

Industrially Applicable Strategies for Mitigating Acrylamide, Furan, and 5-Hydroxymethylfurfural in Food

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ABSTRACT: Acrylamide and furanic compounds, such as furan and 5-hydroxymethylfurfural (HMF), are formed in a variety of heat-treated commercial foods. They are known to be toxic and, according to the International Agency for Research on Cancer, “possibly carcinogenic to humans”. As this gave rise to concern, research was carried out to study their metabolism and toxicity and elucidate the mechanistic pathways of formation. So far, many papers have been published on acrylamide, furan, and HMF occurrence, precursors and mechanisms of formation, toxicity, and detection methods, as well as on possible routes to reduce their levels in food and, thus, consumer intake. This paper reviews the interventions suggested in the literature to mitigate the presence of acrylamide and furanic compounds in food. In particular, some technological measures potentially exploitable at the industrial level are discussed extensively. These are (i) preventive strategies based on the use of asparaginase and thermal input reduction (i.e., low temperature–long time dehydration; dielectric heating) and (ii) removal intervention by means of vacuum treatment, aimed to remove the already formed acrylamide and furanic compounds from the finished product.

KEYWORDS: *heat-induced food toxicants, mitigation strategies, asparaginase, thermal treatment, vacuum technology*

■ INTRODUCTION

Cooking of foods has numerous advantages including destruction of microorganisms, elimination of heat-sensitive toxins, increase in nutrient bioavailability, and development of desired color, flavor, and texture.¹ Heating may be also responsible for the formation of toxicants, among which are acrylamide, furan, 5-hydroxymethylfurfural (HMF), heterocyclic amines, and polycyclic aromatic hydrocarbons. Over the past 10 years, great attention has been directed toward the possible undesired effects of acrylamide, furan, and HMF. Indeed, many papers have been published on the occurrence, precursors, and mechanisms of formation, toxicity, and detection methods of these important heat-induced food toxicants.^{2–12}

Acrylamide, furan, and HMF are generated at high temperatures, the Maillard reaction being the main route of formation. These molecules, however, may present different features. Indeed, acrylamide, which is formed from the reaction between the carbonyl group of sugars and asparagine, is an undesired byproduct of the Maillard reaction that can be found in a variety of low-moisture heated foods, mainly represented by potato derivatives, bakery products, and roasted coffee.¹¹ Furan and HMF are generated by different reactions following multiple routes and involving different precursors and intermediates. Moreover, they can be found in fairly high amounts in almost all heated foods, including jarred foods, cereal derivatives, fruit juices, dried foods, honey, milk, and coffee.^{13,14} It is noteworthy that furan and related compounds are being considered of primary economic importance to the flavor industry as they can significantly contribute to the sensory properties of heated foods.² Table 1 shows the chemical structures and some chemical and physical properties of acrylamide, furan, and HMF.

As acrylamide, furan, and HMF are supposed to have negative consequences to human health,^{16–18} many efforts have been carried out in recent years to find possible strategies able to mitigate their presence in food and thus keep their level as low as

reasonably achievable. A great number of research results on strategies aimed to mitigate acrylamide levels in food can be found in the latest 10 years literature. Many relevant reviews on acrylamide mitigation interventions have been published.^{19–24} Moreover, the published results are summarized together with unpublished information provided by some manufacturers in the so-called acrylamide Toolbox, developed by Food Drink Europe.²⁵ Specifically, the acrylamide Toolbox contains information on potential interventions to be acquired by industry operators to reduce acrylamide exposure. Contrarily to acrylamide, scarce information is available in the literature about the possible routes to mitigate furan and HMF levels in food.⁷ In most cases, information on ways to reduce furan and HMF concentration can be actually drawn from the results of researches dealing with the influence of different composition and process variables on the formation of these two molecules.

In general terms, the ways of mitigating the levels of acrylamide, furan, and HMF in food can be regarded as two conceptually different technological approaches, which are preventive and removal strategies.²⁶ The preventive strategies are aimed to minimize acrylamide, furan, and/or HMF formation during the heating process; the removal interventions are aimed to remove or decompose the already formed molecules in the finished product. Relevant technological strategies of mitigation, classified as preventive or removal interventions, are summarized in Table 2. They can be applied in one or more steps of the food process, as pretreatments, formulation, and process and postprocess interventions. It is noteworthy that the preventive strategies lead to the creation of less favorable reaction

Special Issue: ISMR11 - 100 Years of the Maillard Reaction

Received: November 28, 2012

Revised: April 29, 2013

Accepted: April 29, 2013

Published: April 29, 2013

Table 1. Chemical Structures and Some Chemical and Physical Properties of Acrylamide, Furan, and 5-Hydroxymethylfurfural (HMF) (Adapted from Budavari et al.¹⁵)

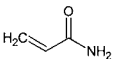

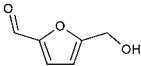
Molecule	Chemical structure	Molecular weight (g/mol)	Boiling point (°C) at 101.3 kPa	Density (g/mL) at 25 °C	Solubility
Acrylamide		71.08	231.7	0.961	Freely soluble in water
Furan		68.07	32.0	1.098	Insoluble in water; freely soluble in alcohol and ether
HMF		126.11	291.5	1.243	Freely soluble in water, methanol, ethanol, acetone, ethyl acetate; soluble in ether, benzene, chloroform

Table 2. Preventive and Removal Strategies of Acrylamide, Furan, and HMF Mitigation in Food, Mechanisms of Action, and Relevant References

process step	type of intervention	mechanism of action	food	toxicant	refs
Mitigation Strategy: Prevention					
pretreatment	dipping in additive solutions	decrease of formation rate	potato derivatives	acrylamide	27, 28
	thermal treatment (blanching)	leaching of precursors	potato derivatives	acrylamide	29, 30
	yeast or lactic acid bacteria fermentation	precursor consumption decrease of formation rate	potato- and cereal-based products	acrylamide	31–36
	use of asparaginase	precursor (asparagine) decomposition	potato- and cereal-based products	acrylamide	37–41
formulation	use of inhibiting or competing ingredients	decrease of precursor concentration decrease of formation rate degradation reactions	potato- and cereal-based products	acrylamide, furan, HMF	42–47
processing	thermal input reduction	decrease of formation rate	potato- and cereal-based products	acrylamide, furan, HMF	48–57
	increase of relative humidity during heating	decrease of formation rate	potato- and cereal-based products, coffee, chicory, jarred foods	acrylamide, furan, HMF	58
Mitigation Strategy: Removal					
postprocessing	physical removal	evaporation or sublimation	potato- and cereal-based products, coffee, chicory, jarred foods	acrylamide, furan, HMF	59, 60
	ionizing radiation	degradation	ready-to-eat foods	furan, HMF	61, 62
	cooking or warming in open vessel	evaporation	potato- and cereal-based products, coffee, chicory, jarred foods	furan, HMF	63, 64

conditions, by acting on precursors and mechanistic pathways; the removal strategies are thought of as postprocess interventions that in principle are not affected by the previous formulation and process steps.

Although total prevention or removal of the formation of these molecules is actually impossible, greater reductions of their levels in food can be achieved by applying, when appropriate, combinations of the above-mentioned mitigation interventions.

Despite the great number of mitigation strategies reported in the literature, only a small number have been implemented by manufacturers in the factory.²⁵ With regard to the preventive tools, a limiting factor to their exploitation is the existence of a close relationship between the formation of acrylamide, furan, and HMF (undesired) and the development of sensory properties of heated foods (desired).

In fact, as the formation of these heat-induced molecules is concomitant with that of color, flavor, and texture, it is very difficult to minimize their generation without compromising the sensory acceptability of the food. In this respect, the present review focuses on a few mitigation strategies that are already or may be potentially applied at an industrial level. In particular, asparaginase pretreatments, low-temperature–long time dehy-

dration, dielectric conventional heating, and postprocess vacuum treatments have been considered. The first three can be regarded as preventive strategies, the latter as a removal intervention. These strategies could actually be compatible with the industrial process. Indeed, they not only exert low impact on the food sensory and nutritional properties but also meet regulatory compliance and are economically sustainable.

■ PREVENTIVE STRATEGIES

Acrylamide Prevention by Asparaginase Pretreatment. The commercial asparaginase obtained by cloning of *Aspergillus oryzae* is now permitted for use in many countries as a preventive tool of acrylamide formation.^{65,66} Asparaginase has been claimed to reduce effectively acrylamide levels by catalyzing the hydrolysis of asparagine into aspartic acid and ammonia, thereby specifically consuming a key precursor for acrylamide formation, with slight or no alteration of the appearance and taste of the final product^{37,67,68} if a proper enzyme dose is used.²⁴ Although in model systems asparaginase caused acrylamide reductions from 85 to 90%,^{37,38,67,68} its efficiency in foods was found to range from 27 to 70%.^{40,41} These differences can be attributable to the ineluctable batch-to-batch variability of

industrial processes as well as to a matrix effect. The batch-to-batch variability can be due to a nonconstancy of raw materials and process parameters and to inhomogeneous distribution of the enzyme in the dough. In fact, the very small quantities of asparaginase incorporated to the other ingredients hardly allow uniform enzyme distribution in the dough to be achieved. With regard to the influence of the food matrix, as the enzyme activity is influenced by the contact with the substrate, factors limiting their mobility would be responsible for incomplete hydrolysis of asparagine and only partial reduction in acrylamide formation. Among these are the food composition and structure. High water contents, by favoring the mobility of reactants and facilitating the contact between enzyme and substrate, were found to promote asparaginase efficacy.^{40,69} Opposite results were exerted by the lipid phase.⁷⁰ Indeed, the percentage of acrylamide reduction in biscuits treated with asparaginase was found to progressively decrease with the increase of fat concentration (Figure 1). It was inferred that the presence of fat contributed to create zones where reactant interactions were hindered.

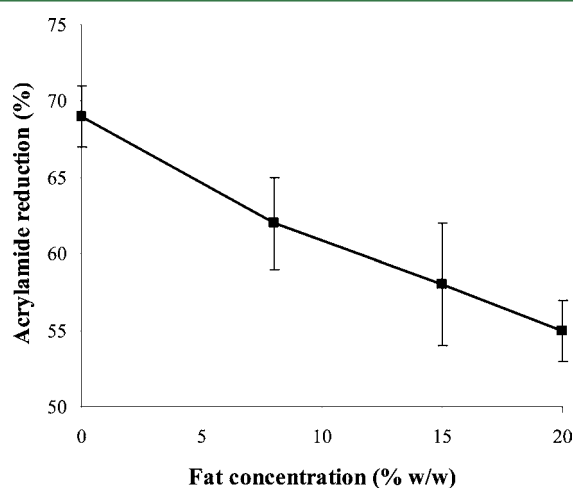


Figure 1. Percentage of acrylamide reduction in short dough biscuits with added asparaginase, as a function of fat concentration (modified from Anese et al.⁷⁰).

Moreover, food structure can greatly affect asparaginase efficacy in reducing acrylamide formation. Significant reductions of acrylamide levels can be obtained by incorporating relatively low asparaginase concentrations (up to 1000 U/kg) to potato- or cereal-based doughs. Indeed, a good enzyme distribution in the system can be achieved. On the contrary, high asparaginase concentrations (>10000 U/L of pre-frying dipping solution) have to be added to the pre-frying dipping solution to obtain a significant reduction of acrylamide in fried potatoes,³⁹ due to a dilution effect and the difficulty of the enzyme to penetrate intact tissues. Therefore, this strategy cannot be realistically applied at the industrial level because it is expensive and time-consuming. However, as already mentioned, any technological operation that favors substrate diffusion and its contact with the enzyme can improve asparaginase efficiency. Blanching of raw potatoes prior to asparaginase treatment is responsible for integrity loss of the tuber cell wall and membrane.^{36,40} Therefore, due to cell decompartmentalization, asparaginase–substrate contact is favored and acrylamide formation prevented more efficiently.

Acrylamide prevention by means of the use of asparaginase may be also favored by reagent compartmentalization.

Acrylamide reduction in cookies with added asparaginase increased from 55 to 65% by using a palm oil–water hydrogel instead of palm oil.⁷⁰ The hydrogel consisted of a three-dimensional gel network of a highly hydrated mesomorphic phase, surrounded by monoglyceride lamellas entrapping oil.^{71,72} Being water-soluble, asparaginase would be confined in the aqueous domain of the hydrogel together with acrylamide precursors. Due to the higher proximity between enzyme and substrate in the hydrogel system, greater acrylamide reductions can be achieved.

Overall results clearly indicate that the use of asparaginase represents a realistic way (i.e., applicable on industrial scale) to prevent acrylamide formation in dough-based products.²⁵ However, one should bear in mind that in order to make this technological tool as efficient as possible, proper process conditions, that is, temperature and incubation time as well as enzyme concentration, should be chosen.⁴¹ The former would allow acrylamide formation to be effectively minimized, whereas an optimal asparaginase dose would avoid off-flavor development due to an excess of ammonia generation from the hydrolysis of asparagine.

Acrylamide, Furan, and HMF Prevention by Thermal Input Reduction. In principle, decreasing the thermal input, that is, the heat amount provided to the sample during heating, represents an effective way of acrylamide, furan, and HMF mitigation.^{20,48,49,53,57} Thermal input reduction can be obtained by applying prolonged heating at lower temperatures, eventually at pressures lower than the atmospheric one (e.g., vacuum frying) or by optimizing the temperature profile of the oven (i.e., higher temperatures at the early stages of heating, when the moisture content is high, followed by lower temperatures in the final ones, when the water content is reduced). However, a decrease of the thermal input must take into account that the desired hygienic and sensory properties must be achieved. Moreover, the longer the heating time, the greater the loss of line efficiency. Alternatively, a reduction of the thermal input can be obtained by means of the application of dielectric (radiofrequency, microwave) heating. This technology allows for rapid and uniform heating because heat is generated within the product due to the frictional interactions of polar dielectric molecules rotating in response to an externally applied AC electric field (Figure 2).⁷³

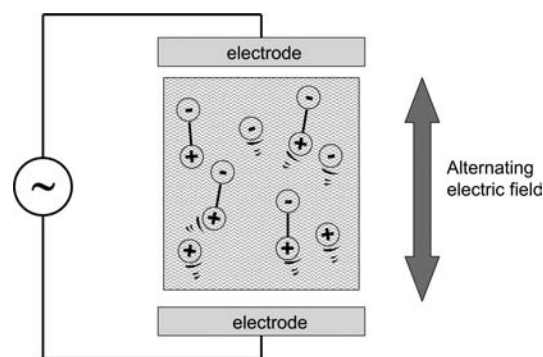


Figure 2. Principle of functioning of dielectric heating.

As water is the target molecule of dielectric heating, heat is generated at any site of the food where water is present. Therefore, foods processed by radiofrequency heating undergo lower thermal input than conventionally processed ones. In particular, radiofrequency-assisted baking processes could represent a feasible approach to keep low the acrylamide

concentration in bakery products, allowing at the same time the desired flavor and color to be developed.^{51,74} As is known, in the final stages of conventional baking, product surface moisture is very low, and considerable time is needed to bake the center because of the low thermal conductivity of the dry foods.⁷⁵ The great amount of heat thus generated is responsible for high acrylamide, furan, and HMF contents. If properly applied, a combination of radiofrequency and conventional heating allows up to 50% acrylamide reduction in bakery products as compared to a conventional only heating,^{51,74} due to the selective action of dielectric heating on water molecules that are still present in the inner part of the product.

Dielectric heating has been found to be effective also in minimizing furan and related compounds. Indeed, microwave-hot air roasting allowed great reduction of furan and HMF formation in black malts as compared to the conventional process.⁵⁵ Similarly, HMF levels in radiofrequency-pasteurized tomato puree were found to be low.⁷⁶

The use of dielectric heating alone or in combination with conventional heating to mitigate acrylamide, furan, and HMF presents numerous advantages. Indeed, it does not require changes in food recipes and can be applied to simultaneously mitigate acrylamide and furan compounds. Moreover, besides the initial costs of investment, dielectric heating allows energy savings.⁵⁵ Other advantages include heating independent of the product thermal conductivity and reduced production floor space requirements. In addition, this technology can be easily adapted to both automated production batch and continuous flow processing.⁷³ To our knowledge, this technology is already industrially exploited to mitigate acrylamide generation in low-moisture bakery products, especially by those food companies that have been using radiofrequency heaters for finishing their bakery products.

REMOVAL STRATEGIES

Acrylamide, Furan, and HMF Removal by Postprocess Vacuum Technology. A promising, not yet industrially exploited, strategy to mitigate acrylamide, furan, and HMF is represented by postprocess vacuum technology. By exploiting their chemical and physical properties, these molecules can be removed from the finished product by proper application of temperature, time, and pressure conditions. According to this technology, the finished product from the heaters is moved to the vacuum step, where the undesired molecules are removed. The results of recent studies^{59,60} showed that the efficacy of the vacuum treatment would depend on different variables, including food composition and water content as well as the nature of the molecule to be removed. Interactions between the undesired molecule and other food components and/or the presence of certain food components (e.g., lipids) would hinder the removal.⁷⁷ For instance, by applying the same combination of temperature, time, and pressure, higher levels of acrylamide were removed from cookies than from potato chips, probably due to a hurdle effect exerted by the superficial fat film of the fried potatoes against the removal.⁵⁹ Due to the viscosity constraint that would limit molecule diffusion through the matrix,⁷⁸ the removal of acrylamide and furanic compounds was not possible from dry foods, such as coffee and cookies, even if the vacuum treatment was carried out at very low pressures for a long time.^{59,60} By contrast, acrylamide, HMF, and furfural escaped quite easily from hydrated food products.^{59,60} Therefore, to make possible the removal of acrylamide, furan, and HMF, food should be subjected to a hydration step before the vacuum process.

Unfortunately, application of the vacuum treatment may be also responsible for the removal of desired flavor compounds.⁶⁰ As shown in Figure 3, the vacuum treatment carried out at 2.7

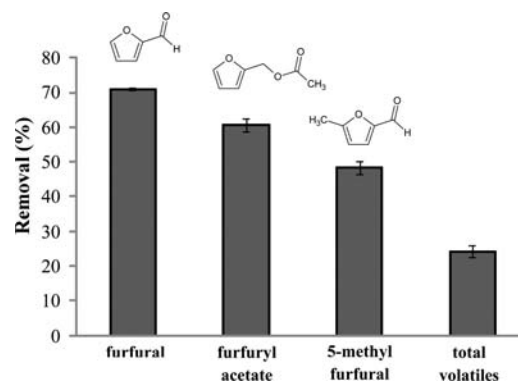


Figure 3. Furfural, furfuryl acetate, 5-methylfurfural, and total volatiles removal from hydrated ($a_w = 0.7$) ground roasted coffee subjected to vacuum treatment (3 min at 2.7 kPa and 60 °C). Furfural, furfuryl acetate, 5-methylfurfural, and total volatiles were determined by SPME-GC according to the method of Lopez-Galilea et al.,⁷⁹ data are the mean of three repetitions \pm SD (Anese, manuscript in preparation).

kPa for 3 min at 60 °C on ground roasted coffee, previously hydrated up to an a_w of 0.7, caused a significant decrease in the headspace furfural, furfuryl acetate, and 5-methylfurfural, as well as total volatile compounds. Furfural removal from hydrated coffee was higher than that of furfuryl acetate, which in turn was greater than that of 5-methylfurfural. Removal of these molecules seemed to be affected by their molecular mass and, especially, chemical polarity. The molecular masses of furfural, furfuryl acetate, and 5-methylfurfural are 96, 110, and 140 Da, respectively, whereas their polarity increases as follows: furfural > furfuryl acetate > 5-methylfurfural. As suggested by Goubet et al.,⁸⁰ the higher the molecular mass of the molecule to be removed, the lower the percentage of removal due to slower diffusion rates of the molecule through the food. Moreover, it can be inferred that molecules having higher polarity would establish stronger interactions with water, making them more prone to leave the product.

Vacuum technology represents a promising technology for the simultaneous removal of acrylamide, furan, and HMF from food. From the industrial application point of view of vacuum technology, one can assume that the finished product from the heaters is moved to a hydration step, carried out by means of a spray of pressurized water before entering the vacuum chamber, where the undesired molecules are removed. As, in principle, this technology can be applied to any finished product, no changes of recipes and process parameters are required. Indeed, to make vacuum technology exploitable at the industrial level, further studies are needed to identify process conditions able to minimize the loss of sensory properties. These could be proper pressure, temperature, and time conditions as well as hydration degree. Alternatively, flavor enrichment of the vacuum-treated foods can represent a further, final, step to be performed just before food packaging. In this regard, the so-called “processed flavors”, that is, heat-induced flavor active compositions,⁸¹ could be used to recreate the same sensory profile the product had before being subjected to the vacuum step.

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Notes

The authors declare no competing financial interest.

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