Editorial

Supercritical Technology Applied to Food, Pharmaceutical, and Chemical Industries

Ádina L. Santana

Grain Science and Industry Department, Kansas State University, 1301 N Mid Campus Drive, Manhattan, KS 66506, USA; adina@ksu.edu

1. Introduction

Everyday life has caused consumers to feel genuine concern about the origin of the products they consume. For this reason, green technologies are required in industrial processing to ensure the development of high-quality products. Supercritical technology is a green methodology that includes multiple types of high-pressure processes that employ substances in conditions next to or above the critical point [1–3].

Supercritical technology has emerged as an environmentally friendly and efficient alternative for use in the preparation of multiple varieties of matrix for the extraction [4–6], fractionation [7,8], and purification of molecules [9]; the transformation of molecules via chemical reactions [10]; particle formation [11]; impregnation [12]; drying [13]; and sterilization [14]. In this Special Issue, fifteen outstanding manuscripts covering novel insights into the theory and practice of supercritical fluid-based processes are published. For more information on this Special Issue, readers are strongly encouraged to visit the website: https://www.mdpi.com/journal/processes/special_issues/Supercritical_Technology (accessed on 6 April 2024).

2. Review Manuscripts

The thermodynamic background [15] of the gas–lattice model and its potential to describe processes at a supercritical state was reviewed by Tovbin [16]. Supercritical fluids possess applications as refrigerant fluids. The optimization of heat transfer using supercritical fluids has been studied via the use of the gas–lattice approach [17,18].

The use of supercritical technology to valorize corn byproducts was reviewed by Santana and Meireles [19], who proposed the use of a novel process according to the biorefinery approach [20–23]. The proposed biorefining method consisted of integrating traditional dry-grinding, performed in an industrial setting, with the supercritical carbon dioxide (SC-CO$_2$)-based extraction of corn-dried distiller’s grains with solubles (DDGS) to obtain an oil that was rich in the carotenoids known as lutein and zeaxanthin. This was followed by the use of pressurized liquid to extract phenolic acids from the semi-defatted corn DDGS, and by the concentration of the extract into precipitated particles.

3. Research Manuscript: Particle Formation Techniques

Particle formation using supercritical technology offers advantages like the control of particle size and morphology, high encapsulation efficiency, and the low degradation of molecules [11,24,25]. In this Issue, Tirado and coworkers [26] modelled the supercritical fluid extraction of emulsions process (SFEE) of a ternary CO$_2$/ethyl acetate/water system in order to design equipment that could meet industrial requirements regarding the permitted quantities of residual organic materials in the leaving streams. In SFEE, SC-CO$_2$ is used to extract the organic phase from an organic phase/water (O/W) emulsion in which the target molecule and its coating material have been previously solubilized. After solvent removal, both compounds precipitate, generating particles that are suspended in the water phase with the aid of a surfactant [27].
4. Research Manuscript: Chemical Reactions

Supercritical transesterification is attractive in comparison with conventional transesterification as a method with which to produce biodiesel since it requires little time and no catalyst [28]. In this Issue, García-Morales and coworkers [29] investigated the potential of alcohols at supercritical state in terms of the transformation of waste beef tallow into fatty acid alkyl esters or biodiesel. We found conversion rates higher than 90% at 335–390 °C for supercritical iso-butanol and at 360 °C for supercritical ethanol.

5. Research Manuscript: Sterilization of Foods

The sterilization of bacteria with SC-CO$_2$ emerged as a method because of the mild temperatures used in comparison to conventional techniques [30,31]. In this Issue, Dacal-Gutiérrez and coworkers [32] observed that the inactivation of *Clostridium* spores in low-moisture honey is not effective when using SC-CO$_2$ at temperatures lower than 70 °C, but that the use of carbon dioxide, modified with cinnamon essential oil, significantly reduced the presence of spores at 60 °C.

6. Research Manuscripts: Removal of Undesirable Compounds

Chiu and coworkers [33] observed that SC-CO$_2$ has good biocompatibility in the decellularization of porcine hide for the reconstruction of an abdominal wall that had been injured by hernia. The decellularization of tissues with SC-CO$_2$ is a pretreatment protocol that is used to remove undesirable tissue and molecules (protein and lipids) for biomedical applications [34].

Náthia-Neves and coworkers [35] studied the extraction of colorants from unripe genipap defatted with SC-CO$_2$. SC-CO$_2$ extraction worked as a pretreatment, ensuring the plant material was suitable for the subsequent ultrasound-assisted extraction of genipin and geniposide with water and ethanol mixtures [36].

7. Research Manuscripts: Extraction of Bioactive Compounds

Supercritical technology is used for the extraction of multiple bioactive compounds, including phenolic compounds [37], carotenoids [38], phytosterols [39], and cannabinoids [40].

Qamar and coworkers [41] employed the half-fractional factorial design to select the best conditions for the SC-CO$_2$ extraction of cannabis flowers and found that the optimal conditions were 45 °C, 250 bar, and 180 min.

Boumghar and coworkers [42] selected the Box–Behnken experimental design to optimize the supercritical fluid extraction of decarboxylated cannabis flower. The authors used decarboxylation to pretreat the raw material in order to increase the affinity of cannabinoids to CO$_2$. The optimal conditions for the extraction of cannabinoids were 55 °C, 235 bar, 2 h, and a CO$_2$ flow rate of 15 g/min.

Popescu and coworkers [43] investigated the potential of oil seeds as modifier to CO$_2$ in order to increase the recovery of carotenoids from tomato slices. After supercritical fluid extraction, two products were obtained: a solid, oleoresin rich in lycopene, and an oil fraction rich in other carotenes and linolenic acid.

Duong and coworkers [44] optimized the recovery of saponins from *Hedera nepalensis* leaves with the use of pressurized liquid extraction. The Box–Behnken design was adopted by the authors to select the extraction time, solvent used, and temperature. The extracts showed antimicrobial activity by inhibiting the growth of three types of bacteria.

Santana and coworkers [45] investigated the effects of the post-acidification of pressurized liquid extracts of sorghum on the concentration of phenolic compounds. The authors observed that acidification considerably improved the concentration of 3-deoxyanthocyanidins and cyanidin, but decreased the concentration of other phenolics, including taxifolin, quercetin, and chlorogenic acid. Sorghum (*Sorghum bicolor* L.) is the fifth most-produced cereal worldwide and is a source of diverse classes of phenolic compounds, including tannins, benzoic- and cinnamic acids, 3-deoxyanthocyanidins, and flavonols [46,47].

Bushnaq et al. [48] proposed a three-step process in order to enhance the yield of sugars from date palm. This was based on (1) the freeze-drying of dates, (2) supercritical fluid extraction with CO$_2$ modified with water, and (3) the spray-drying of a supercritical-based extract. After simulation, the authors reported that a highest rate of sugar recovery can be reached at a CO$_2$–water ratio of 0,07, CO$_2$ flow rate of 31,000 kg/h, 65 °C, and 308 bar. SC-CO$_2$ extraction is useful as a pretreatment technique in the extraction of sugars, allowing manufacturers to remove undesirable compounds of raw material [49].

Before implementing a process in the market, it is important to have knowledge of the economic feasibility of the process. Economic evaluations consider the components involved, the costs of processing, the final products, as well as the economic fluctuations that affect the price of inputs [50–52]. Cruz Sánchez and coworkers [53] extracted lavender flowers with SC-CO$_2$ at 60 °C and 180 and 250 bar, simulated the process for the capacities of 20 L, 50 L, and 100 L. They observed that the cost of manufacturing was lower at 50 and 100 L, and that the price of equipment was the item that most affected the return on equity. The return on equity is a parameter that indicates a process’s profitability [54].

Best and coworkers [55] investigated the economic profitability of the extraction of Maurititia flexuosa pulp using two scenarios: (a) conventional extraction and (b) conventional extraction integrated with SC-CO$_2$ extraction. They concluded that scenario (b) was the most feasible economically, since it was enabled researchers to obtain two types of products—namely, an oil rich in carotenoids, and an extract with high phenolic content.

9. Conclusions

The results obtained from the research published in this Special Issue support the industrial use of supercritical technology via the application of antioxidant extracts in food, pharmaceutical industries, and the medical sector, as well as the conversion of underused fat into value-added fuels. Additionally, the theoretical aspects explored in this Issue, with explorations into thermodynamics, mathematical modeling, and economic evaluation, provide useful information for the optimization of processes and reduction of costs.

Conflicts of Interest: The author declares no conflicts of interest.

References

3. Prasad, S.K.; Sangwai, J.S.; Byun, H.-S. A review of the supercritical CO$_2$ fluid applications for improved oil and gas production and associated carbon storage. J. CO$_2$ Util. 2023, 72, 102479. [CrossRef]
7. Shukla, A.; Naik, S.N.; Goud, V.V.; Das, C. Supercritical CO$_2$ extraction and online fractionation of dry ginger for production of high-quality volatile oil and gingerols enriched oleoresin. Ind. Crops Prod. 2019, 130, 352–362. [CrossRef]
8. Reverchon, E.; De Marco, I. Supercritical fluid extraction and fractionation of natural matter. J. Supercrit. Fluids 2006, 38, 146–166. [CrossRef]
12. Liu, X.; Jia, J.; Duan, S.; Zhou, X.; Xiang, A.; Lian, Z.; Ge, F. Zein/MCM-41 Nanocomposite Film Incorporated with Cinnamon Essential Oil Loaded by Modified Supercritical CO₂ Impregnation for Long-Term Antibacterial Packaging. *Pharmaceutics* 2020, 12, 169. [CrossRef] [PubMed]


32. Dacal-Gutiérrez, A.; Tirado, D.F.; Calvo, L. Inactivation of Clostridium Spores in Honey with Supercritical CO₂ and in Combination with Essential Oils. *Processes* 2022, 10, 2232. [CrossRef]


37. Garmus, T.T.; Paviani, I.C.; Queiroga, C.L.; Cabral, F.A. Extraction of phenolic compounds from pepper-rosmarin (*Lippia sidoides* Cham.) leaves by sequential extraction in fixed bed extractor using supercritical CO₂, ethanol and water as solvents. *J. Supercrit. Fluids* 2015, 99, 68–75. [CrossRef]
40. Qamar, S.; Torres, Y.J.M.; Parekh, H.S.; Falconer, J.R. Effects of Ethanol on the Supercritical Carbon Dioxide Extraction of CBD-Dominant Cannabis Flowers by Supercritical Carbon Dioxide. Processes 2022, 10, 93. [CrossRef]
41. Qamar, S.; Torres, Y.J.M.; Parekh, H.S.; Falconer, J.R. Fractional Factorial Design Study for the Extraction of Cannabinoids from CBD-Dominant Cannabis Flowers by Supercritical Carbon Dioxide. Processes 2022, 10, 93. [CrossRef]
48. Cruz-Sánchez, E.; García-Vargas, J.M.; Gracia, I.; Rodríguez, J.F.; García, M.T. Supercritical CO₂ extraction of lavender flower with antioxidant activity: Laboratory to a large scale optimization process. J. Taiwan Inst. Chem. Eng. 2022, 105404. [CrossRef]
49. Cruz-Sánchez, E.; García-Vargas, J.M.; Gracia, I.; Rodríguez, J.F.; García, M.T. Pilot-Plant-Scale Extraction of Antioxidant Compounds from Lavender: Experimental Data and Methodology for an Economic Assessment. Processes 2022, 10, 2708. [CrossRef]