engineering proceedings

MDPI

Proceeding Paper

Optimizing the Thermal Processing of Honey by Studying the Physicochemical Properties and Its Hydroxymethylfurfural Content †

Sepidar Seyyedi-Mansour [1], Pauline Donn [1], Paula Garcia-Oliveira [1,2], Javier Echave [1], Franklin Chamorro [1], Antia Gonzalez Pereira [1,2], Aurora Silva [1,3], Ana Perez-Vazquez [1], Jesus Simal-Gandara [1], Miguel A. Prieto [1,2,*] and Leila Roufegarinejad [4,*]

1 Nutrition and Bromatology Group, Department of Analytical and Food Chemistry, Faculty of Food Science and Technology, University of Vigo, Ourense Campus, E32004 Ourense, Spain; sepidar.seyyedi@uvigo.es (S.S.-M.); donn.pauline@uvigo.es (P.D.); paula.garcia.oliveira@uvigo.es (P.G.-O.); javier.echave@uvigo.es (J.E.); franklin.noel.chamorro@uvigo.es (F.C.); antia.gonzalez.pereira@uvigo.es (A.G.P.); mass@isep.ipp.pt (A.S.); anaperevazquez00@gmail.com (A.P.-V.); jsimal@uvigo.es (J.S.-G.)
2 Centro de Investigação de Montanha (CIMO), Instituto Politécnico de Bragança, Campus de Santa Apolonia, 5300-253 Bragança, Portugal
3 REQUIMTE/LAQV, Instituto Superior de Engenharia do Porto, Instituto Politécnico do Porto, 4200-072 Porto, Portugal
4 Department of Food Science and Technology, Azad University, Tabriz 5157944533, Iran
* Correspondence: mprieto@uvigo.es (M.A.P.); l.roufegari@iaut.ac.ir (L.R.)
† Presented at the 2nd International Electronic Conference on Processes: Process Engineering—Current State and Future Trends (ECP 2023), 17–31 May 2023; Available online: https://ecp2023.sciforum.net/.

Abstract: Hydroxymethylfurfural (HMF) is a naturally occurring compound that arises because of the thermal processing and storage of honey. The *Codex Alimentarius* Commission has established a threshold of 40 ppm as the upper limit for the HMF content in honey. This research aimed to investigate the impacts of varying heating temperatures (55, 65, and 75 °C) and heating times (10, 20, and 30 min), as well as storage temperatures (25 and 40 °C), over a period of three months. The study employed the response surface methodology (RSM) to evaluate the outcomes. The impacts of the variables mentioned above on the physicochemical properties, including the Lab color factors, pH, and moisture, were determined. Additionally, the concentration of HMF in the samples was analyzed. The prediction model of each treatment was computed. Analyses of the results obtained after the storage periods of 45 and 90 days were conducted. The findings indicated that pH, moisture content, and color were not significantly influenced by temperature, the duration of heat treatment, or the storage time. However, storage temperature considerably impacted both L* and a*. Furthermore, it was observed that all the variables significantly influenced the HMF content, and its concentration increased with the escalation of thermal processing and storage duration. Within the examined samples, the HMF content surpassed the standard limit in the model subjected to heating at 75 °C for 20 min and maintained at 40 °C for 90 days. In contrast, heating at 55 °C for 10 min, followed by storage at 25 °C for 45 days, produced the optimal HMF level.

Keywords: honey; hydroxymethylfurfural; response surface methodology; storage; thermal process

Citation: Seyyedi-Mansour, S.; Donn, P.; Garcia-Oliveira, P.; Echave, J.; Chamorro, F.; Pereira, A.G.; Silva, A.; Perez-Vazquez, A.; Simal-Gandara, J.; Prieto, M.A.; et al. Optimizing the Thermal Processing of Honey by Studying the Physicochemical Properties and Its Hydroxymethylfurfural Content. *Eng. Proc.* **2023**, *37*, 108. https://doi.org/10.3390/ECP2023-14713

Academic Editor: Chi-Fai Chau

Published: 19 May 2023

1. Introduction

Honey is a natural sweet substance yielded by honeybees using plant nectar or living plant secretions, as defined by the *Codex Alimentarius*. It is collected by bees and transformed through combination with specific substances of their own, with the bees depositing, dehydrating, storing and leaving it in the honeycomb to ripen and mature [1].

Honey comprises diverse components, including sugars, water, vitamins, enzymes, minerals, and other constituents. The presence of these compounds, along with enzymes and polyphenolic compounds, has made honey a valuable elixir and food used in traditional medicine for its restorative, antimicrobial, and anti-inflammatory properties. Although honey has numerous nutritional and medicinal properties, improper storage and processing result in some undesirable non-nutritional compounds in this product. In recent years, HMF has gained significant attention as a high-potential toxicological and carcinogenic contaminant in honey [2,3].

HMF is a furanic compound produced through the Maillard reaction or via the direct dehydration of sugar under acidic conditions [4,5]. Two primary metabolic pathways have been identified for 5-hydroxymethylfurfural (5-HMF). The first includes the synthesis of 5-hydroxymethyl-2-furoyl glycine (HMFG), involving the oxidation of the aldehyde functional group to yield 5-hydroxymethyl-2-furoic acid (HMFA), which is subsequently conjugated with glycine. Another metabolic pathway which holds greater significance from a toxicological perspective involves the sulfonation of the allylic hydroxyl function of 5-hydroxymethylfurfural (5-HMF) [6]. 5-Sulfooxymethylfurfural (SMF) is a potent genotoxic with mutagenic effects [7]. The international standard commissions establish the maximum allowable level of HMF in honey at 40 mg/kg [8]. The HMF content in honey is subject to various factors, such as its sugar content, pH, acidity, water activity, diastase and invertase activity and divalent cation concentration [9]. A correlation between the initial pH and HMF contents in honey has been observed. It was predicted that honey samples with a pH of less than 4 may contain approximately 40 mg/kg of HMF, while those with a pH greater than 4 may have HMF contents ranging from 20 to 25 mg/kg. These findings shed light on the interplay between pH and HMF in honey [10,11].

Honey producers heat honey to prevent post-bottling crystallization and delay microbial spoilage. Selecting the best operational parameters, including the time and temperature, is a major challenge in honey production and packaging. The amount of HMF present in honey depends on the duration and temperature of its thermal treatment. As such, its content serves as the primary criterion with which to predict the optimization of this processing. The modeling and identification of a relationship between different heat treatments and storage parameters could be a reasonable approach used to optimize the process and increase the quality and safety of the end product. This study investigated the effects of process conditions (temperature and heating time) and storage conditions on the HMF content and physicochemical properties (Lab color factors, pH, and moisture) of honey to determine the optimal processing and storage conditions.

2. Materials and Methods

Honey samples with a moisture content of 14% and pH of 3.8 were obtained from a local honey-packaging factory. For this purpose, the honey samples were poured into special honey bottles, and to assess the influences of temperature and heating time, a heat treatment was applied with indirect heating using a thermostatic bath at the temperatures of 55, 65, and 75 °C and times of 10, 20 and 30 min. During the investigation, tests were conducted at intervals of 45 days (0, 45, and 90), and three repetitions were performed.

The moisture was determined by measuring the refractive indices at 20 °C with a refractometer, and the corresponding moisture content (%) was calculated according to previous authors [1]. A pH meter was used to measure the pH of the samples [2]. The digital imaging method was used to analyze the samples' color [12]. The measurement of the HMF concentration in the honey was performed using a spectrophotometer, according to the White method [3,13]. The data were analyzed with a Minitab16 using the response surface methodology (RSM) and a central composite design (CCD) (Table 1).

Table 1. Central composite design (using Minitab 16 software).

Run Order	Block	Heating Pasteurization Temperature (°C)	Time of Heating (min)	Storage Temperature (°C)
1	1	65	20	25
2	1	55	30	25
3	1	55	10	25
4	1	75	10	25
5	1	65	20	25
6	1	65	20	25
7	2	75	30	40
8	2	65	20	40
9	2	65	30	40
10	2	65	20	40
11	2	65	10	40
12	2	55	20	40
13	2	75	20	40
14	2	65	20	40

3. Results and Discussion

The pH values of the honey samples during the 45- and 90-day storage periods were 3.7–3.82 and 3.65–3.81, respectively. There was no significant relationship between pH and any of the variables studied. The different times of the thermal process and storage temperature did not significantly affect the moisture values. During the storage periods of 45 and 90 days, the honey samples had average moisture contents of 14.9 and 15, respectively. Only the storage temperature could affect the variables impacting on the *L index. Similar results were also observed regarding the *a index of the honey samples. However, for the *b index, the effect of none of the variables was significant. All the studied factors showed a significant effect on HMF formation ($p < 0.05$), indicating that this content was significantly increased with higher heating times, temperatures, and storage conditions (Figures 1 and 2).

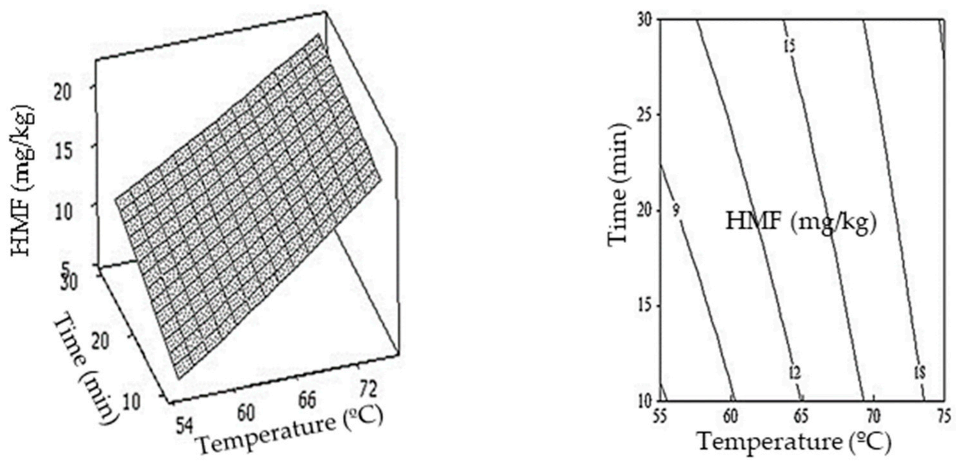

Figure 1. The effect of thermal process temperature on the amount of HMF over 45 days of storage.

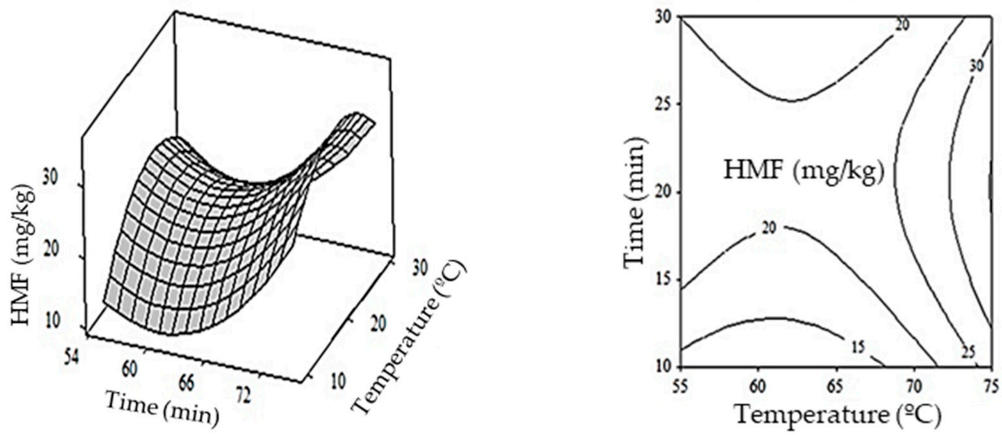

Figure 2. The effect of thermal process temperature on the amount of HMF over 90 days of storage.

4. Conclusions

From the optimization study, it can be concluded that the minimum content of HMF resulted from heating the honey at 55 °C for 10 min and keeping it at 25 °C for 45 days. Therefore, these processing and storage conditions could help one to find the best operation conditions in which to preserve honey's quality and safety.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/ECP2023-14713/s1.

Author Contributions: Conceptualization, S.S.-M. and L.R.; methodology, S.S.-M., P.D., F.C., A.S. and A.P.-V.; software, S.S.-M., M.A.P. and L.R.; validation, M.A.P. and L.R.; formal analysis, S.S.-M., M.A.P. and L.R.; investigation, S.S.-M., P.D., P.G.-O., J.E., F.C., A.G.P., A.S. and A.P.-V.; resources, J.S.-G., M.A.P. and L.R.; writing—original draft preparation, S.S.-M., P.D. and A.G.P.; writing—review and editing, P.G.-O., J.E., J.S.-G., M.A.P. and L.R.; visualization, S.S.-M., M.A.P. and L.R.; supervision, J.S.-G., M.A.P. and L.R.; project administration, M.A.P. and L.R.; funding acquisition, J.S.-G., M.A.P. and L.R. All authors have read and agreed to the published version of the manuscript.

Funding: The research leading to these results was supported by Xunta de Galicia, through the program EXCELENCIA-ED431F 2020/12.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: The research leading to these results was supported by MICINN, supporting the Ramón y Cajal grant for M.A. Prieto (RYC-2017-22891) and by Xunta de Galicia, supporting the pre-doctoral grants of P. Garcia-Oliveira (ED481A-2019/295) and A.G. Pereira (ED481A-2019/0228). The authors express their thanks for the program BENEFICIOS DO CONSUMO DAS ESPECIES TINTORERA-(CO-0019-2021), which supports the work of F. Chamorro.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Codex Alimentarius. Standard for Honey 12-1981. Adopted in 1981. Revised in 1987, 2001. Amended in 2019. pp. 1–8. Available online: https://www.fao.org/fao-who-codexalimentarius/sh-proxy/es/?lnk=1&url=https%253A%252F%252Fworkspace.fao.org%252Fsites%252Fcodex%252FStandards%252FCXS%2B12-1981%252FCXS_012e.pdf (accessed on 4 March 2022).
2. Ünubol Aypak, S.; İnci, A.; Bakırcı, S.; Dereli Fidan, E.; Soysal, M. Comparison of antioxidant activity and hydroxymethylfurfural (HMF) levels in honey taken from hives and markets. *GIDA/J. Food* **2019**, *44*, 86–92. [CrossRef]
3. Kamboj, R.; Sandhu, R.S.; Kaler, R.S.S.; Bera, M.B.; Nanda, V. Optimization of process parameters on hydroxymethylfurfural content, diastase and invertase activity of coriander honey. *J. Food Sci. Technol.* **2019**, *56*, 3205–3214. [CrossRef] [PubMed]
4. Capuano, E.; Fogliano, V. Acrylamide and 5-hydroxymethylfurfural (HMF): A review on metabolism, toxicity, occurrence in food and mitigation strategies. *Lwt* **2011**, *44*, 793–810. [CrossRef]

5. Godoy, C.A.; Valderrama, P.; Boroski, M. HMF Monitoring: Storage Condition and Honey Quality. *Food Anal. Methods* **2022**, *15*, 3162–3176. [CrossRef]
6. Janzowski, C.; Glaab, V.; Samimi, E.; Schlatter, J.; Eisenbrand, G. 5-Hydroxymethylfurfural: Assessment of mutagenicity, DNA-damaging potential and reactivity towards cellular glutathione. *Food Chem. Toxicol.* **2000**, *38*, 801–809. [CrossRef] [PubMed]
7. Durling, L.J.K.; Busk, L.; Hellman, B.E. Evaluation of the DNA damaging effect of the heat-induced food toxicant 5-hydroxymethylfurfural (HMF) in various cell lines with different activities of sulfotransferases. *Food Chem. Toxicol.* **2009**, *47*, 880–884. [CrossRef] [PubMed]
8. Tosi, E.A.; Ré, E.; Lucero, H.; Bulacio, L. Effect of honey high-temperature short-time heating on parameters related to quality, crystallisation phenomena and fungal inhibition. *Lwt* **2004**, *37*, 669–678. [CrossRef]
9. Ajlouni, S.; Sujirapinyokul, P. Hydroxymethylfurfuraldehyde and amylase contents in Australian honey. *Food Chem.* **2010**, *119*, 1000–1005. [CrossRef]
10. Fallico, B.; Zappalà, M.; Arena, E.; Verzera, A. Effects of conditioning on HMF content in unifloral honeys. *Food Chem.* **2004**, *85*, 305–313. [CrossRef]
11. Seyyedi-Mansoor, S.; Roufegari-nejad, L. A Study on the Formation and Safety of HMF in Honey. *Adv. Environ. Biol.* **2014**, *8*, 1228–1231.
12. Yam, K.L.; Papadakis, S.E. A simple digital imaging method for measuring and analyzing color of food surfaces. *J. Food Eng.* **2004**, *61*, 137–142. [CrossRef]
13. Zarei, M.; Fazlara, A.; Alijani, N. Evaluation of the changes in physicochemical and antioxidant properties of honey during storage. *Funct Foods Health Dis.* **2019**, *9*, 593–605. [CrossRef]