**A review on Green Solvents for Sustainable Pigment Extraction from Floral Sources**

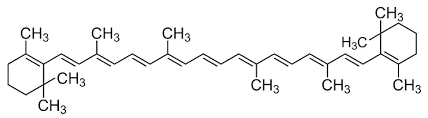
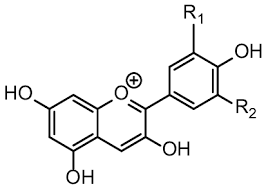
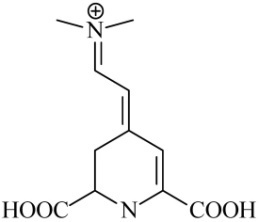
**ABSTRACT**

The study of extracting colorants from flowers appeals to researchers because flowers generate visually appealing results while offering health benefits and industrial utility in food production and medical fields. The usage of modern extraction systems which require toxic organic solvents creates both environmental risks and health concerns. This study evaluates the effectiveness of natural deep eutectic solvents (NADES), ionic liquids, and supercritical fluids as eco-friendly alternatives to toxic solvent systems used for pigment extraction. The solvent systems meet green chemistry protocols while providing more effective solutions that protect health and the environment. Pigment quality remains stable while output increases thanks to extraction methods combined with sonication, microwave and enzyme technologies. New research on carotenoids, anthocyanins and betalains with updated extraction techniques shows how green solvents fulfill their potential role in pigment extraction. The current laboratory evidence shows limitations as scientists struggle to scale up production volumes for profitable operation methods which also adhere to industrial specifications. Making sustainable industrial methods effective for pigment extraction from floral sources needs the resolution of both technical and commercial obstacles.

**Keywords:** Pigment extraction, Green solvents, Deep eutectic solvents (NADES), Sustainable methods, Industrial scalability

**INTRODUCTION**

The things which play importance in both aesthetic and industrial application are the natural pigments e.g. carotenoids, anthocyanins and betalains (Fig 1.) These compounds create a very good visual representation of flowers but also have proven health benefits and are making their way into food, cosmetics and pharmaceutical use. Carotenoids are vital for photosynthesis and antioxidant properties and play a role in human health, (Nabi *et al*., 2023). They are widely used in food industry for color and health effects, as well as in cosmetics for their protective effects of photooxidation. Anthocyanins are important for their rich colors and health benefits, such as anti-inflammatory and anti-cancer properties (Sousa, 2022). The applications of these colorants are in food, cosmetics and textiles, especially in accordance with preference of consumer for natural colorants (Nabi *et al*., 2023).Betalains are naturally red-violet and yellow pigments gaining attention for their effect on health (Qaisar *et al*., 2019). Although the effects of these natural pigments are known; there are difficulties standardizing extraction methods and quality control over the various applications that may restrict their wider use in industries. The conventional extraction methods applied in their production are very expensive, not only because of the enormous amounts of solvents, but also as a result of the accumulation of environmentally harmful waste (Rodríguez *et al.,* 2023). Less stability and limited diversity in terms of color shade are still problems of natural colorants (Delgado et al., 2003). These drawbacks can be optimized by using environmentally friendly and selective extraction processes that provide a balance between potency, safety, and stability of the resulting extracts. One of the six principles of green extraction is the selection of the proper extraction solvent (Chemat *et al.*, 2012), this is also a very important contributor to the selectivity of the extraction method and the stability of the colorant prepared. The present work reviews the use of green solvents for sustainable pigment extraction from flower crops, their environmental impacts and challenges.

**Carotenoids Anthocyanins Betalains**

**Figure 1:** Major pigments found in flower crops

**GREEN SOLVENTS**

Green solvents are environmentally friendly alternatives to conventional solvents used in chemical processes (Bubalo *et al*., 2015). Their main target is minimizing the environmental impact, health hazards and associated safety risks of solvent use in industrial production (Capello *et al.* 2007). Water, ionic liquids, supercritical CO2, biosolvents, organic carbonates and deep eutectic mixtures are some of the green solvents. Selection of green solvents involves their physical properties, chemical reactivity and environmental impact across its entire life cycle (Reichardt, *et al* 1988; Capello, *et al*, 2007). In general, simple alcohols such as methanol and ethanol or alkanes such as heptane or hexane are regarded as environmentally favourable solvents (Capello *et al*., 2007). Green chemistry embraces new rules for the development of sustainable and environmentally benign chemical processes, in which one of the important aspects is the use of green solvents (Bubalo *et al*., 2015)

**PRINCIPLES OF GREEN CHEMISTRY**

Green chemistry is an approach of chemical processes and products which produces and minimises the use and generation of hazardous substances (Abdussalam-Mohammed *et al*., 2020; Ivanković *et al*., 2017). It is founded on several principles that shape the design of environmentally benign substances and processes in their entire life cycle. Preventing waste, maximizing atom economy, using safer chemicals, designing energy efficient processes, incorporating renewable feed stocks, and reducing derivatives are these principles (Abdussalam-Mohammed *et al*. 2020). Applications of green chemistry range from organic, inorganic, biochemistry and polymer chemistry. Catalysis, biocatalysis, and the use of alternative renewable feedstocks, reaction media, and conditions are important key trends in green chemstry (Ivanković *et al*., 2017). Formation of green chemistry principles is related to achieving both economical and environmental protection and this is meant to achieve sustainable development and tackling the global challenges (Mammino, 2022; Hassan & Saleh, 2021).

**SOLVENT SELECTION GUIDE**

Prat *et al*. (2016) has prepared CHEM21 selection guide, which offers a complete ranking of classical and less common solvents according to a safety, health and environmental criteria consistent with global regulations. This work expands on existing efforts, including Sanofi's solvent selection guide that ranks solvents according to four classes, from 'recommended' to 'banned' just like GlaxoSmithKline developed an expanded solvent selection guide of 110 solvents with process safety factors and life cycle assessments (Henderson *et al*., 2011). The objective of these guides is to assist chemists to choose sustainable solvents for the pharmaceutical and chemical processing. Literature reviewing the ecological and economic significance, and application in commercial or academic practice, of green solvents further stress the relevance of such solvents (Nelson, 2003). Taken together, these selection guides constitute an essential first step towards less carbon intensive chemical processes in industrial sectors.

**TYPES OF GREEN SOLVENTS**

Green solvents are environmentally friendly alternatives to petrochemical solvents being developed in accord with green chemistry principles (Benvenuti & Santos, 2021). According to Naziri Mehrabani *et al*. (2022) they are categorized into several types, such as bio sourced solvents, water, ionic liquids, deep eutectic solvents, green synthetic organic solvents and supercritical fluids. This is because these solvents are easily accessible, low toxicity, and possibly reusable (Talele&Shahare, 2020). Green solvents have been employed in various technological fields (e.g. remediation technologies, polymeric membrane fabrication, pharmaceuticals, Benvenuti & Santos, 2021; Talele & Shahare, 2020), and the list is growing with the pressing need for restoration and environmental sustainability. The physicochemical properties of the green solvents have been modeled and classified using the COSMO-RS approach to facilitate the selection of appropriate green solvents in comparison with conventional solvents (Moity*et al*., 2012). In this classification, the possible other solvents listed alongside questionable solvents are identified as potential candidates, while new green solvents should be developed in areas where the alternatives are not shown.

Boiling point is extremely important when choosing green solvents for pigment extraction since these polar, sensitive compounds must be protected. The low boiling points make the extraction process less intensive because it is accomplished at milder temperatures, so the extraction step occurs at lower temperatures than can be used to extract the volatile and thermolabile pigments like carotenoids and chlorophylls. The extraction by novel green chemistry principles promotes safer and more efficient extraction methods as solvents having boiling below about 80°C minimize the loss of volatile compounds during extraction

thereby resulting in high yields of sensitive pigments(Ozel &Göğüş, 2014). For example, supercritical fluids and subcritical water extraction preserve the quality of extracted pigments and run at lower temperatures. Selective pigment extraction can be accomplished with solvents of lower boiling points compared with solvents of higher boiling points ensuring extraction of the desired pigment without accompanying unwanted compounds (Yoshikatsu *et al*., 1980). It has been noted in pigment extraction from microalgae, but in this case the solvents are boiling below 150 °C in order to achieve the simultaneous isolation of chlorophyll and carotenoids (Yoshikatsu *et al*., 1980). Often green solvents with low boiling points are less toxic and environmentally less harmful than traditional organic solvents which is in line with sustainability goals (Viñas-Ospino *et al*., 2023). This transition to these solvents cuts back on exposure to volatile organic compounds (VOCs). However, low boiling points are a virtue but they may impede extraction of high boiling compounds, and a judicious selection of solvents may be needed to achieve a joint optimization of the recovery of compounds and product quality.

**ADVANTAGES OF GREEN SOLVENTS FOR FLOWER PIGMENTS EXTRACTION**

Preserving the natural color and integrity of flower pigments has been demonstrated using green solvents. Natural deep eutectic solvents (NADES) are more stable for natural colorants than are water or ethanol solutions (Dai *et al*., 2014). There is a strong interaction between the natural colorants and the solvent. The flowers have been preserved using modified method which includes ethyl alcohol and polypropylene glycol, color and texture of which are naturally preserved for longer periods (Ito *et al*., 2010). The combination of green solvents like water, supercritical fluids, NADES and ionic liquids with various extraction techniques can optimally stabilise and increase the diversity of natural colorants such as carotenoids, anthocyanins and betalains (Tzanova *et al*., 2024). In addition to these traditional preservation methods using chemicals such as copper sulphate, formalin and silica gel, attempts have been tried to retain natural color pigments in botanical specimens using some chemicals (Verma, 2008). But green solvent approaches have environmentally friendly aspects that can help preserve flower pigments in their brilliance.

Lutein is high in marigold flower and the natural yellow-orange pigment is used as food coloring and nutrient supplement (Kashyap *et al*., 2022, Sowbhagya *et al* 2004). Traditionally, the solvents used for extraction include toxic ones like hexane; they leave residues not suitable for food use (Ballentine, 2020). Nevertheless, there has been research to develop non-toxic and greener extraction process to assure food safety and satisfy consumers' need for natural colorants (Aberoumand, 2011; Kashyap *et al*., 2022). The green solvent 2-methyltetrahydrofuran was shown to extract lutein with yield (2.56%) and purity (97.33%) superior to the conventional methods (Kashyap *et al*. 2022).

There have been recent research into eco friendly practices for pigment extraction from agricultural and floral waste. Recently, natural deep eutectic solvents (NADES) have been a sustainable alternative for the extraction of pigments and antioxidants from agri-processing waste (Kumar & Brooks, 2021). Although vegetable oils are another green solvent choice for carotenoid extraction, they possess food and environmental safety compared to product-based solvents (Tiwari *et al*., 2022). Sustainable carotenoid extraction is feasible through the use of innovative extraction technologies, including ultrasound, microwaves and enzyme assisted, combined with green solvents like ethyl lactate and ionic liquids (Morón-Ortiz *et al*., 2024). These ecofriendly methods also apply to microbial pigments, a new area of interest as possible alternatives to synthetic dyes. Recently, ultrasound assisted extraction and ionic liquid assisted extraction have been employed to extract efficiently and environmentally friendly pigment from microbial sources (Rajendran *et al*., 2023). Nevertheless, the scalability of these laboratory scale processes to industrial applications remains difficult.

Green solvents have been shown promising for preservation and stabilisation of natural pigments from flowers when stored. The various preservation techniques used in preserving natural color pigments in botanical specimens have included treatment with copper sulphate solutions and formalin based mixtures (Verma, 2008). Natural deep eutectic solvents (NADES) have been shown to stabilise pigment molecules of safflower more effectively than in water or ethanol solutions and this is attributed to strong hydrogen bonding interactions (Dai *et al*., 2014). Ethanol at 4–10% concentrations extended the vase life of bouquet flowers by up to 2 days, delaying the wilting and senescent of petals, and maintained higher chlorophyll fluorescence intensity (Hossain *et al*., 2008).

**Table 1: INVESTIGATING GREEN SOLVENTS FOR SUSTAINABLE PIGMENT EXTRACTION FROM FLOWERS**

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Flower** | **Pigment** | **Solvent Used** | **Extraction Method** | **Yield** | **Reference** |
| *Amaranth (A. cruentus)* | Betalains | Water | Solid-liquid extraction | 4.8 mg/g | Howard *et al.*, 2022 |
| *Amaranthus hypochondricus*L*.* | Betalains | Water | Solid-liquid extraction | 0.2–0.4 mg/g 0.2–0.6 mg/g | Lopez *et al.*, 2023 |
| *Bougainvillea glabra* | Betalains | 1:1 Methanol:Water | Solvent extraction | Absorbance = 0.534 at 548 nm | Ibrahim*et al.,* 2019 |
| *Catharanthus roseus* | Anthocyanins | Lactic acid–glucose 1,2-propanediol–choline chloride | Stirring, 40 °C; 30 min | No data | Dai *et al.*, 2016 |
| *Carthamus tinctorius* | Red pigment (Carthamine) | Hydrogen Peroxide (H₂O₂) + Degassed Bicarbonate | Oxidant pretreatment + Degassed solvent extraction | Higher Yield: Effective for red carthamine pigment extraction | Ghorbani *et al.*, 2015 |
| *Clitoria ternatea* L. | Anthocyanins (Ternatins) | Aqueous | Aqueous Extraction | TMAC: 7925.29±36.07 mg/L, Polymeric Color: 25.28±1.14% | Netravati *et al*., 2022 |
| |  | | --- | | Dried Rose (*Rosa spp*.) |  |  | | --- | |  | | Anthocyanins | LacticAcid:Choline Chloride (1:1) | Ultrasound-Assisted Extraction | 826.59 ± 17.35 mg/l | Prakash *et al.,* 2024 |
| *Gomphrena globosa* L | Betalains | Water | UAE; 500 W, 22 min; 5 g/L ratio | 161 mg/g DM | Roriz *et al.*, 2020 |
| *Hibiscus sabdariffa* | Anthocyanins | Citric acid/ethylene glycol with a 1:4 M ratio | MAE; 550 W 50% water (*v*/*v*), 180 s | 3 mg/g DW | Kurtulbas*et al*., 2020 |
| *Hibiscus sabdariffa* L. | Anthocyanins | 39.1% ethanol in water, *v*/*v* | UAE, 296.6 W, 26 min; 30–35 °C; 30 g/L ratio | 24 mg/g DM | Pinela *et al.*, 2019 |
| *Hibiscus sabdariffa* L. | Anthocyanins | 15% acetic acid (*v*/*v*) | Stirring, 25 g/L ratio, ambient temp., 48 h | No data | Segura-Carretero *et al.*, 2008 |
| *Ixora javanica* | Flavonoids, Anthocyanins | Choline chloride + propylene glycol (1:1) | Green ultrasound-assisted deep eutectic solvent extraction | 33 mg quercetin equivalent/g dried sample | Oktaviyanti, N. D., *et al*. (2019) |
| *Rosa ardsrovar* (Red Rose) | Anthocyanins (cyanidin, pelargonidin) | Ethanol | Ultrasound-assisted extraction | 6.79% | Amir-Al Zumahi *et al.,* 2020 |
| *Tagetes erecta* L. | Lutein | ChChl/glucose | No data | 80% 369 mg/g FW | Dharani *et al.*, 2022 |
| *Tagetes erecta* L*.* | Lutein | MeTHF, hexane, tetrahydrofuran, acetone, and 1,4- dioxane | CSE(Conventional Solvent Extraction) | 97.64% | Kashyap*et al.*, 2022 |
| *Tagetes erecta L. (*Marigold*)* | Lutein | Choline chloride + glucose (ChCl+glucose) | Deep Eutectic Solvents (DES) | 368.64 g/kg | Dharani *et al***.** (2022) |

**INTEGRATION WITH SUSTAINABLE EXTRACTION TECHIQUES**

Recent research has brought forward the growing demand for green extraction techniques of bioactive compounds due to environmental concern and a need for the sustainable practice (Muhammad Usman *et al*., 2023). Promising results have been demonstrated by various eco-friendly methods, such as supercritical fluid extraction, ultrasound assisted extraction or enzyme assisted extraction. Traditional solvents, such as deep eutectics solvents or bio based solvents, have shown promise to improve extraction efficiency (Muhammad Usman *et al*., 2023; Beshare Hashemi *et al*., 2022). Synergistic effects of various extraction techniques complementing each other have been combined e.g. ultrasound microwaved assisted extraction to get improved yields with short extraction times (Muhammad Usman *et al*., 2023; Manoj Kumar *et al*., 2023). When scaled up to industrial scale, these green approaches also present cost benefits and high purity potential (Manoj Kumar *et al*., 2023). However, the adoption of these gas detection techniques is hindered by standardization, selectivity and economic viability (Muhammad Usman *et al*., 2023; Naqvi *et al*., 2021).

One approach to reducing both solvent consumption and environmental impact in chemical process is by using green solvents. Compared to conventional solvents, containing and recycling them result in environmental contamination (Desimone, 2002). To overcome these issues, green solvents including ionic liquids, supercritical fluids and biosolvents are developed (Welton, 2015; Shanab *et al*., 2013). Nevertheless, both the technical and commercial performance (Welton, 2015) have to be considered for successful implementation of sustainable processes. Researchers have developed methods to calculate a 'greenness index' based on environmental parameters and solvent amounts (as measured in grams) in order to evaluate and compare solvent greenness (Stewart Slater & Savelski, 2007). With this approach, scientists and engineers can choose alternatives to greener solvents or processes. Solvent free processes and improved recycling protocols have been explored, however, they have limitations and further development and practical application of environmentally benign alternative solvents (Desimone, 2002; Shanab *et al*., 2013) is still required.

**ECONOMIC AND ENVIRONMENTAL IMPACTS**

Temple and floral industries discard flower waste posing environmental problems but they can be transformed into valuable products through green approaches. Several methods have been explored for exploitation of these wastes, such as through solid state fermentation for compost and bioethanol, biogas, dyes, and biosurfactants (biological surface active agents) (Bennurmath *et al*., 2021). The technique of ultra sonication of natural dye extraction has resulted in producing fabrics colors for fabric dyeing and the waste producing residue was further employed in vermicomposting and biochar production (Singh *et al*., 2017).

Sustainable practices in several industries including floriculture require green solvents. The motivation for these solvents is to decrease environmental pollution, energy consumption, and the hazard to health and surrounding environment suffered by traditional organic solvents (Clarke *et al*., 2018; Welton, 2015). As for sustainable solvent alternatives, water, supercritical carbon dioxide and ionic liquids, deep eutectic solvents, bio-derived solvents such as 2-methyltetrahydrofuran and glycerol (Kerton, 2016) are just some examples. The main advantage of these green solvents is that they have lower vapor pressure, reduced flammability and lower toxicity (Kerton, 2016). Moreover, the commercial performance has to be taken into account when environmentally sustainable processes are successfully implemented (Welton, 2015). Like many other properties, solvent free processes and more efficient recycling protocols have been developed, but this has certain limitations that have forced researchers to look for environmentally benign solvents (Desimone, 2002). In terms of chemical production processes, the selection of appropriate solvents can provide a substantial boost to the processes' sustainability and contribute to the more eco-friendly practices in any supply chain that involves floriculture (Clarke *et al*., 2018; Welton, 2015).

**APPLICATION OF FLOWER PIGMENTS EXTRACTED USING GREEN SOLVENTS**

In the textile industry, natural dyes are becoming more popular as eco friendly substitutes of synthetic dyes. Derived from plant sources these dyes are biodegradable, non toxic and ecologically friendly (Jordeva *et al*., 2020). Similar to other textile materials such as cotton, silk and wool, they can be used on most textile materials mordanting them in different ways to improve color fastness (Sanjeeda and Taiyaba, 2014; Gupta, 2019). The natural dyes provided a wide range of color shades and also show fair to excellent fastness properties in the studies (Iqbal & Ansari, 2014). Natural dyes often require mordants to fix, some of which may not be eco friendly (Gupta, 2019); their advantages are that they are non allergic and non carcinogenic. The challenge notwithstanding, use of natural dyes opens avenues for creating new markets and employment, supporting current environmental and economic concerns (Jordeva *et al*., 2020; Ado *et al*., 2015).Cosmetics, food and textiles are being explored to use colors obtained from plant such as anthocyanins, carotenoids, betalains.

**CHALLENGES IN ADOPTING GREEN SOLVENTS**

Different flower pigments are extracted differently depending on their solubility in different solvents. Carotenoids and chlorophylls absorb with characteristic curves in solvents of increasing polarity, for which equations are metrologically developed for their quantitative determination (Lichtenthaler & Wellburn, 1983). Strong hydrogen bonding interactions (Dai *et al*., 2014) account for the improved stability of natural deep eutectic solvents (NADES) for safflower pigments when compared to water or ethanol. Extraction yields and selectivity for natural pigments using alternative solvents such as ionic liquids, eutectic solvents and surfactant solutions are aligned with sustainability goals (de Souza Mesquita *et al*., 2020). Important ecological roles have been played by the three major groups of floral pigments, betalains, carotenoids, and anthocyanins, and they have been targets for biotechnological modification (Grotewold, 2006). However, solvent compatibility, and implementation of novel extraction methods, can help improve recovery and stability of these valuable natural colorants.

However, scaling green solvents for industrial use faces economic challenges, including high costs, complicated product recovery, and recyclability issues (Usman *et al*., 2023; New *et al*., 2022). Additional challenges include biomass feedstock sourcing, cost fluctuations, and limited compatibility with existing processes (Usman *et al*., 2023). To address these challenges, future research should focus on developing alternative feedstock sources, improving solubility and separation techniques, optimizing process parameters, and designing cost-effective equipment (Usman *et al*., 2023). Despite these obstacles, green solvents have the potential to significantly impact industrial chemical processes, offering environmental and health benefits over traditional solvents (Sowbhagyam, 2024).

**EMERGING TRENDS AND FUTURE DIRECTIONS**

Eco friendly alternatives to traditional organic solvents has become a recent focus of novel solvent systems. Cyclopentyl methyl ether (CPME) has surfaced as a versatile, sustainable solvent for biotechnology and biorefineries (de Gonzalo *et al*., 2019). As low cost, easy preparation and renewable raw materials, DESs are a highly desirable class of solvents for a variety of biocatalytic processes (Xu *et al*., 2017). DESs and ionic liquids have been promising to participate in biomedical applications; drug solubilization, pharmaceutical delivery and protein stabilization (Curreri *et al*., 2021). Natural DESs have proven effective in lignocellulosic biomass processing in hydrolyzing of lignin carbohydrate complexes and improving enzymatic digestibility (Satlewal*et al*., 2018). All these novel solvent systems, which are low toxic, biodegradable and recyclable, adhere to sustainable chemistry principles. Research aimed at optimizing their properties and expanding their use in these and other fields such as biotechnology, biorefineries and biomedicine, is ongoing.

By using green solvents, natural plant pigments can be extracted from agri-food waste, which are sustainable alternative to synthetic colorants, which contributes to circular economy (Sharma *et al*., 2022). A practical example is to extract natural dyes from temple floral waste and vegetable peels by ultrasonication and spray drying. The extracted pigments can be used to dye fabrics, residual waste can be further utilized respectively for vermicomposting and biochar production in a closed loop system (Singh *et al*., 2017). In addition to waste management issues, this approach will lead to value added products as well as economies of scale, each generating environmental and economic benefits in the floriculture industry.

**CONCLUSION**

Green solvents offer a promising approach to sustainable pigment extraction in floriculture because not only might they simultaneously increase the extractability of polar and nonpolar compounds, they may also improve recovery rates of valuable bioactive compounds. Natural deep eutectic solvents (NADES) and vegetable oils are used as effective, non-toxic alternatives to conventional solvents retaining natural pigments and functional properties. In addition, ecofriendly extraction techniques, such as ultrasound assisted, microwave assisted, and enzyme assisted methods serve as viable replacements with traditional organic solvent based extraction methods. Although the laboratory studies have been successful, there are still challenges to scaling these methods for industrial applications. An integral part of assessing their overall sustainability will involve the integration of life cycle assessments. With the use of these, green solvents and advances in extraction techniques can open the door to more environmentally friendly, efficient strategies for pigment extraction conforming with the principals of a circular economy in the flower industry.

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Option 1:

Author(s) hereby declare that NO generative AI technologies such as Large Language Models (ChatGPT, COPILOT, etc.) and text-to-image generators have been used during the writing or editing of this manuscript.

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