

Review Article

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Natural Dyes: Extraction and Applications

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ABSTRACT

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Natural dyes are obtained from natural sources such as vegetable, animal or mineral matters. These dyes are generally used in coloration of textiles, food, cosmetics and drugs. This article attempts to reviews the classification of natural dyes according to the method of application available natural dye sources, mordant and its classification, different mordanting methods, extraction methods, dyeing methods and advantages and disadvantages of natural dyes have been discussed.

Introduction

Presently there is a great demand for the use of natural colours throughout the world due to non-biodegradable and carcinogenic nature associated with synthetic dyes (Chungkrang and Bhuyan, 2020). They are not produce any undesired by-products and at the same time they help in regenerating the environment, therefore natural dyes are the safe dyes (Akkewar, 1999; Darwekar *et al.*, 1999). Government of Germany was the first to take initiative to put ban on azo-dyes for manufacturing, dyeing and importing textiles and other consumer goods dyed with these dyes from January 1, 1995 by the act of

German Legislation (Consumer Goods Ordinance). Netherlands followed a ban with effect from August 1, 1996 on similar lines. European Union is likely to impose ban on these toxic dyes shortly. India has also banned the use of specific azo-dyes and under notification “sufficient legal teeth” had been given for taking panel action against those who use these dyes (Kapoor and Pushpangadan, 2001; Singh *et al.*, 2006).

Natural dyes are important since they are better than synthetic dyes in many ways. Undeniably, the natural dyes are healthier products, purely because they do not comprise chemicals damaging to fitness (Baliarsingh *et*

al., 2013). These dyes are collected from nature and no need to apply manufacturing process to prepare them. These dyes are easily decomposed in nature after using and they do not pollute the environment while destroying them after end use (Alam *et al.*, (2020). The natural dyes are used mainly in coloration of textiles, food, drugs and cosmetics. Small quantities of dyes are also used in coloration of paper, leather, shoe polish, wood, cane, candles and such other products requiring coloration (Gulrajani, 2001).

Classification of natural dyes according to the method of application

The natural dyes can be classified into three groups according to the method of application (Gulrajani and Gupta, 1992; Dedhia, 1998; Samanta and Konar, 2012; Jihad, 2014).

Direct or substantive dyes

A dye, soluble in water can be taken up directly by the material to be dyed without any pretreatment, is known as direct or substantive dye. These dyes can be applied directly in a hot aqueous dye solution without the use of fixing agent or mordant. These dyes are mainly used for cellulosic fibers, protein fibers and polyamides. Examples of direct natural dyes are turmeric, annatto, berberry, safflower etc. The direct dyes are further classified into acid dyes and basic dyes.

Acid dyes: These dyes are used for wool and silk and are applied in the acidic medium. They have the structure of either sulphonic or carboxylic group.

Basic dyes: These dyes on ionization give coloured action, which form electrovalent bonds with the carboxylic acid groups of wool and silk. These dyes are applied in neutral to mildly acidic pH.

Mordant dyes: Mordant dyes are those dyes, which have affinity for the mordanted fibres. Most of the natural dyes yield different shades of colours with different mordants. The mordant dyes are classified as monogenetic and polygenetic. Monogenetic dye produce only one colour according to the mordant present on the fiber or applied along with the dye materials. Polygenetic dye produces different colours according to the mordant employed.

Vat dyes: The vat dyes derive the name from the fermented wooden called 'vat' which was used at one time for reducing the dye and converting it into soluble form. The process of the solubilization of the dye is called as 'vatting'. The soluble form of the dye is called as leuco dye. The leuco form of the dye is not only soluble in water but also has an affinity for the natural fibers. The dye colour is pale yellow but the true colour is produced only on treatment with hot soap solution. They include the most ancient of dyes such as indigo and tyrian purple etc.

Disperse dyes: Natural dye has not been classified as disperse dye, however by looking at the structure and solubility characteristics of the natural dyes it is felt that some of the natural dyes can be classified as disperse dyes. Disperse dye has a low relative molecular mass, low solubility to the dye molecule. These dyes have hydroxyl and amino groups which imparts some solubility to the dye molecule. One such dye could be Lawsone. Many other flavones and anthraquinone dyes can qualify to be classified as disperse dyes.

Mordant and its classification: Nearly all natural dyes require mordants to give substantivity. The term mordant has been derived from the Latin word *mordeo*, which means "to bite" or to "take hold of (Gohl and Vilensky, 1983). The majority of natural dyes

need a chemical in the form of metal salts to create an affinity between the fiber and the pigment. These chemicals are known as mordants (Bisht and Goel, 1999; Singh and Srivastava, 2015; Vankar *et al.*, 2009).

Classification

Three types of mordants are used in dyeing textile materials, metallic mordants, tannin and tannic acid and oil mordants (Agarwal and Patel, 2000).

Metal Salts or Metallic Mordants: Several different metal salts can be used for mordanting such as Alum (Potassium aluminum sulphate), Copper (Copper sulphate), Chrome (Potassium dichromate), Iron (Ferrous sulphate) and Tin (Stannous chloride, Stannic chloride)

Tannins and Tannic acid: Tannin and Tannic acid is present in the barks, leaves, fruits and galls of many plants. The word 'tannin' comes from the early French word Tan, which means bark of an oak. The term tannin was introduced by Sequin in the year 1976 to indicate various plants extracts which have capacity to convert hides and skins into leather. Tannins are classified into two categories:

(i) Condensed or flavonoid (Catechol type) tannins based on polymeric flavonoid, obtained from acacia catechu. Tannins such as tannic acid, harda etc are considered as natural mordants.

(ii) Hydrolysable (Pyrogallol type) tannins based on esters of gallic acid, obtained from myrobalan fruit, oak bark, gallnuts, pomegranate rind etc.

In the same plant different parts like bark, nut, leaf and wood may contain different types of tannins (Dwivedi, 1993).

Oil or Oil Mordants: Oil –mordants are mainly used in the dyeing of Turkey red colour from madder. The main function of the oil-mordant is to form a complex with alum used as the main mordant. Since alum is soluble in water and does not have affinity for cotton, it is easily washed out from the treated fabric. The naturally occurring oils contain fatty acids such as palmitic, stearic, oleic, ricinolic etc. and their glycerides. The –COOH groups of fatty acids react with metal salts and get converted into –COOM, where M denotes the metal, for instance in the case of alum it would be Al. subsequently, it was found that the treatment of oils with concentrated sulphuric acid produces sulphonated oils which possess better metal binding capacity than the natural oils due to the introduction of sulphonic acid group, –SO₃H. The sulphonic acid can react with metal salts to produce –SO₃M. The bound metal can then form a complex with the mordant dye such as madder to give Turkey red colour of superior fastness and hue.

Mordanting Methods

Mordanting is classified on the basis of application time of mordants (Gohl and Villensky, 1983). These are pre-mordanting (before dyeing), simultaneous mordanting and post mordanting (after dyeing).

Pre-mordanting: In this method, the textile material is first immersed into the mordant solution for 30 to 60 minutes at the room temperature or higher temperature with a liquor ratio of 1:5 to 1:20. Then dyed the textile material treated with mordant. After dyeing, the dyed material is wash.

Simultaneous mordanting: For this method, the textile material, mordant and dye are immersed in a dyebath simultaneously. For the optimization of dyeing conditions, dyeing process variables can be studied for the

specific fibre-mordant-natural dye system in order to improve the colour yield of textiles. After dyeing, the textile material is wash properly.

Post-mordanting: In this method, the dyeing process is carried out on the textile material without mordant. After dyeing, the dyed fabric is treated in another bath containing the mordant solution. Treatment condition may vary depending on the type of fibre, dye and mordant system being used. After dyeing, wash the textile material properly.

Extraction methods of natural dyes

Extraction of colour materials from natural resources is a complicated process. They need specific technique to extract dye from their original sources. The different techniques for extraction of natural colourants are as follows:

Aqueous extraction: In this method, if the raw material is in dry form, broken into small pieces or powdered by grinding and soaked in water to loosen the cell structure. If the raw material is in wet form then it is chopped into fine pieces or grinded to fine paste. Then boiled and is filtered to remove the non-colouring matters from the dye liquor. Both can be used easily to impart colour in different things.

There are some disadvantages associated with aqueous extraction method like time consuming for extracting colour, high temperature requirement, required large volume of water, heat sensitive colouring substances gets reduced at high temperature low dye yield, only water soluble colouring components can be extracted etc.

Acidic or alkaline extraction: Extraction of dye is done under acidic or alkaline medium. For this dilute acidic or alkaline can also be

used which helps in hydrolysis of glycosides resulting in better extraction. Acidic method is used in extraction of tesu natural dye from tesu flower. Alkaline medium is used for those colour extraction which contain phenolic groups. Extraction of colour from lac insect, annatto seeds, safflower petals are extracted by this method.

Ultrasonic microwave extraction: In this extraction method, raw material is treated in aqueous solution using microwave or ultrasonic rays, so that the high temperature, less time and faster rate of extraction can be done. Colour from butterfly pea and annatto seeds are extracted by using this method.

By fermentation: In this extraction method, micro-organisms present raw materials are used to carry out the fermentation and thus help the extracting method. Colour from turmeric, indigo, annatto seeds extracted by using this method. Time consuming, bad smell due to microbial action, immediate extraction of colour after harvesting are some disadvantages of this method

Enzymatic Extraction: Commercially available enzymes including cellulase, amylase, and pectinase have been used to loosen the binding material present in natural materials like bark, roots or any hard part of plant.

Solvent extraction: Organic solvents like acetone, petroleum, ether, chloroform and ethanol are used in the extraction of natural dyes. Solvent extraction method is more efficient than aqueous method. The yield of dye is good quality, required low temperature and the quantity of water requirement is also less.

Supercritical Fluid Extraction: In this extraction method, a fluid having physical properties between gas and fluid is used. It

has low surface tension, low viscosity and good diffusivity properties which results better interaction with the substrate.

Dyeing methods

Conventional Dyeing System

Textile dyeing started in past carried out conventionally. Textile materials were directly processed with the dye bath at high temperatures. Coloration method for textiles using chemically formed gels with considerable freedom for making color designs and precise pattern prints, and can be used with conventional dyeing and printing equipment was developed. With the time, dyeing also matured with the development of optimization of dyeing parameters, and in recent times advanced technologies evolved like plasma treatment and enzymatic processing etc. (Bohrn and Moggio, 1980).

Advanced Dyeing Systems

Advanced technologies or methods are trending in dyeing in recent times owing to their improved results over the conventional dyeing. Plasma treatment and ultrasonic dyeing methods are technically advanced and sustainability compatible methods used in technologically evolved textile industry. Improved results in ultrasound-assisted dyeing are generally attributed to cavitations phenomena and other mechanical effects are produced such as dispersion (breaking up of aggregates with high relative molecular mass), degassing (expulsion of dissolved or entrapped air from fiber capillaries), diffusion (accelerating the rate of diffusion of dye inside the fiber) and intense agitation of the liquid.

The acceleration in dyeing rates observed by many workers might be the cumulative effects of all these factors (Vajnhandl and Marechal, 2005).

Advantages and disadvantages of natural dyes

Advantages of natural dyes

Usually biodegradable and renewable also
Non-toxic, non-allergic and non-carcinogenic as these are obtained from animals or vegetable matters without any chemical processing
Easy to extract and purify
No disposal problems with natural dye waste
Colours produced by natural dyes are usually soft, lustrous and soothing to the eyes

Disadvantages of natural dyes

Non-standardized colour recipes and methods
Requires skilled workmanship and therefore natural dyes are costly
Applicable to natural fibres only (cotton, wool, silk and linen)
Inadequate fastness properties
Some of the mordants are harmful

Application of natural dyes

Natural dyes are generally applied in industries such as textile, food, leather (Sivakumar *et al.*, 2011), cosmetics (Yusuf *et al.*, 2017), pharmaceutical (Chigurupati *et al.*, 2002) etc.

According to the literature, natural dyes such as Walnut shells (Mirjalili and Karimi, 2013), Onion shells (MIAH *et al.*, 2016; Nurunnesa *et al.*, 2018), *Cassia singueana* (Teklemedhin *et al.*, 2018) *Curcuma longa L.* (Ibrahim *et al.*, 2010), *Senegalia catechu* (Ansari *et al.*, 2018), *Butea monosperma* (Daberao *et al.*, 2016), *Hibiscus Rosa Sinensis* (Bose and Nag, 2012), *Ziziphus jujuba Mill.* (Chungkrang *et al.*, 2018), Orange & Lemon peels (Kumar and Dhinakaran, 2017), *Quercus infectoria Oliv.* (Shahid *et al.*, 2012), *Saraca asoca* and *Albizia lebbbeck* (Baliarsingh *et al.*, 2012) and

so forth produce natural pigments are used in textile dyeing industries.

In addition to their biodegradability and compatibility with the environment, natural dyes have been recently discovered to exhibit other functional properties such as antimicrobial activity (Khan *et al.*, 2012; Shahid *et al.*, 2013), anti-fungal, anti-viral activity (Gupta *et al.*, 2004), insect repellent (Kato *et al.*, 2004; Ali *et al.*, 2013), UV protection (Sun and Tang, 2011; Hou *et al.*, 2013) and deodorizing agents (Lee *et al.*, 2009), dye-sensitized solar cells (Zhou *et al.*, 2011; Narayan, 2012).

Varieties of natural pigments like safflower (Cho *et al.*, 2000), turmeric, quinines (Yusuf *et al.*, 2017), anthocyanins (Wrolstad, 2004), betalains (Azeredo, 2009), chlorophylls (Humphrey, 2004), carotenoids (Mortensen, 2006), tannins (Khanbabaee, 2001) etc. are used in coloration of foods. They have different groups of chemical compounds which may be used directly as colorants or in chemically modified form to produce different hues ranging from green through yellow, orange, red, blue, and violet, depending on the source of colorant or increased stability.

The cosmetic industry now employs many natural dyes due to the fact they will cause fewer side affect than the employment of synthetic dyestuffs but they can also provide extra properties such as UV protection, skin moisturizing and anti-aging (Chengaiyah *et al.*, 2010). Natural pigments obtained from natural resources with active functional components have shown high biological activities and exhibited different properties depending on each kind of pigments which make them excellent colouring materials for natural cosmetics and functional health foods (Yusuf *et al.*, 2017).

In conclusion due to the non-biodegradable and carcinogenic nature associated with the synthetic dyes, people started using natural colours. Textile industry is the main field for application of natural dyes but it can also be used in other fields such as foods, cosmetics, drugs, paper printing etc. There are many research works that has been carried out by many researchers/scientists belonging to industry, small and medium entrepreneurs, social organizations (NGO's), Government organizations, researchers in various educational institutes and research organizations etc. on the revival of natural dyes, but more researches are required to standardize the methods for its application.

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