



Formation and reduction of 5-hydroxymethylfurfural at frying temperature in model system as a function of amino acid and sugar composition



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ABSTRACT

5-Hydroxymethylfurfural (HMF) is formed during heat treatment of carbohydrate-containing foods, especially in a deep-fat frying process. This study aimed to investigate the effect of amino acids on the formation and reduction of HMF from glucose, fructose and sucrose at frying temperature in model systems containing binary mixtures of an amino acid and a sugar in equal concentrations (0.3 M). The results revealed that the formation of HMF from sugars accelerated in the presence of acidic amino acids (i.e. glutamic and aspartic acids). Conversely, the presence of basic amino acids (i.e. lysine, arginine and histidine) led to reduced concentrations of HMF to non-detectable levels in model systems. The results showed that both pH and heating time significantly affected the formation of HMF from fructose in the presence of glutamic acid. In this regard, a higher amount of HMF was formed at lower pH.

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1. Introduction

5-Hydroxymethylfurfural (HMF) forms during thermal treatment of carbohydrate-containing foods in processes, such as roasting (Durmaz & Gökmen, 2011), baking (Zhang et al., 2012) and frying (Göncüoğlu & Gökmen, 2013). HMF is formed from dehydration of hexoses or other sugars (i.e. sucrose) that produce hexoses after hydrolysis (Perez Locas & Yaylayan, 2008). Fig. 1 displays a simple explanation of the formation of HMF through Maillard reaction and acid-catalysed dehydration. The reactive fructofuranosyl cation is produced through hydrolysis of sucrose. It can directly convert into HMF in dry conditions (Perez Locas & Yaylayan, 2008) or convert into fructose in the presence of water or contribute in formation of HMF through the reaction with a free amino group during Maillard reaction. Fructose and glucose are the main precursors of HMF. They can react with amino acids in the early stage of the Maillard reaction and form the corresponding N-substituted amine (Hodge, 1953). After the formation of Amadori rearrangement products, the degradation pathway is pH-dependent. HMF forms from dehydration of sugar through

1,2-enolisation at pH values lower than 7 (Martins, Jongen, & Van Boekel, 2000). Moreover, the acid-catalysed dehydration of sugars through 1,2-enolization also leads to formation of HMF.

A wide range of HMF concentrations has been reported in food products (Capuano & Fogliano, 2011; Petisca, Henriques, Pérez-Palacios, Pinho, & Ferreira, 2014; Pérez-Palacios, Petisca, Melo, & Ferreira, 2013). The major concern about the toxicological effect of HMF has been related to its bioconversion into 5-sulfoxymethylfurfural (SMF) through the sulfonation of its allylic hydroxyl functional group. This conversion is catalysed by sulfotransferases (SULTs) (Capuano & Fogliano, 2011; Surh & Tannenbaum, 1994). SMF can react with DNA and other macromolecules, thereby resulting in toxic and mutagenic effects (Glatt, Schneider, Murkovic, Monien, & Meinel, 2012). European Food Safety Authority (EFSA) confirmed the mutagenicity of SMF (EFSA, 2005). Recently, SMF has been detected in the blood of FVB/N mice after HMF intravenous administration (Monien, Frank, Seidel, & Glatt, 2009).

During deep-fat frying food is immersed in hot oil at 150–190 °C (Choe & Min, 2007). As a result, a number of changes occur in the food due to Maillard reaction, starch gelatinisation, sugar dehydration and protein denaturation. The presence of HMF affects the safety and health of fried foods as well as frying oil due to its migration into the frying medium (Göncüoğlu & Gökmen, 2013).

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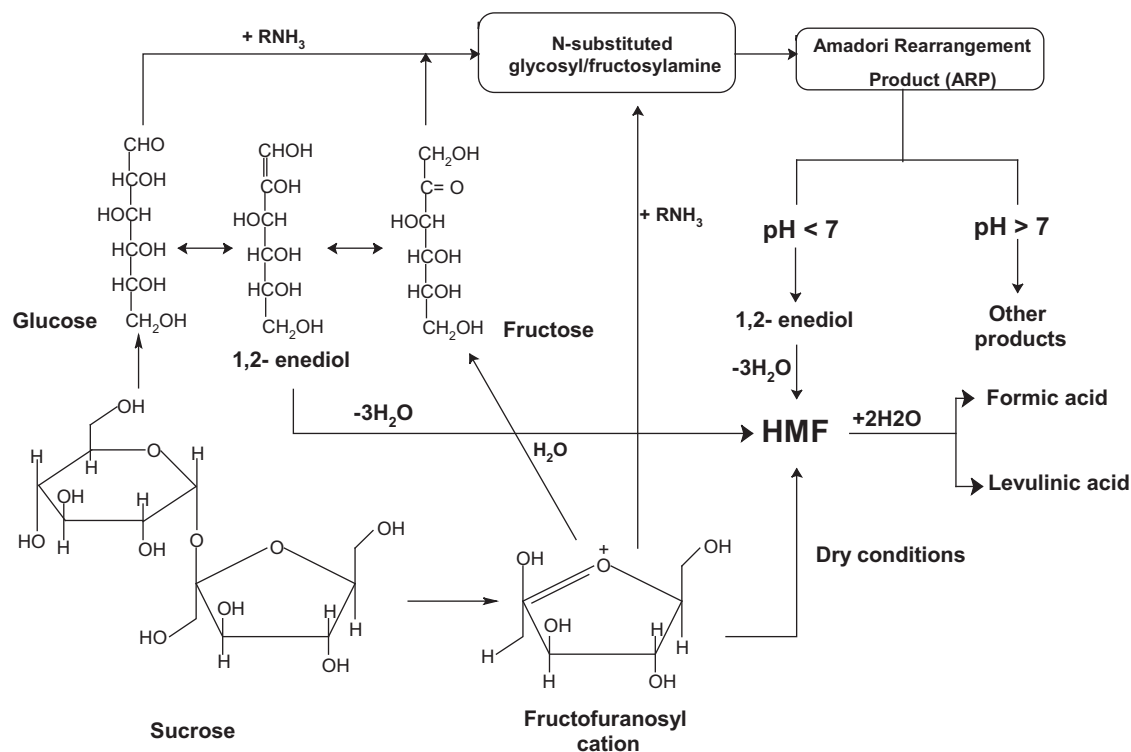


Fig. 1. Schematic representation of HMF formation through Maillard reaction and acid-catalysed dehydration (based on Perez Locas & Yaylayan, 2008 and Hodge, 1953).

HMF can also be formed from sugars that release hexoses after hydrolysis (Kuster, 1990). In this regard, the hydrolysis of these sugars prior to conversion into HMF is one of the most critical factors. This hydrolysis can be affected by pH and temperature (Yaylayan, Ismail, & Mandeville, 1993). Thus, the accumulation of HMF in the food system depends on the type of sugar, pH and thermal processing conditions (Janowski, Glaab, Samimi, Schlatter, & Eisenbrand, 2000). Amino acids contribute to Maillard reaction, but their role in the formation of HMF at frying temperatures has not been identified yet. This study aimed to investigate for the first time the effect of amino acids, sugar composition, pH and heating time on the formation of HMF at frying temperatures in a model system. A clear understanding about the role of amino acids, sugars and frying temperature helps to reduce HMF content in fried food products.

2. Material and methods

2.1. Chemicals and standards

In this study, HMF standard was purchased from Across Organics (Geel, Belgium). Sugars and amino acids, including fructose, glucose, sucrose, alanine, arginine, aspartic acid, cystine, glutamic acid, glycine, histidine, lysine, methionine, proline, serine, tryptophan, tyrosine, asparagine and valine were bought from Sigma Chemicals Co. (St Louis, MO). The purity of all applied amino acids and sugars was above 98%. Methanol (HPLC grade) was obtained from Merck (Darmstadt, Germany). Other solvents and chemicals were chemical grade. Refined, bleached and deodorised (RBD) palm olein was purchased from a local supermarket (Selangor, Malaysia).

2.2. Preparation of various model systems

In the present study, 3 types of model systems were prepared to investigate the effects of various amino acids, pH and heating time

on the formation of HMF. A citrate buffer solution (0.05 M) was prepared by dissolving citric acid monohydrate ($\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$) in deionised water (Lara, del Olmo-Iruela, & García-Campaña, 2013). Subsequently, its initial pH (<2) was adjusted to 4, 5, 6, 7, 8.30 and 10 using sodium hydroxide solution (1 M). To prepare the control samples, a portion of 1.5 mL of the mixture containing 0.3 M of either fructose, glucose or sucrose, citric buffer solution (0.5 mL, pH 6) and palm olein (1 mL) was transferred into 25-mL heat-resistant glass tubes. The binary mixtures of sugars and each target amino acid (0.3 M) and citric buffer solution (0.5 mL, pH 6) and palm olein (1 mL) were prepared in 25-mL heat-resistant glass tubes. To evaluate the effect of pH on the formation of HMF, the binary mixture of fructose and glutamic acid (0.3 M each) was mixed with palm olein (1 mL) and 0.5 mL citrate buffer at different pH values (i.e. 4, 5, 6, 7, 8.30 and 10) in 25-mL heat-resistant glass tubes.

The impact of heating time on the formation of HMF was investigated using model systems containing binary mixtures of fructose and glutamic acids (0.3 M each) in palm olein (1 mL) and 0.5 mL citrate buffer (pH 6). After preparation of various model systems, the tubes were tightly closed with screw caps, and vigorously mixed prior to heating. Then, the tubes were immersed in a well-stirred oil bath containing palm olein (1.5 L) to a depth where the surface of the sample was at least 5 cm below the surface of the oil. The aim was to minimise temperature gradients within the sample. The reaction mixture was heated at $175 (\pm 5)^\circ\text{C}$ for 10 min. The model systems were heated for 4, 7, 10, 13, 16, 19, 22 and 25 min, in order to evaluate the effect of heating time on the formation of HMF. After heating the mixtures were immediately cooled on ice. The experiments were conducted in duplicate.

2.3. Analysis of HMF

Analysis of HMF in mixtures was performed according to a validated method (Ariffin, Ghazali, & Kavousi, 2014). The methanolic extract (20 μL) was injected into a Waters Alliance HPLC system

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