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Review article

A recent (2009–2021) perspective on sustainable color and textile coloration using natural plant resources☆

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ABSTRACT

Fast fashion uses an excessive amount of synthetic dyes and chemical reagents in textile production, while a large quantity of fast fashion apparel and clothes go to the landfill, posting environmental safety concerns. Natural dyes not only produce delicate and subdued shades but also have the potential of novel features to achieve active textile substrate with performance properties such as deodorizing, antioxidant, antimicrobial, antifeedant, UV protection, etc. Developing colored textile products with natural colorants in today's market may enhance consumer interest to an even greater extent. Therefore, finding alternative natural degradable dyes has become one of the leading trends in this field. So far, multiple plants and agriculture byproducts have shown promising results in textile dyeing with increasing sustainability and environmental friendliness. There is no doubt in the general acceptance of natural colorants to be utilized as promising substitutes to synthetic dyes for certain categories of textile products, minimizing the negative impact on the health and the ecosystem. With the continuous advancement of natural dyeing research and technology, the dyes will be elaborated even more with finesse, color yield, stability, and colorfastness. This review gives the present status of natural colorants, natural dyeing and color presentation, natural dyeing methods, technique, and performance, mordants and mordanting for natural dyeing, and selection of suitable Agriculture products/byproducts for natural colorants. We hope to provide readers with specific angles on current natural dyeing applications in the textile and apparel industry.

1. Introduction

Natural plant dyeing is considered as an ecological dyeing technology on textiles or materials alike using dyes from natural plant resources. These dyes are extracted from natural plant parts including stems, roots, flowers, leaves, fruits, and peels. The earliest record to document the use of natural plant dyes can be traced back to 2600 B.C. in China. As archeological evidence indicated, a small number of plants and animal materials were initially used to extract natural dyes back then (Liu et al., 2021). Since the Middle Ages, the cultivation of dye plants and further processing and dyeing became an important economic factor in Europe. For instance, woad (*Isatis tinctoria*) in Germany, and madder (*Rubia tinctorum*) in the Netherlands and southern areas of France were used for fabric dyeing (Meyer, 1997).

More recently, synthetic dyes have replaced natural dyes during the industrial revolution and thousands of these chemical-made colorants are currently prevalent in the market (Hardman and Pinhey, 2009). With more and more advances and well-defined chemical structures, synthetic colorants have more advantages in handling, color performance, and reproducibility than natural dyes. Since then, synthetic dyes have been successfully applied in modern dye houses gradually and globally (Khattab et al., 2020). It is reported that textile industries, all over the globe, produce and use approximately 1.3 million tons of dyes, pigments, and dye precursors that cost around $23 billion (Technique Report 57, 2003). Unfortunately, many of these synthetic dyestuffs, auxiliaries, and dyeing wastes pose a significant threat to human health and the environment in an unsustainable manner. Millions of tons of textile dyeing, printing, and finishing wastes are being discharged into the eco-system annually. Since the millennium, there has been a trend of continued restrictions on synthetic dyeing chemicals, including azo dyes and toxic finishing agents used in textile industries (European Chemicals Agency, 2015; Union et al., 2020). Thus, textile scientists and coloration

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engineers are now in a constant search for greener and healthier dyeing alternatives, even if it is partially. With the trend in mind, natural dyeing is regaining interest and has the potential to be one of the promising technologies to minimize the environmental impact of textile dyeing using tremendous synthetic chemicals (Consumer Awareness for Natural Dyes, n.d.). As endorsed by most of the studies, the advantage of natural dyes lies in their subtle, soft, and elegant colors, together with none-toxic and health benefits, biodegradability, environmental compatibility, and even medical values (Shahid-Ul-Islam and Sun, 2017; Sutrisna et al., 2020; Grifoni et al., 2011; Shahid et al., 2013). Many current perspectives are strived to outline the functional performance of different textiles with natural dyes and functional agents exploited from natural sources (Nambela et al., 2020). In addition, the amount and properties of the wastewater produced in both natural and synthetic dyeing are critical to the eco system. Therefore, research on the comparison of both dyes would provide more supportive evidence for natural dyeing (Atav et al., 2020; Oktav Bulut and Akar, 2012).

Most of the current research efforts are focused on using different methods and sources of natural dyes to increase color payoff and colorfastness properties, e.g., colorant extraction and laboratory dyeing techniques, as well as to explore other important attributes of certain natural ingredients that can provide added benefits to materials. Staying on the edge of current advancement in this area, it is even more imperative in addressing natural dyeing with joint consideration of environmental, socio-economic, and sustainability. Therefore, the essential mission of this review is to screen and suggest suitable plant species for modern commercial textile applications on a relatively reasonable scale from a sustainable point of view. Considering the new motivation for natural colorant textile dyeing, this review article is intended to provide information and discussions regarding natural dyeing and to highlight some outstanding research studies conducted in this realm for the past decade and beyond. We first summarized current published data on natural textile dyeing and provide analysis mostly from publications during the last decade. Then, we identified agricultural products/by-products which have a high potential for colorant extraction and fabric dyeing. The aim is to give the present status of fibers for natural dyeing, dyeing methods and performance, mordanting, and selection of suitable plants, etc. With these endeavors, we hope to inspire further studies on plant growing, dye extraction, dyeing processing, textile and apparel application, and potential end uses of natural dyeing.

2. Fibers used for natural plant dyeing

A variety of natural dyes have been studied for their affinity to natural cellulosic fiber and natural protein fiber fabrics. In general, natural protein fibers, e.g., wool and silk, have a certain degree of affinity with natural dyes and usually form a dyes-fiber combination under the acidic condition, even without the help of mordant, which is usually utilized for better color presentation and colorfastness for protein fibers. In comparison, natural cellulosic fibers, e.g., cotton, usually do not have affirmed affinity to natural dyes, therefore, a mordant and/or pretreatment are needed for a better color presentation. For regenerated fibers like rayon, bamboo, lyocell, and modal fibers, since they share similar chemical composition with natural cellulosic fibers, this category of fibers can also be dyed using the same natural dyeing methods as cotton. Understandably, most studies on natural plant dyeing were applied to natural textile materials. On the other hand, natural dyes are usually considered unsuitable for these synthetic fibers, e.g., polyester (PET) and polypropylene (PP) because they have less affinity and low dyeability due to the high hydrophobicity and crystallinity of the structure.

In recent decades, researchers have started their experiments using natural dyes on synthetic textile materials. Polyamide (PA) fibers were the first being evaluated. Since polyamide fibers contain an amide group (-CONH) to preserve certain moisture regain and comparatively flexible polymer chains, both features are in favor of natural colorant dyeing. In one research article, polyphenolic dyes were extracted from henna leaves, pomegranate rind, and *Pterocarya fraxinifolia* leaves to study their dyeability on PA and PET fiber fabrics (Rahman Bhuiyan et al., 2018). The Fourier transform infrared spectra confirmed the coordination complexes and p–p bonding between the mordants and the dyes (Ebrahimi and Parvinzadeh Gashti, 2016). Madders, areca nut, faba bean, and eucalyptus woods were also evaluated for the dyeing potential on PA fibers (Sadeghi-Kiakhani, 2015; Bhuiyan et al., 2017; Pawar et al., 2018; Erdem Ismal and Yıldırım, 2020; Rossi et al., 2017). In another article, indigo carmine, cochineal carmine, curcumin, and annatto were encapsulated in silica by a sol-gel method and applied in the dyeing of different synthetic fibers including PA and PET. Color change and color transfer of the encapsulated dyes were better compared to the unencapsulated dyes (dos Santos et al., 2018). Besides, waste parts of allium cepa skin, annatto seeds, terminalia chebula, and leaves of macaranga peltate were also utilized as a source of natural colorants to dye PET fibers with good color performance (Pawar et al., 2019; Nakpathom et al., 2019; Lee et al., 2020; Manicketh and Francis, 2020). Recently, He et al. have developed an environmentally friendly coloration process on synthetic fiber using natural colorants, with the assistance of polyphenolic polydopamine anchors, which has achieved good color appearances and acceptable fastness on PET fibers (He et al., 2018). In addition to PET and PA, polyacrylonitrile (Acrylic) could be dyed with colorants from *Opuntia ficus-indica* and cochineal carmine (Guesmi et al., 2012b; dos Santos et al., 2018). More importantly, the fact that synthetic fibers might be reasonably dyed by natural plant colorants will significantly enlarge the scope of application and market due to the domination of synthetic fibers in the current marketplace.

Based on generic fiber types, natural plant dyeing on different generic fibers was summarized in Figure 1. Studies on natural protein fibers account for half of natural dyeing research studies, while natural cellulosic fibers account for more than one-third. Specifically, cotton and wool are representative fibers for natural dyeing where more dyeing potential has been exploited. Although the natural dyeing of natural fibers dominated the past studies, the potential of dyeing synthetic fibers with natural dyes has been exploited with a few attempts.

3. Natural dyeing from a variety of plant sources and color gamut presentation

The interest in using natural dyes from various types of plant sources in textile dyeing has been revived due to their biodegradability, renewability, and environmental-compatibility features (Deveoglu et al., 2012a,b). Generally, natural dyes can be categorized into two main categories: substantive dyes and adjective dyes. Substantive dyes become chemically attached to the fiber without the assist of any other chemicals, e.g., indigo or certain lichens. Adjective dyes require a metal salt to prevent the color from washing off or light bleaching. Most natural dyes are adjective dyes and require the utilization of metal salt solution during the dyeing process. In addition to the traditional classification scheme, natural colorants from plant and animal sources can also be categorized based on their chemical structures such as polyphenols (e.g. anthocyanins, flavanol-quercetin, or curcumin from berry, pomegranate, bark, turmeric), isoprenoids (e.g. iridoids, carotenoids and quinones from gardenia, pumpkin, walnut), heterocyclic compounds (e.g. betalains or indigoids from Indigoid, beetroot, dragon fruit), melanin (e.g. from nut shells), and tetrapyrroles (e.g. from algae; Brudzyńska et al., 2021). In addition, based on the shade, the natural colorant can also be roughly classified as yellow colors from flavonoids, red colors from anthraquinone, purple and blue colors from indigoids, brown and black colors from tannins, and colors dyes from chlorophyll (Zhao et al., 2020). Colorants from fungal and algal have also been studied for textile dyeing (Räisänen, 2019; Weber et al., 2014; Vicente et al., 2020; Azeem et al., 2019). In addition, unlike color matching for synthetic dyes, there have been very limited attempts on natural dyeing using a combination of dyestuffs to achieve satisfactory color matching results (Ding and Freeman, 2017).

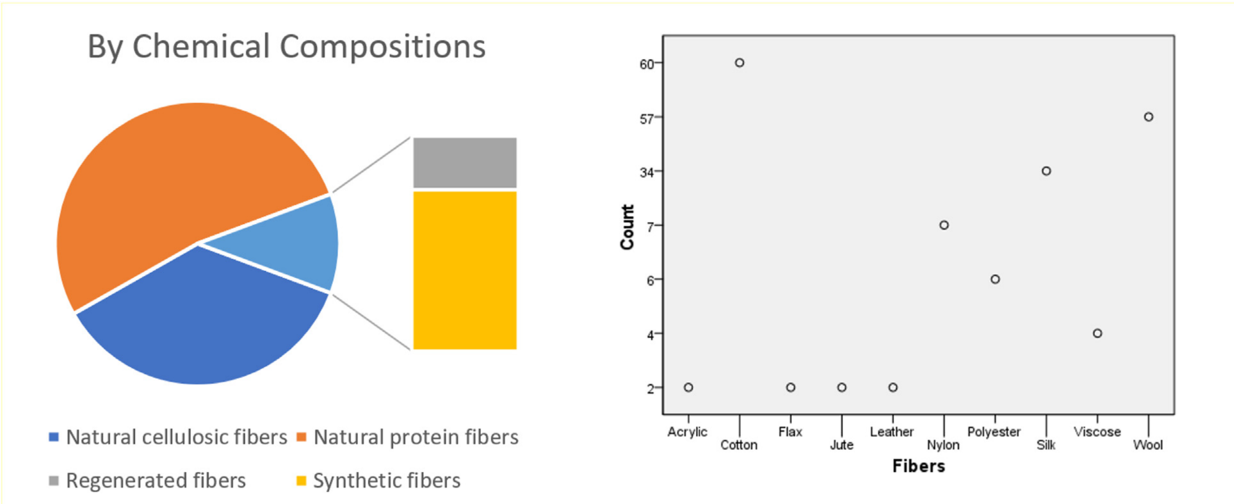


Figure 1. Fibers for natural plant dyeing.

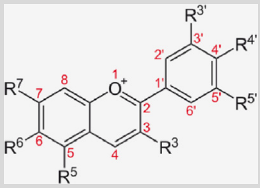
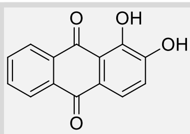
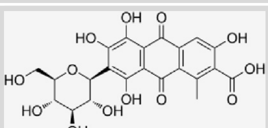
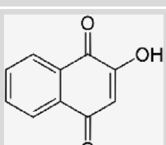
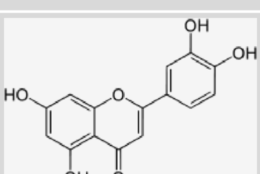
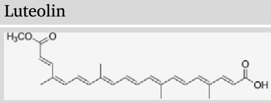
In textile dyeing practices, the K/S (K for Absorption coefficient and S for Scattering coefficient of a dye molecule) value is a critical parameter representing the color depth and color strength of a dye (Burkinshaw, 2015). The higher the K/S value, the deeper colors could be achieved. Literature shows that synthetic dyestuffs in blue or black can easily achieve the K/S values between 20 to 30 on textiles with 2–3% o.w.f., while the K/S value of other hues is comparatively lower (Ahmed et al., 2006). Referring to all reference articles in this review, about 13% of natural dyes were reported to achieve a comparatively darker shade with a K/S value of more than 20. While 41% of natural dyes can merely achieve a K/S value less than 5.0, which indicates very light colors. Since the K/S comparison is more sensible for colors of the same hue, a summary table has been designed with data from the most recent articles, which is shown in Table 1. This table includes the top 10 plants and plants dyestuffs which have been studied based on the frequency. The K/S values of each plant dyestuff on different fibers are listed, reflecting the deepest and strongest color that could be achieved with each dye. With no exception, shades on protein fibers are deeper (K/S $>$ 10.0) than shades on cellulosic fibers for the same color. Surprisingly, synthetic fibers like nylon and polyester can achieve colors with K/S values as high as 31.9. Pantone and Munsell's color indexes are searched using the CIELAB coordinates from research articles for the closest colors in the current Pantone fashion color system and Munsell color system, indicating the range of colors can be achieved with specific natural dye. Pantone indexes are also provided for designers who would like to pursue color design using sustainable natural colors. Further, the major Munsell Hues (H) of these colors achieved are located at Yellow, Yellow-Red, Red, and Red-Purple regions in the Munsell color space with commercially acceptable colorfastness. For these colors, the Munsell Values (V) are mostly ranging from 2 to 8 (0–10 scale), while the highest Chroma(C) can be achieved is about 14. In addition to these colors, there is a limited number of articles that was reported for color in green, in neutral grey colors with Munsell C $\leq$ 2, and in black (adjacent) colors with Munsell V $\leq$ 2 and C $\leq$ 2 (Meisheng Book of Natural Plant Colors, 2021).

In addition to the color depth and color strength, finding more colors to fill the gap in the natural color hue circle has always been a challenge for researchers and practitioners. To understand the color gamut that natural dyeing could achieve, we have selected 17 natural plant dyes from the "MYSUNBIO book of Natural Colors" and dyed cotton fabrics with 5% o.w.f. (MYSUNBIO Book of Natural Plant Colors, 2021). These fabrics were then measured with a spectrophotometer for CIELAB color coordinates. Then, all colors were plotted into CIELAB color space, which is shown in Figure 2. Most of the current commercialized natural colors for textiles are located in the first quadrant of the a*–b* plane, which indicates that the majority of colors from natural plant sources have a red-orange-yellow hue. Indeed, there is an obvious shortage of natural dyes in blue. This phenomenon was explained in that the blue portion of the visible spectrum provides sufficient energy to raise an orbital electron to an excited state and hence force molecules to absorb it, causing these pigments to appear more reddish or greenish (Siddique et al., 2016),. Thereby, if blue colorants from plant sources are reflecting the highest-energy light and absorbing only poorer quality light, then the amount of light that could be effectively taken in by plants in blue is ultimately limited for its growth. It is reported that most biological pigments absorb blue light since its wavelengths are shorter and higher in energy than most colors. Numerically, considering each CIELAB coordinate, a* (red-green scale) is ranged from -15 to 40, b* (yellow-blue scale) is ranged from -10 to 55, and L* (black-white scale) is ranged from 30 to 85, respectively.

4. Functional properties from the natural dyeing

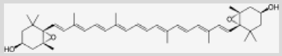
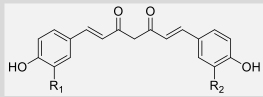
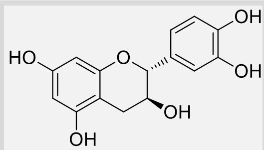
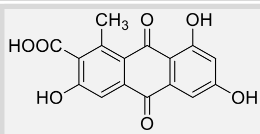
Consumers around the world are longing for textile products that provide special performance or functions like greater comfort and remain hygienic in use. The functional finish is always an exciting treatment where fabrics could achieve such special performance/functional properties during textile wet processing. Usually, the finishing is an additional application process after dyeing. However, some natural plant dyes can provide desirable finishing properties during the same dyeing process. This is a great drive to investigate innovative methods, for the development of hygienic textile products, in textile finishing processes using natural plant extracts/dyes. Some reports are now available on natural colorants for imparting multifunctional properties to textiles such as antioxidant (Kulczyński et al., 2020), mothproof (Nazari et al., 2014), antimicrobial (Singh et al., 2005), insect repellent (Kato et al., 2004), deodorizing (Hwang et al., 2008), UV-protective (Wang et al., 2009), and anti-creasing properties (Sadeghi-Kiakhani et al., 2018b). Some plant dyes have dual effects of color and antimicrobial functions, and they contain antimicrobial active ingredients such as alkaloids, acids, ketones, phenols, and citric acid (Dev et al., 2009). Ellagic acid and polyphenolic compounds in some plant dyes have a measurable antioxidant effect. More natural plant dyes are found to provide UV protective features, which are influenced by many factors such as the structure and physiochemical nature of fiber, dyes, and finishes, fabric thickness, porosity, and moisture content (Gies, 2007). In addition, some natural dyes have insect-repellent and mildew refrain features, e.g., fungicides derived from maple or eucalyptus tree and indigo for medical applications.

Table 1. Top 10 plant dyes in the research articles and their color gamut with Pantone and Munsell index.

Plant names & parts	Main chemical components of color	Max K/S achieved	Pantone Index	Munsell Index	Ref.
Part 1: Top 10 Plants studied for natural dyes.					
Pomegranate (*Punica granatum*) & Peels	 Anthocyanin	~ 14.7 (Hemp) 13.2 (Wo) 9.4 (Co) 6.4 (Lyocell)	19-0405 TCX 8–1112 TCX 18-0724 TCX 18–0510 TCX 17-1129 TCX 17–1113 TCX 17-0929 TCX 17–0636 TCX 17-0517 TCX 16–1133 TCX 16-1126 TCX 16–1120 TCX 16-1118 TCX 16–1108 TCX 16-0928 TCX 16–0737 TCX 16-0730 TCX 16–0726 TCX 15-1116 TCX 15–1040 TCX 15-0719 TCX 15–0533 TCX 15-0522 TCX 14–0925 TCX 14-0721 TCX 13–0919 TCX	10YR/3/2 10YR/6/4 10YR/6/6 10YR/6/8 10YR/7/4 10YR/7/6 10YR/8/6 7.5YR/5/8 7.5YR/6/6 5Y/8/8 2.5Y/4/2 2.5Y/5/4 2.5Y/5/6 2.5Y/6/6 2.5Y/6/4 2.5Y/7/4 2.5Y/8/4 (H/Y ~ YR; V/3–8; C/2–8)	(Ghaheh et al., 2012; Mahmud-Ali et al., 2012; Davulcu et al., 2014; Ajmal et al., 2014; Ebrahimi and Parvinzadeh Gashti, 2016; Benli and Bahtiyari, 2018a; Čuk and Gorjanc, 2017; Rehman et al., 2018; Baseri, 2020; Inprasit et al., 2020; Peran et al., 2020)
Madder (*Rubia tinctorum*) & Roots	 Alizarin	~ 18.4 (Nylon) 15.8 (Wo) 8.8 (Co)	19-1234 TCX 19–1540 TCX 19-1103 TCX 18–1440 TCX 18-1433 TCX 18–1326 TCX 18-1230 TCX 18–1016 TCX 17-1424 TCX 15–1333 TCX 14-1318 TCX	10R/3/8 10R/4/4 10R/4/8 10R/4/8 10R/5/8 10R/6/8 10R/8/10 7.5YR/4/4 5YR/3/4 2.5YR/2/2 2.5YR/7/8 **(H/R-YR; V/2–8; C/2–10)**	(Barani and Maleki, 2011; Zarkogianni et al., 2011; Farizadeh et al., 2010; Deveoglu et al., 2012a,b; Sadeghi-Kiakhani, 2015; Grifoni et al., 2014; Parvinzadeh Gashti et al., 2014; Tayade and Adivarekar, 2016; Sadeghi-Kiakhani et al., 2018b; Fröse et al., 2019; Shahmoradi Ghaheh et al., 2014; Atav et al., 2020)
Cochineal (*Dactylopius coccus*) & dried body	 Carminic Acid	~ 31.9 (PET) 27.7 (Wo) 20.5 (Co)	19-4022 TCX 19–2118 TCX 19-1725 TCX 18–3912 TCX 18-3710 TCX 18–2525 TCX 18-1512 TCX 17–1605 TCX 17-1511 TCX 15–1906 TCX 13-1520 TCX 5025 U	10R/5/2 10R/6/4 10R/8/4 10R/8/6 7.5R/2/2 7.5R/2/6 7.5R/4/4 7.5R/5/4 7.5R/5/4 7.5 R P/2/2 7.5 R P/4/8 2.5R/4/2 **(H/R-RP; V/2–8; C/2–8)**	(Shams Nateri et al., 2016; Sadeghi-Kiakhani et al., 2018a; Zarkogianni et al., 2011; Mehrparvar et al., 2016; Ding and Freeman, 2017; dos Santos et al., 2018; Sadeghi-Kiakhani et al., 2018b; Fröse et al., 2019; Giacomini et al., 2020)
Henna (*Lawsonia inermis*) & Leaves	 Lawsone	~ 22.4 (Wo) 16.5 (Nylon) 9.7 (Co)	19-4008 TCX 19–3911 TCX 19-3909 TCX 19–1432 TCX 19-1034 TCX 19–1018 TCX 19-0822 TCX 19–0814 TCX 17-1129 TCX 17–1125 TCX 17-1052 TCX 17–1044 TCX 17-0935 TCX 14–0936 TCX 13-0922 TCX 732 C	10YR/4/6 10YR/5/6 10YR/8/6 7.5YR/1/2 7.5YR/2/4 7.5YR/5/6 7.5YR/6/8 7.5YR/8/8 5YR/1/2 5YR/2/2 5YR/2/6 5YR/4/8 2.5Y/3/2 2.5YR/1/4 **(H/YR-Y; V/1–8; C/2–8)**	(Zarkogianni et al., 2011; Yusuf et al., Manzoor, 2012; Wang et al., 2016; Bhuiyan et al., 2017; Alebeid et al., 2020a,b; Ebrahimi and Parvinzadeh Gashti, 2016; Shahmoradi Ghaheh et al., 2014)
Weld (*Reseda luteola*) & Leaves	 Luteolin	~ 1.4 (Co) 61.7 (L*, Wo)	16-1133 TCX 16–0737 TCX 16-0730 TCX 15–1217 TCX 13-0608 TCX	5Y/8/4 2.5Y/8/4 2.5Y/7/4 2.5Y/7/6 2.5Y/6/6 (H/Y; V/6–8; C4-6)	(Deveoglu et al., 2012a, b; Ghoranneviss et al., 2011; Sadeghi-Kiakhani et al., 2018b; Fröse et al., 2019), 142
Annatto (Bixa orellana) & seeds	 Bixin and norbixin	~ 23.1 (PET) 5.5 (Co)	18-1235 TCX 18–1244 TCX 17-1347 TCX 17–1327 TCX 16-1164 TCX 14–1133 TCX	5YR/5/6 5YR/3/6 2.5YR/4/6 2.5YR/5/8 2.5YR/8/12 2.5YR/6/14 (H/YR-R; V/3–8; C6-14)	(Savvidis et al., 2013; Chattopadhyay et al., 2014; dos Santos et al., 2018; Kesornsit et al., 2019; Nakpathom et al., 2019)

(*continued on next page*)

Table 1 (*continued*)

Myrobalan (*Terminalia*) & fruits/seeds	 Violaxanthin	~ 19.0(Wo) 6.8 (Leather)	18-1018 TPG 17–1128 TCX 17-1052 TCX 17–1045 TCX 17-1022 TCX 16–1118 TCX 161 U 15–1125 TCX	10YR/4/4 7.5YR/7/6 7.5YR/6/6 7.5YR/5/6 5YR/4/8 2.5Y/6/4 2.5Y/5/4 (H/YR-Y; V/4–7; C/4–8)	(Shabbir et al., 2016; Song et al., 2017), 138, (Rather et al., 2019; Adeel et al., 2019; Sinha et al., 2016)
Turmeric (Curcuma longa) & root	 Curcumin	~ 22.6 (PET) 8.8 (PA)	19-3911 TCX 19–0840 TCX 19-0815 TCX 19–0612 TCX 18-1160 TCX 18–0935 TCX 18-0627 TCX 17–1113 TCX 17-1009 TCX 17–0949 TCX 16-1149 TCX 14–0756 TCX	10YR/3/4 10YR/5/8 10YR/6/4 7.5YR/2/2 5R/1/2 5YR/5/10 2.5Y/3/4 2.5Y/4/4 2.5Y/8/12 (H/Y-YR-R; V/1–8; C/2–12)	(Bhatti et al., 2010; Tayade and Adivarekar, 2016; Ma et al., 2020; Shahmoradi Ghaheh et al., 2014; Čuk and Gorjanc, 2017; dos Santos et al., 2018)
Tea (Camellia sinensis) & Leaves	 Catechins	~19.4(Wool) 9.5(Cotton)	16-1326 TCX 16–1317 TCX 15-1119 TCX 15–1114 TCX 15-0628 TCX 14–1213 TCX 14-1113 TCX 13–0919 TCX 12-0605 TCX 12–0418 TCX 11-0701 TCX	10YR/6/4 10YR/6/6 10YR/7/4 10YR/7/6 10YR/8/4 10YR/8/6 5YR/6/6 7.5YR/8/6 7.5YR/9/4 5Y/7/4 5Y/8/4 2.5Y/7/6 (H/YR-Y; V/6–9; C/4–6)	(Čuk and Gorjanc, 2017; Shahid-ul-Islam et al., 2018a,b; Sukemi et al., 2019; Shahmoradi Ghaheh et al., 2014; Bonet-Aracil et al., 2016; Deveoglu et al., 2012a,b)
Lac (Kerria lacca) & secretes	 **Laccaic acid**	~ 31.8 (L*, Silk)	19-1245 TCX 19–1101 TCX 17-1516 TCX 15–1906 TCX	10R/5/6 7.5R/7/6 5YR/2/2 2.5YR/4/8 (H/R-YR; V/2–7; C/2–8)	(Chimprasit et al., 2019; Kesornsit et al., 2019; Sombatdee and Saikrasun, 2020; Tayade and Adivarekar, 2016)
Part 2: Other plants studied for natural dyes					
Chestnut; Logwood*; Sticta coronate*; Walnut; Citrus/Orange; Indigo; Purple-fleshed sweet potato; Almond shell; Buckthorn; Goldenrod; Tectona grandis L.; *Macaranga Peltata*; *Monascus; Opuntia ficus-indica; Lavende; Pterocarya fraxinifolia; Bixa orellana seed; Fallopia japonica;* Areca nut; Marigold flower; Camphor leaves; Faba bean husk; Gallnut; Alkanet roots*; Lxora coccinea L.; Blackcurrant*; Hibiscus flower; Mulberry (Morus rubra); *Eupatorium odoratum leaves; Flowers of Tabebuia argentea*; Mangrove bark; *Saraca asoca; Albizia lebbeck; Acacia cyanophylla; Helichrysum arenarium*; Quince leaves; Hazelnut; *Rubia cordifolia roots; Rheum emodi L.; Alkanna*; Brazilwood; Sandalwood; Safflower; Amur corktree*; Chrysanthemum boreale; Artemisia*; Walloon oak acorn caps; Chicken gizzard; *Alpinia blepharocalyx; Thyme; Pseudomonas species*; Alloon oak*; Onosma echoides*; Gromwell; Bastard hemp; Rose; Rosemary; *Acacia arabica* bark; *H. italicum* (Roth); *G. Don* (curry plant); *D. gnidium L.* (daphne); *C. scolymus L.* (artichoke); *Sesbania aculeata*; Calico leaves; Dates; Persimmon; *Spathodea campanulata* flower; Oak bark extract*; Cassia Singueana* bark; Avocado seed; *Commiphora* of the *Burseraceae* shrub; *Yerba mate; Camellia oleifera Abel* shell; Coffee; Pine cone; Peanut skin; Pineapple; *A. nilotica*; *A. vasica*; *Cladophora glomerata L. (Chlorophyta)* alga; *M. anisopliae*; Ketapang leaf; *Ficus amplissima* Smith; *Coccus laccae* insect; Fungal; Bark of *Araucaria columnaris*; Leaves of *Macaranga peltate*; *Averrhoa bilimbi*; Sappanwood; Mango leaf; Blueberry, etc.					

Articles related to the natural dyes' functional performance finishing are carefully reviewed and summarized in Table 2. The top two functions that natural plant dyes can bring to textiles are antimicrobial, and UV protective performance. Specifically, the natural plant dyes can inhibit a broad range of bacteria including Gram-positive bacteria (e.g. *Staphylococcus aureus*) and Gram-negative bacteria (e.g. *Klebsiella pneumonia*;

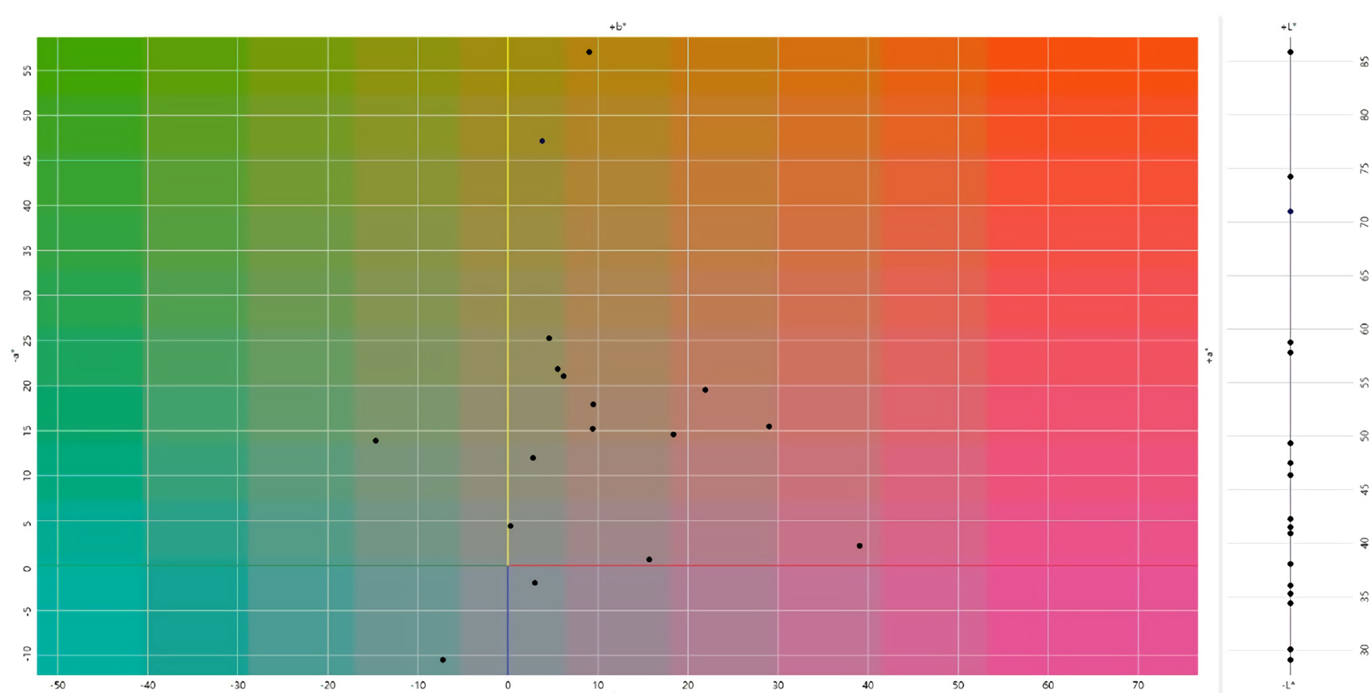


Figure 2. Color coordinates of 17 natural plant dyes in a* and b* plane and L* axis of the CIELAB color space (Courtesy from and credit to MYSUNBIO®).

Table 2. Accumulated data of function performance provided by natural dyeing.

Functions	Plants	Ref.
Antibacterial/ Antimicrobial	Quince leaves, Tectona grandis L. leaves extract, Rheum emodi L., henna, pomegranate, thyme, gallnut extract, gromwell, Pterocarya fraxinifolia, Bixa orellana seed, Acacia arabica bark, Chinese gall, green tea, coffee, pinecone, pineapple peel waste, pomegranate rind, buckwheat hull, Croton urucurana Baill. bark, green tea, turmeric, Commiphora of the Burseraceae shrub, Saraca asoca and Albizia lebbeck, Citrus grandis Osbeck, henna, Walnut Sheel, Cochineal, madder, weld, onion skin, pineapple peel waste	(Cerempei et al., 2016; Shahid-ul-Islam et al., 2018a,b; Khan et al., 2012; Alebeid et al., 2020a,b; Davulcu et al., 2014; Rehman et al., 2018; Lee et al., 2015; Hong et al., 2012; Ebrahimi and Parvinzadeh Gashti, 2015; Chattopadhyay et al., 2015; Zhang et al., 2014; Shahid-ul-Islam et al., 2018a, b; Hong, 2018; Lee et al., 2018; Sheikh et al., 2019; Baseri, 2020; Inprasit et al., 2020; Peran et al., 2020; Zhang et al., 2020; dos Silva et al., 2020; Shahmoradi Ghaheh et al., 2014; Lee et al., 2017; Baliarsingh et al., 2012; Yi and Yoo, 2010; Yusuf et al., 2012; Ghaheh et al., 2012; Sadeghi-Kiakhani et al., 2018b; Pawar et al., 2019; Sheikh et al., 2019; Inprasit et al., 2020; Lee et al., 2020; Haji, 2012)
UV protective	Henna, Bixa orellana seed, Acacia arabica bark, Marigold (Tagetes erecta), Peanut skin roasted, Pineapple peel waste, A. nilotica, buckwheat hull, Golden shower pods, Neem bark, Andaman satinwood leave, Burma padauk bark, Sappan, Acacia bark, Croton urucurana Baill bark, Equisetum arvense L., Oak Bark Extract, curcurmin, H. italicum (Roth) G. Don (curry plant), R. peregrina L. (wild madder), D. gnidium L. (daphne), L. stoechas L. (wild lavender) and C. scolymus L. (artichoke), Green/Red/Black Tea	(Yadav et al., 2019; Chattopadhyay et al., 2015; Čuk and Gorjanc, 2017; Shabbir et al., 2018), 108, (Sheikh et al., 2019; Rather et al., 2019; Zhang et al., 2020; Rungruangkitkrai et al., 2020; dos Silva et al., 2020; Park and Park, 2020; Jia et al., 2017; Čuk and Gorjanc, 2017; Grifoni et al., 2014; Bonet-Aracil et al., 2016; Baseri, 2020)
Antioxidant	Purple-fleshed sweet potato, Tectona grandis L. leaves extract, green/red/ black tea, coffee, A. nilotica, Equisetum arvense L. (Equisetum)	(Koh and Hong, 2017; Shahid-ul-Islam et al., 2018a,b; Bonet-Aracil et al., 2016; Hong, 2018; Rather et al., 2019; Park and Park, 2020)
Deodorizing	Amur Corktree, gallnut extract, Commiphora of the Burseraceae shrub, Pinecone	(Farizadeh et al., 2010; Lee et al., 2015; Lee et al., 2017; Lee et al., 2018)
Anti-creasing	Cochineal, madder and weld	(Sadeghi-Kiakhani et al., 2018b)
Mothproof	Madder	(Nazari et al., 2014)

Hong et al., 2012). One example of UV protective benefit from *Flos sophorae* dyes is that the dyed silk fabric has a UPF of 60 (Wang et al., 2009). There are other functions that natural dyes can bring to fabrics, including deodorizing, anti-creasing, and mothproof (Farizadeh et al., 2010; Nazari et al., 2014; Koh and Hong, 2017; Sadeghi-Kiakhani et al., 2018). The flame-retardant property was also reported in the past but there was not much progress in recent decades. In general, there is great potential for antimicrobial and UV protective properties from natural plant dyes, while other functions like antioxidant and deodorizing are some new areas to explore.

5. Special treatment of fabrics for natural dyeing

There are great potential to explore modern technology for natural textile dyeing by making the dyeing process more cost, time, and energy effective. In order to enhance the color performance of naturally dyed fabrics, textile scientists and colorists made endeavors to apply a physical or chemical treatment to textile dyeing, which are summarized in Table 3. These special treatments are usually categorized as wet treatment and dry treatment. Examples of wet treatment include surface modification agents containing cationic and anionic groups to treat

Table 3. Accumulated data of special treatment for natural dyeing.

Specialty Treatment	Plants	Fibers	Prominent Features	Ref.
Chitosan	Lac, Cochineal, Onion, Tea, Madder, Weld	Cotton, Silk, Wool	Metal mordants replacement, Biodegradability, non- toxicity, and antimicrobial	(Mehrparvar et al., 2016; Bonet-Aracil et al., 2016; Verma et al., 2017; Shahid-ul-Islam et al., 2018a,b; Sadeghi-Kiakhani et al., 2018b; Chimprasit et al., 2019)
Chemical Modification	Areca nut extract; *Phellodendron amurense Rupr*;	Cotton, Wool, Polyester, Nylon	Molecular level, Improved colorfastness More efficient dyeing process on time, cost, and energy	(Pawar et al., 2018; Kim and Park, 2007; Oktav Bulut and Akar, 2012)
Plasma	Almond shell, madder, Rose/ Rosemary/Lavender/Mate tea, (Fallopia japonica) rhizome, Cotton pods (top), Arnebia euchroma (middle), Harmal seeds (bottom), pomegranate peel, grape leaves	Wool, Cotton, Bamboo	Surface medication, No industrial waste and shortening dyeing process Reduce the dyeing temperature Improve absorbency and dyeability	(Erdem Işmal et al., 2013; Barani and Maleki, 2011; Deveoglu et al., 2012a, b; Gorjanc et al., 2016; Haji, 2020; Peran et al., 2020; Haji and Payvandy, 2020; Peran et al., 2020)
Ultrasonic or microwave	Coconut coir, turmeric, green tea	Silk, Wool, Tencel	Better dye diffusion Lower dyeing temperature Improved color strength Faster and high efficiency	(Adeel et al., 2020; Ma et al., 2020; Sukemi et al., 2019)
Ozone	Pomegranate peel, onion skin, nutshell, orange tree leaves, alkanet root	Cotton	Alternative for mordants Improved dyeability Improved colorfastness	(Benli and Bahtiyari, 2018a),
Gamma/UV Radiation	Turmeric, chicken gizzard, pomegranate, calico leaves	Cotton, Silk	Improved cross linking, grafting, and dye uptake Improved resistance to shrinkage and wrinkling Enhanced water repellency and color strength	(Bhatti et al., 2010; Batool et al., 2013; Ajmal et al., 2014; Khan et al., 2014)

cotton and wool fabrics (Kim and Park, 2007; Gorjanc et al., 2019). The treatment with these agents offered improved color, colorfastness, and functional characteristics of dyed fabrics to a significant extent, along with an eco-compatibility feature of the treatment (Oktav Bulut and Akar, 2012). Some studies exploited the potential applicability of chitosan, a naturally occurring biopolymer with distinct chemical and biological properties, as a surface modification agent to improve dyeing performance on cotton (Verma et al., 2017), silk (Chimprasit et al., 2019), and wool fabrics (Mehrparvar et al., 2016) with increased binding sites and better dye absorption. Other studies also showed the pre-treatment of fabrics with chitosan increased antibacterial and antioxidant activity on both cotton and wool fibers (Shahid-ul-Islam et al., 2018a,b; Sadeghi-Kiakhani et al., 2018b).

For the dry treatment, various finishing treatments of textile fabrics by modern surface modification techniques are considered to be superior to traditional chemical modification methods and related to a more environmentally friendly textile processing methodology. Examples of dry treatments include plasma, ozone, and radiation treatments. The low-temperature plasma (LPT) treatment of fibers improves the surface characteristics of fibers while the bulk properties of the fibers are not affected (Erdem Işmal et al., 2013; Barani and Maleki, 2011; Deveoglu et al., 2012a,b; Gorjanc et al., 2016; Haji, 2020; Peran et al., 2020; Haji, n.d.; Haji and Payvandy, 2020). Ultrasonic radiation can be used to maximize the extraction and dyeing process via the cavitation effect (Vinod et al., 2010). Ozone is a strong oxidizing agent and can be used in many chemical reactions. It has generally been examined as a bleaching process as well for the modification of fibers before dyeing (Benli and Bahtiyari, 2018a,b). Gamma radiation treatment of fabric may facilitate shrinking and wrinkling resistances of fibers, improve the rate of dye uptake, fixes more dye on fibers as well as improve the shade of the dyed fabric, which not only improves the extraction of colorant but also deepens the color of the extract (Bhatti et al., 2010; Batool et al., 2013; Ajmal et al., 2014; Khan et al., 2014). There are much more materials used in addition to chitosan like other biopolymers, alginates, cyclodextrins, gums as well as their derivatives, and they were not included in this review. Overall, there is significant potential to reduce the ecological impact of existing processes with the introduction of these new technologies of textile dyeing and finishing without compromising eco-safety standards. It is no doubt that modern technology will bring more promising possibilities for natural dyeing applications.

6. Mordants and classification for natural dyeing

The term “mordant” comes from the Latin “mordere”, which means “to bite”. A mordant is a chemical substance used to bind dyestuffs on fabrics by forming a coordination complex with the dyestuffs, which then affixes to the fibers (IUPAC, 1997). Over the centuries many interesting substances were used as mordants to ensure dyeing colorfastness, including arsenic and other deadly chemicals (Llewellyn, 2005). In history, most dyers mordanted their yarns and fabrics before dyeing them. Alum and iron were extensively utilized as mordants in Egypt, India, and Assyria, as there are many alum deposits in the Mediterranean region (Manhita et al., 2011). In addition to generating affinity between dye and fibers, the use of mordants also alters the lightness, hue, and chroma of certain dyes. That is, with the same dye, different mordants may be darker, brighter, or even drastically change the final hue of the dyed materials (Yi and Cho, 2008). Natural protein and cellulose fibers need to be mordanted differently because of their structural and chemical composition.

In rural areas where these metal agents were not widely available, plants extracts were also used as mordants, especially those that have a natural ability to extract such minerals, such as club moss, which could be used for dyeing fabrics, intensifying colors, improving colorfastness, or achieving special properties Three mordanting techniques - pre-mordanting, simultaneous mordanting, and post-mordanting have been developed over the history to produce beautiful shades with improved dyeing performance. The use of mordants in natural dyeing has provided textiles with durable functional properties and boosted their applications in textiles for various end-uses including sportswear, fashion apparel, medical sector, and carpet industries (Manian et al., 2016).

As discussed, the mordants have dominated the natural textile dyeing and played a critical role in achieving colors with certain permanence. The accumulative results of mordants studied in recent studies are detailed in Table 4. It shows that traditional mordants, alum compound, ferrous compound, and copper compound, are still the most utilized ones which provide the most effective mordanting performance and cost. These three types of mordant account for about the majority (two-thirds) of studies in the time frame of this review. The metallic mordants are used to apply to natural fibers dyeing only. In addition, recent researchers have started to exploit the potential in dyeing synthetic textiles materials like nylon, polyester, acrylic, etc., with the assist of mordants. Besides, regenerated textile materials, e.g., lyocell and Bamboo fibers, were also being evaluated for their natural dyeing potential with mordants. For metallic mordants, progress has been made towards novel metallic salt or fewer metal ions to be used. Some minority mordants are also being evaluated as substitutes to traditional mordants with better performance regarding color and colorfastness. In addition to metallic mordants, chemicals like rare earth chlorides, together with their environmental traits, were employed as a substitute mordant for the natural dyeing which greatly reduced the ionic concentration in natural dyeing. There were a few attempts using a combination of different metallic mordants for dyeing (Kumaresan, 2016; Dehnavi et al., 2016). All in all, though new mordants have experimented for better coloration performance of textiles, traditional alum, ferric, and copper mordants are still dominate in the natural dyeing applications.

On the other side, the application of metallic mordanting in natural dyeing applications has been a major concern in recent years because of the consumer's awareness of the toxic and carcinogenic consequences associated with metal mordants (Shahid et al., 2013). Therefore, there has been an increasing demand to explore alternative environmentally benign agents for mordanting of textiles. Considering this voice, bio-mordants extracted from medicinal plants have been introduced as an interesting alternative to metal mordants because of their remarkable properties. As shown in Table 5, the summarization of bio-mordants and their usage in natural textile dyeing is presented based on the mordant sources, chemical composition, and generic fiber types. In the past, Bio-mordant sources are plants with either high tannin content or high metal hyperaccumulating. Bio-mordants can be applied to cellulose fibers such as cotton and linen which usually involve the use of baking soda or tannins to create an alkaline dyebath. Vegetable tannins are water-soluble polyphenolic compounds from oak galls, chestnut, lemon, and turmeric) which were used in dyeing cellulosic fibers as they attach well to the plant fibers. Chlorophyll extracted from different plant sources has also been successfully employed as bio-mordant (Guesmi et al., 2013). In addition to cellulosic fibers, there were several attempts on natural dyeing of protein fibers and synthetic fibers. Mordants for protein fibersare usually applied in acidic dyebaths. Cream of tartar, citric acid, and oxalic acid are bio-mordant which are used to assist the dyes in taking to the protein fibers. Some other novel bio-mordants are extracted from whey, milk powder, mango bark, aloe vera, sodium alginate, and sodium carbonate. Bio-mordanting provides a truly sustainable natural dyeing solution since it eliminates the use of metal mordants relating to their potential health issues. The use of natural bio-mordant would be another area of advancement for future natural dyeing.

7. Natural dyeing market and application

Natural textile dyeing is traditionally done by artisans and craftsmen on a very limited scale. Based on the concept by Hartl and Vogl (Hartl and Vogl, 2003), with a few modifications, a structure of four technological levels of natural textile dyeing was developed and shown in Table 6,

Table 4. Metallic mordants utilized for natural dyeing.

Name	Fibers	Key Features	Ref.
Alum Compound - $KAl(SO_4)_2$; $Al_2(SO_4)_3$; $AlCl_3$; $Al(NO_3)_3$	Wool	Brightening mordants; Strong affinity for both cellulose and protein fibers; Form an insoluble complexes; Enhanced color depth; Keep original shade; Improve light and wash colorfastness; Less toxicity than others.	(Zarkogianni et al., 2011; Yi and Yoo, 2010; Mansour, 2010; Farizadeh et al., 2010; Erdem Işmal et al., 2013; Deveoglu et al., 2012a,b; Ghaheh et al., 2012; Khan et al., 2012; Shahid et al., 2012; Guesmi et al., 2012a; Yusuf et al., 2012; Deveoglu et al., 2012a,b; Tutak and Benli, 2012; Ghouila et al., 2012; Ebrahimi and Parvinzadeh Gashti, 2016; Sadeghi-Kiakhani, 2015; Nazari et al., 2014; Zhang et al., 2014; Shabbir et al., 2016; Islam and Mohammad, 2018; Shabbir et al., 2018; Shahid-ul-Islam et al., 2018a,b; Lee et al., 2018; Yan et al., 2019; Fröse et al., 2019; Rather et al., 2020; Adeel et al., 2020; Peran et al., 2020; Haji, 2020; Zhang et al., 2020; dos Silva et al., 2020; Jabar et al., 2020)
	Cotton; Flax; Jute; Ramie; Bamboo; Lyocell; Viscose		(Vankar and Shukla, 2011; Zarkogianni et al., 2011; Arroyo-Figueroa et al., 2011; Yi and Yoo, 2010; Farizadeh et al., 2010; Bhatti et al., 2010; Rehman et al., 2013; Batool et al., 2013; Velmurugan et al., 2013; Savvidis et al., 2013; Tutak and Benli, 2012; Chattopadhyay et al., 2015; Davulcu et al., 2014; Zhao et al., 2014; Grifoni et al., 2014; Swami et al., 2014; Chattopadhyay et al., 2014; Dehnavi et al., 2016; Benli and Bahtiyari, 2018a; Ding and Freeman, 2017; Nakpathom et al., 2017; Adeel et al., 2017; Rehman et al., 2018; Lee et al., 2018; Sadeghi-Kiakhani et al., 2018a,b; Azeem et al., 2019; Mir et al., 2019; Faisal and Chafidz, 2019; Kumbhar et al., 2019; Fröse et al., 2019; Jaffer et al., 2019; Giacomini et al., 2020; Inprasit et al., 2020; Manicketh and Francis, 2020; Phan et al., 2020; dos Silva et al., 2020; Lohtander et al., 2020; Sinha et al., 2016; Benli and Bahtiyari, 2018a)
	Silk		(Vankar and Shukla, 2011; Vinod et al., 2011; Yi and Yoo, 2010; Farizadeh et al., 2010; Vinod et al., 2010; Punrattanasin et al., 2013; Deveoglu et al., 2012a,b; Wang et al., 2013; Baliarsingh et al., 2012; Mahmud-Ali et al., 2012; Deveoglu et al., 2012a,b; Torgan et al., 2015; Ajmal et al., 2014; Tayade and Adivarekar, 2016; Dehnavi et al., 2016; Jia et al., 2017; Vankar et al., 2017; Giacomini et al., 2017; Yin et al., 2017; Khan et al., 2018; Lee et al., 2018; Yan et al., 2019; Adeel et al., 2019; Rungruangkitkrai et al., 2020)
	Nylon; Polyester; Acrylic		(Hunger et al., 2005; Guesmi et al., 2012b; Sadeghi-Kiakhani, 2015; Shams Nateri et al., 2016; Ebrahimi and Parvinzadeh Gashti, 2016; Erdem Ismal and Yıldırım, 2020; Erdem Ismal and Yıldırım, 2020; Nakpathom et al., 2019)
Ferrous Compound - $FeSO_4$; $FeCl_3$	Wool	Dulling mordants; Highest achievable K/S values; Form coordination complex – chelating; increase fastness of any colors; Grey to black shades; Prefer to cellulosic fibers.	(Zarkogianni et al., 2011; Yi and Yoo, 2010; Farizadeh et al., 2010; Erdem Işmal et al., 2013; Rehman et al., 2013; Ghaheh et al., 2012; Khan et al., 2012; Yusuf, M. et al., 2012; Mahmud-Ali et al., 2012; Tutak and Benli, 2012; Ghouila H. et al., 2012; Ebrahimi and Parvinzadeh Gashti, 2016; Li et al., 2016; Shabbir et al., 2016; Shabbir et al., 2018; Shahid-ul-Islam et al., 2018a,b; Rather et al., 2019; Yan et al., 2019; Fitz-Binder and Bechtold, 2019; Rather et al., 2020; Adeel et al., 2020; Glogar et al., 2020; dos Silva et al., 2020)
	Silk		(Yi and Yoo, 2010; Punrattanasin et al., 2013; Wang et al., 2013; Baliarsingh et al., 2012; Mahmud-Ali et al., 2012; Tayade and Adivarekar, 2016; Patil and Datar, 2016; Patil and Datar, 2016; Jung, 2016; Yasukawa et al., 2017; Jia et al., 2017; Khan et al., 2018; Rather et al., 2019; Adeel et al., 2019; Rungruangkitkrai et al., 2020)
	Cotton; Flax; Jute; Lyocell		(Batool et al., 2013; Yi and Yoo, 2010; Tutak and Benli, 2012; Chattopadhyay et al., 2015; Davulcu et al., 2014; Zhao et al., 2014; Swami et al., 2014; Khan et al., 2014; Chattopadhyay et al., 2014; Li et al., 2016; Benli and Bahtiyari, 2018a; Yasukawa et al., 2017; Ding and Freeman, 2017; Nakpathom et al., 2017; Adeel et al., 2017; Rehman et al., 2018; Azeem et al., 2019; Sheikh et al., 2019; Mir et al., 2019; Faisal and Chafidz, 2019; Kumbhar et al., 2019; Giacomini et al., 2020; dos Silva et al., 2020; Benli and Bahtiyari, 2018a)
	Nylon; Polyester; Acrylic		(Guesmi et al., 2012b; Erdem Ismal and Yıldırım, 2020; Nakpathom et al., 2019; Erdem Ismal and Yıldırım, 2020)

(*continued on next page*)

Table 4 (*continued*)

Name	Fibers	Key Features	Ref.
Copper Compound - $CuSO_4$; $CuCl_2$	Cotton; Flax; Lyocell	Dulling mordants; Normally much stronger than those of iron; Higher dye uptake than others; Eco and toxicity concern.	(Yi and Yoo, 2010; Kobayashi et al., 2010; Batool et al., 2013; Tutak and Benli, 2012; Davulcu et al., 2014; Swami et al., 2014; Khan et al., 2014; Benli and Bahtiyari, 2018a; Yasukawa et al., 2017; Nakpathom et al., 2017; Adeel et al., 2017; Rehman et al., 2018; Amemiya and Nakanishi, 2018; Azeem et al., 2019; Mir et al., 2019; Kumbhar et al., 2019; Manicketh and Francis, 2020; Benli and Bahtiyari, 2018a; Yadav et al., 2019)
	Wool		(Zarkogianni et al., 2011; Ghoranneviss et al., 2011; Yi and Yoo, 2010; Farizadeh et al., 2010; Erdem Işmal et al., 2013; Ghaheh et al., 2012; Tutak and Benli, 2012; Ghouila, H. et al., 2012; Ebrahimi and Parvinzadeh Gashti, 2016; Amemiya and Nakanishi, 2018; Yan et al., 2019; Rather et al., 2020; Adeel et al., 2020)
	Silk		(Yi and Yoo, 2010; Punrattanasin et al., 2013; Wang et al., 2013; Baliarsingh et al., 2012; Mahmud-Ali et al., 2012; Torgan et al., 2015; Ajmal et al., 2014; Tayade and Adivarekar, 2016; Yasukawa et al., 2017; Jia et al., 2017; Yan et al., 2019)
	Nylon		(Shams Nateri et al., 2016; Erdem Ismal and Yıldırım, 2020; Erdem Ismal and Yıldırım, 2020)
Stannous Compound - $SnCl_2$; $SnSO_4$; $SnCl_4$;	Wool	Brightening mordants brighter than others; Improve colorfastness; May cause stiff hand; Loss tensile strength of fiber.	(Zarkogianni et al., 2011; Ghaheh et al., 2012; Khan et al., 2012; Shahid et al., 2012; Ghouila et al., 2012; Shabbir et al., 2016; Shabbir et al., 2018; Shahid-ul-Islam et al., 2018a,b; Adeel et al., 2020; Barani and Maleki, 2020; Dehnavi et al., 2016)
	Silk		(Punrattanasin et al., 2013; Baliarsingh et al., 2012; Mahmud-Ali et al., 2012; Tayade and Adivarekar, 2016; Torgan et al., 2015);
	Cotton; Flax; Lyocell		(Davulcu et al., 2014; Torgan et al., 2015; Zhao et al., 2014; Swami et al., 2014; Wang et al., 2016; Rehman et al., 2018; Azeem et al., 2019; Kumbhar et al., 2019; Phan et al., 2020)
	Nylon		(Shabbir et al., 2016; Shams Nateri et al., 2016; Erdem Ismal and Yıldırım, 2020; Erdem Ismal and Yıldırım, 2020)
Potassium Compound - $K_2Cr_2O_7$	Wool	Brightening mordants; Health and toxicity concern.	(Ghaheh et al., 2012)
	Cotton; Lyocell		(Swami et al., 2014; Benli and Bahtiyari, 2018a; Rehman et al., 2018)
	Silk		(Mahmud-Ali et al., 2012; Ajmal et al., 2014; Tayade and Adivarekar, 2016)
Other minor Compounds:*	Cotton; Acrylic; Wool; Silk; Nylon.	Various, overall improve color depth and/or colorfastness	(Zarkogianni et al., 2011; Yi and Yoo, 2010; Farizadeh et al., 2010; Wang et al., 2013; Guesmi et al., 2012a,b; Torgan et al., 2015; Parvinzadeh Gashti et al., 2014; Cerempei et al., 2016; Yasukawa et al., 2017; Khan et al., 2018; Erdem Ismal and Yıldırım, 2020; Yan et al., 2019; Faisal and Chafidz, 2019; Adeel et al., 2020; Peran et al., 2020; Zhang et al., 2020; Jabar et al., 2020)

* Zinc [$ZnSO_4$; $ZnCl_2$; $Zn(BF_4)_2$], Calcium [$Ca(OH)_2$; $CaCl_2$, $Ca(CH_3COO)_2$,CaO], Cobalt [$CoSO_4$], Magnesium [$MgSO_4$], Manganese [$MnSO_4$), Lanthanum Oxide [La_2O_3], Rare earth Chlorides [$ReCl_3$,$NdCl_3$, $ZrOCl_2$], Silver nitrate [$AgNO_3$], Ammonium sulfate [$(NH_4)_2SO_4$], Sodium acetate [$C_2H_3NaO_2$], Praseodymium chloride [$PrCl_3$], Kunipia-F Nano clay [$NiSO_4$].

reflecting the current marketplace of natural textile dyeing. Handicraft level, lab/studio level, small business level, and industrial level are operated differently for different market levels and end-uses. Presently, more and more eco-conscious companies and brands have put efforts into promoting plant dyeing at different levels. For example, major brands like Patagonia are experimenting with ways to incorporate it into their product line. Patagonia has a "Clean Color" fashion collection, which uses natural dyestuff from mulberry, Carmine, pomegranate, indigo, etc., (Clean Color Collection, 2019). Spoonflower supplies naturally dyed colors for 900+ fabrics, home décor, and wallpapers (Natural Dyes Designs, 2019). Nudie Jeans uses "Dyer's Woad" to achieve blue colors on denim fabrics. Celebrities like Gisele Bündchen are rocking naturally dyed clothes on the press circuit (Loewe, 2019). Many middle to small clothing and fashion businesses are now starting to embed natural dyestuffs in the design, series, and collections (In, 2019).

The application of natural dyeing to textile fiber is complex and challenging. To promote the natural dyeing application, there are some fundamental characteristics relating to natural dyeing that the industry ought to put their efforts to educate the general public, minimizing potential biases toward natural dyeing products. These characteristics are dyeing uniformity, color yield, and process selection, and they will be discussed separately. Firstly, macromolecules such as sugar and pectin are difficult to eliminate during colorants extraction from many plants' dyes, which might cause leveling issues of dyeing. Fabrics for natural dyeing should be fully pre-treated before dyeing which is a critical but often overlooked step in archiving dyeing uniformity. Fibers absorb natural dyes at normal temperature, and plant dye liquor should be added to the dye bath slowly at room temperature to achieve a more uniform color surface. Secondly, color yield is another consideration when the color performance of natural dyes is evaluated. In textile coloration, CIELAB colorimetric properties of dyed fabrics are normally reported to represent the color on a piece of dyed fabric. In addition, as mentioned in the previous section, the K/S value is used to determine the depth of color of dyed fabric. Color performance is evaluated mainly by colorfastness to sunlight, laundering, and crocking. As a common practice, the color yield of natural dyeing in protein fibers is comparatively higher than in cellulosic fibers. Fabric structure and yarn count have a significant impact on the color yield of natural dyeing, e.g., the more complex and tighter the fabric, the lower the color yield. As such, dyeing parameters like liquid ratio and pH value affect the color yield drastically. Lastly, the dyeing profile, dyeing equipment, and dyeing method are mostly based on the fiber component of textile material. Thereby, dyeing stage plays a vital role in the natural dyeing application. The dyeing stages include stock/top dyeing (loose fiber dyeing), yarn dyeing, piece dyeing, tie-dyeing, form dyeing, and hang dyeing, etc. It is more challenging to dye fabrics with multiple fiber mixes than only one

Table 5. Bio-mordants for natural plant dyeing.

Source of Mordants	Chemical Composition	Fibers	References
Memecylon scutellatum; Gallnut; Chicken gizzard leaves; Algal; Lemon Chestnut; Gallnut; Pomegranate	Tannic Acid ($C_{76}H_{52}O_{46}$)	Cotton/Flax/Hemp/Jute	(Chairat et al., 2011; Mansour and Heffernan, 2011; Hong et al., 2012; Chattopadhyay et al., 2015; Grifoni et al., 2014; Khan et al., 2014; Chattopadhyay et al., 2014; Lee et al., 2017; Adeel et al., 2017; Phan et al., 2020; Sinha et al., 2016; Mansour and Heffernan, 2011; Batool et al., 2013; Azeem et al., 2019; Mir et al., 2019; Jaffer et al., 2019; Manicketh and Francis, 2020)
Acacia nilotica; Henna; Pomegranate; Turmeric; Rosemary; Thuja; Valex; Myrobolan		Wool	(Lee et al., 2017; Hong, 2018; Alebeid et al., 2020a,b; Adeel et al., 2020; Erdem Işmal et al., 2014);
Turmeric; Pomegranate; Lemon		Silk	(Ajmal et al., 2014; Tayade and Adivarekar, 2016; Giacomini et al., 2017; Vinod et al., 2011; Vinod et al., 2010; Adeel et al., 2019; Adeel et al., 2020; Manicketh and Francis, 2020; Liu and Bai, 2012)
Lemon, Gallnut		Polyester	(Nakpathom et al., 2019; Manicketh and Francis, 2020)
Henna, Pomegranate, *P. Fraxinifolia*		Nylon	(Ebrahimi and Parvinzadeh Gashti, 2016)
Citric acid	($C_6H_8O_7$)	Wool	(Deveoglu et al., 2012a,b; Zhang et al., 2020)
		Nylon/Elastane	(Erdem Ismal and Yıldırım, 2020)
		Cotton	(Deveoglu et al., 2012a,b)
		Nylon	(Erdem Ismal and Yıldırım, 2020)
Tartaric acid/Oxalic acid	$C_4H_6O_6$; $H_2C_2O_4$)	Nylon/Elastane	(Erdem Ismal and Yıldırım, 2020)
Cream of tartar	($C_4H_5KO_6$)	Cotton/Viscose/Ramie/ Wool/Bamboo	(Fröse et al., 2019)
Whey protein	bovine serum albumin & lactalbumin	Cotton	(Baseri, 2020)
Sodium alginate	$C_6H_9NaO_7$	Cotton	(Phan et al., 2020)
Skimmed milk powder	Protein, Lactose; Fat; Ash	Rayon	(Park and Park, 2020)
Sodium carbonate	Na_2CO_3	Wool/cotton	(Deveoglu et al., 2012a,b)
Mango bark and Aloe vera	N/A	Leather	(Berhanu and Ratnapandian, 2017)

component. For example, for a mélange fabric made of silk and flax fiber, since no natural dyes have an affinity with both fibers, to achieve a multicolor effect, two-baths and two-step processes should be employed to dye both fibers properly. Besides, some plant dyes cannot be used simultaneously with other dyes due to compatibility issues. In this case, more complicated dyeing techniques are needed.

8. Selection of the U.S. Plant species suitable for modern natural dyeing

The number of colorants found in nature is enormous, but the majority of them are not suitable for textile dyeing. Usually, natural plant for textile dyeing are selected from non-food products and these plant species can be processed for colorants. Therefore, it is important to screen and choose appropriate plant species, of which are suitable for contemporary cultivation practices as well as for sustainable textile dyeing on a business scale. Natural dyeing researchers and practitioners have made progress towards this direction.

Some preliminary studies on plant species were conducted for plant dyes. In 1989, according to an investigation by Hofmann, about a thousand plant species might be used for textile dyeing (Hofmann, 1989). While some of them are known well and had a long history of application, still many species are not commercially important despite of notable color performance. Thereby, application trials were piloted on industrial plants to screen and select species. In Germany, more than a hundred plant species are assessed for their suitability for modern cultivation systems, yields, and the dyeing quality of these species. Among these plants, 19 species were concluded as useful for cultivation and dyestuff production (Bechtold and Mussak, 2009). In addition, madder (*Rubia tinctorum*), weld (*Reseda luteola*), Canadian goldenrod (*Solidago canadensis*), dyer's chamomile (*Anthemis tinctoria*), and dyer's Knotweed (*Polygonum tinctorium*) are considered to be suitable for future dye plant cultivation and processing (Hartl and Vogl, 2003). In the U.S., India, and China, indigo is widely used as a dye plant and commonly employed for jeans in the natural dyeing industry. Field trials in California showed that indigo could be cultivated successfully in wide areas for one or two years

Table 6. Natural textile dyeing levels and features.

Feature/ Level	Handcraft level	Lab/Studio level	Small business level	Industrial Level
How	Simple equipment and consumer chemicals (e.g., pots, gas cookers, home washer and dryer, vinegar, backing soda)	Lab size equipment and some chemicals (e.g., dyeing kier, lab dyeing machinery, water bath, pH meter, acetic acid, mordants)	Small to mid-size specialized equipment and chemicals (e.g., Kettles, Boiling Can, Centrifugal Drier, Heat Pump, ventilation)	Mid to Large Scale industrial Equipment/ Machinery with automatic control/digital communication; bulk industrial chemicals
Where	Home (kitchen or garage)	studio or laboratory	conventional dyeing mill with workshops	Modern dyeing mill
What	Mianly natural protein fibers (e.g., wool and silk, as they are easy to dye)	Limited dyeing possibility. Can dye natural protein and cellulosic fibers, (e.g., silk, wool, cotton, flax). The quantity and color quality could not be guaranteed fully.		Various materials of natural or synthetic fibers/ material (e.g., wool, silk, cotton, linen, hemp, leather, accessories made of horn, nutshells, mother-of-pearl)
Typical Products	Personal usage: T-shirt, scarves, pullovers, socks, etc.	Small batches of a commercial product of woven or knits etc. scarf, kids' wear; bedding, towels, napkins, socks, disposable wipes, carpets, Hair Dye, cosmetic, stationary.		Full spectrum of regular apparel or textiles, and other natural colorants applications

(FiberShed – indigo, 2021). Overall, the suitability of these plants for coloration provision has been investigated and they have a higher potential to meet the requirements of modern dyehouses and the textile industry (Bechtold and Mussak, 2009).

Even there were much progress in the past two decades, it must bear in mind that the annual yields of the majority of plants in these study works are much limited. Then, considerations should be made not only on the color process and color quality of natural plant dyes, but also on sustainability and cost with desirable color quality. While most of the articles provided very vague replies with weak support, the economic and sustainability aspects of the natural dyeing technology should have been evaluated thoroughly. Understandably, the economic and sustainability dimensions of natural dyeing are complex issues. However, there has been a trend to explore the potential of full utilization of plant waste from an agricultural perspective recently. There are increased interests in discovering innovative, close-loop solutions for reducing agricultural byproducts, which would bring negative impacts on our environments otherwise. Based on a report by the Environmental Protection Agency, agriculture/food by-products consist of approximately 22% of total municipal solid waste generated in the United States, and 95% of this type of waste is ultimately landfilled (Environmental Protection Agency, 2018). The idea of finding new sustainable solutions to reduce agricultural waste is on the rise so that existing resources can be repurposed in an innovative approach to an even greater extent. The under-utilized byproducts, e.g., berry, grape, pumpkin, pomegranate, and orange pomace waste, etc., contain natural colorants that might be used as a potential supplementary or substitute to synthetic ones. Research into the agricultural byproducts as natural colorants could provide benefits for both agricultural industries and the fashion industry with a creative sustainable solution.

The U.S. has the largest agricultural economy in the world. The plantation of natural plants and economic crops not only promotes the rapid development of the rural economy but also generates a large amount of agriculture, botanic, and forest wastes, which has a negative impact on the ecological environment and affects the local environment (Rossi et al., 2017). Waste disposal techniques have been developed in recent years, which are usually limited to simple utilization of heat energy and solvent (e.g., water; Rather et al., 2019). Therefore, efforts have been put in maximizing the utilization of biodegradable natural dyes from agriculture products or byproducts and forest wastes with rich colors. Recent technology has been employed to extract and separate natural colorants quickly and uniformly with expected purity. The successful implementation of innovative technology provides a novel approach to the utilization of agricultural wastes (Baseri, 2020; Kumar et al., 2019; Erdem Işmal et al., 2013; Sukemi et al., 2019; Rossi et al., 2017).

To seek potential U.S. plants candidates for natural colorants, we utilized the most current statistical data from the USDA Census of Agriculture in 2017 (USDA-NASS, 2017) and the 2019 US Economic Research Service by USDA (USDA-Cash receipts by states, 2020) as major considerations that are summarized in Table 7.

Byproducts and wastes from agriculture products in this table provide a strong pool for plant dye sources and reflect the potential candidates that could be utilized for sustainable natural textile dyeing. To make a

Table 7. Corps yield and value ranking of top potential plants for textile dyes in the U.S.

Corps and Yield of Top Potential Plants (2017 Census)				Commodity Ranking by Dollar Value (2019)		Plant Parts	Known Functional Property	Potential for Industrial application
Commodity group	Crop	Acres	Yield (cwt)	Receipts ($1K)	Ranking			
Vegetables, Melons, and Potatoes	Onions, dry	163,982	543.8	1,001,986	31	Skin	n/a	Agriculture (or Ag.) waste; less percentage on mass (skin); not all onions have color; low potential.
	Pumpkins	93,563	220.2	180,190	71	Whole, except seeds	Anti-oxide	moderate yield; good color presentation; mainly for food; low potential.
	Ginger root	266	n/a	n/a	>100	Whole	Antibacterial	limited yield; mainly for food/beverage; excellent color presentation; functional performance; moderate potential.
Specified fruits and nuts	Coffee	9,308	3,530	52,742	89	Whole	Deodorizing	High yield; mainly for food/beverage; functional performance; moderate priority.
	Grapes	1,136,155	14,720	5,719,758	14	Skin	n/a	Very high yield; Ag. waste; less percentage on mass (skin); moderate potential.
	Persimmons	4,685	n/a	n/a	>100	Whole, except seeds	n/a	limited yield; mainly for food; low potential.
	Pomegranates	31,472	n/a	n/a	>100	Peel	Antibacterial	Ag. waste; limited yield; excellent color presentation; functional performance; moderate potential.
	Oranges, all	602,830	n/a	1,765,347	23	Peel	UV-Protective	Ag. waste; High yield; good color presentation; functional performance; high potential.
	Tangerines	77,701	n/a	700,977	39			
	Almonds	1,266,160	2,270	6,094,440	11	Shell	Antibacterial	Ag. waste; high yield; excellent color presentation; functional performance; high potential.
	Hazelnuts	70,091	1,720	84,480	81		n/a	
	Walnuts	419,706	3,760	1,286,410	25		n/a	
Berries	Blackberries/ Cranberries	210,623	6,517.6	n/a	>100	Whole fruit	n/a	High yield; mainly for food; excellent color presentation; moderate potential.
	Blueberries	153,258 (dos Silva et al., 2020)	6,100	934,705	33			
Others	Safflower	n/a	n/a	44,776	94	Flowers	n/a	Limited yield; none-food; excellent color presentation & value; moderate potential.

sensible selection from the current pool of natural colorants, the major factors to be addressed are availability and abundance, extraction/ coloration techniques, color performance and specification, production feasibility and efficiency, and potential market/end-use products. Bear these criteria in mind, we can learn from Table 7 that there are several plant species highlighted for the sorting process. They are pomegranates(peel), oranges/tangerines(peel), almonds(shell), pumpkins (peel and meats), coffees(fruit), walnuts(shell), hazelnuts(shell), and safflower(flower). The promising results obtained by using these plant species in textile industries create an opportunity for farmers to produce such crops with current scale but more benefits. The byproducts of these plants could be repurposed or reused as a source of natural plant dyes to minimize waste disposal and environmental issues. These byproducts can simply be obtained from a local farm, its production is green, and residual products are biodegradable and environmentally friendly. An additional consideration for natural dyeing is the eco-friendly characteristics of natural dyeing as conventional farming uses much energy, water, and agrochemicals. For natural plants for dyeing, it is ideal that farming processes comply with organic certifications such as Oeko-Tex, Organic Trade Association, or California Certified Organic Farmers (Dawson, 2012), though the cost will be a challenge indeed. Besides, organic fibers and dyes have become an attraction for many brands and eco-consciousness consumers. The combination of organic fiber with natural plant dyestuffs will be a sparkling drive for the choice of green and sustainable textiles. In addition, the progress of natural plant dyeing is based on efforts from many parties involved - more research on technology aspects such as extraction, purification, dyeing, and mordanting methods; more studies on plants cultivation and product processing with added value; more support from the end-use market for environment-conscious textile brands and retail company.

9. Conclusion and perspectives

There is no denying that there is much argument related to natural dyes and their applications, while no natural dye is absolutely sustainable. Color performance and quality, economy, eco, and health considerations are several fundamental criteria for a suitable natural plant dye. However, colorants from natural sources could be utilized more broadly in the textile industries as a potential alternative to some synthetic dyes, even starting with a small scale for specific categories. Almost all naturally derived colorants related to their practice for social and aesthetic application provides unique knowledge about society's ever-changing needs. Based on this appeal, creativity can be designed from naturally dyed textiles and innovations of industrial applications can be implemented with precious natural resources. Nowadays, as so many textiles need to be dyed and printed, natural-derived colorants would offer a simple and safe alternative to synthetic ones in some types of textile products, where natural dyeing offers a decent quality product with added value and higher eco-compatibility in the marketplace. Thereby, from this review article, specific conclusions are summarized below:

- Cotton and wool are two representative fibers for natural dyeing where more dyeing potential has been exploited. Recent efforts have been made towards regenerated and synthetic fibers.
- Current natural dyes family can be used to easily achieve dark colors with Munsell $V < 2$ and strong colors with Munsell $C > 10$ on generic textiles, within commercially acceptable colorfastness tolerance. Finding more colors to fill the gap in the color hue circle in the blue region would be a great challenge for textile researchers and practitioners.
- There are great potential for both antibacterial and UV protective properties from many current natural colorants, while other functions like antioxidant and deodorizing are some new areas of interest.
- Modern technology like plasma and gamma radiation may bring more promising possibilities for natural dyeing applications.
- Though new mordants experimented for better coloration performance of textiles, traditional alum, ferric, and copper mordants are still the major players in this field.
- Bio-mordanting provides a truly sustainable natural dyeing solution since it eliminates the use of metallic mordants relating to potential health concerns. It will be a main stream of research for natural textile dyeing in time to come.
- A structure of technological levels of natural textile dyeing application was introduced based on the current natural textile dyeing market.
- For the U.S. plants/crops, agriculture byproducts from pomegranates (peel), oranges/tangerines (peel), almonds (shell), pumpkins (peel and meats), coffees (fruit), walnuts (shell), hazelnuts (shell), and safflower (flower) are highlighted for natural textile coloration.

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Author contribution statement

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Additional information

No additional information is available for this paper.

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