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Use of non-conventional methods for the extraction of value-added products from citrus waste

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Abstract
Oranges, grapefruit, lemons, limes, and mandarins are just a few of the citrus fruits that are grown most frequently worldwide. Nearly half of the weight of fresh fruit is wasted in the citrus processing business, which generates enormous amounts of trash. Orange juice results in a significant quantity of waste generation. The bioactive chemicals are typically extracted from orange peel using traditional extraction methods. carotenoids are hydrophobic and unstable at higher temperatures, under the influence of light and oxygen, and under other external conditions, their usage in food, cosmetic, and pharmaceutical goods is restricted. The use of safer and less harmful substitutes has begun to be addressed, and it has been a priority to reduce the negative effects that solvents routinely used in industry have on the environment and human health. The major classes of carotenoids, chlorophyll, anthocyanin, and betalains are represented by the pigments that the UAE is stated to have extracted from by-products. Conventional techniques that use organic solvents have a same negative environmental impact as citrus waste because they demand lengthy extraction times, a lot of energy, and other resources.

Key words: solvent extraction, ultrasound extraction, citrus, oranges, green-extraction, pigments

Introduction

Oranges, grapefruit, lemons, limes, and mandarins are just a few of the citrus fruits that are grown most frequently worldwide. Brazil is the world's top exporter of citrus fruits that have been processed, particularly frozen orange juice concentrate. Due to rising consumer demand, their cultivation and production is increasing every year. Nearly half of the weight of fresh fruit is wasted in the citrus processing business, which generates enormous amounts of trash.[1] Orange peel is a source of many polyphenols, carotenoids, dietary fibre, sugar, essential oils, ascorbic acid, and considerable amounts of some trace components.[2] Due to the fact that orange peel, which is made up of flavedo and albedo, makes up over a quarter of the total fruit mass, the high production of orange juice results in a significant quantity of waste generation. Citrus trash also contains other carotenoids. Orange peel extracts offer a variety of pharmacological properties that are crucial for the prevention of many human diseases as a result of their chemical makeup. The aforementioned bioactive chemicals are typically extracted from orange peel using traditional extraction methods. The green approach led to the development of various extraction techniques that are quick, effective, and prevent the degradation of thermolabile chemicals. In orange peel, alpha-carotene, beta-carotene, lutein, zeaxanthin, and -cryptoxanthin are the most prevalent carotenoids.[3] The recovery of these natural value-added elements from citrus trash, such as fibre, bioactive compounds, additives, and pigments, has improved. Depending on the kind of solvent used and the extraction method, the total carotenoid concentration ranges from 11 to 204 mg of -carotene equivalents per 100 g of dry weight. They are typically extracted using acetone, ionic liquids, and a mixture of hexane and isopropanol, but these methods can be hazardous to the environment.

The oils are increasingly used as solvents since they are non-toxic and non-irritating. Increased immunity and a decreased risk of developing degenerative illnesses including cancer, Alzheimer's disease, and cardiovascular disease are linked to an

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organism's intake of carotenoids.[4] Because carotenoids are hydrophobic and unstable at higher temperatures, under the influence of light and oxygen, and under other external conditions, their usage in food, cosmetic, and pharmaceutical goods is restricted. Encapsulating carotenoids in various delivery systems is one approach to solving the aforementioned issue. For the encapsulation and release of carotenoids, a variety of biopolymers were used as carrier materials, including starch, cyclodextrins, carrageenan, pectin, alginate, etc.[5] Natural pigments called carotenoids have grown in importance in the food business as a replacement for rapidly dated artificial colours. Natural compounds are now more in demand than their synthetic counterparts, and there has been more focus on the differences between natural and synthetic products in recent years. The advent of the "Green Chemistry" concepts has changed how both academia and industry construct chemical processes as a result of growing environmental and sustainable development concerns over the past few decades. The use of safer and less harmful substitutes has begun to be addressed, and it has been a priority to reduce the negative effects that solvents routinely used in industry have on the environment and human health. Alternative extraction methods with "greener" and "more sustainable" credentials have been developed by researchers.[6] Ultrasound is a new technology that has been extensively researched in the food industry to enhance procedures and outcomes.[7] The passage of acoustic waves and acoustic cavitation generate several mechanisms that are responsible for variations in the original matrix of the sample when ultrasound is applied to a product with solid or fluid properties. By extracting substances of interest, such as natural pigments, one can make use of the effects of ultrasound to benefit from by-products.

Extraction of Natural Pigments From By-Products Using Ultrasound Assist

By-products may be utilized as a fresh sample or one that has already been conditioned. Sample preparations frequently involve freezing or drying using hot-air, sun, or freeze-drying techniques.[8] The dried materials are next ground or chopped, then sieved to continue with UAE. After the samples are ready, they are combined with a solvent and put through the UAE process to produce extracts rich in natural pigments. These extracts can then be employed in a variety of food, medicinal, and cosmetic applications as dried pigment or concentrated extract. The major classes of carotenoids, chlorophyll, anthocyanins, and betalains are represented by the pigments that the UAE is stated to have extracted from by-products.[9]

Carotenoids extraction from by-products using ultrasound assist

Lipophilic pigments known as carotenoids, which are primarily generated by plants, algae, and some creatures like arthropods and salmonids, are what give things their distinctive yellowish, orange, and reddish hues.[10] Both xanthophylls, which contain oxygen in their chemical formula, and carotenes, which do not, are two major categories of carotenoids. Carotenogenesis of carotenes and xanthophylls is governed by the transcript genes, which are primarily regulated by light and temperature. Many other natural carotenes and xanthophylls are synthesised from lycopene, which is regarded as the first colored carotenoid in the process. Alpha carotenes are produced from lycopene, and whereas beta-carotene is transformed into lutein, Alpha-carotene is changed into a variety of different compounds, including cryptoxanthin, zeaxanthin, antheraxanthin, capsanthin, violaxanthin, and neoxanthin. Due to their ability to act as antioxidants and as precursors of vitamin A, carotenoids are crucial functional metabolites in the human body and have been linked to significantly lower rates of cancer, cardiovascular disease, and age-related macular degeneration. Conventional techniques that use organic solvents have a same negative environmental impact as citrus waste because they demand lengthy extraction times, a lot of energy, and other resources.[11] The difficulties of traditional extraction techniques, the accessibility of citrus waste,

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and the growing desire to minimize environmental impact have all boosted interest in the proper treatment of this sort of trash. As a result, research into unconventional technologies known as "green extraction methods" has become more important.

Experimental bases for Ultrasound Assisted Extraction

Using waves with a frequency exceeding 10 MHz, ultrasonic application, also known as ultrasound assisted extraction (UAE), is a cutting-edge method.[12] Initially, it was intended to preserve food, but in the past ten years, it has also been employed to extract beneficial components (mainly polyphenols). Due to the technique' simplicity, benefits like shorter extraction times, higher extract yields, and the use of water as a solvent rather than organic solvents have been mentioned, all of which lower environmental hazards. UAE is regarded as a "green extraction process" due to these factors.[13]

Citrus fruit residues have been used to extract bioactive substances and pectins using the cavitation and disruptive capabilities of USN. In numerous investigations, the extraction of carotenoids, phenolic compounds, and pectins from citrus waste by USN is described along with the impact of the parameters utilized. The method's optimization and a study design are usually incorporated in USN studies. The response surface methodology (RSM) is highly helpful in this area. Since carotenoids are naturally occurring fat-soluble pigments that give citrus fruits their distinctive yellow, orange, and red hues as well as their roles in photosynthesis and photoprotection, the peel is primarily the material employed in the extraction of carotenoids from citrus waste.[14] Its 40 terpenoid carbons, made from isoprenoids, are connected by double and single bonds that make up its structure. Some types of carotenoids are more closely associated to polar solvents like acetone or a-polar solvents like hexane because of their structure.[15] These solvents are extremely hazardous and challenging to get rid of. Studies on the effectiveness of extracting phenolic components from citrus residues utilising UAE provide information on this topic.

The primary and secondary metabolites in plants are phenolic compounds, which have more than 9000 known chemical configurations. The two primary types of polyphenols in citrus species are flavonoids and phenolic acids.[16] However, the main phenolic chemicals found in orange peel are flavonoids.[17] Several studies have detailed the effectiveness of UAE against CE and the parameters examined to recover phenolic chemicals from citrus waste. For instance, The most sensitive parameters for the extraction of phenolic acids from tangerine peel (Citrus unshiu Marc) using USN are time, temperature, and the power of the equipment. The scientists found that increasing the extraction duration (10–40 min), power (3.2–30 W), and low temperatures (15^0C and 30^0C) enhanced the yield of the extracts.[18] They demonstrated the effectiveness of UAE against CE (maceration) and showed that they could achieve better results with shorter extraction times. Similar to this, Khan et al. compared the extraction of polyphenols from orange peel using UAE and CE (Citrus sinensis L.).[19] They demonstrated that the particle size of 2 cm^2 favored a greater yield, recovering 38% and 41% more of naringenin and hesperidin, respectively, in comparison to the sample treated with CE, even though it is true that they did not identify the factors that significantly influenced the results. The efficiency of UAE against CE in the extraction of polyphenols (Citrus reticulata L.) from mandarin peels was then compared by Safdar et al. in 2017.[20] They used methanol 80% as the solvent and found that the UAE samples had the greatest concentration of TPC (3248 mg GAE/100 g). Using ethanol 80% as the solvent, they also demonstrated that the main component in orange peel.

Citrus waste's primary component of the cell wall is pectin, a complex carbohydrate that is chemically organized as a galacturonic acid polymer with a variable amount of methyl ester groups and abundant in citrus peel. For use as a byproduct, this chemical is frequently

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extracted using traditional techniques. Numerous research demonstrate that using an unorthodox methodology, like UAE, can produce good extraction outcomes. For instance, Bagherian et al. employed USN treatment to extract grapefruit skin pectins (albedo).[21] They were able to extract 17.92% of the pectins with a 25-minute extraction period utilising acidified water with a pH of 1.5 and 0.1 N HCl as the extracting agent. Wang et al.[22] also studied the effectiveness of ultrasound aided heating extraction (UAHE) for extracting pectins from grapefruit peel in comparison to traditional heating extraction (CHE). As an extraction solvent, deionized water that has been pH-adjusted by 0.5 M HCl was utilized. In comparison to pectin extracted by CHE, the pectin yield of grapefruit peel treated by UAHE was 16.34% higher. The authors noted that power intensity and extraction temperature, as well as their interactions with power intensity and sonication time, both had an impact on the pectin yield. Additionally, the pectins extracted by UAHE had a degree of esterification, polyphenols, and flavonoids concentration of 65.52%, 4.21 g GAE/mg, and 1.76 g RE/mg, respectively.

Conclusion

The United Nations' sustainable goals by 2030 are in line with the present emphasis in reducing industrial and food waste. Researchers have been more interested in employing ecologically friendly technology to extract bioactive chemicals from food waste, such as citrus leftovers. These substances have potential for re-use in the food, pharmaceutical, and cosmetic industries. It is well recognized that bioactive substances and pectin can be recovered from citrus waste using traditional procedures. However, because organic solvents are used, conventional procedures need a large time and energy investment, making them an undesirable choice.

The replacement of unconventional approaches, on the other hand, has demonstrated a significant number of benefits in a framework of green extraction for the extraction of high biological value chemicals from citrus by-products. The majority of researchers who use unconventional approaches conduct an optimization study since certain parameters can affect the outcomes depending on the properties of the chosen technologies and samples. The most important factors to take into account when choosing the best technology are the type of waste and the molecules that need to be removed. However, further research is required on the bio- accessibility, bioavailability, and validation of high biological value chemicals found in citrus waste.

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