Advancements in Natural Dyes Extraction

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The dyeing and finishing step represents a clear hotspot in the textile supply chain as the wet processing stages require significant amounts of water, energy, and chemicals. In order to tackle environmental issues, natural dyes are gaining attention from researchers as more sustainable alternatives to synthetic ones. The extraction of colorant from natural sources is a fundamental step in preparing purified natural dyes, as a plant's matrix contains only a small percentage of dye, usually in the range of 0.5–5%, and several other constituents such as water-insoluble fibers, carbohydrates, protein, chlorophyll, and tannins, among others. The selection of the most suitable extraction technique should be based on the evaluation of the nature and solubility of the dyeing materials.

natural dyes
textile
aqueous extraction
solvents
alkaline extraction
ultrasound-assisted extraction
microwave-assisted extraction
enzymatic extraction
supercritical fluids

1. Introduction

The global textile industry is responsible for having a serious environmental impact across the entire supply chain, with remarkable greenhouse gas emissions (over 3.3 billion metric tons per year) [1], significant land and water consumption, pollution of the soil, air and water, and increasing waste production.

The current linear system uses large amounts of resources, which creates significant negative impacts on ecosystems and people. It is estimated that every year, about 98 million tons of non-renewable resources are consumed, including, for example, fertilizers to grow natural fibers, oil to produce synthetic fibers, and chemicals used in different stages of textile production [2].

The dyeing and finishing steps represent a clear hotspot in the textile supply chain as the wet processing stages require large volumes of water to be heated and are especially energy intensive. The greenhouse gases emitted from burning fossil fuels to generate the heat and electricity required in these stages of textile production account for their high contribution to the climate impact. According to the UN Environment Programme (UNEP) report on sustainability and circularity in the textile value chain, the contribution of dyeing and finishing to the climate impact of the textile field is accountable for 36% of the entire textile supply chain [3]. The consumption of water is also a major issue, and finishing processes consume great amounts of water, for example to prepare dye baths and wash fabrics after the dyeing process. It is estimated that every kg of cotton requires around 125 L of water to be dyed and finished [4]. Moreover, the dyeing and finishing stage relies heavily on hazardous chemicals and represents a hotspot in terms of carcinogenic human toxicity and a hotspot for non-carcinogenic human toxicity because of the use of detergents, dyes, and water-repellent agents [5].

While promising innovations are emerging to reduce the impact of the dyeing step, such as low water or waterless dyeing processes and chemical-free technologies [6], there is a renewed interest in the topic of natural dyes as a more sustainable alternative to synthetic dyes and a way to reduce the usage of chemicals and the impact on the environment.

2. Aqueous Extraction

Aqueous extraction is a traditional method in which dye matter is usually first reduced to small pieces or powdered and then immersed in water to loosen the cell structure and improve the efficiency of the process. The dye solution is obtained by boiling and then filtered. The extraction and filtration process can be repeated several times [7].

Aqueous extraction is a sustainable and safe technique, and the extract can be easily applied to textiles. The disadvantages are its long extraction time, the large amounts of water required, and the low dye yield as only the water-soluble dye components are extracted. Moreover, sugars and other water-soluble components are extracted along with the dye. Yields of heat-sensitive dyes are reduced at high temperatures [8].

Pervaiz et al. [9] performed a simple extraction in aqueous medium to obtain dye through the valorization of marigold (*Tagetes erecta* L.) waste flowers by soaking petals for several hours, boiling them, leaving the solution to cool, and then filtering it through filter paper. Different extraction conditions were compared, and it was reported that the maximum extraction yield was obtained when extraction was performed at 40 °C for 40 min.

3. Solvent Extraction

Similar to aqueous extraction is extraction with organic solvents such as ethanol or methanol or a mix of solvents, which allows for a higher extraction yield, however. Lower temperatures can be used, limiting chances of degradation. Water/alcohol extraction can extract both water-soluble and water-insoluble components. Moreover, the solvents can be easily removed through distillation to be reused. Disadvantages include the presence of toxic residual solvents. Additionally, the extracted material is not readily soluble in water; the co-extraction of other substances such as chlorophylls and waxy materials could occur [8].

Al-Alwani et al. [10] compared the effectiveness of nine solvents, namely n-hexane, ethanol, acetonitrile, chloroform, ethyl ether, ethyl acetate, petroleum ether, n-butyl alcohol, and methanol to extract natural dyes from cordyline, pandan, and dragon fruit (respectively *Cordyline fruticosa, Pandannus amaryllifolius,* and *Hylocereus polyrhizus*), assessing the best extraction conditions. The results obtained showed that the most suitable solvents for the dye extraction of the plants considered were methanol, ethanol, and water.

4. Alkali or Acid Extraction

Similar to the previous techniques, extraction under alkali or acid conditions can facilitate the hydrolysis of glycosides with higher extraction yields—as many dyes are in the form of glycosides. Alkaline extraction is particularly suitable for dyes containing phenolic groups, which are soluble in alkali conditions. A disadvantage of this extraction technique is that alkaline conditions could damage the dyeing matter, as many dyes are pH sensitive [11].

Alkaline extraction of natural dye from grape pomace, a by-product of wine production, was performed using sodium hydroxide and compared to aqueous extraction. The results showed that a higher extraction yield was obtained through alkaline extraction. Moreover, different extraction conditions (sodium hydroxide concentration, grape pomace amount, extraction duration, and temperature) were studied to optimize the dyeing process of wool fabric with extracted dye [12].

5. Ultrasound- and Microwave-Assisted Extraction

In ultrasound-assisted extraction (UAE) and microwave-assisted extraction (MAE) the dye matter is treated with water or other solvents in the presence of ultrasounds or microwaves. These processes provide a better extraction yield, lower extraction temperatures, lower solvent usage, and less time and energy consumed. The possibility of using lower temperatures is more suitable for heat-sensitive molecules [13].

Ultrasounds are defined as mechanical waves characterized by a frequency above 20 kHz (human hearing range). The waves can propagate in solids, liquids, and gasses through compression and rarefaction cycles. When high intensity waves propagate in a liquid medium, the negative pressure during the rarefaction phase is stronger than the force attracting molecules together, causing molecules’ dispersion and the formation of cavitation bubbles. These bubbles grow until they collapse, generating the phenomenon known as cavitation, with an increase in temperature and pressure. Cavitation is an important mechanism exploited in the ultrasound-assisted extraction of bioactive compounds. As a matter of fact, the collapsing of bubbles causes a series of mechanisms such as erosion or pore formations, which can ultimately facilitate the breaking of plant matrix cells and the release and solubilization of compounds of interest [14].

Several authors have used ultrasound-assisted extraction to study the potentiality of new dye sources, investigating the best process conditions [15][16][17] for application on different textile substrates—natural, regenerated, and synthetic substrates.

Wizi et al. [18] individuated sorghum husk (an agro-industrial by-product) as a promising source of natural dyes for textiles due to the high amounts of phenolic colorants which are tightly bound to the cell walls, however, and therefore difficult to extract. Ultrasound technology was employed in combination with microwaves to increase the extraction efficiency. Moreover, the effects of different solvents were investigated. The extracts were subsequently used to dye wool and cotton. The results showed that a higher color strength was obtained when the extraction was performed with 70:30 ethanol:water mixture (*v*/*v*) with HCl. The dyed wool and cotton fabrics showed good fastness values to washing, crocking, and light. Additionally, it was noticed that extracts with different solvents led to different shades on wool and cotton fabrics.

Microwaves are included in the electromagnetic spectrum and are characterized by wavelengths ranging between 0.001 and 1 m and frequencies ranging between 0.3 and 300 GHz. Compared to conventional heating, heating through microwaves shows higher efficiency (up to 50%), resulting in significantly inferior energy consumption [19].

Unlike traditional heating techniques in which heat is transferred from the equipment to the solution, microwaves allow for direct heating of the solution, resulting in a faster process with a lower temperature gradient. Moreover, microwave-assisted extraction allow for the significantly lower consumption of organic solvents [20].

In the microwave-assisted extraction (MAE) of plant metabolites, the main mechanism induced by microwaves is the generation of heating and dipole rotation in organic molecules of the plant matrix. This leads to an increase in kinetic energy and friction between the ions, eventually causing the breaking of hydrogen bonding, as well as facilitating the penetration of the solvent in the vegetal matrix [21], resulting in a significant reduction in organic solvent consumption.

A better extraction yield was obtained through the microwave-assisted extraction of cinnamon bark as dye matter for biomordanted silk fabric. It was observed that by using an aqueous medium, microwave extraction enhanced the color strength (K/S). The type of medium greatly affected the results.

Chemat et al. [22] discussed the potential of ultrasound-assisted extraction in combination with microwave-assisted extraction (MAE), as microwave irradiation provides fast and efficient extraction but inhomogeneous heating. Combination with ultrasounds was presented as a solution to overcome this issue.

6. Enzymatic Extraction and Fermentation

Extraction through enzymes is considered an environmentally friendly technique to extract active compounds from plant matrices, avoiding the use of solvents. Enzymes act as catalysts and are used to extract, modify, and synthetize natural active compounds [23].

Appropriate enzymes such as cellulase, amylase, and pectinase are used to decompose plant tissues under mild conditions, helping the release of active compounds and increasing the speed of extraction. Temperature and pH are the main factors that affect the activity of enzymes [8]. These techniques are particularly suitable for hard plant materials such as the bark and roots [24].

Tiwari et al. [25] compared enzymatic extraction with pectinase and cellulase, ultrasound extraction and enzyme-assisted extraction, and the enzyme-mediated ultrasonic-assisted extraction of natural colorants from pomegranate rind. The dyeing behavior of te extracted dyes on cotton and wool was also investigated, and it was found that the combination of enzymes and ultrasounds gave the highest results in terms of color yield.

7. Supercritical Fluids

During the last two decades, supercritical fluids have gained popularity in the extraction of organic compounds from plant matrices due to several advantages [26].

Supercritical fluids are defined as substances above their critical pressure and temperature which possess properties of both liquids and gasses. The critical values depend on the specific substance. When a gas is above its critical temperature and pressure, it is compressed in a supercritical fluid and is characterized by a density similar to that of a liquid, a viscosity similar to that of a gas, and a diffusion coefficient between liquids and gasses. Thanks to these properties, supercritical fluids possess high solvating power and diffusivity and low viscosity and surface tension. These characteristics allow for fast mass transfer in supercritical fluids. Consequently, in extraction with supercritical fluids (SFE) the mechanism of penetration of the solvent into the matrix is facilitated, resulting in a fast and efficient extraction process. Another advantage of supercritical fluids is that the changing pressure, temperature, and density affect the solubility of these substances [27]. Additionally, the density of the fluid can be modified by altering the pressure and temperature values. Therefore, in the extraction process with supercritical fluids, the solvent's strength can be also regulated by modifying the various parameters. Supercritical fluids' extraction comprises two main steps, which are the solubilization of the extract in the supercritical solvent and the consequent separation of the extract from the solvent. In the first stage, the absorption of the supercritical solvent by the plant matrix causes the swelling of its cellular membranes and a decrease in mass transfer resistance. At the same time, the extracted compounds are solubilized and move to the external surface of the cell. Afterwards, the solubilized compounds are transported from the surface of the cell to the solvent. Finally, in the last stage, they are removed from the supercritical solvent [28][29].

Several solvents can be used as supercritical fluids, such as carbon dioxide, ethane, ethene, methanol, nitrous oxide, n-butene, n-pentane, sulfur hexafluoride, and water, although carbon dioxide (CO_2) is the most commonly used, and it is estimated that more than 90% of all supercritical fluid extractions (SFEs) are carried out with CO_2. This is due to several reasons: firstly, carbon dioxide is non-toxic and non-flammable, and it is considered safe for human health and the environment in terms of manipulation and between certain value ranges. Secondly, it is available at high purity, at relatively low cost, and is easy to remove from the extract. Moreover, it has a low critical pressure and temperature (73 atm and 31.2 °C, respectively), factors that favor the preservation of the bioactive compounds contained in the extracts [28][30].

The main disadvantage of the use of CO_2 as a supercritical solvent is its low polarity, which makes it an efficient solvent for the extraction of compounds with no or low polarity but ineffective at extracting polar compounds. Nevertheless, the addition of small amounts of so-called modifiers—polar organic solvents, such as methanol—has been proven an adequate strategy to improve the extraction efficiency of CO_2, amplifying its extraction range with the inclusion of more polar compounds [27][31].

In order to perform selective extraction of the desired compounds only, the complex interplay between thermodynamics (solubility compounds to be extracted or of the undesired compounds) and kinetics (mass transfer resistance) must be considered. In this context, microscopic analysis is a useful tool that allows for the identification of the mass transfer resistance in the structure of the matrix [32].

Overall, supercritical fluid extraction presents many advantages: it is an efficient process in terms of high yields and low extraction times, requires low or room temperatures, and uses solvents generally recognized as safe (GRAS); the extract does not contain residual solvent; and it is possible to directly couple the extraction process with analytical chromatographic techniques such as gas chromatography (GC) or supercritical fluid chromatography (SFC) [33][34].

The main disadvantages include the high cost of the technology, the inefficient extraction of polar substances [8], and risks to workers. As a matter of fact, systems that use fluids in supercritical conditions operate at very high pressure, which is much higher than atmospheric pressure. This aspect represents a potential danger for the workers involved in the process and requires the adoption of preventive safety measures and risk analysis [35].

Kabir et al. [36] used supercritical fluids to carry out an innovative dyeing process for PET fabric with curcuminoid dyes from turmeric, in which the simultaneous extraction of natural dyes and the dyeing process itself were carried out in the same supercritical bath using carbon dioxide. As for the parameters used, the temperature, pressure, and time were respectively set at 150 °C, 20 MPa, and 1 h. The resulting samples were compared with samples obtained through conventional dyeing with ethanol-extracted dyestuff, and it was found that the PET samples dyed with a supercritical carbon dioxide technique were characterized by the highest dye exhaustion, K/S, and fastness properties.

References

1. Quantis. Measuring Fashion: Insights from the Environmental Impact of the Global Apparel and Footwear Industries. Full Report and Methodological Considerations. 2018. Available online: https://quantis.com/wp-content/uploads/2019/11/measuringfashion_globalimpactstudy_quantis_2018.pdf (accessed on 24 July 2023).
2. Ellen MacArthur Foundation. A New Textiles Economy: Redesigning Fashion’s Future. 2017. Available online: https://ellenmacarthurfoundation.org/a-new-textiles-economy (accessed on 3 June 2023).
3. UN Environment Programme (UNEP). Sustainability and Circularity in the Textile Value Chain; Global Stocktaking: Nairobi, Kenya, 2020.
4. Turley, D.B.; Horne, M.; Blackburn, R.S.; Stott, E.; Laybourn, S.R.; Copeland, J.E.; Harwood, J. The Role and Business Case for Existing and Emerging Fibres in Sustainable Clothing: Final Report to the Department for Environment; Food and Rural Affairs (Defra): London, UK, 2009.

5. Sandin, G.; Roos, S.; Peters, G. Environmental Assessment of Swedish Clothing Consumption—Six Garments, Sustainable Futures. 2019. Available online: https://core.ac.uk/download/pdf/270109142.pdf (accessed on 3 June 2023).
6. Maxwell, D.M.; Andrew, L.; Ryan, J. The State of the Apparel Sector 2015 Special Report—Water a Report for the Global Leadership Award in Sustainable Apparel; The Sustainable Business Group: London, UK, 2015.
7. Merdan, N.; Seyda, E.; Mujgan, N.D. Ecological and sustainable natural dyes. In Textiles and Clothing Sustainability: Sustainable Textile Chemical Processes; Springer: Berlin/Heidelberg, Germany, 2017; pp. 1–41.
8. Saxena, S.; Raja, A.S.M. Natural dyes: Sources, chemistry, application and sustainability issues. In Roadmap to Sustainable Textiles and Clothing: Eco-Friendly Raw Materials, Technologies, and Processing Methods; Springer: Berlin/Heidelberg, Germany, 2014; pp. 37–80.
9. Pervaiz, S.; Mughal, T.A.; Khan, F.Z.; Hayat, S.; Aslam, A.; Shah, S.F. Environmental friendly leather dyeing using Tagetes erecta L. (Marigold) waste flowers. Int. J. Biosci. 2017, 10, 382–390.
10. Al-Alwani, M.A.; Mohamad, A.B.; Kadhum AA, H.; Ludin, N.A. Effect of solvents on extraction and adsorption of natural dyes extracted from Cordyline fruticosa and Hylocereus polyrhizus. Asian J. Chem. 2014, 26, 6285.
11. Gupta, V.K. Fundamentals of natural dyes and its application on textile substrates. In Chemistry and Technology of Natural and Synthetic Dyes and Pigments; Books on Demand: Norderstedt, Germany, 2019.
12. Baaka, N.; Ticha, M.B.; Haddar, W.; Hammami, S.; Mhenni, M.F. Extraction of natural dye from waste wine industry: Optimization survey based on a central composite design method. Fibers Polym. 2015, 16, 38.
13. Adeel, S.; Habib, N.; Arif, S.; ur Rehman, F.; Azeem, M.; Batool, F.; Amin, N. Microwave-assisted eco-dyeing of bio mordanted silk fabric using cinnamon bark (Cinnamomum verum) based yellow natural dye. Sustain. Chem. Pharm. 2020, 17, 100306.
14. Kumar, M.; Dahuja, A.; Tiwari, S.; Punia, S.; Tak, Y.; Amarowicz, R.; Kaur, C. Recent trends in extraction of plant bioactives using green technologies: A review. Food Chem. 2021, 353, 129431.
15. Ma, X.; Wei, Y.; Wang, S.; Zuo, X.; Shen, B. Sustainable ultrasound-assisted ultralow liquor ratio dyeing of cotton fabric with natural turmeric dye. Text. Res. J. 2020, 90, 685–694.
16. Sadeghi-Kiakhani, M.; Hashemi, E.; Gharanjig, K. Inorganic nanoparticles and natural dyes for production of antimicrobial and antioxidant wool fiber. 3 Biotech 2019, 9, 456.
17. Qadariyah, L.; Mahfud, M.; Sulistiawati, E.; Swastika, P. Natural dye extraction from teak leves (Tectona Grandis) using ultrasound assisted extraction method for dyeing on cotton fabric. In

MATEC Web of Conferences; EDP Sciences: Lisses, France, 2018; Volume 156, p. 05004.

18. Wizi, J.; Wang, L.; Hou, X.; Tao, Y.; Ma, B.; Yang, Y. Ultrasound-microwave assisted extraction of natural colorants from sorghum husk with different solvents. Ind. Crops Prod. 2018, 120, 203–213.
19. Tylor, M.; Atri, B.S.; Minhas, S. Development in Microwave Chemistry; Evalueserve: Zürich, Switzerland, 2005; pp. 5–7.
20. Chaturvedi, A.K. Extraction of Nutraceuticals from Plants by Microwave Assisted Extraction. Syst. Rev. Pharm. 2018, 9, 31–35.
21. Akhtar, I.; Javad, S.; Yousaf, Z.; Iqbal, S.; Jabeen, K. Microwave assisted extraction of phytochemicals: An efficient and modern approach for botanicals and pharmaceuticals. Pak. J. Pharm. Sci. 2019, 32, 223–230.
22. Chemat, F.; Rombaut, N.; Sicaire, A.G.; Meullemiestre, A.; Fabiano-Tixier, A.S.; AbertVian, M. Ultrasound assisted extraction of food and natural products. Mechanisms, techniques, combinations, protocols and applications. A review. Ultrason. Sonochem. 2017, 34, 540–560.
23. Gardossi, L.; Poulsen, P.B.; Ballesteros, A.; Hult, K.; Švedas, V.K.; Vasić-Rački, Đ.; Carrea, G.; Magnusson, A.; Schmid, A.; Wohlgemuth, R.; et al. Guidelines for reporting of biocatalytic reactions. Trends Biotechnol. 2010, 28, 171–180.
24. Hou, X.; Chen, X.; Cheng, Y.; Xu, H.; Chen, L.; Yang, Y. Dyeing and UV-protection properties of water extracts from orange peel. J. Clean. Prod. 2013, 52, 410–419.
25. Tiwari, H.C.; Singh, P.; Kumar Mishra, P.; Srivastava, P. Evaluation of various techniques for extraction of natural colorants from pomegranate rind—Ultrasound and enzyme assisted extraction. Indian J. Fibre Text. Res. 2010, 35, 272.
26. Pourmortazavi, S.M.; Hajimirsadeghi, S.S. Supercritical fluid extraction in plant essential and volatile oil analysis. J. Chromatogr. A 2007, 1163, 2–24.
27. Mitra, S. Sample Preparation Techniques in Analytical Chemistry; John Wiley & Sons, Inc.: Hoboken, NJ, USA, 2003.
28. Da Silva, R.P.; Rocha-Santos, T.A.; Duarte, A.C. Supercritical fluid extraction of bioactive compounds. TrAC Trends Anal. Chem. 2016, 76, 40–51.
29. Brunner, G. Supercritical gases as solvents: Phase equilibria. In Gas Extraction: An Introduction to Fundamentals of Supercritical Fluids and the Application to Separation Processes; Steinkopff-Verlag: New York, NY, USA, 1994; pp. 59–146.
30. Elmaaty, T.A.; Abd El-Aziz, E. Supercritical carbon dioxide as a green media in textile dyeing: A review. Text. Res. J. 2018, 88, 1184–1212.

31. Björklund, E.; Sparr-Eskilsson, C. Extraction, Supercritical Fluid Extraction. In Encyclopedia of Analytical Science; Elsevier: Amsterdam, The Netherlands, 2005; pp. 597–604.
32. Reverchon, E.; De Marco, I. Supercritical fluid extraction and fractionation of natural matter. J. Supercrit. Fluids 2016, 38, 146–166.
33. Borges, M.E.; Tejera, R.L.; Díaz, L.; Esparza, P.; Ibáñez, E. Natural dyes extraction from cochineal (Dactylopius coccus). New extraction methods. Food Chem. 2012, 132, 1855–1860.
34. Herrero, M.; Cifuentes, A.; Ibañez, E. Sub-and supercritical fluid extraction of functional ingredients from different natural sources: Plants, food-by-products, algae and microalgae: A review. Food Chem. 2006, 98, 136–148.
35. Iovine, A.; Leone, G.P.; Larocca, V.; Di Sanzo, G.; Casella, P.; Marino, T.; Musmarra, D.; Molino, A. Risk analysis of a supercritical fluid extraction plant using a safety software. Chem. Eng. 2020, 79, 79–84.
36. Kabir SM, M.; Hasan, M.M.; Uddin, M.Z. Novel approach to dye polyethylene terephthalate (PET) fabric in supercritical carbon dioxide with natural curcuminoid dyes. Fibres Text. East. Eur. 2019, 27, 65–70.

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