International Journal of Biological Macromolecules 117 (2018) 179–188

Contents lists available at ScienceDirect

International Journal of Biological Macromolecules

journal homepage: http://www.elsevier.com/locate/ijbiomac

Antimicrobial cellulosic hydrogel from olive oil industrial residue

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ARTICLE INFO

Article history:
Received 24 April 2018
Received in revised form 21 May 2018
Accepted 24 May 2018
Available online 25 May 2018

Keywords:
Acrylamide
Olive stone residue
Grafting
Silver nanoparticles
Antimicrobial hydrogel

ABSTRACT

The cellulose-based antimicrobial hydrogel was prepared from seed and husk cellulosic fibers of olive industry residues by load silver nanoparticles (AgNPs) onto grafted acrylamide monomer (Am) cellulosic fibers. The grafting approach was the free radical mechanism by utilizing ceric ammonium nitrate (CAN) as initiator in aqueous medium and *N,N* methylene bisacrylamide (MBAm) as a cross linker. The effect of different grafting conditions on the properties of produced hydrogels has been studied by determining the grafting parameters, i.e. concentration of Am, MBAm, grafting time and temperature to optimize grafting yield (G %), grafting efficiency (GE %), and swelling %. Characterizations of the obtained hydrogels were performed through monitoring swelling behavior, FTIR spectroscopy, SEM, and EDX. AgNPs were grown into the prepared hydrogel. Hydrogel/AgNPs were characterized by FT-IR spectroscopy, X-ray diffraction (XRD) and scanning electron microscopy (SEM). The hydrogel loaded AgNPs exhibit high efficient antimicrobial activity against *Staphylococcus aureus*, *Pseudomonas aeruginosa*, and *Candida albicans*.

1. Introduction

Olive stone was produced as a lignocellulosic byproduct during olive oil extraction and its major constituents are hemicelluloses, cellulose and lignin in addition to proteins, fat, phenols, free sugars as well as polios. Olive stone consists of two main portions husk and seed residues, from which cellulosic fibers can be produced by different delignification processes. The olive fruit is consisting of skin and pulp. Both of skin and pulp represent the husk of olive fruit. And the stone, that contains the seed [1].

Utilization of olive stone was limited for production of energy by combustion or activated carbon. In addition to furfural production, and plastic filler as well as its uses as animal nutrition and resin manufacture [2]. Cellulose, the major component of this lignocellulosic material, is an organic polysaccharide consisting of D-glucose units linked to each other by β (1 → 4) glucosidic linkage [3]. Each D-glucose possess hydroxyl groups (-OH) at positions C2, C3 and C6 which typically reacted as secondary and primary alcohols, respectively. Many modification processes have been reported in the literature to improve the properties of the native cellulose. Grafting is one of the most imported approached utilized for cellulose modification. Grafting of cellulose is a striking study because its products could possess any number of required properties [4] [5]. Grafting has been performed by several methods, but free radicals are the most convenient methods as they generate radicals on the cellulosic backbone before grafting [6–8]. It can be performed with different initiator systems which are widely used to synthesize graft copolymers and they include ammonium persulfate, potassium persulfate, benzoyl peroxide, azobisisobutyronitrile and ceric ammonium nitrate [9–11]. To the best of our knowledge, no work has been directed towards grafting of acrylamide using ceric ammonium nitrate as initiator and in the presence of MBAm to form hydrogel onto olive oil by-products and then loaded with AgNPs by impregnation of grafted cellulose in $AgNO_3$ solution followed by using trisodium citrate solution. Antimicrobial materials have a great significance in many areas including medical applications, water purification systems, textiles and food packaging. AgNPs have been known to have strong antimicrobial activity. The loading of AgNPs was widespread in many scientific research due to it have much attention to control infections [12] and also have antibacterial properties than bulk silver as a result of high surface area which leading to incorporating more nanoparticles inside the bacteria and stop its action [13]. In addition, AgNPs were nontoxic to human cells [14].

The current study focused on using olive stone for production of antimicrobial hydrogel by loading AgNPs onto grafted cellulosic fibers which extracted from seed or husk portions of olive stone. Optimization of the grafting condition was achieved by attempt to investigate different grafting conditions. The study aim to make characterization of the prepared

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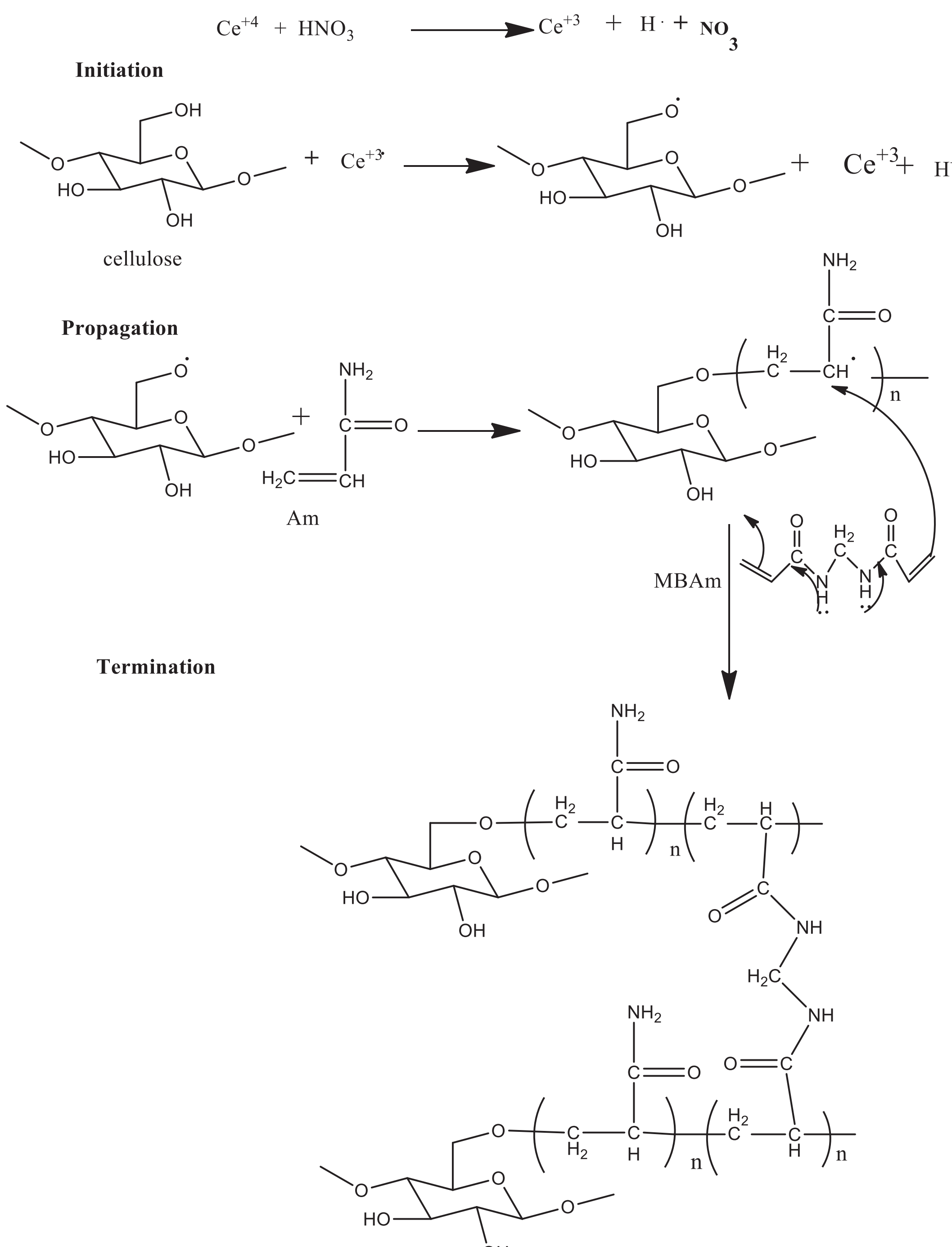
https://doi.org/10.1016/j.ijbiomac.2018.05.179
0141-8130/

hydrogels by FTIR, SEM, EDX, and XRD. The antimicrobial activity of the prepared hydrogels were tested against Staphylococcus aureus, Pseudomonas aeruginosa, and Candida albicans.

2. Experimental

2.1. Materials

Olive stone collected from industrial by-product of olive oil factory in Egypt. The main two portions (seed and husk) were separated by using the flotation technique, where husk has lower density than that of seed. The ratio between them was found to be 1.3:1.0 (Seed: husk as wt/wt). Acrylamide (Am) was purchased from alpha Chemika. *N*,*N* methylene bisacrylamide (MBAm) and silver nitrate ($AgNO_3$) were purchased from Acros Organic. Ceric ammonium nitrate (CAN) was purchased from Merck. Buffer solutions with pH range of 3 to 10 were used to study the effect of pH on swelling of hydrogels. The following buffer solutions were used; pH 10 ($NaHCO_3$/NaOH), 0.1 mol/L of $NaHCO_3$ was titrated with 0.1 M of NaOH solution to achieve pH 10) and pH 3 (H_3PO_4/NaOH), 0.1 mol/L of H_3PO_4 was titrated with 0.1 M of NaOH solution to achieve pH 3, the pH values were checked by a pH-meter.



Scheme 1. Preparation of hydrogel from cellulose.