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NATURAL COLOURANTS: A REVIEW

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ABSTRACT

Natural materials like plants, animals, minerals, insects, bacteria, and fungi can be used to make natural dyes, but the majority of dyes are derived from plant components like leaves, bark, flowers, fruits, and roots. Natural dyes have some unique qualities, such as calming colors, biodegradability, safety, non-hazardousness, antimicrobial resilience, biodegradability, etc. Plant parts and occasionally their by-products are needed as raw materials in the extraction procedure for natural dyes (Iqbal & Ansari, 2021). Many different things have been colored with natural dyes, including leather, fur, silk, cotton, wool, and other natural materials (Cristea & Vilarem, 2006).

Keywords: Natural dye, Sources, History, Application.

1.0 INTRODUCTION

" Pigments are essential to existence on earth."(Britton,1995).
Natural colorants and dyestuffs are a significant class of non-wood forest products used in the manufacture of confections, other food items, textiles, cosmetics, medicines, leather, paper, paint, ink, and other goods(Křížová, 2015). The textile business now faces environmental protection difficulties because it uses many chemicals for dyeing and printing. These substances are dangerous for both humans and the ecosystem. There are a number of problems with synthetic colors. Some synthetic dyes that are even mutagenic and carcinogenic have been outlawed. Interest in natural dyes has increased in this age of environmentally conscious consumers, largely as a result of studies showing that these dyes have benefits for both human health and the environment (Singh & Srivastava, 2017).
Natural dyes can be found readily and are safe for the environment. Natural dyes are gaining popularity because they are thought to produce dyed fabrics and apparel that are friendly to the earth. The ecological and cultural legacy of the world still heavily depends on natural dyes and colorants. Since many industrialized countries are promoting natural (green) products and sustainable lifestyles, there seems to be a shift in societal attitudes toward natural dyes as interest in sustainability grows (Deka et al., 2014).

2.0 HISTORY

Humans are drawn to the stunning colors found throughout nature. Natural dyes have been used for coloring. food, cave walls, textiles, leather, and everyday items since the dawn of civilization. There are numerous sources of plants, animals, insects, and minerals that can be used to produce dyes and pigments. As long a human civilization, coloring has been a practice. The existence of dyed textiles during archaeological excavations at various locations around the globe is proof that dyeing was a common practice in prehistoric civilizations. The use of dyestuffs is first documented in writing in China in the year 2,600 BC. Nearly 3,000 years ago, cochineal was used in Mexico and Peru as a cloth and paint dye (Křížová, 2015).
In Europe, cloth artifacts from the Prehistoric period have alizarin, purpurin, and indigo in them. These were discovered in the Late Bronze Age on textiles found in Chinese Yang Hai. (Kramell et al., 2014).
They learned how to dissolve gold and silver in deserts from the advanced knowledge of dyers who used these same minerals in textile dyeing. The majority of our knowledge of pre-Hispanic dye methods comes from ethnohistorical commentary, observation of contemporary traditional dyeing, and chemical analysis (SALTZMAN, 1978).
To achieve the broad variety of colours seen in pre-Hispanic fabric samples, it is certain that early dyers used chemicals such as mordants.

Most Dyes do not react immediately with fibre and must be combined with mordants (positively charged metal ions) to form a bond between the fibre and the dyestuff. The Andeans mine natural deposits of alum (potassium aluminium sulphate) and iron (in the form of ferrous sulphate); words for these minerals can be found in the local languages. The Quechua language people call alum colpa or alcaparosa. Because deposits of these two minerals are frequently discovered together, iron sulphate is also known by these terms. Alum is known as millu in Aymara. Mio and patchu are terms for brown or black muds; stagnant mud, typically rich in iron, is used to darken or even blacken textile dyes. The dyed cloth is buried in mud for a period of time to enable the reaction to occur (de Mayolo, 1989).

2.1 Pigment of ancient Egypt

The Egyptians were well-versed in a variety of mineral colours (i.e., inorganic pigments) derived from natural ores (Skelton, 1999). Many of these pigments were still used recently, but their use is decreasing due to toxicity concerns.

Orpiment (CI Pigment Yellow 39:1): Although the mineral is not found in Egypt, the pigment (probably from Persia) was used in writing inks as early as the Old Kingdom. (2900–2475 BC). The tiny bag containing the pigment was discovered in Tutankhamun's tomb (1325 BC), evidently as a cosmetic. It is lemon yellow in color and has a mica-like sparkle, making it perfect for imitating gold (Fitzhugh, 1997).

2.2 Natural dyes and colorants in the world today: a crossroads between vanishing ancient knowledge and new applications:

The discovery and use of natural dyes and pigments added to the preservation of the age-old bond between humanity and nature, which could now be revived and enhanced. Natural dyes and colorants are an important part of the world's ecological and cultural heritage; their selection and use to produce colors is shared by all civilizations1. Archaeological findings over the last century have largely contributed to demonstrating that, since the dawn of humanity, the search for sources of dyes and pigments has coexisted with the selection of food and medicinal plants and animals. Natural colorants have historically played an important role in economic and cultural exchanges between countries (D Cardon, 2010).

The 19th century saw a significant turning point in this lengthy history: as organic chemistry advanced in Europe, developed nations began to increasingly use synthetic dyes and pigments made from fossil resources like coal tar and oil. They started a societal revolution and were inexpensive and simple to use. People worldwide have come to take colors for granted due to the world have come to take colors for granted as a result of the mass synthetic coloring of common plastics, textiles, paints, cosmetics, and food (Cardon, n.d.).

3.0 CLASSIFICATION OF NATURAL DYES:

Various categories have been used to categorize natural pigments. The primary factors used to categorize natural dyes are the places where they are produced, how they are applied to fabrics, and their chemical composition.

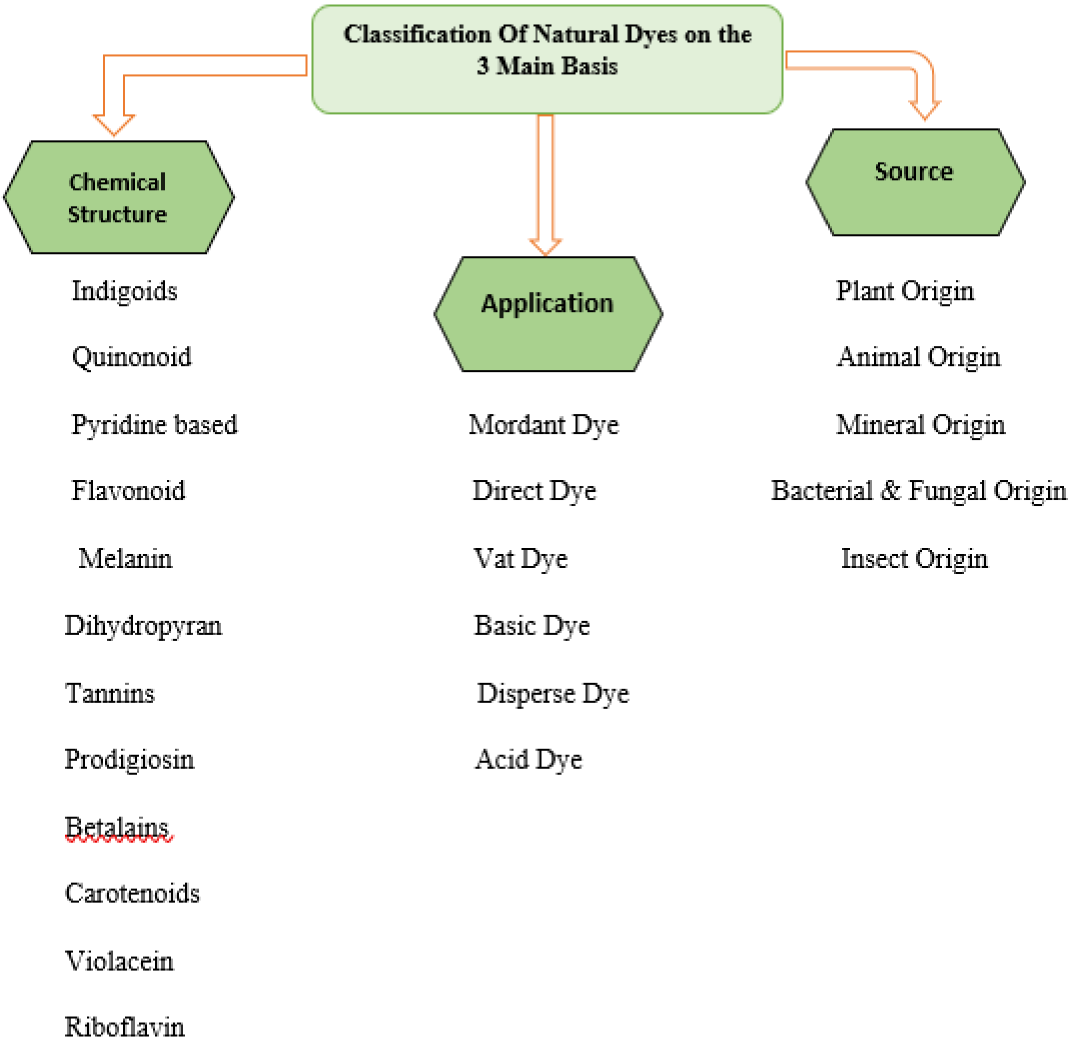

Figure:1 Schematic representation for classification of Natural Dyes.

3.1 Natural colourants classified on the basis of Main Sources

Different sources can be used to get the natural dye. Natural dyes have been used for centuries and can be made from a variety of plant components, such as roots, bark, leaves, flowers, and fruit. (Saravanan et al.,2013). These colourants can be categorised as :1) Plant-based natural colors, such as those made from berries, flowers, bark, leaves, and seeds (e.g., catechu, Indigofera, myrobalan, pomegranate). 2) Cochineal and lac, natural insect-based colours. 3) Animal-derived natural dyes received from molluscs, cuttlefish, snails, and shellfish. 4) Mineral-based natural dyes are received from clay, ochre, and malachite (Singh & Srivastava, 2015).

Based on their source, natural colours can be divided into five groups: plant, animal, mineral, microbial & fungal and insect (R Jihad, 2014) (Yusuf et al., 2017).

3.1.1 Plant Origin

Colorful pigments and dyes can be found in various plant portions like roots, nuts, and flowers. Henna (orange-red) is made from henna plant fronds, Catechu (brown) - from resin (sticky material from acacia tree vegetation, Fustic (yellow) - is made from the fustic tree's bark derived from the leaves and stalks of the indigo plant, indigo (blue) derived from the

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centre (core) of the log wood tree, The two most prevalent ones are saffron (yellow) and turmeric (violet), both of which come from the stigmas of common crocuses (Korankye,2010). One source of natural colourant that is categorised under plant sources is native plant life. They are readily accessible throughout the nation and can be regarded as free dyes because they are grown for other purposes. The main source of natural colorants are plants, and nearly all of their components, including stems, leaves, fruits, seeds, and pills, are used to obtain natural colour as well as their antimicrobial, antifungal, bug repellent, deodorant, disinfectant, and other therapeutic properties. Up to 500 plant types were found to be potential sources of dyes. These dyes are made from different tree parts. (Flower, bark, seeds, leaves, and roots). Not only are these plant-based sources replaceable, but they are also biodegradable. They also have medicinal and good health advantages (R Jihad, 2014).

3.1.2 Animal Origin

Tyrian purple was primarily produced by red-mouthed rock shells, and the research attributes their extinction to the ocean's warming surface waters. The most regal and revered of all ancient dyes, those made from Levantine marine snails of the family Muricide, are the Phoenician purples and biblical blues. These molluscs might have been used to make the regal purple pigment (Koren, 2005).

3.1.3 Mineral origin

The term "natural dye," which encompasses all dyes derived from natural resources, including vegetable dyes as well as minerals, is more suitable because minerals are used to fix or enhance the fastness of vegetable dye. Additionally, some elements are used to provide a colouring substance. Serum, bovine dung, cow urine, and egg albumin are a few examples (R Jihad, 2014). This Mineral Pigments created from coloured clays and earth oxide, chrome green from a chromium and oxygen compound, chrome red from a chromium and lead compound, chrome yellow from a chromic acid and lead compound, and Prussian blue from an iron and cyanide complex (Rosenberg, 2008).

These natural colours include many pigments derived from inorganic metal salts and metal oxides. The following are the most significant natural pigments: By their colors, natural colourants of mineral sources can be further categorized.1) Red pigments made from minerals include Realgar, Red Ochre, Red Lead, and Cinnabar.2) Due to the wide spectrum of yellow colours they produce, Yellow Ochre (Ram Raj), Raw Sienna, Orpiment, and Litharge (Massicot) are all considered yellow pigments. A variety of hydrated forms of iron oxide, especially the mineral limonite, are present in the yellow ochre, giving it its yellow colour. (3) Green colours include Terre-Verte (Green Earth), Malachite, and Vedgiris. Terre-verte has been the most popular among them since ancient times. (4) Azurite and ultramarine blue are both blue dyes. Lapis lazuli, a mineral and semi-precious stone, contains the intense blue pigment known as ultramarine blue (Lajward in Hindi). India's miniature artworks have made use of it. During the fourteenth and fifteenth centuries, lapis lazuli was brought from Afghanistan to India. (5) White Color include White lead, white zinc, and chalk (white lime). One of the varieties of calcium carbonate is chalk. It is frequently utilized in artworks. Chalk is a pigment that has been used since very early times and is found in limestone formations. Conch shell white is favoured by painters in India and is thought to have unique qualities. (6) Among the list of black colours are Charcoal Black, Lamp Black, Ivory Black, Bone Black, Graphite, Black Chalk, and Terre-noire (Black Earth). As a black pigment, finely powdered charcoal has been used frequently (Yusuf et al., 2017).

3.1.4 Insect origin:

The main sources of natural dyes are insect secretions and dried insect body parts. Shellfish, for example, supply the colouring agent. Lac and cochineal are two examples of natural dyes made from insect carmine that produce colours that are identical. It is harvested during the red rain season by laying fabric on the ground beneath infested trees (Verma & Gupta, n.d.).

3.1.5 Bacterial & Fungal origin:

There are many different genera that make pigments, including Streptomyces, Nocardia, Micromonospora, Thermomonospora, Actinoplanes, Microbispora, Streptosporangium, Actinomadura, Rhodococcus, and Kitasatospora The highest pigment production was recorded for the genus Streptomyces (Conn & Conn, 1941). Many species of this genus, like Streptomyces griseus, Streptomyces griseoviridis, Streptomyces coelicolor (Darshan & Manonmani, 2015), Streptomyces cyaneus (Petinate et al., 1999), Streptomyces vietnamensis (Zhu et al.,2007), Streptomyces peucetius (Arcamone, 1998), Streptomyces echinoruber

(Gupta et al., 2011), and Streptomyces shaanxiensis (Lin et al.,2012) have been published to generate pigments.

A wide variety of pigments, including carotenoids, melanin, violacein, prodigiosin, pyocyanin, actinorhodin, and zeaxanthin, are also produced by bacteria, much like fungi (Ahmad et al., 2012; Venil et al., 2013).

3.2 Natural colourants classified on the basis of Applications:

3.2.1 Vat Dyes: Indigo is a water-insoluble pigment that is first dissolved in water before being applied. Natural indigo is solubilized with the aid of sodium hydroxide and sodium hydrosulphite. The cellulosic fibre receives it after solubilization, and after dyeing, the colour is developed by oxidation with hydrogen peroxide. The dye indigo serves as an example of the Indigoid family of Vat dyes.

3.2.2 Direct Dyes: The direct dyeing technique can be used with natural dyes that are water-soluble, have long, planar molecules, and contain conjugated (single- and double-bond) bonds. The dye compounds may include sulphonic, amino, and hydroxyl groups. Direct colouring can be used to apply annatto, Harda, pomegranate, and turmeric. It is possible to improve dye depletion by using table salt. Keep the colouring temperature at 100°C.

3.2.3 Acid Dyes: The dyeing of wool and silk with saffron is done by the acid dyeing method. The presence of common salt in a dye bath produces a levelling effect. The dye molecule has sulphonic or carboxylic groups in its structure, which produce an affinity for wool and silk fibers. The dyeing is done at an acidic pH of 4.5 to 5.5. After dyeing, the fastness is enhanced with tannic acid.

3.2.4 Basic Dyes: The dye molecules dissolve in the water at an acidic pH and then create coloured cations. The -NH2 groups in the dye molecules combine with the -COOH groups in wool and silk. By adding acetic acid, the dye bath's pH is maintained between 4 and 5 (Samanta et al., 2020).

3.2.5 Mordant Dyes: By adding a mordant, a substance that strengthens the bond between dye and fiber, a dye or colorant can be bound to a substance for which it would otherwise have little or no affinity. All dyes that are capable of forming a complex with the metal mordant are now included in the traditional definition of "mordant dyes", which has been expanded. Most of these dyes produce a variety of hues and tones when combined with various mordants (Yusuf et al., 2017).

3.2.6 Disperse Dyes: Disperse dyes, which color polyester and acetate fibres, are water-insoluble dyes. In contrast to the history of natural dyeing, the principle of dispersed dyeing is relatively new. However, it is believed that some of the natural dyes, including lawsone, juglone, lapachol, and shikonin, can be categorized as dispersed dyes due to their molecular similarities and solubility traits (Yusuf et al., 2017).

3.3 Mordanting Method:

In conjunction with natural colorants, so-called mordants are used to increase the substantivity of those colours towards fabrics, some metal salts, or other chemicals or compounds. The application times of pre-, meta-, and post-mordanting mordants are used to categorize mordanting.

3.3.1 pre-mordanting:

Pre-mordanting is the term for treating textile materials with mordants before dyeing, which gives the textile material exclusive, ample time, and sites to bond to the mordants.

3.3.2 Meta-mordanting/Simultaneous Mordanting:

By complexing with one another, this type of manufacturing significantly wastes resources, including dye and mordant. Three different kinds of complexation—between textiles and mordants, textiles and dyes, and dyes and mordants—occur, overloading the ecosystem with dye effluent and endangering sustainability concerns.

3.3.3 post-Mordanting:

This technique involves first applying dye or colourant to the bare textile material, followed by the process of mordanting. This method is primarily used to increase the shade range of textile materials by complexing dye molecules on the surface of the mordant. The colour fastness may not be fastened using this technique (Yusuf et al., 2017).

4.0 Extraction of Dye from Plant Material:

Drying or Grinding

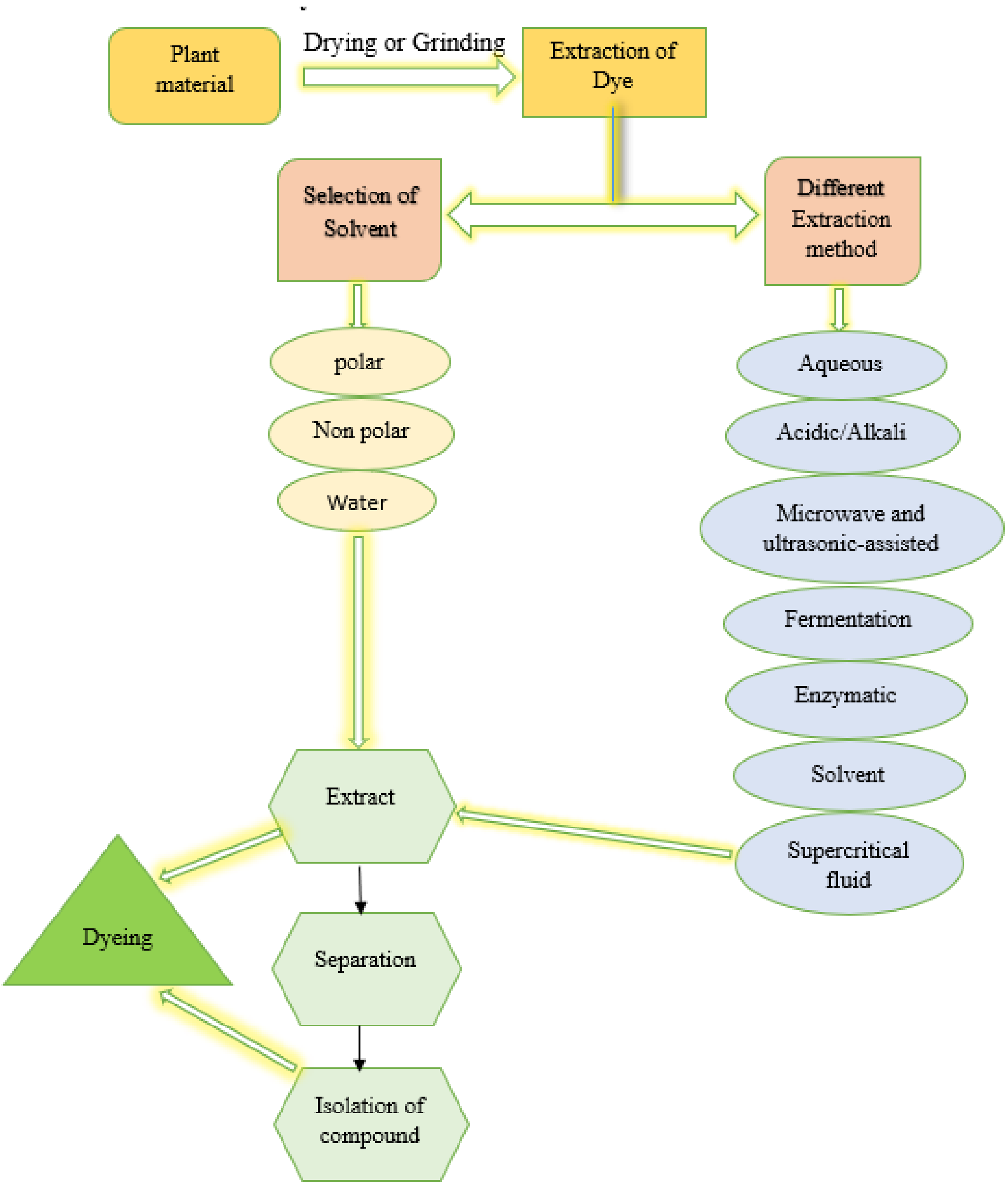

Figure:2 Schematic representation for extraction of natural colorants from plant material.

Figure 2 shows that, the first stage in the extraction process is to prepare the plant material so that it is ready to be extracted. This preparation includes gathering the plant material, drying it, and grinding it to create a homogenous mixture and increase the surface area for the solvent's contact time. The next and most crucial stage is choosing the appropriate solvent based on the makeup of the compounds that need to be isolated or extracted. Polar solvents like methanol, ethanol, or ethyl acetate can be used to extract hydrophilic compounds, and dichloromethane or a 1:1 combination of dichloromethane and methanol can be used to extract lipophilic compounds. Depending on the polarity and thermal stability of the target molecule, various techniques, such as sonification, heating under reflux, Soxhlet extraction, and others, are frequently used. Due to their advantages in terms of yield and ease of extract collection, some contemporary methods are also used for extraction, including solid-phase micro-extraction, supercritical-fluid extraction, pressurized-liquid extraction, microwave-assisted extraction, solid-phase extraction, and surfactant-mediated techniques. The results of extraction are typically mixtures of compounds that need to be further separated using methods like adsorption chromatography, thin-layer chromatography (TLC), and high-performance liquid chromatography. (HPLC). Then, substances should be identified using spectroscopic methods like ultraviolet (UV), Fourier-transform infrared (FTIR), etc (S.Sasidharan et al.,2011)(Pilz & Bart, 2011; Saxena & Raja, 2014).

5.0 APPLICATION:

In dye-sensitized solar cells, which use cell sensitization to convert visible light into energy, dye is used as a sensitizer. These cells' abilities rely on the dye. An essential use for natural dye is sensitization. By using straightforward extraction techniques, natural dyes are replacing costly chemical synthesis methods and lowering the high cost of metal complex sensitizers (Richhariya et al., 2017).

Titanium dioxide-based semiconductors are utilized in the photoelectrochemical electrochemical process of water splitting. Due to their affordability and environmental friendliness, natural dye sensitizers are used to increase the efficacy of photoelectrochemical electrochemical reactions. Dye sensitizers show quick electron injection and delayed backward reactions. Natural pigment sensitizer must be protected by a layer, such as a conductive polymer layer because it is unstable in solution (Jaafar et al., 2017).

In dye-sensitized solar cells (DSCs), anthocyanin and chlorophyll are naturally occurring sensitizers that are extracted from the troll flower and cypress foliage. A UV-Vis absorption measurement revealed that, in comparison to each dye alone, the combination of two dyes allowed a greater and wider absorption in the wavelength range of 300 nm–700 nm. According to FTIR findings, anthocyanin is chemically adsorbed onto the TiO2 film, whereas chlorophyll is physically adsorbed. The mixture's optimal ratio of the two dyes was discovered to be 2:5, resulting in just enough charge transfer driving force and little energy waste. The photovoltaic efficacy was noticeably enhanced when this mixture was used as a adsorbing sensitizer as opposed to a single dye sensitization system (Nan et al., 2017).

Lawsone is the name for henna. It is a reddish-orange pigment that has been used to dye textiles, as well as skin, hair, and other body parts. Henna has attracted a lot of scientific attention recently for use in textile dying, largely because of how well it works in harmony with nature. Henna has no negative environmental effects and a low chemical reactivity (Bhuiyan et al., 2017).

A successful antimicrobial finishing of wool fibres was achieved as a result of research into the impact of Rheum emodi L. dye and its dyed wool yarns activity against two bacterial (Escherichia coli and Staphylococcus aureus) and two fungal (Candida albicans and Candida tropicalis) species (Khan et al., 2012).

Silk cloth has been dyed using natural dye made from plants like Black carrot, Hibiscus, Delonix, Plumeria, Combretum, Ixora, and Bischofia (Vankar, 2017).

Rheum emodi, Gardenia yellow, and curcumin are three natural yellow dyes that have been effectively used to simultaneously dye and functionalize silk to give textiles UV protection properties (Rungruangkitkrai et al., 2012).

Usually known as Bixa Orellana, annatto is a natural pigment. It is a yellow-orange dye made from this plant's seeds. It is environmentally friendly and highly biodegradable, with low toxicity. The primary substances present in all parts of this plant are carotenoids, apocarotenoids, terpenes, terpenoids, sterols, and aliphatic compounds, which are said to exhibit a variety of pharmacological activities. Annatto is used in a variety of industries, including solar cells, leather, food, and textiles (Shahid-ul-Islam et al., 2016).

Comparative studies of the deodorizing performance of textiles coloured with natural colourant extracts revealed that pomegranate outperformed gardenia and Cassia tora in terms of deodorizing power. L., the pomegranate skin, and the coffee sludge. (Young- Hee Lee et., al, 2015).

Natural dye has a lengthy history of use in wool dying. Today, some fresh materials are also used for dying clean wool. Wool can be dyed with Celosia, Nerium, Hollyhock, Hibiscus mutabilis, Caryatia, Tegetus, Rambutan, and Curcuma. Alkalines weaken animal proteins like wool, which dye best in acidic environments. Natural dyes that are used are mainly used to color wool in acid dye baths (Vankar, 2017).

A specific nation has its own fundamental laws and acceptable criteria for artificial colour additives in food, which can vary from one nation to the next. Natural-origin colours have gained acceptance over time, and scientific groups have focused more on developing greener substitutes because they are typically more widely accepted.

5.1 Natural dyes Use in textiles industries

Despite this, the majority of natural dyes have poor to middling lightfastness, whereas synthetic dyes have excellent to very good lightfastness.(Yoshizumi & Crews, 2003) According to Vankar et al., Beilschmiedia fagifolia (also known locally as Loto sheng) leaf extract can be used to dye cotton (Vankar et al.,2008). They have made B. fagifolia aqueous extracts and dyed cloth using a sonicator method. According to the authors, pretreatment with a 1-2% metal mordant and the use of a 5% plant extract led to the best results by ensuring adequate fastness. In a separate study, Shah and Datta coloured cotton textiles with floral dye made from marigold flowers (Saha & Datta., 2008). The use of Lac dyes on various fibres has also been the subject of some research (Chairat et. al., 2004). By Javali et al., annatto, a natural dye, had been applied to mulberry silk (Javali et. al., 2009).

6.0 CONCLUSION:

Natural dyes have drawn researchers in conventional and diversified uses due to the increased demand for eco-friendly materials made from sustainable resources. These researchers have created efficient eco-friendly and cleaner process technologies (Shahid et al., 2012).The use of natural dyes is steadily gaining popularity on the international market, and the creation of textiles that are eco-friendly and naturally dyed is a great way to protect the earth from harmful synthetic dyes. The colour derived from raw plant materials is known to be extremely sensitive to the conditions of food processing, but, generally, an eco-friendly standard that is paid safely to reconsideration of technological parameters, with a greater emphasis on their effects on colour stability, is advised and could be a promising alternative to artificial colourants (Hill, 1997).Before natural dyes, the quick-acting, cheap synthetic dyes pose a serious concern. However, naturally derived colourants' non-toxic, non-carcinogenic, biodegradable, and environmentally friendly qualities found their own way into the hearts of health-conscious customers despite being more expensive (Shahid et al., 2013).

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