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Radiation synthesis of superabsorbent polyethylene oxide/tragacanth hydrogel

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ABSTRACT

A new superabsorbent hydrogel has been prepared from tragacanth and polyethylene oxide (PEO) by gamma radiation at room temperature. Tragacanth solutions with different concentrations (1%, 3% and 5%) have been blended with 5% aqueous solution of PEO at a ratio of 1:1 and irradiated at doses 5–20 kGy. The properties of the prepared composite hydrogels were evaluated in terms of the gel fraction and the swelling behavior. An unexpected growth of the gel fraction was observed in PEO/tragacanth hydrogels irradiated at 5 kGy. Incorporation of 5% tragacanth into the aqueous PEO increased significantly the swelling percent of the hydrogels to more than 14,000% and thus makes it a superabsorbent material.

1. Introduction

Superabsorbent hydrogels have a structure in which water-soluble polymers are made insoluble by some means of crosslinking. These materials exhibit the ability to absorb water and swell, but do not dissolve in water (Buchholz et al., 1998).

In recent years, the attention of many researchers has been focused on the preparation of hydrogels with large amount of water absorption and improved wet strength for different purposes like adapting them for medical purposes (Pitarresi et al., 2002), entrapment of drugs (Martellini et al., 1998), personal hygiene products(Iskra, 1991; Kellenberger, 1992), agricultural uses (Siemer et al., 1993), construction industry (Yamaguchi et al.,1987; Yooda, 1994), food packaging (Tatsuo et al., 1995), electronic and cables (Sheu, 1992) and cosmetics (Siller Jackson et al., 2003).

Natural polymers such as polysaccharides represent an interesting class of materials that can be explored in combination with synthetic polymers for developing composite hydrogels. These kinds of hydrogels are known since at least mid-1990s and they have applications in biosensors, enzyme immobilization, cell encapsulation and drug-delivery devices. Hyaluronic acid-based hydrogel was synthesized by reacting hyaluronic acid with polyethylene oxide (PEO) via Michael-type addition reaction and has applications in tissue engineering scaffolds (Noh et al., 2006; Kim et al., 2007). Composite gels of carboxymethylcellulose and polyethylene oxide have been made to separate healing tissues and reducing adhesions between them (Liu et al., 2002). Other composite hydrogels are chitosan/PEO (Khalid et al., 1999) and kappa-carrageenan/PEO that was prepared by gamma irradiation (Tranquilan-Aranilla et al., 1999).

Tragacanth is a natural polymer that is composed of poly-D-galacturonic acid and bassorin. It is obtained through incisions in the stem of the small bushes of *Astragalus gummifer*, native to the highlands of Asia Minor from Turkey to Afghanistan, but it is produced chiefly in Iran (Columbia Encyclopedia, 2005). The galactronic acid part of tragacanth is water soluble and has a high molecular weight which gives highly viscous solutions. Bassorin, the other part of tragacanth, is water insoluble but swells to a gel. As a result, tragacanth is almost insoluble in water, but swells to form a stiff gel. It has a wide application in pharmaceuticals, cosmetics, industrial textile sizing and as a thickening agent in foods (Wilks, 2001). Generally, polysaccharides degrade under ionizing radiation and their molecular weight decrease (Nagasawa et al., 2000).

Polyethylene oxide (PEO) has rarely been used as an absorbent polymer, because it is a highly crystalline polymer and water absorption is therefore limited. By radiation crosslinking of aqueous solutions of high-molecular-weight polyethylene oxide, it forms a hydrogel with absorbent properties (Rosiak et al., 1995; Stringer et al., 1996).

By using a combination of the PEO and tragacanth a more economic hydrogel with good superabsorbent properties could be produced. This paper presents the preliminary results on the

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properties of PEO/tragacanth composite hydrogel prepared by gamma radiation.

2. Experimental

2.1. Materials

PEO with molecular weight of 200,000 was supplied by Aldrich Co. Tragacanth was obtained from Sigma Company with approx. 95% purity. Distilled water was used for the preparation of hydrogels.

2.2. Preparation of PEO/tragacanth blends

Various concentrations of tragacanth (0%, 1%, 3%, 5%) and PEO (5%) solutions were prepared by dissolving in water. They were blended at a ratio of 1:1 and poured into the glass tubes. After removing oxygen from the samples in an ultrasonic bath, they were irradiated by Co-60 Gamma cell-220 with a dose rate of 1.418 kGy/h, at different doses of 5–20 kGy.

2.3. Measurements

2.3.1. Gel fraction

The hydrogels were dried to constant weight at 40 °C (W_0). The dried gels were put in a soxhlet system and extracted with distilled water for 6 h to remove the sol fraction. Then the gels were dried to a constant weight (W_g). The gel fraction was calculated as follows:

$$\text{Gel fraction (\%)} = W_g/W_0 \times 100$$

2.3.2. Swelling behavior

The dried gels were immersed in distilled water at room temperature. The swollen gels were periodically weighed after the excess surface water was removed with a filter paper. The procedure was repeated until there was no further weight increase. The swelling percent was determined according to

$$\text{Swelling (\%)} = (W_t - W_d)/W_d \times 100$$

where W_d is the weight of dried gel before swelling and W_t is the weight of the swollen gel at time t.

3. Results

3.1. Gel fraction

Fig. 1 shows the variation of the gel fraction of PEO/tragacanth hydrogels as a function of the irradiation dose (5–20 kGy) and varying amount of tragacanth in the hydrogels. It is observed that upon irradiation, more gel is formed in the samples with tragacanth as compared with the homopolymer (without tragacanth).

It is known that tragacanth is a natural polysaccharide which degrades on irradiation like the common natural polymers (IAEA, 2002), while PEO crosslinks in aqueous medium by irradiation. When the mixture of tragacanth and PEO is irradiated, an unexpected growth of the gel fraction was observed. Addition of 1% tragacanth resulted in a drastic increase of the gel fraction to 81% at dose of 5 kGy, while the PEO homopolymer gel showed only 15% gel fraction at this dose. This could be due to the formation of an interpenetrating network (IPN) with chemical crosslinking of PEO and physical crosslinking of tragacanth.

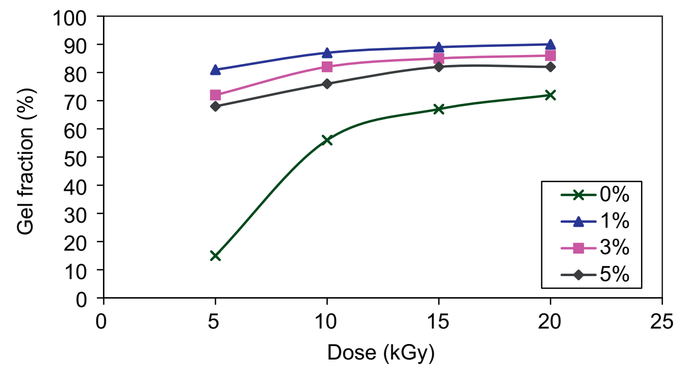

Fig. 1. Gel fraction of PEO with various concentration tragacanth (0%, 1%, 3%, 5%) hydrogels as a function of dose.

Similar results have been reported for other polysaccharide, carrageenan, while forming hydrogel with acrylic acid (Francis et al., 2004) and poly(diallyldimethylammonium chloride) (Jing et al., 2001) by gamma radiation processing.

By addition of tragacanth percent from 1% to 5% in PEO/tragacanth blend, irradiation of these samples resulted in degradation and reduced crosslinking density in the blend and therefore a decreasing gel fraction.

3.2. Swelling behavior

The swelling behavior of blends made of 5% PEO and various amounts of tragacanth (0%, 1%, 3%, 5%), irradiated at 5–20 kGy are shown in Fig. 2(a–d). The swelling increases with time, but after some times reaches a limiting equilibrium value. For each of the blends, increasing the irradiation dose was accomplished with decreasing of the swelling behavior of the hydrogel. This is due to the formation of more crosslinks and a tighter structure at high irradiation doses. Crosslinking hinders the mobility of the polymer chain and hence it reduces the penetration of water in to the hydrogel structure.

The homopolymer gel of PEO formed at 5 kGy was too loose and it had a weak structure so that after soxhlet process there was no firm gel for swell test (Fig. 2a). The homopolymer gels formed at higher doses (10–20 kGy) swell up to 4000% after many days. By increasing the amount of tragacanth in the blends, the swelling percent of the hydrogels increased significantly (Fig. 2c–d). Especially when the hydrogel with 5% tragacanth was irradiated at low dose of 5 kGy the swelling reached more than 14,000% at the early time of immersion in water (Fig. 2d). This result is in agreement with lower gel formation in the blend with 5% tragacanth as shown in Fig. 1. It was hard to dissolve more than 5% tragacanth in water due to its limited solubility and high viscosity.

The effect of tragacanth concentration on the gel fraction accompanied by maximum swelling of hydrogels which were irradiated at 5 kGy is shown in Fig. 3. By increasing the amount of tragacanth from 1% to 5%, the gel percent levels off from 80% to 60%. This could be due to degradation of tragacanth upon irradiation and probably physically screening the approach of radical sites that form crosslinks. According to Fig. 3, although increasing of tragacanth in the blend caused the gel percent to decrease, the swelling behavior of the hydrogel increased up to 14,000% because of ease in penetration of water into the looser network structure. It can be concluded that although increasing of tragacanth in the blend causes a decrease of the gel fraction, the swelling behavior of the hydrogel was improved. Similar behavior is observed for swelling of carrageenan/PEO blend irradiated at 10 kGy (Tranquilan-Aranilla et al., 1999).