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Comparative study of the effects of antioxidants on furan formation during thermal processing in model systems

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ABSTRACT

Furan, a possible carcinogen, is commonly produced by thermal processing in a number of heated foods. In this study, the effects of several natural and synthetic antioxidants on thermally induced furan formation in ascorbic acid, linoleic and linolenic acid model systems were investigated, and results demonstrated that not all of the antioxidants tested showed mitigating effects on furan formation. For the ascorbic acid model system, chlorogenic acid was found to be the most efficient antioxidant in suppressing furan formation. For the linoleic and linolenic acid model systems, the most significant reduction (92% in the former and 80% in the latter) was observed in the case of model systems with butylated hydroxytoluene. In addition, the effects of antioxidants on the kinetics of furan formation were investigated by adding chlorogenic acid into the ascorbic acid model system and sesamol into both linoleic and linolenic acid model system, and the results showed that the mitigating effects of antioxidants might decline with increasing heating time. To describe the kinetics of furan formation from these model systems, power function models were established by non-linear regression and the results indicated that the proposed models could successfully fit the experimental data ($R^2 > 0.988$).

1. Introduction

Furan, a colorless aromatic heterocyclic compound with high volatility, had been found to exist in various food systems for a considerable period of time (Maga, 1979). Nevertheless, it had not come to so wide public attention until it was classified as a possible human carcinogen by several government agencies including the International Agency for Research on Cancer (IARC) of the World Health Organization (WHO) (IARC, 1995) and the US Department of Health and Human Service (NTP, 1993). It has also been considered as a carcinogenic compound that might act via a DNA-reactive genotoxic metabolite by the risk assessment of the Joint FAO/WHO Expert Committee on Food Additives (JECFA) (JECFA, 2011). Thenceforward, the occurrence, toxicity, exposure level and formation mechanism of furan were explored by many scholars and government agencies (Crews & Castle, 2007; EFSA, 2011; Seok et al., 2015; Shen et al., 2015).

The US Food and Drug Administration (FDA) reported that relatively high levels of furan could be found in heat-processed foods especially in jarred and canned foods (FDA, 2009). In our previous study, 11 categories of heat-processed foods (including 133 kinds of food samples) in China were analyzed, and levels of furan in soy sauce samples (128.8 ng/g) had been found to be higher than those in coffee samples (60.6 ng/g, Nie, Huang, Zhang, et al., 2013).

Considering the extensive occurrence of furan in thermal-processed foods and its possible carcinogenic risk to human, it is necessary to take reasonable measure to mitigate the presence of furan in foods. On the basis of available literatures, many pathways of the formation of furan are possible during food processing (Crews & Castle, 2007; Fan, 2005; Locas & Yaylayan, 2004; Mariotti et al., 2012), and several nutritional ingredients such as carbohydrates (Fan, 2005; Limacher, Kerler, Davidek, Schmalzried, & Blank, 2008; Mariotti et al., 2012), amino acids (Locas & Yaylayan, 2004), PUFAs (Becalski & Seaman, 2005), ascorbic acid (Fan, 2005; Limacher, Kerler, Conde-Petit, & Blank, 2007; Locas & Yaylayan, 2004) and carotenoids (Crews & Castle, 2007) were identified to be potential precursors of furan. Possible formation mechanisms from those precursors to furan during food thermal-treatment process

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were elucidated: 1) thermal degradation of carbohydrates or reacting with amino acid through Maillard reaction (Fan, 2005), 2) thermal degradation of specific amino acids (Locas & Yaylayan, 2004), 3) thermal oxidation of PUFAs, ascorbic acid and carotenoids (Locas & Yaylayan, 2004; Maga, 1979) (Fig. 1). In food, however, these reaction pathways are closely related, for example, lipid oxidation products or intermediates can influence the course of the Maillard reaction and vice versa, and furan could result from both reaction pathways (Adams, Bouckaert, Van Lancker, De Meulenaer, & De Kimpe, 2011).

Presently, some strategies have been used to mitigate the furan concentrations in heated foods, such as decreasing precursor concentration, high pressure-high temperature treatment (Palmers, Grauwet, Kebede, Hendrickx, & Van Loey, 2014), ionizing radiation (Fan & Mastovska, 2006), and post-process vacuum treatment (Anese, Manzocco, Calligaris, & Nicoli, 2013). However, because of the close relationship between furan formation and the generation of desired flavors of heated foods, more research is needed on the reduction of furan during food processing without loss of flavors (Limacher et al., 2008). Furthermore, microbial safety standards may also limit the implementation of some mitigation strategies such as lowering heating times and temperatures.

Antioxidants are naturally present in foods, such as polyphenols, flavonoids and tocopherols, in addition, synthetic antioxidants, such as butylated hydroxytoluene (BHT) and propyl gallate (PG), are widely used in food industry to prolong the shelf life of foods. Antioxidants can effectively scavenge free radicals and chelate transition metals, thus slowing-down progressive autoxidative damage and production of hazardous substance (Brewer, 2011; Tsuzuki, 2011). Previous model studies have suggested that linoleic/linolenic acid had the highest potential to produce furan upon thermal treatment, followed by ascorbic acid (Becalski & Seaman, 2005). In this study, the three typical model systems (ascorbic acid, linoleic/linolenic acid) were developed to evaluate the effects of antioxidants on furan formation during thermal processing.

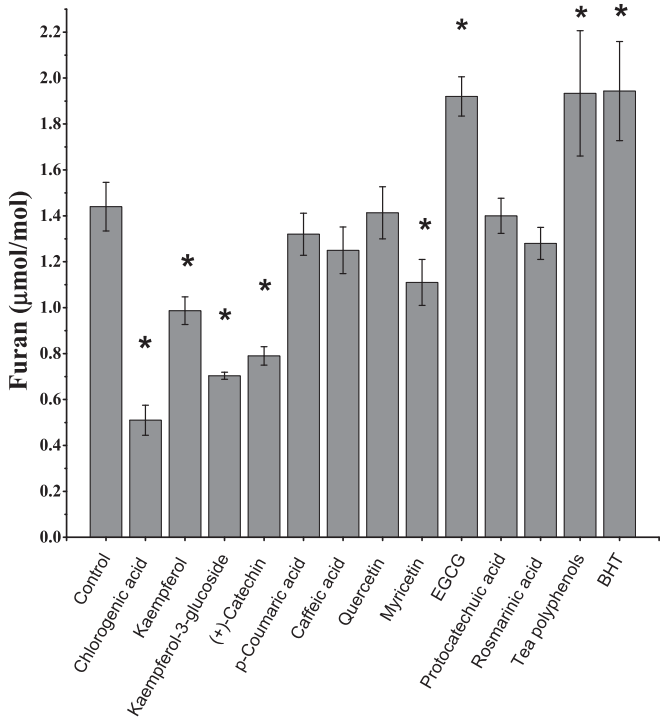

Fig. 1. Effects of antioxidants on furan formation in the ascorbic acid model system. Data represent means ± SD of three independent experiments, bars with * are significantly different from the control group ($p < 0.05$).

2. Materials and methods

2.1. Chemical and reagents

L-ascorbic acid (AA, ≥99%), linolenic acid (≥99%), linoleic acid (≥99%), furan (≥99%), d_4-furan (≥98%), chlorogenic acid (≥95%), kaempferol (≥97%), kaempferol-3-glucoside (≥97%), (+)-catechin (≥95%), *p*-coumaric acid (≥98%), caffeic acid (≥98%), quercetin (≥95%), myricetin (≥98%), epigallocatechin gallate (EGCG, ≥98%), protocatechuic acid (≥98%), rosmarinic acid (≥95%), sesamol (≥98%), α-tocopherol (≥98%), butylated hydroxytoluene (BHT, ≥98%), propyl gallate (PG, ≥98%) and ethanol (HPLC-grade) were purchased from Sigma-Aldrich (St. Louis, MO, USA). Disodium hydrogen phosphate ($Na_2HPO_4{\cdot}12H_2O$), sodium dihydrogen phosphate ($NaH_2PO_4{\cdot}2H_2O$) and tea polyphenols (≥98%) were obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China), and other chemicals were of analytical grade. According to the quality report provided by the manufacturer, tea polyphenols contain 70% catechins and EGCG is the major catechin and accounts for 40% of the tea polyphenols. Ultrapure water was obtained from the Milli-Q gradient A10 water purification system (Millipore Corp., Bedford, MA, USA).

2.2. Preparation of model system solutions

The effects of pH on furan formation in the ascorbic acid, linoleic and linolenic acid model systems have been investigated in our previous study (Nie, Huang, Hu, et al., 2013; Shen et al., 2015), and results have shown that furan formation from these precursors was significantly influenced by pH. So far, it is unclear why pH affected furan formation in model systems, and we speculate that acid and alkaline may respectively donate and accept protons and act as catalyst or inhibitor for the formation of furan. In this study, we investigated the effects of antioxidants on furan formation at neutral pH, and $Na_2HPO_4{\cdot}12H_2O$ (50 g/L, pH 8.7) and $NaH_2PO_4{\cdot}2H_2O$ (50 g/L, pH 4.6) solutions were used to prepare the sodium phosphate buffered solution (pH 7.0). The further study on the effects of antioxidants on furan formation in acidic and alkalic conditions is under way in our laboratory.

Preparation of ascorbic acid solution: 3.52 g of ascorbic acid (0.02 mol) was weighed into a 100 mL volumetric flask, and then the sodium phosphate buffered solution at pH 7.0 was added to obtain the final volume of 100 mL.

Preparation of linoleic and linolenic acid solutions: 180 μL (0.6 mmol) of linoleic acid was added into a 100 mL volumetric flask, and then the sodium phosphate buffered solution at pH 7.0 was added to obtain the final volume of 100 mL. The linolenic acid model system was prepared at the same way.

2.3. Effects of antioxidants on furan formation

The stock solutions of antioxidants were prepared in absolute ethanol at a concentration of 10 mg/mL. Besides, antioxidant solutions at a concentration sequence of 1.0, 10^{-1}, 10^{-2}, 10^{-3} and 10^{-4} mg/mL were obtained via diluting the stock solution with sodium phosphate buffered solution at pH 7.0, gradually. These solutions were stored at 4 °C in a refrigerator for further use.

Aliquots of the model system solutions (5 mL) were transferred into 20 mL headspace vials, and 50 μL antioxidant solutions at different concentrations were added respectively into headspace vials as the tested group. Model system solutions with 50 μL sodium phosphate buffer were used as control group (no antioxidant). For the ascorbic acid model system spiked with chlorogenic