion containing 0.5% w/w different doses irradiated GT that are depicted in Fig. 2.

As illustrated in Fig. 2, there are determined two different regions for all systems, namely, a linear viscoelastic region, in which G' and G'' were constant, and a nonlinear region, in which both moduli started to decrease by increasing strain, tending to crossover. The strain sweep tests were performed at a constant frequency of 1 Hz. The limiting values of strain (γ_L), tan δ and τ obtained within the LVE range are presented in Table 2. The limiting values of strain (γ_1) were 5.8 and 10.3 for NIGT and GT 3, respectively and >600% for both GT 5, GT 10. Frequency sweep tests of 10% w/w oil in water emulsions containing 0.5% w/w different doses irradiated GT are presented in Fig. 3. As mentioned before, systems linear viscoelastic range has been determined by a strain sweep test, but in order to characterize its structure, frequency sweep test has been done at a certain strain below critical strain.

In fact, this test obtains information about the effect of colloidal droplets interactions on emulsions structure strength.

Discussion

A steady state rheological property is an indicator of shear rates dependency of emulsion flow properties. Shear thinning behavior that is observed for different doses irradiated GT may be as a result of elongation and alignment of flocs or water-swellable fraction of GT with shear rate field (Balaghi et al., 2010). Apparent viscosities for these emulsions at all shear rates, as a function of irradiation dose followed the order of NIGT>GT 3>GT 5>GT 10. According to the stocks law, systems lower viscosity will enhance the upward movement of emulsion droplets and increase the emulsion instability (Dickinson, 2009). By respect to Table 1, the consistency coefficient decrease that is perceived by increasing the irradiation dose indicates that irradiation treatment decrease droplet interaction and will attenuate the emulsion structure. Flow behavior index is an indicator of shear rate dependency of systems. The increasing trend in the flow behavior index with the increasing irradiation

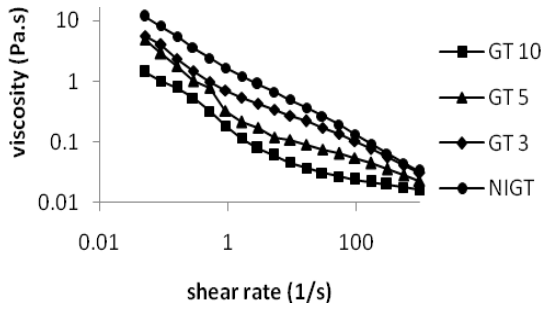

Fig. 1. Effect of shear rate on the apparent viscosity of emulsion containing different dose irradiated GT (0.5%, w/w) at 25 ˚C

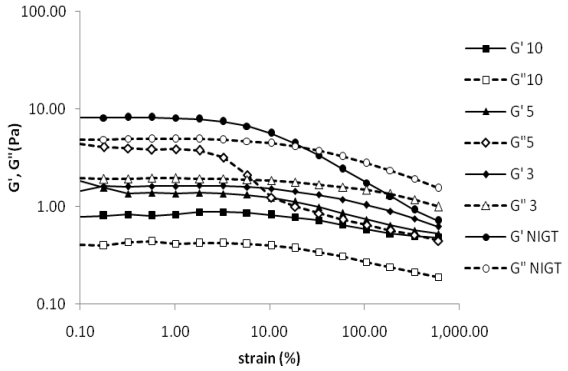

Fig. 2. Strain sweep of moduli G' and G" for 10% w/w oil in water emulsion containing 0.5% w/w different doses irradiated GT at 25 ˚C

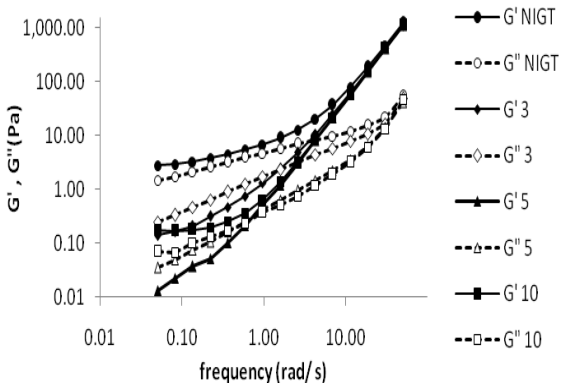

Fig. 3. Frequency sweeps of moduli G' and G" for oil in water emulsion containing 0.5% w/w different doses irradiated GT at LVE range at 25 ˚C

Table 1: Parameters of Power-law emulsion containing 0.5% w/w different doses irradiated GT

Irradiation dose (kGy)	Parameters			
	m (pa.s^n)	n	R^2	S (n- 1)
0	1.88^{a*}	0.42^d	0.99	0.5
3	0.808^b	0.536^c	0.99	0.792
5	0.221^c	0.679^b	0.99	0.575
10	0.076^d	0.764^a	0.99	0.499

* Means with different letters within the same column differed significantly (p< 0.05)

Table 2: Structural strength G'_{LVE}, limiting value of strain γ_{LVE} and loss-tangent value tan δ_{LVE} in the linear viscoelastic range, yield stress at the limit of the LVE range and flow-point stress with corresponding for emulsion containing irradiated GT over different doses of Gamma- irradiation (0, 3, 5 and 10 kGy), as determined by strain sweep tests at 25 ˚C and a frequency of 1 Hz

Irradiation dose (kGy)	parameters				
	G'_{LVE}(Pa)	γ_L (%)	tanδ_{LVE}	τ_y (Pa)	τ_f (Pa)
0	7.8^{a*}	5.8^c	0.654^c	0.287^c	1.32
3	1.62^b	10.3^b	1.19^a	0.70^b	-
5	0.75^c	>600^a	1.05^b	>3.35^a	-
10	0.564^d	>600^a	0.48^d	>3.35^a	-

* Means with different letters within the same column differed significantly (P< 0.05)

dosage indicate that systems containing gums which were exposed to higher doses of irradiation, showed less pseudoplasticity and were nearing newtonian behavior. By regard to Fig. 1, systems containing NIGT are more dependants to shear rate than those containing higher doses irradiated GT. Since the shear rate dependency of viscosity

among linear polymers is lower than that of branched polymers (Hwang and Kokini, 1991), and flocculated droplets show absolutely shear thinning behavior (Farzi et al., 2013), in our investigation it seems that irradiation treatment change the gum structure in a way that decrease the polymer molecule branches. This assumption is in compliance with other studies that have been shown the irradiation treatment decrease the apparent viscosity of konjac glucomannan, agar, alginate, gum arabic and carrageenan in an aqueous solution as a function of dosage (Aliste et al., 2000; Blake et al., 1988; Dogan et al., 2007).

In oscillatory testing instruments, samples are subjected to harmonically varying stress or strain. Strain sweep test, the preliminary step in the analysis of oscillatory flow data, determine the linear viscoelastic region of emulsions. As mentioned before, there are two regions in strain sweep tests graphs. The lengths of the LVE range indicated that both storage and loss moduli amounts were independent of the oscillation strain that is called reversible elasticity.

By regard to Fig. 2, it can be seen that both moduli decreased by increasing the irradiation dose that is may be due to the reduction of the effective volume fraction of the emulsion (McClements, 2005). Additionally, results indicated that emulsion containing NIGT had the highest dynamic moduli at linear viscoelastic range and storage modulus dominate loss modulus. It has been reported that systems which have higher storage modulus than loss modulus in the LVE range exhibits certain rigidity. However, this is typical for solids or stable pastes but much dispersion such as coatings or foodstuffs that exhibit low viscosity flow behavior at medium and high shear rates also have G' greater than G" in the LVE range. Increasing the irradiation dose up to 5 kGy decrease the moduli amount in order to dominance of loss moduli over storage ones which means systems containing irradiated GT at 3, 5 kGy behave like sol. However, increasing the irradiation dose up to 10 kGy decrease the moduli amount but decrease the loss modulus more than storage one. In other words, in systems containing 10 kGy irradiated GT, storage modulus dominates loss modulus and behaves like a gel, but this gel structure is weaker than that has been made in NIGT containing systems. It is assumed that irradiating up to 10 kGy breaks the gum structure in a way that increases the active unit and decreases the systems strength. So, smaller active units will cover the oil droplets and make a weak structure. Breakage that is occurred during irradiating, however increase the gum anchoring ability but is unable to create structured network to make a stable emulsion. This inability may be as a result of decreasing the continuous phase viscosity. As mentioned before, GT has two fractions, water soluble Tragacanthin and water swellable Basorin. Water swellable fraction is responsible for the formation of aggregates in the system, due to its hydrophobicity and higher amount of methoxyl groups, so increase the continuous phase viscosity and enhance the emulsion stability. Increasing the irradiating dose may affect the instability by destructing the gum insoluble fraction (Balaghi eta al., 2011; Alijani et al., 2011).

Considering Table 2, it is clear that systems with higher γ_L represent longer linear viscoelastic ranges which imply a higher stability of the viscoelastic material under the strain amplitude. Previous studies indicated that the linear viscoelastic region is dependant to gums soluble to insoluble fractions (Balaghi et al., 2011). In other words, gums with higher soluble to insoluble fraction have higher limiting values of strain. So, increasing irradiation dose will enhance the soluble to insoluble fraction and decrease the systems strength.

Frequency sweep test provides information about the effect of colloidal droplets interactions on emulsions structure strength. In a frequency sweep test, measurements are made over a range of 0.05-50 Hz oscillation frequencies at constant oscillation amplitude 0.8% at 25 $^{\circ}$C.

As indicated in Fig. 3, it seems that systems containing NIGT with high amount of large hydrodynamic radius Bassorin show the dominance of elastic behavior that is due to that Bassorin adsorbs water and swells and creates a gel. Increasing the irradiation doses up to 5 kGy will cause a transition from a predominantly viscous response at lower frequencies (G">G') to a predominantly elastic response at higher frequencies (G'>G") so these two moduli cross over at middle frequencies. This means that increasing the irradiation dose makes the system behaves like an entanglement network without any physical and chemical bonds (Chamberlain and Rao, 2000). However, regularly weaker networks have higher crossover frequency, but in our investigation weaker structure have lower one, to the extent that by increasing the irradiation dose from 3 to 5 kGy, this crossover point shifted from 1.58 Hz to 0.63 Hz. Surprisingly, systems containing 10 kGy irradiated GT behaves like a gel in all frequency range but its elastic modulus is drastically lower than that of NIGT containing emulsion. Since by increasing the frequency dependency of the elastic modulus makes the material behave more fluid-like, by considering the Fig. 3, it is clear that increasing the irradiation dose changes the gums containing emulsion to the sol behavior.

According to the frequency sweep datum, it seems that increasing the irradiation dose breaks the gums insoluble fraction to the extent that is unable to create a structure system.

Conclusion

The results demonstrated that increasing the irradiation dose decreases the emulsion viscosity. It also changed the oscillatory behavior of systems containing different doses irradiated GT. Therefore, irradiation treatment will change

GT function. These changes may make the gum more applicable in some systems. So, irradiation treatment must be done at certain dose by considering its desirable application.

Conflicts of interest

The authors declare that they have no conflict of interest.

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