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Effects of dough formula and baking conditions on acrylamide and hydroxymethylfurfural formation in cookies

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Abstract

The effects of dough formula and baking conditions on the formations of acrylamide and hydroxymethylfurfural (HMF) were studied in a cookie model system. Increasing the sugar concentration in the dough formula increased acrylamide formation during baking at 205 °C for 11 min. The effect of sugar on acrylamide formation was more pronounced for glucose than for sucrose, expectedly. Addition of citric acid into dough formula comprising sucrose increased the susceptibility of acrylamide formation, while it decreased acrylamide formation in the dough formula comprising glucose. Decreasing the pH of dough formula increased the tendency to surface browning and the formation of hydroxymethylfurfural in cookies during baking. The results suggest that a cookie with acceptable texture and colour, but having less than 150 ng/g of acrylamide, can be manufactured by lowering the baking temperature and avoiding reducing sugars in the recipe.

Keywords: Acrylamide; HMF; Cookie; Baking; Browning; Maillard reaction

1. Introduction

Acrylamide formation was found to occur during a thermal process, by Maillard reaction, of reducing sugars with asparagine at temperatures above 120 °C (Friedman, 2003; Mottram, Wedzicha, & Dodson, 2002; Stadler et al., 2002; Yaylayan, Wnorowski, & Perez-Locas, 2003). The formation of key intermediates responsible for acrylamide formation is determined by the concentrations and types of sugars and amino acids present. These intermediates also react with other amino acids to form brown-coloured products and flavour compounds. Meanwhile, some chemical indicators, such as hydroxymethylfurfural (HMF), for assessing the quality of thermally processed foods, also form (Berg & van Boekel, 1994; Gökmen & Acar, 1999; Gökmen & Şenyuva, 2006a; Morales, Romero, & Jiménez-Pérez, 1997). Thus, the formation of acrylamide from asparagine is one of a number of competing processes. The yield of acrylamide is sensitive to both the composition of food and the conditions which are known to promote the Maillard reaction.

The major concern of food producers is to reduce the acrylamide content of foods (concerning the health impact), but to keep quality parameters unaffected by the adjusted processing conditions (concerning the economic impact). Some results clearly indicate that additions of acids, amino acids, or proteins seem to reduce acrylamide formation (Jung, Choi, & Ju, 2003; Kita, Bråthen, Knutsen, & Wicklund, 2005; Pedrechi, Moyano, Kaack, & Granby, 2005; Rydberg et al., 2003). However, it is unclear whether these pretreatments impart undesirable changes to the finished product, because the desired consequences of the Maillard reaction share intermediates with acrylamide formation.

The aim of this work is to understand the effects of dough formula (type and concentrations of sugars and pH) and the baking process (temperature and time) on acrylamide and HMF formations in cookies.

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2. Materials and methods

2.1. Materials

Acrylamide and HMF were purchased from Sigma (Diesenhofen, Germany). A solution of $^{13}C_3$-acrylamide (1 mg/ml) in methanol was purchased from Cambridge Isotope Laboratories (Andover, MA, USA). Sodium thiosulfate pentahydrate, hydrobromic acid, hydrochloric acid, potassium bromide (all AnalaR grade) and bromine (99.8%) were purchased from Merck (Darmstadt, Germany). Methanol, acetonitrile, glacial acetic acid, potassium hexacyanoferrate and zinc sulfate (all AnalaR grade) were purchased from Merck (Darmstadt, Germany). Ultra pure water was used throughout the experiments (MilliQ system, Millipore, Bedford, MA, USA).

Oasis HLB (1 ml, 30 mg) solid phase extraction cartridges and the Atlantis dC_{18} analytical column (4.6 × 300 mm 5 μm) were supplied by Waters (Milford, MA, USA). The capillary column (InnoWax, 30 m × 0.25 mm i.d., 0.15 μm film thickness) was purchased from Agilent Technologies (Palo Alto, CA, USA). Macro-spin PVDF centrifuge filters (0.45 μm) were purchased from Alltech (Deerfield, IL, USA).

Carrez I solution was prepared by dissolving 15 g of potassium hexacyanoferrate in 100 ml of water, and Carrez II solution by dissolving 30 g of zinc sulfate in 100 ml of water.

2.2. Preparation of cookies

Flour and shortening were supplied by local producers, and other ingredients were purchased from local supermarkets. The model cookies were prepared according to a recipe described in AACC (American Association of Cereal Chemists) Method 10-54, with some modifications, in order to study the effects of type and concentrations of sugars, pH and baking conditions on acrylamide and HMF formation. Recipes are listed below.

Recipe 1: 80 g of wheat flour, 32 g of shortening, 0.8 g of non-fat dry milk, 1 g of salt, 0.8 g of sodium bicarbonate, 0.4 g of ammonium bicarbonate, 17.6 ml of deionized water and sucrose (10, 15, 20, 25, 30 or 35 g), respectively.

Recipe 2: 80 g of wheat flour, 32 g of shortening, 0.8 g of non-fat dry milk, 1 g of salt, 0.8 g of sodium bicarbonate, 0.4 g of ammonium bicarbonate, 17.6 ml of deionized water, 10 g of sucrose and glucose (0, 5, 10, 15, 20 or 25 g).

Recipe 3: 80 g of wheat flour, 32 g of shortening, 0.8 g of non-fat dry milk, 1 g of salt, 0.8 g of sodium bicarbonate, 0.4 g of ammonium bicarbonate, 17.6 ml of deionized water, 35 g of sucrose and citric acid (3, 1 or 0 g, corresponding to dough pH of 3.28, 4.37 and 7.40, respectively).

Recipe 4: 80 g of wheat flour, 32 g of shortening, 0.8 g of non-fat dry milk, 1 g of salt, 0.8 g of sodium bicarbonate, 0.4 g of ammonium bicarbonate, 17.6 ml of deionized water, 10 g of sucrose, 25 g of glucose and citric acid (3, 1 or 0 g, corresponding to dough pH of 3.28, 4.37 and 7.40, respectively).

Recipe 5: 80 g of wheat flour, 32 g of shortening, 0.8 g of non-fat dry milk, 1 g of salt, 0.8 g of sodium bicarbonate, 0.4 g of ammonium bicarbonate, 17.6 ml of deionized water and 35 g of sucrose.

Recipe 6: 80 g of wheat flour, 32 g of shortening, 0.8 g of non-fat dry milk, 1 g of salt, 0.8 g of sodium bicarbonate, 0.4 g of ammonium bicarbonate, 17.6 ml of deionized water, 10 g of sucrose and 25 g of glucose.

The ingredients were thoroughly mixed according to the recipe. Dough was rolled out to disks, after which trays of these disks were then baked in the oven. Four dough disks were produced from each recipe. Dough disks of Recipe 1–4 were baked at 205 °C for 11 min. Dough disks of Recipes 5 and 6 were baked at different temperatures and times (5, 10, 15 and 20 min at 160 and 180 °C; 5, 10 and 15 min at 200 and 210 °C and 5, 8 and 10 min at 230 °C).

2.3. Measurement of acrylamide

The cookies were ground, and 2 g were taken for analysis. Acrylamide was analyzed as the dibromo derivative by gas chromatography–mass spectrometry (GC–MS), using the method of Castle, Campos, and Gilbert (1991) with some modifications. The extracting medium was methanol (20 ml), rather than water, because addition of water to the ground cookies resulted in a thick slurry, rendering extraction difficult (Gökmen, Şenyuva, Acar, & Sarıoğlu, 2005). $^{13}C_3$-acrylamide (500 ng) was added to the extract as the internal standard, along with 15 ml of brominating reagent. The bromination was allowed to proceed overnight at room temperature.

The brominated extract (1 μl) was injected onto an Agilent 5973 GC–MS system (Agilent Technologies, Palo Alto, CA) in splitless mode at 200 °C. Helium carrier gas flow rate was maintained at 1 ml/min. An InnoWax capillary column was used. The oven temperature was 80 °C, rising at 10 °C/min to 250 °C and followed by holding for 10 min. The transfer line was held at 250 °C and the ion source at 180 °C. Electron impact mass spectra were obtained at 70 eV. The mass spectrometer was operated in selected ion monitoring mode. Four ions were used to characterize brominated $^{13}C_3$-acrylamide (m/z 108, 110, 153, and 155), and another four ions were used to characterize brominated acrylamide (m/z 106, 108, 150, and 152). The ion m/z 152 was used to quantify brominated acrylamide.

Stock solution of acrylamide, at a concentration of 1.0 mg/ml, was prepared in methanol. Working standards were prepared daily by diluting the stock solution to concentrations of 0.01, 0.02, 0.05, 0.10, 0.25, 0.50 and 1.0 μg/ml with methanol. They were brominated prior to GC–MS analysis using the procedure described above. The limit of detection and the limit of quantitation for acrylamide were 15 and 50 ng/g in cookies, respectively. Signal response was linear over a concentration range between 50 and 1000 ng/g of acrylamide. The coefficient of variation was 10% or lower for three repetitive measurements of acrylamide in cookies.