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A Comprehensive Study on Natural Dye From Red Onion Skin

Ayush Vinod Hate and Dr. Namita More
D. G. Tatkare Mahavidyalaya, Mangaon, Raigad, Maharashtra

Abstract: *Red Onion Skin Synthetic dyes and chemicals create an enormous impact on environmental pollution both in textile manufacturing and after the product's lifetime. Biobased plant-derived colorants and mordants have great potential for the development of more sustainable textile dyeing processes. Colorants isolated from biomass residues are renewable, biodegradable, and usually less harmful than their synthetic counterparts. Interestingly, they may also bring additional functions to the materials. However, the extraction and purification of the biocolorants from biomass as well as their dyeing efficiency and color fastness properties require a more thorough examination. Here, we extracted Red onion skin (Allium Cepa L) skins to obtain polyphenolic flavonoids and anthocyanins as biocolorants, characterized the chemical composition of the mixture, and used a quartz crystal microbalance and thin films of cellulose nanofibrils to study the adsorption kinetics of dyes onto cellulose substrates in situ. The effect of different mordants on the adsorption behavior was also investigated. Comparison of these results with conventional dyeing experiments of textiles enabled us to determine the interaction mechanism of the dyes with substrates and mordants. Chitosan showed high potential as a biobased mordant based both on its ability to facilitate fast adsorption of polyphenols to cellulose and its ability to retain the purple color of the red onion dye (ROD) in comparison to the metal mordants FeSO4 and alum. The ROD also showed excellent UV-shielding efficiency at low concentrations, suggesting that biocolorants, due to their more complex composition compared to synthetic ones, can have multiple actions in addition to providing aesthetics..*

Keywords: Red Onion skin, Synthetic Dye, Quercetin, Natural Dyeing, Textile, Biocolorants

I. INTRODUCTION

Natural dyes have been used for centuries to add value, individuality, and even social status to textiles. In the Roman Empire, for example, only the aristocracy and eventually emperor Gaius Julius Caesar were allowed to wear a rare purple dye obtained from sea snails. This precious dye was one of the most expensive and strived trade goods in history. (1) The tradition of natural dyeing nearly disappeared after the development of synthetic dyes in the mid-19th century. Synthetic dyes replaced natural ones in the modern textile finishing industry as they were cheap and easy to produce and apply. However, current aims toward biobased products and sustainable processes have changed the picture. Most synthetic dyes are derived from petrochemical sources, and their production may require hazardous chemicals posing environmental challenges. The textile industry produces yearly over 200 000 tons of harmful substances released into effluents. (2) These pollutive processes have rekindled academic and commercial interest in natural dyes to bring the textile industry up to date with modern environmental and sustainability standards.

Sustainable textile production can benefit from natural (and biobased) dyes because they are of renewable origin, biodegradable, and often less harmful compared to synthetic ones. (3) However, the efficiency of their extraction and purification needs to be considered in detail as well as how to firmly attach them to the fabrics in a sustainable manner. In a recent study, different extraction and purification methods of natural dyes were compared, and the results suggest that the final form of liquid dye was the most feasible, as the drying phase increases energy consumption considerably. (4) EU legislation recommends liquid-type colorants whenever convenient to use to decrease the health hazards caused by the dusting of powder-form dye. (5)

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Aside from color, the extracts derived from natural sources have also been reported as attractive alternatives to add functionality to materials in a variety of applications, including medicinal products, (6) coatings, (7) and smart packaging. (8−10)

In this work, an analytical study was carried out to gain a more detailed understanding of the dyeing process on cellulosic materials. Quartz crystal microbalance with dissipation monitoring (QCM-D) was employed to detect the in situ adsorption in real time, allowing mordant and dye adsorption kinetics and adsorbed amounts to be determined. A natural dye obtained from red onion skins (*A. cepa*) and the efficiency of chitosan as a mordant was investigated. The adsorption kinetics and optical properties of CNF thin films dyed directly with the red onion dye (ROD) were compared to films mordanted with a conventional metal ($FeSO_4$) or chitosan. In addition, the ROD was structurally and chemically characterized using nuclear magnetic resonance (NMR) spectroscopy, highperformance liquid chromatography combined with high-resolution mass spectrometry (HPLC-HRMS), and Fourier transform infrared spectroscopy (FTIR). The surface-sensitive dye adsorption studies were correlated to industrial scale simulating dyeing experiments of cotton fabrics with color-fastness testing. Furthermore, ROD's potential as an effective UV-shielding agent for functional textiles was demonstrated by UV–visible (UV–Vis) spectrophotometry of cellulose-dyed films. The scheme of this work is shown in Figure 1.

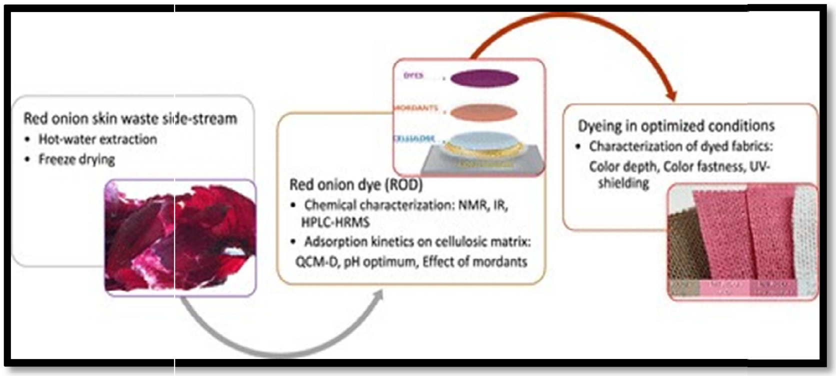

***Figure:-** Scheme of the workflow on the dyeing method development of the red onion dye (ROD) extract on cellulosic fibers.*

II. LITERATURE REVIEW

Some of the long history of shellfish purple is recounted. Understanding how the dye is produced from molluscs, the composition of the pigment and the chemistry involved has only been achieved as a result of the advances in analytical chemistry in the twentieth century, but some mysteries remain. Other diverse aspects such as the taste of the molluscs, the actual colour of the dye, the smell associated with the production and even the application in photography contribute to the fascination of the subject.

The fashion industry is facing increasing global scrutiny of its environmentally polluting supply chain operations. Despite the widely publicized environmental impacts, however, the industry continues to grow, in part due to the rise of fast fashion, which relies on cheap manufacturing, frequent consumption and short-lived garment use. In this Review, we identify the environmental impacts at critical points in the textile and fashion value chain, from production to consumption, focusing on water use, chemical pollution, CO 2 emissions and textile waste. Impacts from the fashion industry include over 92 million tonnes of waste produced per year and 79 trillion litres of water consumed. On the basis of these environmental impacts, we outline the need for fundamental changes in the fashion business model, including a deceleration of manufacturing and the introduction of sustainable practices throughout the supply chain, as well a shift in consumer behaviour—namely, decreasing clothing purchases and increasing garment lifetimes. These changes stress the need for an urgent transition back to 'slow'fashion, minimizing and mitigating the detrimental environmental impacts, so as to improve the long-term sustainability of the fashion supply chain.

Biocolourants have been investigated as alternatives to synthetic dyes. However, natural origin per se is not a label of harmlessness and research is needed to obtain safe dyes. We studied the cytotoxicity of the extracts from fungal (*Cortinarius semisanguineus*, *Tapinella atrotomentosa*) and plant (*Tanacetum vulgare, Salix phylicifolia*) sources and the woollen fabrics dyed with the extracts. Cytotoxicity in vitro using hepa-1 mouse hepatoma cells for 24 h and 72 h exposure was observed as the highest tolerated dose. All biocolourants produced intensive colour on fabrics with fastness properties from moderate to good. The *Salix* and *Cortinarius* samples did not show any cytotoxic effects, whereas the *Tanacetum* and *Tapinella* samples had slightly higher test values but were not interpreted as being significantly toxic. Higher than zero values of the undyed fabrics showed the importance of examining their toxicity as well. It was found that the cytotoxicity of the samples dyed with the biocolourants did not differ significantly from the undyed wool fabric. The concentrations of dyes used in the assays were very low, imitating the dose of the user. In addition to colouring properties, natural dyes may have pharmaceutical and antibacterial properties which would enhance the interest in using them in products for added value.

Fruit and vegetables contain molecules that have particular colors, which can potentially be an environmentally attractive substitute for their synthetic counterparts in (non-)food applications. The most sustainable source for such natural colorants would be by the valorization of by-products from the fruit and vegetable industries, but qualitative and quantitative characteristics of food by-products for this purpose remain scarce. Natural dyes also show mediocre stability and affinity toward textile fibers, which questions their potential feasibility for application and level of sustainability to overcome these issues. This review describes three dye classes (i.e., anthocyanins, quinones, and carotenoids) along with their occurrence, mass, and concentration in by-products that are generated from agricultural losses as well as the fruit and vegetable processing industries. To tackle the shortcomings of natural dyes on fibers, several application techniques were collected from the literature. A discussion on technoeconomic potential and environmental sustainability is included. The latter is done by including a life cycle assessment (LCA) to investigate the environmental impact of extracting anthocyanins, quinones, and carotenoids from fruit and vegetable processing by-products and their subsequent application to the dyeing process. The mapping of by-products for each natural dye class illustrates the vast availability of agro-food residues (>0.1 Mt annually in the EU-28) with a natural dye content of up to 56 kg/t DW for anthocyanins, 18 kg/t DW for quinones, and 593 kg/t DW for carotenoids. Metallic mordants are mostly favored for improving the fixation of natural dyes but entail potential environmental issues. Greener approaches, such as biomordants and enzymes, still show room for improvement, chemical modification methods might also guarantee dye fixation, though questionable in environmental sustainability.

Dye Extraction:-

Red onion skins were added to a reactor in an onion skin-to-water mass ratio of 1:20. Water-soluble compounds were extracted with constant stirring (250 rpm) at 80 °C for 60 min. To remove the remaining insoluble particles, the extract was filtered twice (VWR filter paper with a 12–15 μm pore size) followed by centrifugation (4500g, 20 min). The filtrate was then freeze-dried and ground to obtain ROD in a yield of ca. 6 g/L. The purple powder was stored in a cold room (16 °C) in sealed plastic vessels covered with aluminum foil to prevent light degradation.

III. CHARACTERIZATION

High-Performance Liquid Chromatography Mass Spectrometry (Hplc-Ms):-

To trace the different flavonoid components in ROD, HPLC analysis was performed using an Agilent HPLCVWD (Santa Clara, CA) equipped with a diode array UV detector. Detection wavelengths were 254, 420, and 530 nm. Components of the mixture were separated with a Phenomenex Luna C18 column (3 μm, 100 Å, 150 mm × 4.6 mm) using a 0.8 mL/min flowrate with mobile phases A (15% MeOH in acetonitrile) and B (3% formic acid in MQ-water). The concentration of the sample was 2 mg/mL, and the injection volume was 10 μL. The gradient elution started at 97% B and was decreased to 88.5% B within 40 min, kept at 88.5% B for 10 min, and decreased to 84.5% B within 20 min, whereafter B was decreased from 84.5 to 77.0% in 15 min. At the end of the run, B was returned to 97% within 5 min where the column was equilibrated for 3 min before each run. A commercial standard was used to identify quercetin by comparison of its retention time and UV spectrum.

UV-Shielding Tests:-

The UV-shielding effect of ROD was evaluated using self-standing CNF films. Briefly, the CNF suspension (100 mL, 0.8 wt %) was poured on a poly(vinylidene fluoride) (PVDF) membrane filter (hydrophilic PVDF, 142 mm membrane with a 0.45 µm pore size, Dupore), and the water was removed using air pressure filtration equipment integrated with a tripod chamber (inner diameter of 12 cm, height 8.5 cm) under 2.5 bar overpressure for 45 min. The formed CNF wet film was then ambient dried at 23 °C and 50% relative humidity for 72 h under a load of 5 kg. Then, the CNF-dried film was removed from the PVDF membrane. (31)

The CNF films were dyed with ROD liquor, keeping the fiber-to-dye liquor mass ratio at 50 g/L (1:20). The dyeing was conducted with the same methodology used in the adsorption measurements. First, CNF films were immersed in 1% acetic acid solution (pH 4) for 30 min, then samples were moved to mordant solutions (chitosan and FeSO4 5 g/L) for 30 min, followed by rinsing with 1% pH 4 acetic acid. Next, the samples were immersed in the dye liquor of concentrations 0.01, 0.1, 0.25, 0.5, and 1.0% of dye on the weight of the fiber (owf) for 60 min, followed by washing in 1% pH 4 acetic acid and distilled water (pH 6).

The optical transmittance between 400 and 800 nm was measured at room temperature using a diffuse reflectance accessory coupled to a UV–Vis–NIR Agilent Cary 5000 spectrometer (Agilent, CA).

Textile Dyeing:-

To remove textile auxiliaries and impurities prior to dyeing, each textile sample (10 g) was washed in 1% (v/v) acetic acid solution (pH 2.8) in a fabric-to-dye liquor mass ratio of 1:20 at 40 °C for 30 min whereafter rinsed with water.

The fabric samples were premordanted with alum, FeSO4, or chitosan. The premordanting and dyeing conditions are explained in Figure S1 and Table S1. Mordanting was carried out in 0.5 g/L chitosan solution in a fabric-toliquor mass ratio of 1:20 at 80 °C for 30 min, whereas with 8 g/L alum solution and 3 g/L FeSO4 solution, a fabric-to-liquor mass ratio of 1:10 at 50 °C for 60 min was used. All samples were rinsed twice with water for 10 min after mordanting to remove the unfixed mordant. The dyeing with ROD liquor (0.1 g/L) was carried out in a fabric-to-liquor mass ratio of 1:20 at 50 °C for 95 min. An Original Hanau Linitest equipment (Hanau, Germany) was used to simulate the industrial winch dyeing machine.

Color Strength and Light Fastness of Red Onion Dyed Fabrics:-

Alterations of the fabric surface after premordanting and dyeing were evaluated by comparing the FTIR spectra of untreated, pretreated, and dyed fabric samples. A Bruker α-P FTIR instrument with software Bruker Opus 6.5 (Billerica, MA) was used.

The color of the dyed sample was measured as the recommendation of the color scale, CIE L*, a*, b*, and reflectance values, using a Konica Minolta (Tokyo, Japan) CM-2600d spectrophotometer (illuminant D65, CIE 10° observer). Specular Component Included (SCI) values were recorded. This type of color evaluation measures the total appearance independent of surface conditions. (32)L*, a*, and b* refer to the three axes of CIE color space: L* representing the lightness axis and obtaining values from zero for perfect black to 100 for white; a* presenting both the hue and the chroma of the red–green axis where a* > 0 describes the redness, and a* < 0, the greenness; and b* representing the blue–yellow axis where b* > 0 indicates the yellowness, and b* < 0, the blueness of the color. (32) The total color difference, ΔE, between the two samples can be defined in terms of a difference in these three components (eq 1)

IV. CHEMICAL CHARACTERIZATION OF THE RED ONION DYE AND DYED FABRICS:-

The concept of directly employing natural plant extracts after extraction in hot water is a cost-effective way to acquire natural dyes. However, these natural dye extracts are not pure substances; instead, they contain a complex mixture of flavonoids and other water-soluble compounds. To identify the main components of the ROD mixture, HPLC and high-resolution ESI-QTOF-MS as well as NMR were employed. Figure 2. comprises the chemical structures of the main flavonoids, as well as the HPLC chromatograms of ROD at wavelengths 254, 420, and 530 nm. The chromatogram obtained at 254 nm shows two major absorption peaks, of which the latter one, at retention time (RT) 108.3 min, was also detected at 420 nm. This signal is attributed to flavonol quercetin (1) as also confirmed by the correlation to the RT of the pure quercetin standard (data not shown). The NMR spectra (Figure S2a–c) also confirmed quercetin as the main phenolic compound in the ROD. Carbohydrate analysis of onion extracts revealed glucose and pectin galacturonic acid as the main carbohydrates.

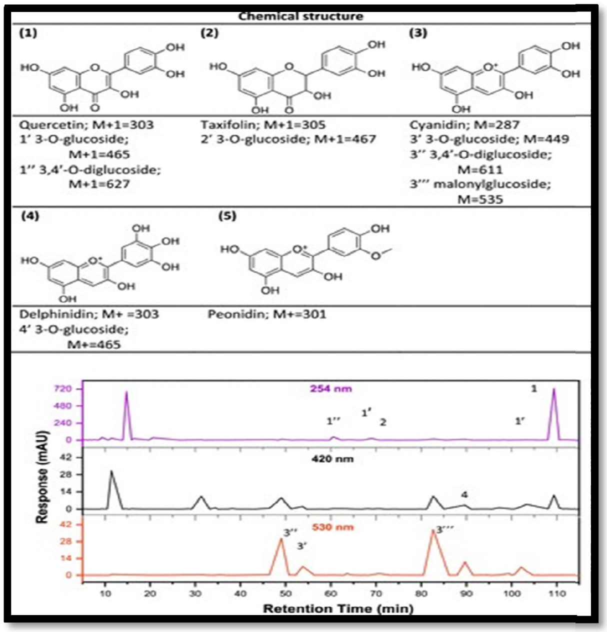

Figure 2:- HPLC Chromatograms And Qualitative Identification Of The Main Flavonoids In ROD Detected At 254, 420, And 530 Nm.

In ESI-QTOF-HRMS positive ion mode, the intense peak at RT 108.3 min with [M + H]+ 303.0508 was found, and in comparison to the quercetin [$C_{15}H_{10}O_7$] + H+ calculated exact mass 303.0499, the mass error was only 3 ppm, further confirming the presence of quercetin. The HRMS analysis revealed the mono- and diglucosides of quercetin at 59.7, 67.4, and 103.8 min. Another flavonoid compound with the observed mass [M+H]+ 305.0656 could be detected at 71.1 min in small intensity and was interpreted to be taxifolin (2) [$C_{15}H_{12}O_7$] + H+ with a calculated mass 305.0671 and mass difference of 1.6 ppm. The chromatogram obtained at 530 nm revealed many small peaks suggesting the presence of several compounds that absorb in the long wavelengths, mainly anthocyanins such as cyanidin (3). (33) We note that some of the characteristic peaks of anthocyanins also absorb at 530 and 420 nm in the UV region.

V. METHODOLOGY

Dyeing with red onion skins is a straightforward process that involves extracting the pigment from the onion skins and applying it to fabric. Here's a step-by-step methodology for natural dyeing using red onion skins:

Materials Needed:

Red onion skins (the outer layers)
Fabric or yarn (natural fibers like cotton, wool, or silk work best)
A mordant (alum, vinegar, or iron are commonly used)
A large pot or dyeing vessel
A strainer or cheesecloth
Water
Wooden or plastic spoons/stirrers (avoid metal, as it can react with the dye)
Gloves (optional, to avoid staining your hands)

Step-by-Step Method:

Prepare the Fabric/Yarn:

Wash the fabric or yarn thoroughly to remove any oils, dirt, or chemicals that might interfere with the dyeing process.

Pre-mordant the fabric (this step helps the dye adhere better). A common mordant for onion skin dyeing is alum, which can be dissolved in water and used to soak the fabric for about 30 minutes to an hour.

Alternatively, vinegar or an iron mordant can be used, depending on the color effect you desire.

Prepare the Dye Bath:

Collect the red onion skins. You'll need about 1–2 cups of onion skins for a moderate-sized batch of fabric.

Fill a large pot with water and add the onion skins. Bring the water to a simmer (not a full boil) and let it cook for about 45 minutes to an hour. This helps to extract the pigment from the skins.

Strain out the onion skins using a cheesecloth or fine mesh strainer, leaving the dye liquid in the pot.

Dyeing the Fabric/Yarn:

Add the pre-mordanted fabric or yarn into the dye bath. Ensure the fabric is completely submerged and stir gently to ensure even dyeing.

Let the fabric soak in the dye bath for at least 30 minutes, but you can leave it longer (up to 2 hours) for deeper, richer colors.

The color will likely appear lighter when wet, so don't be discouraged if it looks faint at first. The final color will intensify as the fabric dries.

Rinsing and Drying:

Once the fabric has reached the desired color, remove it from the dye bath and rinse it in cold water until the water runs clear.

Hang the fabric to dry in a shaded area to preserve the color (sunlight can cause fading over time).

Optional Post-Treatment for Color Variation:

You can alter the color slightly by using different mordants. For example:

- **Iron** can darken the dye and create more muted, earthy tones.
- **Vinegar** may produce a pinker shade.
- **Alum** often gives a more vibrant, yellowish tone to the dye.

Experimenting with different mordants or combining them can lead to a wider range of color results.

Procedure:-

Prepare the Fabric (Pre-mordanting):

Wash the fabric or yarn thoroughly to remove any dirt or impurities.

Prepare a mordant solution. If using alum (a common mordant), dissolve 1 tablespoon of alum in 4 cups of water. Soak your fabric or yarn in the mordant solution for 30 minutes to 1 hour. This step helps the dye bond with the fibers.

After soaking, rinse the fabric in cold water and wring out the excess.

Prepare the Dye Bath:

Collect the outer skins of red onions (about 1–2 cups for a moderate dye bath).

Place the onion skins in a large pot or dyeing vessel.

Add enough water to fully submerge the onion skins.

Bring the pot to a simmer, allowing the skins to boil gently for 45 minutes to 1 hour. Stir occasionally.

After simmering, strain out the onion skins using cheesecloth or a fine mesh strainer. You now have your dye bath.

Dye the Fabric:

Add the pre-mordanted fabric or yarn into the dye bath.

Ensure the fabric is fully submerged. Stir gently to ensure even coverage of the dye.

Let the fabric soak in the dye bath for 30 minutes to 2 hours. The longer the fabric stays in, the more intense the color will become. You can check the color periodically by removing a small section and rinsing it to see how it looks when dry.

For a lighter color, remove the fabric earlier; for deeper tones, leave it longer.

Rinse the Fabric:
Once you've reached your desired color, remove the fabric from the dye bath.
Rinse the fabric in cold water until the water runs clear. This removes excess dye and helps set the color.

Dry the Fabric:
Hang the fabric to dry in a shaded area, as sunlight can cause colors to fade more quickly. Allow the fabric to air dry completely.

Optional: Experiment with Post-Treatment:
You can experiment with different mordants to alter the color:
o **Iron** will darken the color, creating more muted tones (a rusty brown or grayish hue).
o **Vinegar** can add a pinkish tint to the fabric.
o **Alum** creates brighter yellowish tones.
You can also try over-dyeing with another natural dye for a layered color effect.

VI. CONCLUSION

The future of natural dyeing from red onion skins is promising, with opportunities for growth in sustainability, innovation, and education. By tapping into this potential, the industry could reduce its environmental footprint while promoting more ethical and sustainable practices in fashion, agriculture, and crafts.

By modeling the traditional dyeing process in QCM-D, this study for the first time analytically evaluates the interaction mechanisms underlying dye absorption and binding to cellulosic substrates, expanding the fundamental understanding of natural dye adsorption. The adsorption of ROD onto CNF thin films and the ζpotential measurements revealed pH 4 as optimal for ROD adsorption to cellulose. Adsorption curves and dyeing experiments demonstrated that chitosan, due to its cationic nature in acid pH, can significantly increase dye adsorption onto cellulosic films or fabrics, cementing its potential as a natural mordant. The adsorption of ROD onto chitosan-mordanted CNF films was very rapid and metal salts could be avoided. Efficient UV blocking required furthermore only 0.5 and 1% owf of the concentration of the dye and mordant solutions, respectively. Importantly, chitosan retained the original dye color, with a slight shift to red/pink shades, whereas $FeSO_4$ and alum-mordanted samples completely changed the obtained color to green shades due to metal ion–dye complexation. The combination of plant-based natural dyes and mordants derived from biomass side streams, such as chitosan, is a promising approach to fulfill the textile industry in seeking for alternatives to hazardous chemicals and auxiliaries in their processes. Studies show that agricultural and food processing wastes provide adequate streams for valorization of natural dyes and biomordants, especially when applied on a niche scale. (4)

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