



Antioxidant and antimicrobial activity of Maillard reaction products from xylan with chitosan/chitooligomer/glucosamine hydrochloride/taurine model systems



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ABSTRACT

The structure, UV absorbance, browning intensity, fluorescence changes, antioxidant activity and antimicrobial assessment of Maillard reaction products (MRPs) derived from xylan with chitosan, chitooligomer, glucosamine hydrochloride and taurine model systems were evaluated. The results revealed that all MRPs had similar infrared spectra and molecular structures. MRPs from different model systems on the UV absorbance at 294 nm after heated 90 min and browning intensity at 420 nm showed the similar law: xylan-*taurine* > xylan-*glucosamine hydrochloride* > xylan-*chitooligomer* > xylan-*chitosan*, and the order of DPPH scavenging activity of MRPs was as follows: xylan-*chitosan* > xylan-*chitooligomer* > xylan-*glucosamine hydrochloride* > xylan-*taurine*, which revealed that the properties of MRPs were closely related to molecular weight of model systems. Moreover, the highest radical scavenging activity of MRPs from xylan with chitosan/chitooligomer/glucosamine hydrochloride/taurine model systems was 65.9%, 63.7%, 46.4% and 42.5%, respectively.

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1. Introduction

The Maillard reaction (MR) also named as nonenzymatic browning, is a chemical reaction involving the condensation between a carbonyl group of reducing sugars, aldehydes or ketones, and an amino group of amino acids, proteins or any nitrogenous compound (Hodge, 1953). MR is the main reaction responsible for the transformation of precursors into colourants and flavour compounds during food processing and preservation (Totlani & Peterson, 2007), which can be influenced by many factors, including reactant concentration, temperature, time of heating, initial pH, and the characteristics of reactants. There are many researches focused on the systematic and comprehensive studies of different model systems. For example, MRPs from four sugars (glucose/fructose/xylose/ribose) and twenty amino acids showed different antioxidant properties, and MRPs from fructose-amino acid models revealed the similar UV–Vis absorbance and antioxidant activity with MRPs from glucose-amino acid models, MRPs from ribose-amino acid models showed the similar UV–Vis absorbance and antioxidant activity with MRPs from xylose-amino acid models (Yu, Zhao, Hu, Zeng, & Bai, 2012). The various biological activities of MRPs from sugar (fructose and glucose) and 20 amino acid

model systems were investigated by vitro antioxidant, α -glucosidase inhibitory, antihypertensive, and antiproliferative activities measurements. The results have shown that MRPs derived from the fructose-amino acid model system showed higher biological activities than glucose-amino acid model system. Especially, sugar-tryptophan and -tyrosine MRPs exerted higher biological activities than the other MRPs (Hwang, Kim, Woo, Lee, & Jeong, 2011). Considering the good antioxidant and antibacterial activity of MRPs in the food industry, there has been an increasing interest in the utilisation of MRPs as preservative.

Xylan polysaccharides are the main hemicellulose components of secondary cell walls next to cellulose in wood and various other plants such as grasses, cereal, and herbs (Ebringerova & Heinze, 2000). It consists of a β -1,4-linked xylose backbone which may be substituted at the 2'-OH or 3'-OH with other molecules such as acetyl groups, 4-O-methyl glucuronyl groups, or arabinose (Dodd & Cann, 2009). Due to xylan-type polysaccharides source dependent diversity, branching, and varying chemical composition, their application potential has not been fully commercial development by contrast with other natural polysaccharides. During the latest decade, great efforts have been devoted to develop high-added products of xylan-type polysaccharides such as xylan based nanocomposite films (Peng, Ren, Zhong, & Sun, 2011), porous foams (Kohnke, Lin, Elder, Theliander, & Ragauskas, 2012), gels from cross-linked xylan (Peng, Zhong, Ren, & Sun, 2012) and

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surfactants (Wei, Shrestha, Tu, & Adhikari, 2011). Chitosan, the deacetylated derivative of chitin, is another nontoxic and biodegradable polymer widely existing in nature. Chitosan and its derivative are widely used in the field of biomedicine (Wang, Zhang, Hu, Yang, & Du, 2008), water treatment (Ngh, Ab Ghani, & Kamari, 2005), food science technology (No, Park, Lee, & Meyers, 2002), cosmetic and polymer science (Wang, Du, & Fan, 2005). In addition, a reactive amino group on the structure units of chitosan makes chitosan much easier to be modified by chemical reaction than cellulose (Zeng, He, Li, & Wang, 2012). Therefore, the chemical reactions between the hemiacetal hydroxyl groups on xylan chains and the amino groups on chitosan or chitosan derivatives make their MR possible. The objective of this current was to study antioxidant activity, antimicrobial activity and MR kinetics of MRPs derived from xylan with chitosan/chito oligomer/glucosamine hydrochloride/taurine model systems under similar experimental conditions. In this work, MRPs from glucosamine hydrochloride, xylan-glucosamine hydrochloride and xylan-aurine model systems were reported for the first time.

2. Experimental

2.1. Chemicals

The hemicellulose used was xylan isolated by alkaline extraction of corn cobs (poly β -D-xylopyranose [1 \rightarrow 4]) with a molecular weight of 3.24×10^3 Da detected by dynamic light scattering (DLS) and purchased from Shanghai Hanhong Ltd. (China). Chitosan was obtained from Zhejiang Yuhuan Ltd. (China) with a deacetylation degree of 80% and a molecular weight of 210 KDa. Taurine (C₂H₇NO₃S, Mr = 125.5 Da) was obtained from Qianjiang Yongan Pharmaceutical Co. Ltd. Glucosamine hydrochloride (C₆H₁₃NO₅·HCl, Mr = 215.63 Da) was supplied by Hubei Qianjiang Huashan Aquatic Food and Product Co. Ltd. Chito oligomer was obtained by enzyme hydrolysis of chitosan with immobilised neutral protease. The neutral protease derived from *Bacillus subtilis* 1.398 was a product of Ningxia XiaSheng Industry Co. Ltd. (China). The polymerisation degree of chito oligomer was between 2 and 5 studied by matrix-assisted laser desorption/ionisation time-of-flight mass spectrometry (MALDI-TOF-MS). 2,2-diphenyl-1-picrylhydrazyl (DPPH \cdot) was purchased from Sigma-Aldrich. The other chemicals used were of analytical grade obtained from Sinopharm Chemical Reagent Co. Ltd.

2.2. Methods

2.2.1. Preparation of Maillard reaction products (MRPs)

MRPs were synthesised according to the method described by Wu et al. (Wu, Du, Hu, Shi, & Zhang, 2013) with some modifications. Chito oligomer, glucosamine hydrochloride, taurine and xylan were dissolved in distilled water as 1:1 M ratio of xylan (0.1 M) and amino group (0.1 M). After adjusting the pH value to 9.0 using sodium hydroxide (1 M), the mixture was then transferred to a flask and refluxed in an oil bath at 100 °C for 4 h. The samples of the heated mixture were removed at 5, 15, 30, 60, 90, 120, 150, 180, 210, and 240 min, then immediately cooled in an ice-water bath for analysis. Chitosan could not be dissolved in distilled water. Thus, chitosan was prepared in 1% (v/v) glacial acetic acid with pH 4.0 at a concentration of 1% (w/v) and filtered to remove insoluble residue. The equimolar (1.0×10^{-3} M) amounts of xylan (4 g) then added with 75 mL of 5.6×10^{-6} M chitosan solution. MRPs derived from the xylan-chitosan model system were prepared in same way as described above. For comparison, xylan, taurine, glucosamine hydrochloride, chito oligomer and chitosan were respectively heated under the same experimental conditions as controls. All aqueous MRPs were prepared in triplicate.

2.2.2. Structure analysis of MRPs

MRPs of xylan with chitosan/chito oligomer/glucosamine hydrochloride/taurine model systems at reaction time 120 min were freeze-dried and grounded to powder. FT-IR spectra of taurine, glucosamine hydrochloride, chito oligomer, chitosan and their MRPs with xylan were recorded with KBr discs in the range of 4000–400 cm⁻¹ on Nicolet-170 SX spectrophotometer. X-ray diffraction was recorded by a Rigaku Kmax-r AX diffractometer with scanning scope of 5–40° scanning speed of 2°/min, using Cu K α radiation.

2.2.3. Spectrophotometric analysis

The UV absorbance and browning of the sample was measured according to the literature (Wu et al., 2013). The heated solutions were appropriately diluted with distilled water for spectrophotometric analysis and antioxidant activity. UV absorbance and browning intensity were measured at 294 nm and 420 nm, respectively. The fluorescence intensity was measured at an excitation wavelength of 343 nm and an emission wavelength of 415 nm on a Hitachi-F4500 fluorescence spectrophotometer.

2.2.4. Scavenging of free radicals

The DPPH radical scavenging activity of taurine, glucosamine hydrochloride, chito oligomer, chitosan and their MRPs with xylan heated for different time was estimated according to the methods described herein (Brand-Williams, Cuvelier, & Berset, 1995) with slightly modifications. An aliquot (0.1 mL) of the heated solutions was added to 2.0 mL of 0.1 mM DPPH \cdot in ethanol. The mixture was shaken vigorously and then kept at room temperature for 30 min under the dark for protection from light. The absorption was measured using Unico UV-2000 at 517 nm. The percentage of DPPH radical scavenging activity was calculated as follows:

$$[1 - (A_s - A_r)/A_0] \times 100,$$

where A_0 was the absorbance of 0.1 mM DPPH \cdot , A_s was the absorbance of 0.1 mM DPPH \cdot with the solutions, and A_r was the absorbance of ethanol with solutions. There were three replicate samples in each group for the measurements of free radical scavenging.

2.2.5. Measurement of reducing power

The reducing power of taurine, glucosamine hydrochloride, chito oligomer, chitosan and their MRPs with xylan heated for

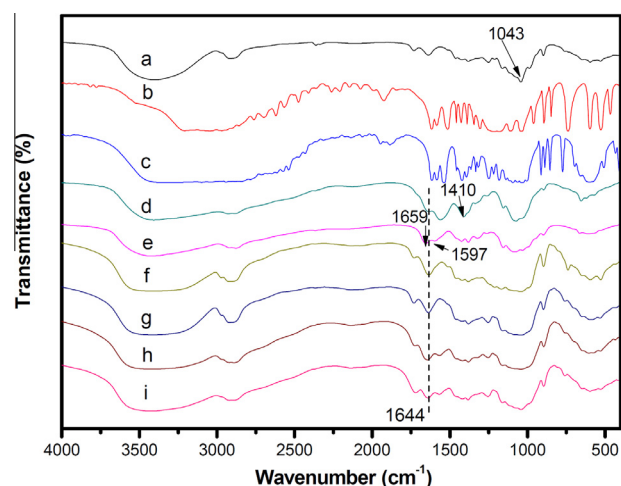


Fig. 1. FT-IR spectra of taurine, glucosamine hydrochloride, chito oligomer, chitosan and their MRPs with xylan at reaction time 120 min. (a) Xylan; (b) taurine; (c) glucosamine hydrochloride; (d) chito oligomer; (e) chitosan; (f) xylan-aurine; (g) xylan-chito oligomer; (h) xylan-chitosan; (i) xylan-chitosan.

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