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Published in:
Current Research in Green and Sustainable Chemistry

DOI:
10.1016/j.crgsc.2022.100340

First published: 27/10/2022

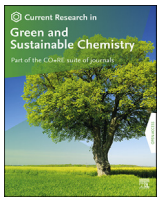
Document Version
Publisher's PDF, also known as Version of record

Current Research in Green and Sustainable Chemistry 5 (2022) 100340

Contents lists available at ScienceDirect

Current Research in Green and Sustainable Chemistry

journal homepage: www.elsevier.com/journals/current-research-in-green-and-sustainable-chemistry/2666-0865



Greener technologies in agri-food wastes valorization for plant pigments: Step towards circular economy

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ARTICLE INFO

Keywords:
Agri-food waste
Green valorization strategies
Plant pigments
Sustainable food production
Green circular economy

ABSTRACT

The need for food has drastically increased with the world population's unprecedented growth, which consequently raised agricultural production. The increased agricultural and food production as a whole has resulted in the generation of enormous amounts of food waste, which is now posing a threat to both the environment and humanity. To address these issues, it is now crucial to persuade people to use promising green technologies to manage and valorize agri-food waste into valuable food additives. The recovery of value-added pigments from agri-food waste as natural food colorants is one of the new business prospects being created by approaches influenced by the circular economy model that have been continuously growing globally. These natural pigments are expected to substantially impact the development of functional foods and offer a wealth of bio-therapeutic potential. The production of naturally safe food pigments from the agri-food waste offers a greener approach within circular economy concept while avoiding the use of synthetic-petro-based colourants from the food chain. This also promotes the incorporation of natural food pigments obtained through sustainable resources. This state-of-the-art graphical review focused on the recent advances in green valorization technologies of agri-food waste to exploit natural plant pigments and their relation to sustainable food production and green circular economy.

1. Introduction

The generation of agri-food waste is a global concern that doesn't seem to be decreasing, continuously creating problems for the economy, environment, human health, and society. The carefully monitoring the food chain and making better use of renewable resources, can be an effective strategy in the food waste management field [1]. Reducing food waste generation, utilizing the food waste for reuse, and valorising food waste for the recovery of natural bioactive compounds are the key pillars of the food waste management system (Fig. 1). As such, by lowering the environmental effects of resource usage and increasing food waste utilization, the circular economy has the potential to protect the valuable and scarce non-renewable resources [2]. Agri-food wastes and biomass must be transformed into a variety of end products and bioactive substances, including food additives, feedstock, biopolymers, biobased chemicals, bioenergy, and biofuels, in order to functionalize sustainable bioeconomy model [3].

Artificial colorants are frequently developed from petro-based sources to use in food, pharmaceutical, and cosmetic industry applications. Their production and application has detrimental impacts on human and environmental health due to their chemical origin. The switch from chemical based synthetic colorants to natural bioactive pigments produced from plants is therefore essential. Researchers have therefore been inspired to develop novel pigments that are green, natural, non-toxic, safe for human and environmental health with Generally Recognized As Safe (GRAS) status [4], and should be produced from renewable and sustainable resources. Recent research has concentrated on the valorization of agri-food wastes as a sustainable resource of natural pigments while keeping in mind notions from the 'circular economy' model. Food waste, especially from fruits and vegetable processing industry provides a

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https://doi.org/10.1016/j.crgsc.2022.100340
Received 5 August 2022; Received in revised form 29 September 2022; Accepted 22 October 2022
Available online 27 October 2022

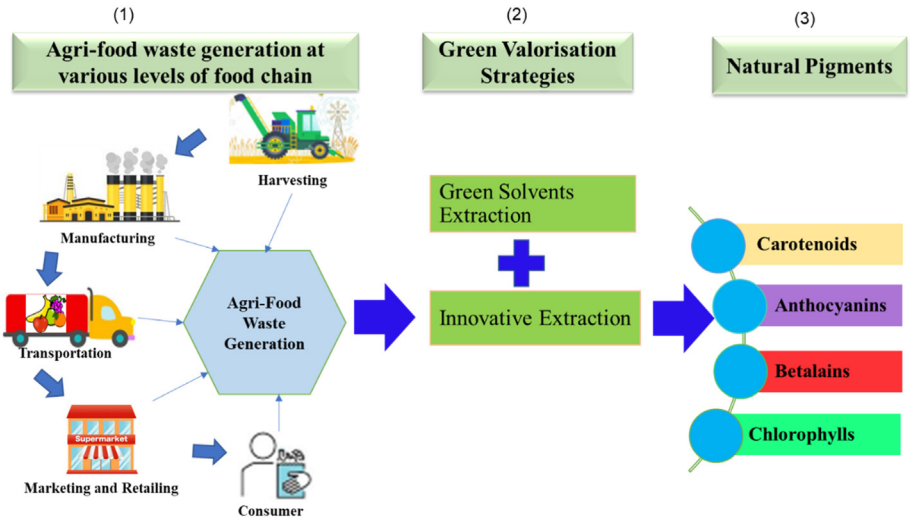

Fig. 1. Graphical representation for the concept of agri-food waste generation and green valorization strategies for the production of natural pigments. 1) Food waste is the term used to describe the loss of perishable food that occurs in the supply chain throughout post-harvest, distribution, transportation, processing, wholesale, retailing, market, food service, and household. This food waste is an effective potential renewable resource for the production of different bioactive compounds. Several studies established the cutting-edge technologies used to recover natural bioactive pigments and other desirable compounds from agri-food waste, 2) Green extractions methods that consume less energy, permit the use of alternative solvents, guarantee the production of safe and high-quality bioactive compounds, can be a better option to valorize the pigment potential of agri-food waste, 3) In response to consumer demand, significant new research have been conducted to investigate nature-derived safe food additives i.e., natural pigments with possible health benefits. The rising consumer's awareness towards safe food and ingredients, supports the extermination of synthetic food additives/pigments from the food chain due to their ill-effects on human and environmental health and motivate to evolve plant derived natural pigments. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

wide array of natural pigments (secondary metabolites), including carotenoids (yellow to orange), anthocyanins (blue to purple), chlorophylls (green), and betalains (dark red to pink) as shown in Fig. 1. Besides providing the colour to various food preparations, the natural bioactive pigments are potent antioxidants that have positive impacts on human health. Natural pigments production has great interest in food applications by enhancing the aesthetic appeal, consumer demand and market potential of the food products. The need for natural bioactive pigments driven by health-conscious consumers, because of being safe to human health, green (natural) nature, non-toxic, and having GRAS status. Therefore, it is essential for both economic and environmental reasons to produce bioactive pigments in a sustainable manner in order to guarantee their continued usage for present and future generations and to enable a viable rise in their use.

2. Traditional *versus* green extractions

Traditionally, natural bioactive pigments have been extracted from the food waste using time-consuming, expensive, and unsustainable approaches called conventional methods. The conventional extraction methods have several disadvantages as shown in Fig. 2. To overcome these disadvantages of traditional/conventional extractions, the novel or non-conventional extraction technologies are continuously being investigated. In this regards, green extraction technologies emerged as an

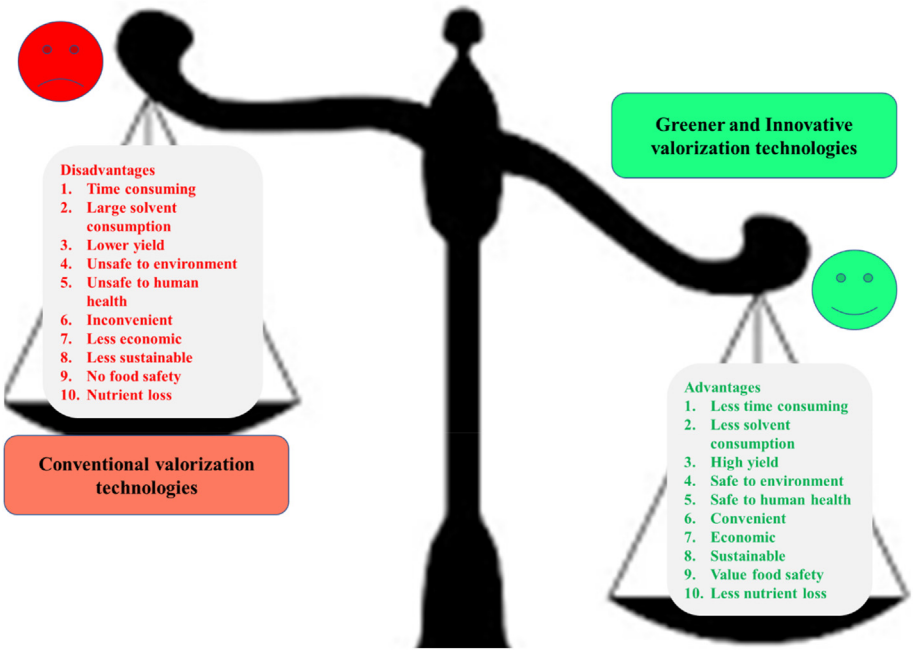

Fig. 2. Advantages of green and innovative extraction over conventional extraction technologies. The conventional extraction methods have several disadvantages, such as time-consuming, high solvent consumption, low extraction yield, high energy consumption, unsafe to human and environmental health. To sustain and support the green chemistry and green circular economy concept, green extraction technologies have several advantages, a) utilizing innovative technologies – novel, b) using substitute green solvents – green, c) decrease energy usage – economic, d) generation of by-products instead of waste – supports zero waste concept, e) less solvent & time consuming, provide high yield – efficieient, f) preserve the bioactivity of the extracted compound without producing contaminants & lesser nutrient loss – food safety, g) safe to human and environmental health. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

effective substitute to replace the traditional extractions, by exploiting their enormous advantages and potential in food, pharmaceutical and cosmeceutical fields. With the ability to boost the extraction yield while reducing extraction time, energy use, and solvent consumption, these technologies proved to be a viable substitute for conventional extraction methods.

Application of volatile organic solvents in phytopigment extractions like hexane, methanol, and chloroform are widely utilized in various industrial sectors, but due to their volatile and toxic nature, they pose harmful impacts on the handling workers (unsafe to human) and environment (environmental toxicity). Alternative or green solvents are thus needed to replace toxic solvents, as more environmentally friendly ones. Currently, ionic solvents (ILs), deep eutectic slovents (DESs), ethanol, water, edible oils, inert gases (CO_2) etc., are generously preffered by the scientists to perform green extractions for natural pigments. The benefits of the green substitutes to the traditional organic solvents, include better extractant (enhance extraction yield, reduce the time of extraction, increased thermal stability of the target compounds), safe to human (no harmful impacts on human health) & environmental health (increase process safety due to a reduction in the solvents' negative environmental effects), and more economic (can be recovered and reuse).

3. Green valorization technologies for plant-derived natural pigments

A variety of cutting-edge strategies have been explored for the extraction of potential bioactive food pigments from agri-food wastes, including ultrasonic assisted extraction [5], ohmic heating [6], microwave assisted extraction [7], high voltage discharges [8], pulsed-electric field assisted extraction [8], pressurized liquid extraction, light stresses, supercritical fluid extraction [9], enzyme assisted extraction [10], high pressure processing [11], and ionizing radiation etc., as shown in Fig. 3. The plant derived bioactive pigments such as carotenoids [7], anthocyanins [12], chlorophylls, lutein [9], and betalains [10], have been exploited from agri-food wastes using green extractions that showed a higher yield and enhanced stability of these natural pigments as compared to conventional extractions.

For the conservancy of green circular economy, green solvents like water [10], ethanol [13], edible vegetable oils [5,14], ionic liquids [15], deep eutectic solvents [16], aqueous solutions of bio-based surfactants, gases, supercritical fluids, and glycerol, etc. are gaining tremendous attraction in the area of greener extractions of natural bioactive pigments. For an environmentally friendly extraction of lipophilic colors, non-polar solvents with GRAS status, including cyclopentyl methyl ether, 2-methyl tetrahydrofuran, dimethyl carbonate, ethyl lactate, and ethyl acetate, can be used [17]. Ionic liquids and deep eutectic solvents stand out as potential replacements for volatile organic solvents among the alternative solvents because of their non-volatile, flammable and non-toxic nature. Depending on the ionic composition, ionic liquids can incur different properties and can be used to extract both hydrophobic (carotenoids) as well as hydrophilic (flavonoids and anthocyanins) compounds [18]. The ability to recycle and reuse is another benefit of ILs, helping to reduce its carbon footprint in the extraction processes and thus facilitating green circular economy concept [19]. There is a need to explore more green solvents or solvent free advanced extraction technologies to sustain the circular bioeconomy model in bioactive extraction field. Adopting multi-stage extraction technologies or integrated approaches, that are supported by various physico-chemical processes and designed specifically to extract a range of natural pigments, can be another advantageous opportunity in this field.

4. Green pigments valorization of agri-food waste and sustainable green circular economy

Environmental, economic, and social sustainability are regarded as the three core elements supporting the ideal sustainability [2,20]. The circular economy has been used to convert the value chain from linear to closed-loop and increase the efficacy of resource consumption in order to balance the economic, environmental, and societal costs caused by the current linear resource use [20]. Stimulating the shift to a circular economy to green circular bioeconomy is a top priority for the world, which encourages the promotion of resource-efficient and sustainable policies for long-term socio-economic and environmental advantages. In this regard, the valorization of food wastes and by-products can assist in fulfilling the Sustainable Development Goals (SDGs) of the UN, such as Goal 12 for effective resource management to actualize the attainment of green circular bio-economy principles, as shown in Fig. 4. The impression can reduce the environmental burden by managing, reducing, or recycling food waste to produce incredibly important bioactive natural chemicals [7]. Development of novel green extraction technologies of food waste valorization for natural pigments, further supports the SDGs' Goal – 9 (SDG 9 – Industry, Innovation, and Infrastructure), which can further be stated as "Inclusive and sustainable industrial development has been incorporated, together with resilient infrastructure and

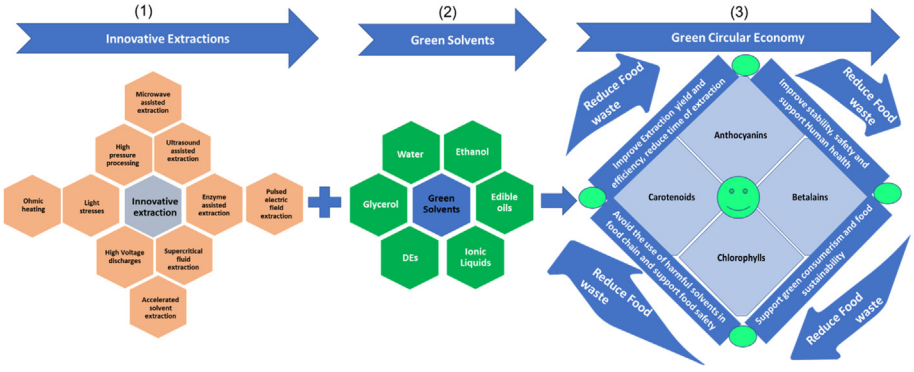

Fig. 3. Different integrated valorization approaches with innovative technologies and green solvents to achieve circular bioeconomy model. 1) The emerging innovative technologies can be integrated or applied in the combinatorial approaches synergized with green solvents to sustain the transition of 'circular economy' model to 'green circular bioeconomy' model. 2) To work under the auspices of green chemistry and green circular economy concepts, researchers are exploring green solvents alternative to conventional harmful solvents due to their ill-effects on human and environmental health. 3) The use of integrated approaches synergized with green solvents to extract natural pigments from agri-food waste, enhance the naturalness and stability of food pigments; improve the technological aspects such as increase the extraction yield, reduce the time of extraction, and reduce the solvent use or solvent free; promote the use of green solvents that harmless to human and environmental health, which consequently supports the circular bioecomomy. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

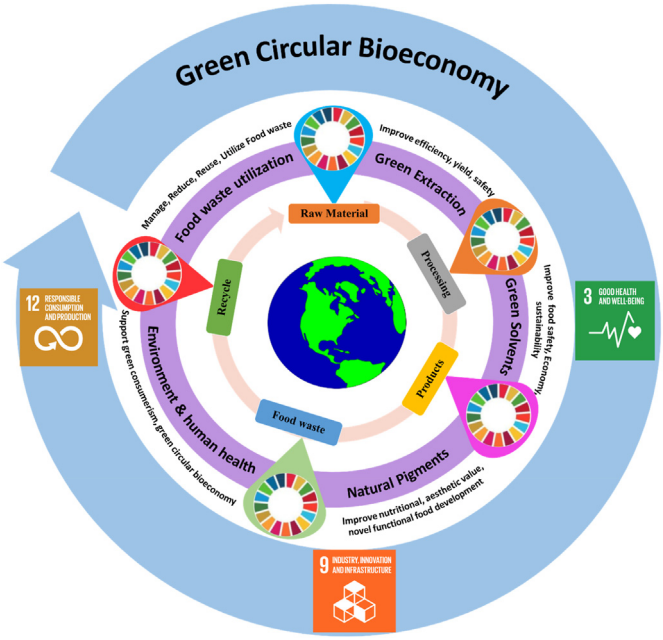

Fig. 4. Food waste utilization using green extractions supports green circular economy model. The utilization of agri-food waste as a renewable resource for the exploitation of natural bioactive pigments by using integrating innovative valorization strategies synergized with green solvents, providing all essential benefits to the *food chain ecosystem*, including, a) manage, reduce, reuse, and utilize the food waste – reduce food waste burden on environment; b) improve process efficiency & yield, and safety to human and environmental health – utilizing green extraction technologies; c) facilitate sustainable development of novel functional foods with enhanced biofunctional attributes by incorporating natural pigments (enriched antioxidants); d) support food economy – enhance market potential by improving aesthetic value of food; and, e) support SDGs 3, 9, and 12, food safety, food security, food nutrition, food economy, food sustainability under the aegis of circular bioeconomy model. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

innovation". Even if the sustainability concept is frequently brought up by authors, speakers, and governments when it comes to industrial processes and services, the lack of a precise definition continues to be a significant barrier to effective communication between regulatory bodies, business, academia, and governments [2]. The implementation of the green circular economy would increase and broaden the markets for bio-based goods. The key tenet of the green circular economy is the compatibility with the biorefineries that make it easier to recycle and reuse the materials in order to build holistic, inclusive and sustainable strategies to resource utilization [20].

This holistic approach further supports the SDGs' 'Goal 3 – Good Health and Well-being' by developing functional and nutraceutical foods enriched with agri-food waste derived bioactive pigments, which facilitate the generation of novel or future foods with enhanced nutritional value, aesthetic value, antioxidant potential, and safety (human and environmental health). On the basis of exploiting their potent antioxidant nature, the agri-food waste derived natural pigments play various biological roles to support human health, including anti-inflammatory (by cytokine signalling pathway), antioxidant & anti-cancerous (through free radical scavenging activity), antimicrobial, cardioprotective, antithrombotic (by using mitogen-activated protein kinase mechanism), promoting ocular and neural health, and preventing non-communicable diseases (by using cyclo-oxygenase mechanism) [4]. Plethora of research have been explored on different green extraction technologies to exploit their extraction potential for natural pigments from agri-food waste and incorporate these pigments in food products to enhance their nutritional, sensorial, textural and antioxidant potential. Moreover, several studies discovered the micro/nano encapsulated delivery systems of natural pigments aiming to enhance their stability in food formulations, as the stability is the vital aspect of natural pigments in aqueous food formulations while incorporation and food processing. In order to protect the natural pigments from numerous processing and environmental risks, such as moisture, oxygen, temperature, light, etc., encapsulation technologies are able to entrap the pigments in biopolymeric coatings to stabilize them in the food system. Several researchers studied the potential of various encapsulating techniques such as spray drying, emulsification, electrospinning-nanofibers, freeze-drying, or complex coacervation etc. [21–24], to preserve and improve the bioactivity, colour, *in vitro* & *in vivo* bioavailability, and stability of various natural pigments. Incorporating agri-food waste-derived natural bioactive pigments into food products can promote the sustainable development of functional foods. However, there are limited studies contemplated incorporating encapsulated-natural pigments from agri-food waste in food formulations. Thus, there is need to explore effective resource utilization, novel encapsulation strategies, feasibility, qualitative and quantitative analysis during pigment incorporation in food, regulatory guidelines and consumer perception on food-waste derived natural pigment's incorporation in food formulations, that can ultimately support the green circular economy concept.

5. Conclusion and future prospects

Natural bioactive pigment's extraction requires intricate procedures which can be carried out using combination of various methods, that can be suitable for the extraction of a variety of pigments and green solvents. Green extraction ideas and practices are recognized with great prospects for achieving the industrial sector's sustainability targets as well as possible partnering in overcoming the present challenges. To achieve the green circular economy or sustainable circular bioeconomy model in the area of agri-food waste valorization for bioactive pigments, there is a need: a) to discover newer food waste resources of pigment potential, b) to establish & explore more advanced green valorization technologies with enhanced extraction yield, low solvent consumption or solvent free strategies, c) to reconnoitre novel green solvents with improved efficiency, and d) redesign the integrated or combinatorial approaches to support post-extraction zero-waste concept and generate minimal greenhouse gas emissions (GHGs). According to the literature review, ILs, eutectic solvents, edible oils and surfactants are suitable replacements for conventional organic solvents, in the extraction of natural colors from various agri-biomass waste matrices. However, in case of green solvents, the assessment of effective process integration should be necessary, particularly concerning solvent recycling and life cycle analysis of the entire process, as these will have a significant impact on the assessment of the economic and environmental impact, ultimately resulting in the sustainability of the colorant application in food system. Additionally, combining work on the development of balanced strategies and collaboration between academia and industry is required to establish sustainable approaches of natural pigment's extractions and applications. To generate and obtain high value-added (profitable and highly effective) products (natural pigments from agri-food waste), these balanced strategies must incorporate effective procedures. In the end, public policies, regulatory and ethical aspects emphasizing industrial sustainability must be debated, with the need for competitive production of natural colorants being claimed.

CRediT authorship contribution statement

Minaxi Sharma: Conceptualization, Writing – original draft, Data curation, Validation, Visualization, Writing – review & editing. **Kandi Sridhar:** Validation, Visualization, Data curation, Writing – review & editing. **Vijai Kumar Gupta:** Validation, Visualization, Data curation, Writing – review & editing. **Praveen Kumar Dikkala:** Visualization, Data curation, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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