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Research Article • DOI: 10.2478/ftee-2023-0033 FTEE • 31(4) • 2023 • 30–37

Blossoming Beauty: Enhancing Natural Fibres with Calendula Officinalis L. Flower Dye and Assessing Color Fastness with Microbial Properties

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Abstract
Mordants are materials used to fix long periods of colour on selected fabrics or fibres. The current research aimed to dye natural fibres like palm leaf (*Borassus flabellifer* L.), korai grass (*Cyperus pangorei* L.), banana fibre (*Musa accuminata* L.), screw fine fibre (Pandanus), sisal fibre (*Agave sisalana* Perrine), and pineapple fibres (*Ananas comosus* L.) with *Calendula officinalis* plant flower dye. The dye was fixed to the selected fibre material with the help of different mordants. The *Calendula officinalis* flower dye produced different colours, like black, brown, orange, sandal, yellow, etc., on the dyed materials. The different colour formations on the dyed materials are based on the mordents used to fix the dye. The mordants tonify the different colours in the same dyestuff of *Calendula officinalis* flower dye. Mordant-based dyed natural fibres have good fastness properties against light and water. *Calendula officinalis* dyes have potential activity against the skin infection-causing organism *Pseudomonas aeruginosa*.

Keywords
Calendula officinalis, fastness, fibres, mordant, natural dye.

1. Introduction

Natural dyes are dyes that are derived from plants, insects, minerals, or other natural sources [1]. These dyes have been used for thousands of years to colour fabrics, yarns, and other materials [2]. Some examples of natural dyes include the following: plant dyes, which are obtained from various parts of plants, such as leaves, roots, bark, and fruits, examples of which include indigo (from the indigo plant), madder (from the madder root), and woad (from the woad plant) [3, 4]; insect dyes, which are obtained from insects, such as cochineal (from the cochineal insect) and lac (from the lac insect) [5]; mineral dyes, which are obtained from minerals, such as ochre (from iron oxide) and azurite (from copper carbonate) [6, 7]. Natural dyes are popular for their bright and long-lasting colours, as well as for the fact that they are non-toxic [8], eco-friendly [9], and biodegradable [10]. However, natural dyes are often less colourfast and lightfast than synthetic dyes, meaning that they may fade or change colour over time or when exposed to light [11].

Natural dyes are extracted largely from plant parts like leaves, flowers, stems, wood, fruit, seeds, etc. India's expertise in natural dyes dates back to a very old era. There are about 450 dye-producing plants found in India [12]. The most famous vegetable dyes are madar, indigo, and magitha [13–15]. The identification and usage of synthetic dyes entered the textile industry in the nineteenth century [16]. High colour values and cost-effective synthetic dyes caused a rapid decline in the use of natural dyes. Synthetic dyes are highly toxic and hazardous to environments, including human skin and lungs [17]. But natural dyes are low-toxic, less polluting, less health-hazardous, non-carcinogenic, and non-poisoning. However, natural dyes have some limitations; one is fastness [13].

Mordants are chemical compounds that are used in the process of dying to improve the colorfastness and adhesion of natural dyes to fibres, textiles, and other materials [18]. They serve as a link between the dye molecules and fibres, forming a strong binding that increases the dye's endurance and stability. Mordants can also change the tint and colour of the final colour generated. Mordants commonly used include alum, iron (ferrous sulphate), copper (copper sulphate), tin (stannous chloride), tannic acid, oxalic acid, and others. When working with natural dyes, mordants are very important since they improve the overall quality and endurance of the dyed material. Dye-coated fabrics, yarns, and fibre products fade with exposure to light or washing out. Mordants are materials used to fix long periods of colour on fabric or fibres. The current research aimed to dye natural fibres like palm leaf (*Borassus flabellifer* L.), korai grass (*Cyperus pangorei* L.), banana fibre (*Musa accuminata* L.), screw fine fibre (Pandanus), sisal fibre (*Agave sisalana*), and pineapple fibres (*Ananas comosus* L.) with *Calendula officinalis* plant flower dye.

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2. Experimental

2.1. Natural fibres

The selected natural fibres—palm leaf, korai grass, banana fibre, screw fine fibre, sisal fibre, and pineapple fibre—were collected from local traditional craft workers in the Kanyakumari district, Tamil Nadu, India.

2.2. Preparation of natural fibres

Before dying, the fibres were soaked with soft water for 30 minutes and then washed with freshwater.

2.3. Preparation of natural dye

Calendula officinalis plant flowers were collected from agricultural land in the Tirunelveli District of the Therkumadathur village in the Kadayam region, Tamil Nadu, India, latitude: 8°49'21.432"N and longitude: 77°23'14.096"E The collected flowers were shade-dried for two weeks. After drying, the flower petals were removed. The natural dye was extracted from the dried petals by an aqueous extraction process [19].

2.4. Dyeing procedure

The processed plant fibres were soaked in the extracted dye solution and heated at 95 °C for 30 minutes [20].

2.5. Mordanting

After dyeing the mordants, sodium hydroxide (salt), sodium bicarbonate (soda salt), oxalic acid, tannic acid, ferrous ammonium sulphate (FAS), potassium alum, tin metal, and tamarind were added.

2.6. UV/Vis spectral study

The natural dye and dye with different mordant mixtures prepared were analysed by a UV/Vis spectrophotometer -2203 [21].

2.7. Light fastness study

The dyed natural fibres were analysed for lightfastness using the standard method (Gupta, 1999). The grayscale was used to measure the lightfastness of the dyed fibres [22].

2.8. Wash fastness study

The dye-coated natural fibres were washed in 100 mL of tap water, to which 0.1 g of commercially available detergent was added. This setup was kept under normal room conditions and stirred for 15 minutes using a magnetic stirrer. After stirring, the water used for washing was analysed for its optical density by a UV/Vis spectrophotometer [23].

2.9. Antibacterial study

The test organism was purchased from NCIM, Pune. The test was carried out using the well diffusion method [24].

3. Results and discussion

Calendula officinalis flower dye-coated fibres have different colours like black, brown, orange, sandal, yellow, etc. (Figure 1). The mordants play an important role in fixing the colour to the fibres. The mordants toned the different colours in the same dyestuff, *Calendula officinalis* flower dye e.g., the selected natural fibres (except korai grass) attains a sandal colour based on oxalic acid mordant. Ferrous sulphate gives a blackish colour to natural fibres. Potassium alum creates a yellow colour in screw pine fibres and an orange colour in banana and sisal fibres. Brown colour is attained in pineapple, screw pine, sisal, and banana fibres with the use of tamarind (Figure 1).

The salt and soda salt mordant did not make any changes in the dyed fibres. It is all the same adding the mordant to the dyed fibres or not (Figure 1). The role of the mordant is to fix the colour to the fibres permanently and tone the dyed fibres colour. After mordanting, dye absorption changed in the UV spectra based on the mordant used for dyeing (Figure 2).

Absorbance at 358.4 nm, 387.2 nm, and 257.6 nm was present in the aqueous extraction of *Calendula officinalis* flower dye. After dyeing the fibres, the dye has three absorption peaks at 363.2 nm, 387.2 nm, and 257.6 nm. The salt-added dye has the same absorption range at 358.4, 387.2, and 257.6nm. The soda-salt-added dye has three absorption peaks at UV ranges of 257.6 nm, 348.8 nm, and 387.2 nm. Oxalic dye has three absorption peaks: one in the visible region at 401.6 nm and two in the UV region at 363.2 nm and 257.6 nm, respectively. Tannic acid as a mordant in the dye has three major absorptions at 358.4 nm, 257.6 nm, and 387.2 nm. With ferrous sulphate added as a mordant, the dye has a single absorption peak in the UV region located at 243.2 nm. Potassium alum mixed dye has three absorption peaks at 406.4 nm, 363.2 nm, and 257.6 nm. Tin metal used as dye has three absorptions at 401.6, 257.6, and 363.2nm. Natural mordant tamarind used as dye has three major absorptions located at 257.6 nm, 358.4 nm, and 387.2nm.

The limitations of natural dyes are that they are easily faded by light and wash out. The mordants used on the dyed fibres are more stable to light and water than those without mordanting (Table 1). When oxalic acid is used in sisal fibres, ferrous sulphate in palm leaf and banana fibres, tin in banana and pineapple fibres, and alum in banana fibres, they have excellent fastness properties against light.

Dyed palm leaf fibres have good lightfastness properties with the use of sodium hydroxide (salt), oxalic acid, tannic acid, potassium alum, tin metal, and tamarind mordants. Korai grass fibres have good colour stability against light with the help of soda salt and FAS mordants. Banana fibres have good lightfastness with the help of oxalic acid and tamarind mordants. Natural dye-coated screw pine fibres have good light fastness with the addition of soda salt,

*Fig. 1.*Calendula officinalis *flower dye coated natural fibres*

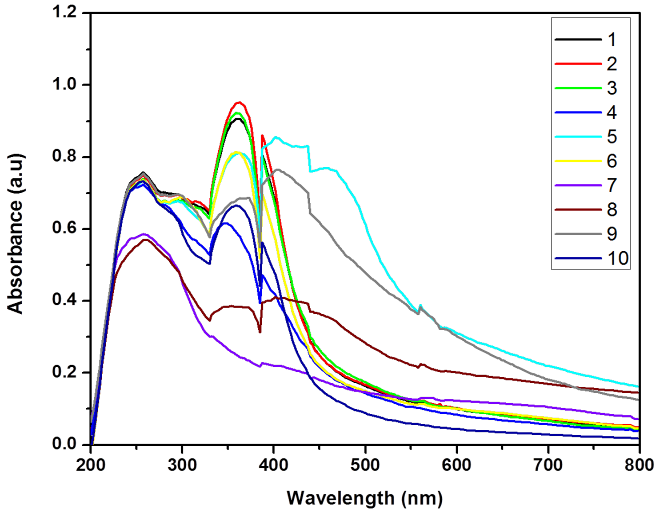

Fig.2. UV spectral data of dye with different mordants. 1. Calendula officinalis *flower dye extracted by aqueous method; 2.* Calendula officinalis *flower dye coated fibres without mordanting; 3. dye mixed with sodium chloride; 4. dye mixed with sodium bicarbonate; 5. dye with oxalic acid; 6. dye with tannic acid; 7. dye with ferrous sulphate; 8. dye with tin metal; 9. dye with potassium alum; 10. dye with cooking tamarind*

FAS, alum, and tamarind. Sisal fibres dyed with the use of mordants such as soda salt, tannic acid, ferrous sulphate, potassium alum, and tin metal have good lightfastness properties. Pineapple fibres dyed with oxalic acid, potassium alum, and tamarind mordants have good lightfastness properties (Table 1).

The natural dye-coated fibres were analysed for their washing properties by a UV/Vis spectrophotometer (2203) in the range of 200–800 nm. Figure 3 indicates spectral data of water used for the 1st-5th washings of dye-coated fibre materials without mordanting. The dyed fibres are easily washed out by water. The presence of many intensive peaks in the UV and visible regions of the spectral data confirmed that without mordant, the dyed fibres have poor wash fastness properties.

The wash fastness property of salt-based mordanting dye-coated fibres is represented in Figure 4. The colour was easily removed by washing with water. There are a number of absorbance peaks present in the spectral data. In the 3rd-5th washes, the absorption peak value is very high, which confirms the poor wash property.

Figure 5 indicates the wash fastness property of soda-salt-based dyed natural

S. No	Fibre	Without	Salt	Sodium bi carbonate	Oxalic acid	Tannic acid	Ferrous (FAS)	Tin	Alum	Tamarind
1	Palm leaf	2-3	3-4	2-3	3-4	3-4	4-5	3-4	3-4	3-4
2	Korai	2-3	2-3	3-4	2-3	2-3	3-4	1-2	2-3	2-3
3	Banana	3-4	2-3	2-3	3-4	3-4	4-5	4-5	4-5	3-4
4	Screw pine	2-3	2-3	3-4	2-3	2-3	3-4	2-3	3-4	3-4
5	Sisal	2-3	3-4	2-3	4-5	3-4	3-4	3-4	3-4	2-3
6	Pineapple	3-4	2-3	2-3	3-4	1-2	2-3	4-5	3-4	3-4

Table 1. Light fastness study of Calendula officinalis *flower dye coated natural fibres*

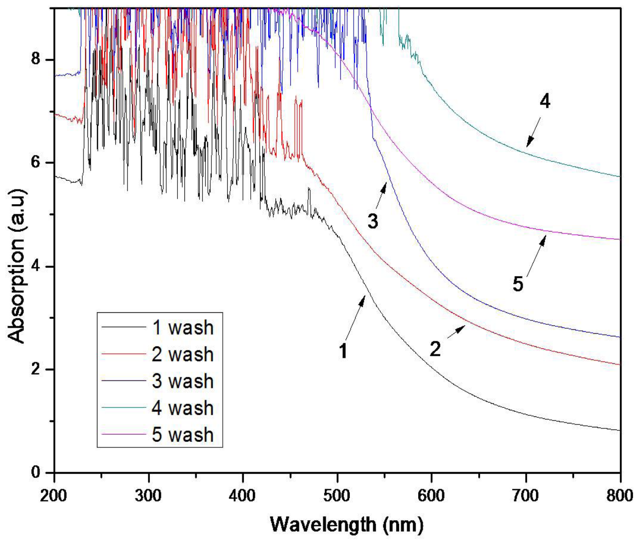

Fig.3. Wash fastness study of Calendula officinalis *flower dye coated natural fibres without mordanting*

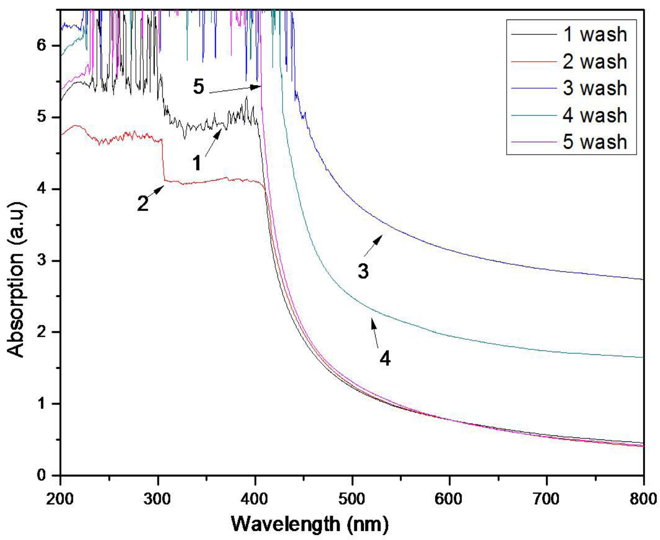

Fig. 4. Wash fastness study of salt mediated mordanting of Calendula officinalis *flower dye coated natural fibres*

fibres. These fibres are moderately washed out by the water. The 3rd wash absorption range is high compared to the 1st, 2nd, 4th and 5th water washes.

Figure 6 indicates spectral data of the wash fastness study of oxalic acid-mixed natural dye-coated fibres. These fibres have good wash fastness properties. The absorption peak range is high in the fifth wash.

The wash-fastness property of tannic acid-based dyed natural fibres is shown in Figure 7.

The tannic acid-based dyed fibres are rather washable with water. In the third wash, the dyed fibres easily washout. The FAS-mediated natural dye-coated fibres have excellent wash fastness properties, as shown in Figure 8.

The spectral data do not have any absorption peak in the visible region of the 1st to 5th washes (Figure 8). Figure 9 indicates spectral data of the wash fastness study of potassium alum mixed with natural dye-coated fibres.

These fibres have excellent wash fastness properties. The spectral data do not have any absorption peak in the visible region of the 1st to 5th washes (Figure 9). Figure 10 indicates the washing fastness property of the tin metal-based dyed natural fibres. These fibres have good wash fastness properties. The 3rd and 4th wash absorption ranges were high compared to those of the 1st, 2nd, and 5th.

Figure 11 indicates spectral data of the wash fastness study of natural mordants of tamarind mixed with natural dye-coated fibres. These fibres have excellent fastening properties. The spectral data do not have any absorption peak in the visible region of the 1st to 5th washes.

Colour fastness refers to the resistance of a colour to fading or changing when exposed to various environmental factors such as light, washing, and rubbing. The measured absorption is the amount

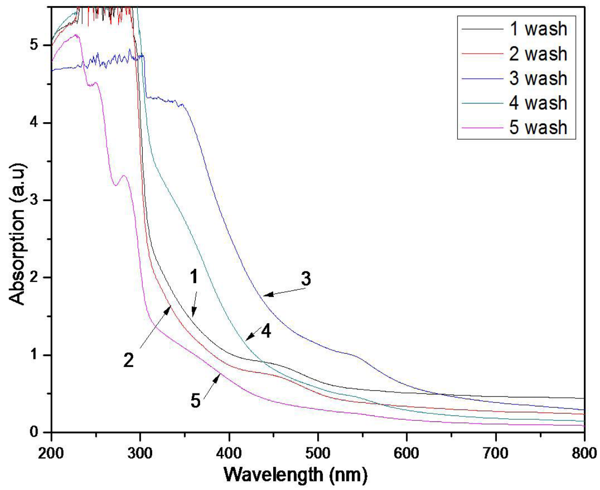

Fig. 5. Wash fastness study of soda salt based mordanting of Calendula officinalis *flower dye coated natural fibres*

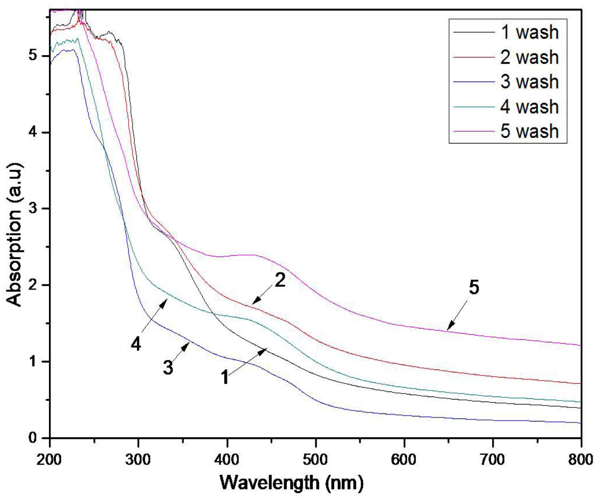

Fig. 6. Wash fastness study of oxalic based mordanting of Calendula officinalis *flower dye coated natural fibres*

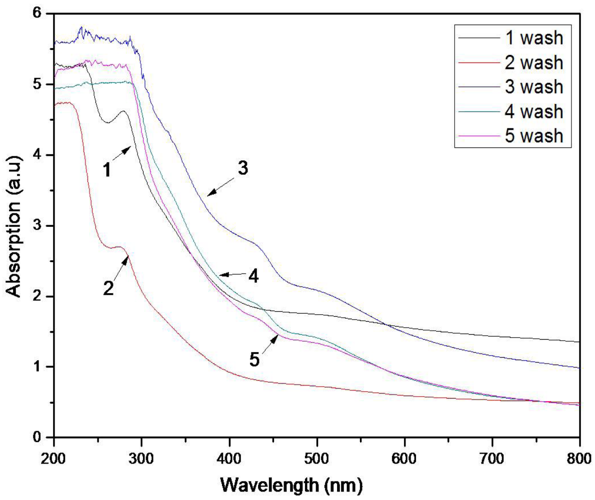

Fig. 7. Wash fastness study of tannic acid mordanting of Calendula officinalis *flower dye coated natural fibres*

of light that a material or substance has absorbed, typically expressed as a percentage. There is a relationship between colour fastness and measured absorption in that the more a colour absorbs light, the less likely it is to fade or change. This is because the absorbed light energy is converted into heat, which is then dissipated, reducing the overall impact of light exposure on colour.

The *Calendula officinalis* flower dye highly inhibits the skin infection-causing organism *Pseudomonas aeruginosa* (Figure 12).

Skin infection by *Pseudomonas aeruginosa* refers to a condition where the bacterium *Pseudomonas aeruginosa* causes an infection on the skin. *Pseudomonas aeruginosa* is a versatile and opportunistic pathogen known for causing various types of infections in humans, particularly in individuals with compromised immune systems, wounds, or underlying medical conditions. The *Calendula officinalis* dye-coated fibre-based product was extracted to prevent the skin infection caused by *Pseudomonas aeruginosa.*

4. Conclusion

The use of *Calendula officinalis* flower dye to cover natural fibres produced an array of appealing colours such as black, brown, orange, sandal, and yellow. The use of mordants was critical in anchoring these colours to the fibres, improving colour retention. Mordants were used as agents to blend different colours from the same *Calendula officinalis* dye source. Oxalic acid mordant produced a sandal colour transition in selected natural fibres, while ferrous sulphate produced a blackish tint. Meanwhile, potassium alum gave screw pine fibres a yellow colour and banana and sisal fibres an orange colour. When pineapple, screw pine, sisal, and banana fibres were treated with tamarind, they turned a rich brown colour.

The absorption study of *Calendula officinalis* flower dye revealed the existence of substantial peaks at 358.4

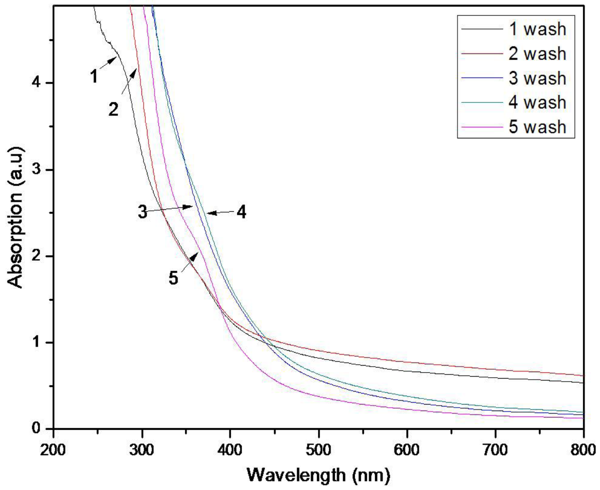

Fig.8. Wash fastness study of FAS based mordanting of Calendula officinalis *flower dye coated natural fibres*

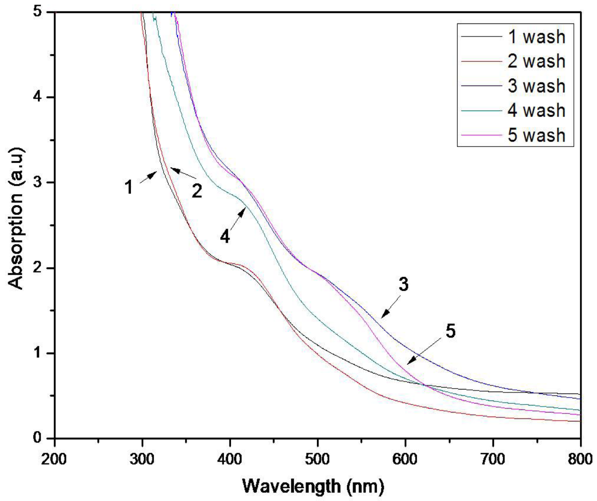

Fig. 9. Wash fastness study of potassium alum mordanting of Calendula officinalis *flower dye coated natural fibres*

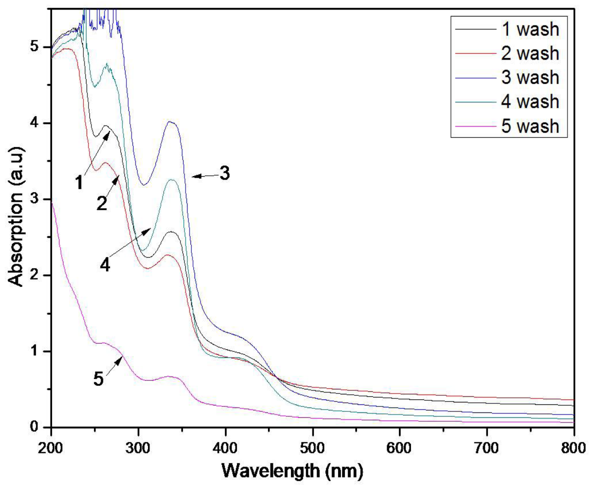

Fig. 10. Wash fastness study of tin metal based mordanting Calendula officinalis flower dye coated natural fibres

nm, 387.2 nm, and 257.6 nm. The dyed fibres had absorption maxima at 363.2 nm, 387.2 nm, and 257.6 nm after the dyeing procedure. Notably, different mordants caused modest differences in the absorption peaks, confirming the importance of mordanting to dye absorption behaviour.

Despite the enchantment of the results, natural dyes have limitations, such as fading when exposed to light and washing. However, the addition of mordants significantly improved colour stability in the face of these problems. Furthermore, a UV/Vis spectrophotometer evaluation of washing qualities revealed the sensitivity of the dye-coated fibres without mordant to washing, as intense peaks in the UV and visible regions proved their poor performance. Mordanted dye-coated fibres, on the other hand, displayed different degrees of wash fastness. Mordants such as sodium hydroxide, oxalic acid, tannic acid, potassium alum, tin metal, and tamarind have shown outstanding colour stability against water washing, confirming mordants' protective effect.

The study also discovered that the *Calendula officinalis* flower dye had strong inhibitory capabilities against the skin infection-causing bacterium *Pseudomonas aeruginosa*. This demonstrates that it might be effective in preventing skin infections brought on by this bacterium.

In essence, the study emphasises the aesthetic and scientific union of using *Calendula officinalis* flower dye to enhance natural fibres with a variety of colours, driven by mordant orchestration. While issues such as colour fading and washability continue in natural dyes, the strategic use of mordants offers a viable answer. Furthermore, the outcomes of the study point to the potential of *Calendula officinalis* dye as an antibacterial agent. Overall, this investigation opens up possibilities for both aesthetic and practical applications, bridging tradition and innovation.

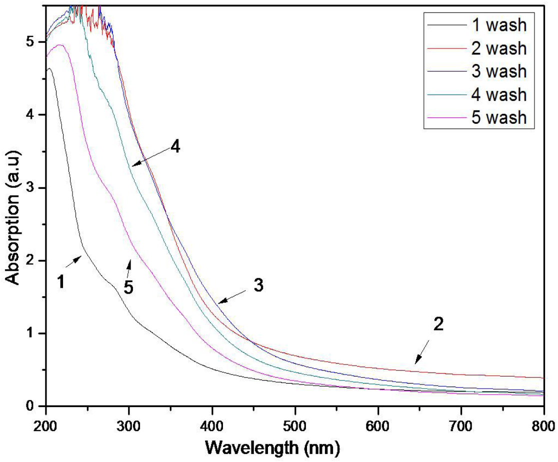

Fig. 11. Wash fastness study of tamarind mediated mordanting Calendula officinalis *flower dye coated natural fibres*

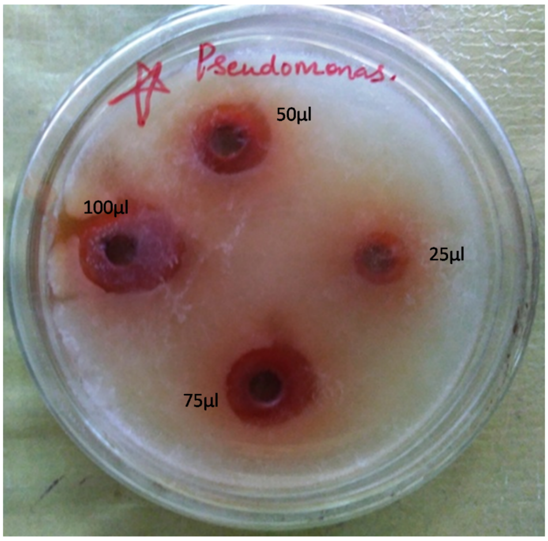

Fig. 12. Antibacterial activity of Calendula officinalis *flower dye*

Conflicts of interest

The authors declare that there is no conflict of interest regarding the publication of this manuscript.

Acknowledgement

The authors extend their appreciation to the Research Supporting Project No. (RSPD2023R708), King Saud University, Riyadh, Saudi Arabia.

References

1. Lizamoni C, Smita B. Natural Dye Sources and its Applications in Textiles: A Brief Review. *Int. J. Curr. Microbiol. App. Sci.* 2020; 9(10): 261-269. doi: https://doi.org/10.20546/ijcmas.2020.910.034.
2. Anastasia Fröse, Karolina Schmidtke, Tobias Sukmann, Irén Juhász Junger, AndreaEhrmann. Application of natural dyes on diverse textile materials. *Optik* 2019; 181: 215-219.https://doi.org/10.1016/j.ijleo.2018.12.099
3. Mariselvam R, Athinarayanan G, Ranjitsingh AJA, Usha Raja Nanthini A, Rajapandiyan K, Alshatwi AA. Extraction of Dyes from *Petrocarpus santalinus* and Dyeing of Natural Fibres Using Different Mordants. *FIBRES & TEXTILES in Eastern Europe* 2018; 26, 5(131): 20-23. DOI: 10.5604/01.3001.0011.7312.
4. Mariselvam R, Ranjitsingh AJA, Mosae Selvakumar P, Rajapandiyan K, Alshatwi AA. Eco friendly natural dyes from *Syzygium cumini (L)* (Jambolan) fruit seed endosperm and to preparation of antimicrobial fabric and their washing properties. *Fibers and polymer* 2017; 18(3): 460-464.
5. MacVean, C. Lacquers and Dyes From Insects. In: Capinera, J.L. (eds) Encyclopedia of Entomology. Springer, Dordrecht 2008. https://doi.org/10.1007/978-1-4020-6359-6_1939.
6. Har Bhajan Singh, Kumar Avinash Bharati, in Handbook of Natural Dyes and Pigments, 2014.
7. Siddall R. Mineral Pigments in Archaeology: Their Analysis and the Range of Available Materials. *Minerals* 2018; 8(5): 201. https://doi.org/10.3390/min8050201.
8. Mumal S, Mona V, Lalita L. Eco-friendly surface modification of natural fibres to improve dye uptake using natural dyes and application of natural dyes in fabric finishing: A review. *Materials Today: Proceedings* 2021; 43(5): 2868-2871, https://doi.org/10.1016/j.matpr.2021.01.078.

9. Bharat P, Pragnya K. Sustainable dyeing and printing with natural colours vis-à-vis preparation of hygienic viscose rayon fabric.Sustainable *Materials and Technologies*. 2019; 22: e00116. https://doi.org/10.1016/j.susmat.2019.e00116.
10. Bechtold T, Turcanu A, Ganglberger E, Geissler S. Natural dyes in modern textile dyehouses — how to combine experiences of two centuries to meet the demands of the future?. *Journal of Cleaner Production*. 2003; 11(5): 499-509. https://doi.org/10.1016/S0959-6526(02)00077-X.
11. Kumar Samanta, Ashis, Nasser S. Awwad, and Hamed Majdooa Algarni, eds. 2020. "Chemistry and Technology of Natural and Synthetic Dyes and Pigments," September. IntechOpen. doi:10.5772/intechopen.83199.
12. Siva R. Status of natural dyes and dye yielding plants in India. *Current Science*2007; 92: 916-925.
13. Chengauah B.Mallikarjuna Rao K. Mahesh Kumar K Alagusundaram M, Madhusudhana Chetty C.Medicinal importance of natural dyes – a review. *Int. J. PharmTech Res* 2010; 2(1): 144-154.
14. Bechtold T, Turcanu A, Ganglberger E, Geissler S. Natural dyes in modern textile dyehouses – how to combine experiences of two centuries to meet the demands of the future? *J. Clean. Prod* 2003; 11: 499-509.
15. Goodarzian H, Ekrami E. Extraction of dye from Madder plant (*Rubia tictorium* L.) and dyeing of wool. *World Appl. Sci. J* 2010; 9(4): 434-436.
16. Komboonchoo S, Bechtold T. Natural dyeing of wool and hair with indigo carmine (C.I. Natural Blue 2), a renewable resource based blue dye. *J. Cleaner Prod*2009; 17: 1487-93.
17. Singh RV. Colouring plants – An innovative media to spread the message of conservation. *Down to Earth* 2001; 20: 25-27.
18. Balachandran B, Sabumon PC. Scope of Natural Dyes and Biomordants in Textile Industry for Cleaner Production. In: Muthu, S.S. (eds) *Novel Sustainable Process Alternatives for the Textiles and Fashion Industry*. Sustainable Textiles: Production, Processing, Manufacturing & Chemistry. Springer, Cham. 2023. https://doi.org/10.1007/978-3-031-35451-9_4.
19. Mariselvam R, Ranjitsingh AJA, Kalirajan K. Antifungal activity of different natural dyes against traditional products affected fungal pathogens. *Asian Pacific J. Trop. Biomed* 2012; 2: S1461-S1465.
20. Mariselvam R, Ranjitsingh AJA, Kalirajan K, Nanthini AUR, Athinarayanan G, Mosae Selvakumar P. Extraction of natural dyes from *Curcuma longa, Trigonella foenum graecum* and *Nerium oleander*, plants and their application in antimicrobial fabric. *Industrial Crop. Prod* 2015; 70: 84-90.
21. Guinot P, Gragadennec A, La Fisca P, Fruchier A, Andary C, Mondolot I.*Serratula tinctoria*, a source of natural dye: flavonoid pattern and histolocalization. *Industrial Crop. Prod* 2009; 29: 320-325.
22. Gupta D. Fastness properties of natural dyes. Part II: Effect of chemical structure on light fastness. *Colourage*1999; August: 41-46.
23. Schmidt A, Bach E, Schollemeyer E. The dyeing of natural fibers with reactive disperse dyes in supercritical carbon dioxide. *Dyes Pigment* 2003; 56: 27-35.
24. Mariselvam R, Ranjitsingh AJA, Kalirajan K. Anti-microbial Activity of Turmeric Natural Dyeagainst Different Bacterial Strains.*Journal of Applied Pharmaceutical Science* 2012; 2(6): 210-212.