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A novel photo-grafting of acrylamide onto carboxymethyl starch. 1. Utilization of CMS-g-PAAm in easy care finishing of cotton fabrics

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ABSTRACT

The photosensitized grafting of vinyl monomers onto a range of polymeric substrates has been the subject of particular interest in the recent past. Carboxymethyl starch (CMS)-poly acrylamide (PAAm) graft copolymer (CMS-g-PAAm) with high graft yield was successfully prepared by grafting of acrylamide onto CMS using UV irradiation in the presence of the water soluble 4-(trimethyl ammoniummethyl) benzophenone chloride photoinitiator. CMS-g-PAAm with nitrogen content of 8.3% and grafting efficiency up to 98.9% was obtained using 100% AAm, a material: liquor ratio of 1:14 and 1% photinitiator at 30 °C for 1 h of UV irradiation. The synthesis of CMS-g-PAAm was confirmed by FTIR and Nitrogen content (%). Surface morphology of CMS and surface morphological changes of CMS after grafting with AAm were studied using SEM. Thermal properties of both CMS and CMS-g-PAAm were studied using TGA and DSC. To impart easy care finishing to cotton fabrics, aqueous formulations of: CMS-g-PAAm, dimethylol dihydroxy ethylene urea (DMDHEU), CMS-g-PAAm-DMDHEU mixture or methylolated CMS-g-PAAm were used. Cotton fabrics were padded in these formulations, squeezed to a wet pick up 100%, dried at 100 °C for 5 min, cured at 150 °C for 5 min, washed at 50 °C for 10 min and air-dried. CRA (crease recovery angle) of untreated fabrics and fabrics finished with a mixture of 2% CMS-g-PAAm and 10% DMDHEU or methylolated CMS-g-PAAm (10% formaldehyde) were: 136°, 190°, 288° respectively. Increasing the number of washing cycles up to five cycles results in an insignificant decrease in the CRA and a significant decrease in RF (releasable formaldehyde) of finished fabric samples. The morphologies of the finished and unfinished cotton fabrics were performed by SEM.

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1. Introduction

Starch is naturally occurring, abundant, renewable, biodegradable, biocompatible and low cost polymeric carbohydrate bearing three hydroxyl groups per its repeated anhydroglucose unit. The abundance of these hydroxyl groups imparts hydrophilic properties to the polymer. On the other hand, the linearity, mobility, and hydroxyl groups of starch permits the formation of hydrogen bonding between the parallel/adjacent chains leading to the formation of “crystalline structure”. As a result, the affinity of the polymer to water is reduced. Due to its high molecular weight, starch acquires “film forming” properties. However, the films are rigid and require good lubricant. In addition, Starch is a biodegradable polymer but this makes it susceptible to rot by microorganisms. These combinations between the advantages and the disadvantages of the starch properties make modification of starch an essential need before application in different industries. Modified starches were developed to overcome one or more of the shortcomings of starch and thus expand the usefulness of starch for a number of industrial applications (Chiu & Solarek, 2009; El-Sheikh, 1999a; Zhang, 2001).

Among the chemical modifications of starch are hydrolysis (Kim, Park, Kim, & Lim, 2013; Murthy, Johnston, Rausch, Tumbleson, & Singh, 2011), oxidation (El-Sheikh, Ramadan, & El-Shafie, 2009; El-Sheikh, Ramadan, & El-Shafie, 2010), etherification (El-Sheikh, 2010; Lee et al., 2010), esterification (Shogren & Biswas, 2006; Tarvainen et al., 2004), crosslinking (Gui-Jie, Peng, Xiang-Sheng, Xing, & Tong, 2006; Nagasawa, Yagi, Kume, & Yoshii, 2004) and graft copolymerization (Hebeish, Bayazeed, El-Alfy, & Khalil, 1988; Hebeish, El-Rafie, Sayed, Drag, & Ramadan, 2015).

The carboxymethylation (etherification reaction) of starch successively increases the water solubility. At higher DS, CMS is cold-water soluble and its solution is clear. The importance of the carboxymethylation process arises from the promising properties of the carboxyl group in applications such as chelation, ion-exchange, polyanion flocculation, and the acidity function as well as the solution properties that include thickening, gelation,

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water absorption, adhesion, and film forming (El-Sheikh, 1999c, 2010; Tijsen, Voncken, & Beenackers, 2001).

An important advantage of graft copolymerization is the fact that starch and synthetic polymer are held together by chemical bonding rather than existing merely as a physical mixture. The two dissimilar polymers, therefore, tend to be more intimately associated. Separation of the two polymer phases is less likely to occur.

The synthesis of polymer-starch composites has been investigated by a number of authors. These composites have been synthesized in order to add new attributes to the starch without any major change to its natural characteristics and so to expand the market for such starch-based products. Many of these composites have important industrial uses such as thickening agents (Lu, Yao, Lin, Cao, & Chen, 2003), waste water treatment (Dragan & Apopei, 2011; Güçlü et al., 2010; Janaki et al., 2012; Zheng, Hua, & Wang, 2010), sizing agents (Hebeish, Aly, El-Shafei, & Zaghloui, 2008; Mostafa, 1997; Zhang & Li, 2003), superabsorbents (Guo, Liu, & Gong, 2013; Li, Xu, Wang, Chen, & Feng, 2009; Saboktakin, Maharramov, & Ramazanov, 2009; Teli & Waghmare, 2010; Zhang, Li, & Wang, 2006), bioadhesion, drug delivery (Al-Karawi & Al-Daraji, 2010; Ameye et al., 2001; Geresh et al., 2004; Mahkam, 2010; Simi and Abraham, 2006), nanoparticles synthesis (Das et al., 2015; El-Sheikh, 2015; El-Sheikh, El-Gabry, & Ibrahim, 2013; El-Sheikh & Ibrahim, 2014 Tudorachi & Chiriac, 2013; Wang et al., 2012) and oilfield (Zhang, 2001; Zhang & Chen, 2002, 2003).

The photosensitized grafting of vinyl monomers onto a range of polymeric substrates has been the subject of particular interest in the recent past. Among the various methods allowing the generation of radicals, e.g. γ irradiation, redox chemical initiation, heating, dye-sensitized reaction and UV light-induced grafting, sensitized reactions are more attractive. UV light-induced grafting allows the use of larger wavelengths of excitation, thus increasing the possibility of potentially valuable commercial utilization (Hérold & Fouassier, 1980). However, this kind of reaction requires knowledge of the photo reactivity of the initiator with respect to the chemical nature of the monomer and of the macroradical. Since polymers absorb light below 300 nm, photosensitizers or photoinitiators that absorb near UV or visible light have been generally added to formulations. The excited molecule may then split and/or react with the subsequent formation of reactive radicals that are capable of attacking the polymeric backbone (Merlin & Fouassier, 1980). Free radical generation in cellulose, by photosensitized excitation, has shown clearly that UV irradiation, both in photosensitized and unsensitized experiments, results in chain scission and radical generation on the glucosidic ring (Bellobono, Calgari, Leonardi, Selli, & Paglia, 1981). By proper choice of the sensitizer, the scission of glucosidic bonds can be reduced and radical generation can be enhanced, i.e. a lower degradation of the polymer and an increase in the rate of photochemical reaction.

With the development of efficient water-soluble photosensitizers for polymerization reactions, attention has been given to grafting reactions in largely aqueous media, involving hydrophilic substrates.

Benzophenone compounds are powders that can absorb and dissipate UV radiation. Benzophenone compounds are utilized as a part of bath items, cosmetics items, hair items, sunscreens, and healthy skin items. They save the skin from the unsafe impacts of the sun.

Bottom et al. (Bottom, Guthrie, & Green, 1985a; Bottom, Guthrie, & Green, 1985b) created and estimated the productivity of a novel, water-dissolvable photosensitizer, 4-(trimethyl ammonium methyl) benzophenone chloride, and utilized it for the photoinitiated graft copolymerization of 2-hydroxyethyl acrylate onto cellulose. El-Sheik and collaborators (El-Sheikh, 1999b; El-Sheikh & Guthrie, 1997) further used this productive photoinitiator in the graft copolymerization of acrylic acid onto both starch and CMS with an effective graft yield and in the photooxidation of starch (El-Sheikh et al., 2009). Recently, El-Sheikh (El-Sheikh, 2014) used the water soluble 4-(trimethyl ammoniummethyl) benzophenone chloride photoinitiator for the first time in the photoreduction of silver ions to silver nanoparticles (AgNPs) in presence of CMS as a stabilizing agent and utilizing the prepared AgNPs to impart cotton, wool and acrylic fabrics antibacterial properties (El-Sheikh et al., 2013; El-Sheikh & Ibrahim, 2014).

Grafting of AAm onto starch was studied by many authors. Different chemical initiation systems such as potassium persulfate (Lu et al., 2003; Nakason, Wohmang, Kaesaman, & Kiatkamjornwong, 2010), ammonium persulfate (Li, Liu, & Wang, 2005; Zhang, Chen, & Wang, 2006), ceric ammonium sulfate (Eutamene, Benbakhti, Khodja, & Jada, 2009) and ceric ammonium nitrate (Hao, Chang, Duan, & Zhang, 2007) were used. Physical initiation systems such as γ irradiation (Kaur, Misra, Barsola, & Singla, 1993) and microwave (Singh, Tiwari, Pandey, & Singh, 2006) were also utilized. Synergism of microwave radiation and ceric ammonium nitrate system was also applied (Banerjee et al., 2012).

To the knowledge of the authoress, the water-soluble photoinitiator 4-(trimethyl ammoniummethyl) benzophenone chloride and UV irradiation system was not used to initiate graft copolymerization of AAm onto starch or starch derivatives.

In this work, 4-(trimethyl ammoniummethyl) benzophenone chloride/UV irradiation system was used for the first time to initiate graft copolymerization of AAm onto CMS to produce CMS-g-PAAm graft copolymer. The produced CMS-g-PAAm and the grafting process were assessed by measuring the nitrogen content and the grafting efficiency. Optimum conditions that produced highest nitrogen content and reaction efficiency were selected. Final product at which optimum conditions were applied at was further characterized using SEM, FTIR, TGA and DSC.

It is thought that water soluble starch composite bears both carboxyl and methylol groups can efficiently act as an easy care finishing agent for cotton fabric. Here, the produced CMS-g-PAAm was methylolated using different concentrations of formaldehyde solution. CMS-g-PAAm, DMDHEU, CMS-g-PAAm-DMDHEU or methylolated-CMS-g-PAAm were applied to cotton fabrics to impart easy care finishing properties. Optimum conditions that produce finished cotton fabrics with highest CRA were studied. Final finished cotton fabric was further characterized for its morphological properties using SEM.

2. Experimental

2.1. Materials

100% bleached, cotton fabric was supplied by Misr El–Beida Dyers, Egypt. The fabric was not subjected to any type of finishing treatments. The fabric was washed with a solution containing 5 g/l sodium carbonate and 5 g/l non–ionic detergent at boil for 3 h. It was then rinsed with hot and cold water and left to dry in air at room temperature.

Native maize starch (NS) was kindly supplied by the Egyptian Company for Starch and Glucose Manufacture, Cairo, Egypt.

4-(trimethyl ammoniummethyl) benzophenone chloride was supplied by The associated Octel Ltd., Widnes, Great Britain, and used without further purification.

Monochloroacetic acid, hydrochloric acid, acetic acid, sodium hydroxide, sodium carbonate monohydrate, ethanol, acrylamide, ammonium chloride, boric acid, formaldehyde solution (36.5%) were of laboratory grade.

Egyptol, a non–ionic detergent of technical grade chemicals, was provided by The Egyptian Company for Starch, Yeast and Detergents, Alexandria, Egypt.

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