Food Chemistry 243 (2018) 239–248

Contents lists available at ScienceDirect

Food Chemistry

journal homepage: www.elsevier.com/locate/foodchem

Impact of fluorescent lighting on the browning potential of model wine solutions containing organic acids and iron

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ARTICLE INFO

Chemical compounds studied in this article:
L-Tartaric acid (PubChem CID: 444305)
L-Malic acid (PubChem CID: 222656)
Succinic acid (PubChem CID: 1110)
Citric acid (PubChem CID: 311)
Sodium L-lactate (PubChem CID: 23664767)
Acetic acid (PubChem CID: 176)
Iron (PubChem CID: 23925)
(−)-Epicatechin (PubChem CID: 72276)
Ethanol (PubChem CID: 702)

Keywords:
Iron(III) carboxylate
Photochemistry
Oxidation
Carbonyl compound
Flavan-3-ol
Polymerization
Xanthylium cation ethyl ester

ABSTRACT

Model wine solutions containing organic acids, individually or combined, and iron(III), were exposed to light from fluorescent lamps or stored in darkness for four hours. (−)-Epicatechin was then added, and the solutions incubated in darkness for 10 days. Browning was monitored by UV–visible absorption spectrophotometry and UHPLC-DAD. The pre-irradiated solutions containing tartaric acid exhibited increased yellow/brown coloration compared to the dark controls mainly due to reaction of the tartaric acid photodegradation product glyoxylic acid with (−)-epicatechin to form xanthylium cation pigments. In these solutions, browning decreased as the concentrations of organic acids other than tartaric acid increased. Xanthylium cations were also detected in the pre-irradiated malic acid solution. However, in the malic acid, succinic acid, citric acid and lactic acid solutions, any coloration was mainly due to the production of dehydrodiepicatechin A, which was largely independent of prior light exposure, but strongly affected by the organic acid present.

1. Introduction

Browning of finished white wines is largely due to the polymerization of phenolic compounds, leading to the formation of products with an increased absorbance at wavelengths in the visible region (Li, Guo, & Wang, 2008; Singleton, 1987). Studies in white wines have provided evidence that the main phenolic compounds contributing to browning are the flavan-3-ols (+)-catechin and (−)-epicatechin, and their derivatives, all of which originate from the skins and seeds of grapes (Fernández-Zurbano, Ferreira, Escudero, & Cacho, 1998; Fernández-Zurbano et al., 1995; Simpson, 1982).

There are several pathways whereby flavan-3-ols can be converted to pigments in wine. Flavan-3-ols and other 1,2-dihydroxyphenolic compounds can bind iron(III), forming an unstable complex that can degrade into a semiquinone radical and iron(II) (Danilewicz, 2003). Semiquinone radicals can disproportionate or be oxidized to form *o*-quinones (Danilewicz, 2003). Flavan-3-ols have two nucleophilic positions (C6 and C8) that can attack *o*-quinones in a conjugate addition reaction. The nucleophilic addition of the flavan-3-ol C8 to the *o*-quinone results in the production of a colorless dimer, which can undergo subsequent reactions to form yellow pigments (Guyot, Cheynier, Souquet, & Moutounet, 1995; Guyot, Vercauteren, & Cheynier, 1996). For example, one pigment that can be generated in this manner is dehydrodiepicatechin A (Supplementary Fig. 1).

Alternatively, flavan-3-ols can undergo nucleophilic addition to aldehydes that may be present in wine, such as glyoxylic acid, a tartaric acid oxidation product (Fulcrand, Cheynier, Oszmianski, & Moutounet, 1997). The reaction of a flavan-3-ol with glyoxylic acid leads to the production of different colorless dimers (structural isomers), which undergo dehydration and oxidation to form yellow xanthylium cations (Es-Safi, Guernevé, Fulcrand, Cheynier, & Moutounet, 2000) (Supplementary Fig. 1). The colorless dimers also react with ethanol to

Abbreviations: DAD, diode array detection; LMCT, ligand-to-metal charge-transfer; LOD, limit of detection; LOQ, limit of quantification; MS, mass spectrometry; Q-TOF, Quadrupole-Time of Flight; UHPLC, ultra high performance liquid chromatography; UV, ultraviolet

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http://dx.doi.org/10.1016/j.foodchem.2017.09.093
Received 18 April 2017; Received in revised form 10 September 2017; Accepted 18 September 2017
Available online 19 September 2017

form ethyl esters, which give rise to the corresponding xanthylium cation pigments (Es-Safi et al., 2000) (Supplementary Fig. 1). Other aldehydes that have been shown to react with flavan-3-ols to generate the corresponding xanthylium cation pigments include furfural and 5-(hydroxymethyl)furfural (Es-Safi, Cheynier, & Moutounet, 2000) (Supplementary Fig. 2). In addition, certain aldehydes such as glyoxal (Es-Safi, Cheynier, & Moutounet, 2003) and L-xylosone (Barril, Clark, Prenzler, Karuso, & Scollary, 2009) (Supplementary Fig. 2) have been shown to react with flavan-3-ols to generate colorless intermediates that are ultimately converted to the same xanthylium cations derived from glyoxylic acid.

Previous studies in model wine solutions containing tartaric acid and (+)-catechin showed that the types of pigments produced (Oszmianski, Cheynier, & Moutounet, 1996), and their rates of formation (George, Clark, Prenzler, & Scollary, 2006), were dependent on the concentrations of the transition metal ions, iron and copper, which act as oxidation catalysts (Danilewicz, 2003). A study in model wine solutions containing iron(III), (+)-catechin and either tartaric acid or acetic acid demonstrated that the organic acid strongly influenced the oxidation of the flavan-3-ol and the production of pigments (Danilewicz, 2014). After addition of (+)-catechin, in the tartaric acid solution, iron(III) remained in the form of iron(III) tartrate. However in the acetic acid solution, the concentration of iron(III) acetate decreased and an iron(III) phenolate complex was formed and subsequently degraded (Danilewicz, 2014). It was proposed that tartrate anions interacted strongly with iron(III), and thus prevented iron(III) from interacting with (+)-catechin, whereas acetate anions interacted less strongly with iron(III), and this allowed iron(III) to oxidize (+)-catechin (Danilewicz, 2014).

White wine color development can be influenced by exposure to UV–visible light (Grant-Preece, Barril, Schmidtke, Scollary, & Clark, 2017). A study in Chardonnay wine supplemented with flavan-3-ols demonstrated that UV–visible light exposure accelerated browning (Dias, Smith, Ghiggino, & Scollary, 2012). It was also shown that xanthylium cations of the same type produced from glyoxylic acid and (+)-catechin were among the main pigments produced in a Chardonnay wine supplemented with (+)-catechin exposed to light under oxidizing conditions (Dias, Clark, Smith, Ghiggino, & Scollary, 2013). A study in a model wine solution containing tartaric acid, without added iron, found that aging the solution for six months with periodic exposure to sunlight increased its browning potential (Clark & Scollary, 2003). After addition of (+)-catechin and incubation in darkness, the aged solution exhibited increased yellow coloration compared to a freshly prepared solution (Clark & Scollary, 2003). Prior to (+)-catechin addition, glyoxylic acid was not detected in the aged solution or the freshly prepared solution. However, exposure of a freshly prepared solution to sunlight for three days induced the production of glyoxylic acid, and this was not observed in an equivalent solution stored in darkness (Clark & Scollary, 2003). Further investigations revealed that a model wine solution prepared in a similar manner contained iron at a low concentration (10 ± 5 μg/L), and that in the light-exposed solution, iron(III) tartrate was degraded via light-induced ligand-to-metal charge-transfer (LMCT), resulting in oxidation of the tartrate ligand and the production of various compounds including glyoxylic acid (Clark, Dias, Smith, Ghiggino, & Scollary, 2011; Clark, Prenzler, & Scollary, 2007). It is possible that glyoxylic acid was not observed in the solution aged for six months with periodic sunlight exposure because it was degraded under these conditions and/or its concentration was below the limit of detection (Clark & Scollary, 2003).

In our previous study, model wine solutions containing tartaric acid, malic acid, succinic acid, citric acid and lactic acid, individually or combined, and iron(III), were added to clear glass wine bottles, which were sealed and either exposed to light from fluorescent lamps or stored in darkness for four hours (Grant-Preece, Barril, Schmidtke, & Clark, 2017). Light exposure increased dissolved oxygen consumption and caused the degradation of the organic acids to a range of different carbonyl compounds that were not detected in the dark controls (Grant-Preece et al., 2017) (Supplementary Fig. 3). In the irradiated solutions in which dissolved oxygen was almost completely consumed, namely the tartaric acid solution, the 18 and 2.2 mmol/L lactic acid solutions and the 2.6 mmol/L citric acid solution, a substantial decrease in the concentration of iron(III) was observed (Grant-Preece et al., 2017). The aim of the current study was to determine the effect of exposure to light from fluorescent lamps on the browning potential of the model wine solutions. This was achieved by adding (−)-epicatechin to the pre-irradiated solutions and the dark controls and then incubating the solutions in darkness. Color development was monitored by UV–visible absorption spectrophotometry and UHPLC-DAD.

2. Materials and methods

2.1. General

Acetic acid (> 99.8%), anhydrous citric acid (≥99.5%), L-malic acid (≥99%) and succinic acid (≥99.5%) were obtained from Fluka (Switzerland). (−)-Epicatechin (96.3%), glyoxylic acid monohydrate (98%), iron(III) sulfate hydrate (97%), sodium L-lactate (~98%) and L-tartaric acid (99.5%) were obtained from Sigma-Aldrich (USA). Iron(II) sulfate heptahydrate (≥99.0%) was obtained from Biolab (Australia). All glass and plastic labware was soaked in 10% (v/v) nitric acid for at least 12 h and then rinsed thoroughly with water filtered through a Millipore Milli-Q water purification system, with a resistivity of 18.2 MΩ.cm. All solutions were prepared using Milli-Q water.

2.2. Model wine solutions

A solution of each organic acid at 18 mmol/L and additional solutions of succinic acid, citric acid and lactic acid at wine-like concentrations (i.e. 6.8, 2.6 and 2.2 mmol/L, respectively) were prepared in 12% (v/v) aqueous ethanol. Solutions containing all the organic acids at 18 mmol/L or the wine-like concentrations (18 mmol/L tartaric acid, 18 mmol/L malic acid, 6.8 mmol/L succinic acid, 2.6 mmol/L citric acid, 2.2 mmol/L lactic acid) were also prepared. The pH of the solutions was adjusted to 3.2 ± 0.1 by adding 1 mol/L sodium hydroxide or 0.5% (v/v) sulfuric acid. Samples (200 mL) of the solutions were added to 250 mL Schott bottles. An iron(III) stock solution (0.5050 g/L) was prepared by dissolving iron(III) sulfate hydrate in water acidified to pH 3.1 ± 0.1 with sulfuric acid. Immediately before irradiation/storage in darkness, the 200 mL samples were aerated by rapid stirring for 1 min using a magnetic stirrer. An aliquot (2 mL) of the iron(III) stock solution was then added to each aerated sample to achieve an iron(III) concentration of 5.0 mg/L (0.090 mmol/L). These solutions were exposed to light or stored in darkness as outlined below. In addition, an 18 mmol/L acetic acid solution was prepared in the same manner as the other model wine solutions and used to prepare solutions containing either glyoxylic acid (0.4 mmol/L), iron(II) or iron (III) (5.0 mg/L, 0.090 mmol/L). These solutions were treated as described in Section 2.4.

2.3. Irradiation and storage in darkness

Solutions were irradiated or stored in darkness as described previously (Grant-Preece et al., 2017). Briefly, after aeration and addition of iron(III), the solutions were transferred into 187 mL clear glass wine bottles, which were sealed with screw caps leaving no headspace, and the dark control bottles were covered in aluminum foil. All bottles were stored 5 cm from two Philips Alto TL-D 36W/865 cool daylight fluorescent lamps at 20.7 ± 0.5 °C for four hours. At this position, the photosynthetic photon flux density (400–700 nm) measured using a LI-COR Biosciences LI-250A light meter and quantum sensor was 150 μmol/m^2/s. The transmission spectrum of the bottle glass indicated that wavelengths below 300 nm were absorbed, with transmittance