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# A Review On Extraction Of Dye From *Terminalia catappa* Hull: A Substitute To Synthetic Dyes

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**Abstract***:* Natural substances are the biodegradable compounds which are obtained from natural resources unlike synthetic or man-made substances that are synthesized artificially mimicking the natural substances. The utmost reason for the extended usage of such synthetic substances is, they are more affordable than the naturally derived substances. One such example for xenobiotic substances is dye. Dyes are basically coloured substances which chemically bind to substrate and impart color to it. These synthetic dyes find its applications majorly in paper, cosmetic, textile and food industries. But due to its vast and continuous usage, some synthetic dyes have become harmful to both living beings as well as to the environment. This paper reviews the detrimental effects of synthetic dyes and essential requirements for substituting the synthetic dye with that of natural dye. The natural dye discussed in this paper is extracted from the hull of Terminalia catappa fruit (An Indian almond fruit). Further, the composition of T. catappa fruit, importance of flavonoids, one of the major plant secondary metabolites present in the extract and different extraction methods that can be opted for extracting the natural dyes are discussed.

***IndexTerms* - Anti microbial activity, eco friendly, extraction techniques, mordants, natural dyes, phenolic compounds, substitute, synthetic dyes, *Terminalia catappa* fruit hull.**

## I. INTRODUCTION

The history of dye dates back to the Neolithic period. Earlier, dyes were made with natural sources using water and oil for decorating skin, jewelry, clothing and were used on caves for painting or to signify some particular information. Dyes are basically a chemical substance which is responsible for imparting color to the substrate to which they bind. This color imparting property may occur either in the presence or absence of mordants. Before the origin of synthetic dyes, natural dyes were majorly extracted from plant sources like fruits, flowers, stem, roots, barks, leaves, wood as well as from animal sources like molluscs. In 1856, teenager William Perkin while trying to make quinine in his home lab, accidently discovered a dye and it was named as mauve, a purplish hue. This revolutionized fashion, medicine and chemical industries as well as led to a drastic shift from natural dyes to synthetic dyes. The development of mauveine led to competition between different countries for the production of colors and provided a greater opportunity to the chemical industries. Synthetic dyes were further classified into different types of dyes such as acid, basic, substantive, mordant, vat, reactive, disperse, azoic, sulfur dyes etc [1]. Since synthetic colors are less expensive and were available in different shades, it found its enormous application in pharmaceutical, textile industries, food processing, plastics, cosmetics, photographic and paper industries. In textile industries along with the synthetic dyes, different mordants were combined in dyeing of clothes to enhance the color imparting property as mordant fixes dye to fibers as well as new hues could be made by combining them [2]. On contradictory, synthetic dyes are non-biodegradable and based on the studies carried out, some of the synthetic food dyes (Example: Brilliant blue, indigo carmine, Erythrosine, Allura red, Tartrazine, Sunset yellow etc) are carcinogenic which has potential to cause great harm to living beings as well as causes tingling effect, sniffling effect, watery eyes, allergies, food poisoning, hyper acidity and hyper sensitivity [3]. In addition, release of industrial wastewater into the water bodies which utilizes synthetic dyes majorly azo dyes affects the environment adversely. As a result, increases BOD and COD value and hence causes disturbance in the natural cycle of amphibian biota, diminishes the ability of light to penetrate into water bodies and ultimately leads to death of marine lives [4].

Due to the increase in the adverse effects of synthetic dyes, most of the people who are health conscious and environmentally friendly are substituting such dyes with natural dye. These are biodegradable compounds and obtained from natural sources. Shifting back to natural dye has given many perks to current situations. Some of the vital benefits of non-synthetic dyes are these are not destructive to the environment hence disposing them do not create pollution, they help to obtain a soft hues or soothing shades. Some non-synthetic dyes, like that of carmine used in lipsticks, would not cause harmful or health problems when ingested [1]. In addition, natural dyes can also be used in food processing and cosmetic industries. (Example: Annatto extract, Anthocyanins, Caramel, Beet juice color, Aronia extract, β-carotene etc) Furthermore, natural dyes have a wide application in textile industries. The color enhancing property as well as different shades of hues can be obtained by using different mordants such as tannins, oil type mordants, iron, potash alum, sulfates etc by using either pre mordanting, post mordanting or simultaneous mordanting methods [2][5]. As a result, utilization of synthetic dyes can be minimized. The scope and awareness of natural dyes is increasing day by day, therefore detailed studies are carried out to explore these dyes. It is required to categorize the dye extracted from natural resources and are basically classified as (a) based on sources of origin-Plant origin, Insect/Animal origin, Mineral origin; (b)Based on solubility- Soluble natural dye and Insoluble natural dye; (c)Based on substantively-Substantive natural dye and Adjective natural dye; (d)Based on color formed in the presence or absence of mordant- Monogenetic natural dye and Polygenetic Natural dye; (e)Based on chemical constitution-Indigoids, Berberine, Carotenoids, Quinones, Flavonoids, Dihydropyran based dyes, Betalains and Tannins [1][6].

Most of the existing natural dyes are already a substitute to some of the synthetic dyes. *Terminalia catappa*, an Indian almond and an ornamental tree popularly known as "desi badam" (a well-known herb in Ayurvedic system) is one of the natural resources through which a natural dye can be extracted and can contribute in replacing the usage of synthetic dyes. *Terminalia catappa*, also known as country almond, sea almond, tropical almond and Malabar almond, grows mainly in the tropical regions of Asia, Africa, and Australia. In India, these trees are found in Maharashtra, Karnataka, Tamilnadu, Andhra Pradesh and Kerala. It is an angiosperm, monoecious and belongs to the leadwood tree family, Combretaceae. The height of the tree varies from 20 to 45 meter and shows a strong resistance to drought, wind and salt. This tree almost grows throughout tropical areas, usually growing wide on the beach and roadside as a road shade. However, currently these trees are not exploited intensively even though the production rate of tropical almond fruits is high. In traditional Ayurveda, the extract of leaves was used in preparation of medicinal lotion for leprosy, scabies and was taken for stomach ache and headache. Later on, based on the studies carried out and on analyzing the composition of extract it was found that the leaves and fruit of *Terminalia catappa* tree or any of its parts have anticancer, antidiabetic, antimicrobial as well as antioxidant properties. It is also used in an aquarium to lower the pH and heavy-metal content of the water. Indian almond fruit is edible and classified as drupe with fleshy mesocarp and endocarp containing seed. The fruit drupe is 5-7 cm long, 3-5.5 cm broad and unripened fruit is green in color whereas ripened fruit is red or yellow in color.

**Table No. 1 Composition in *Terminalia catappa* hull**

| PULP CONTENT | COMPOSITION |
|---|---|
| Crude protein | 8.75±0.01% |
| Ash | 4.79±0.17% |
| Crude fibre | 3.10±0.03% |
| Fat | 0.51±o.2% |
| Carbohydrates | 82.85±0.23% |
| Organic matter | 95.21% |
| Phosphorous | 22.566±0.012mg/100 |
| Vitamin C | 797.95±0.11 mg/100g |
| Vitamin A | 0.38±0.03 mg/100g |
| Oxalate | 396.645±0.017 mg/100g |
| Phytic acid | 4.675±0.014 mg/100g |
| Phenolic compounds | 244.33 GAE/g |
| Alcoholic compounds | 142.84 GAE/g |
| Tannin | 44.00±0.003 mg/100g |
| Flavonoids | 51.67 mg/100g |

Values are average ± standard deviation of triplicate determinations

Table No.1 depicts the proximate values about the composition of pulp [7][8][9]. On analysing the composition, it can be inferred that the natural dye can be extracted from the fruit hull as it contains flavonoids and pigments such as Violaxanthin, Leutin, Zeaxanthin and Anthocyanin.

## II. IMPORTANCE OF PHENOLIC COMPOUNDS

Secondary metabolites such as phenolic compounds which are produced in the shikimic acid of plants and pentose phosphate through phenylpropanoid metabolization [10] contains several benzene ring, with one or more hydroxyl group and vary from simple phenolic molecules to highly polymerized compounds [11]. The most prominent secondary metabolites found in plants are the phenolics. These polyphenols contain quite a few ranges of compounds like simple flavonoids, complex flavonoids, coloured anthocyanins and phenolic acids [12]. The phytochemical compounds like phenolics obtained by the consumption of fruits green leaves and vegetables show a positive impact, especially due to their antioxidant activity [13]. Some recent research shows that the inhibition of α-glucosidase and α-amylase is induced by phenolic compounds and therefore is helpful in the treatment of type-2 diabetes [14].

One of the sub-classes of phenolic phytochemicals, anthocyanins is blue, purple or red pigments found in plants, in particular fruits, flowers, and tubers. Anthocyanin appear as red pigment in acidic conditions while blue pigment in alkaline conditions. Blue, purple, and red -coloured anthocyanins pigments are extracted from flowers, fruits, and vegetables and are traditionally used as dye and food colorant. Additionally used as natural colorants, several of the anthocyanin rich flowers and fruits have been conventionally used as medicine to treat various diseases. Plant anthocyanins have been broadly studied for their therapeutic values. Anthocyanins have the properties such as anti-microbial, anti-inflammatory, anti-diabetic, anti-obesity, and anti-cancer effects and to prevention of cardiovascular diseases (CVDs) [15]. For that reason, anthocyanins obtained from edible plants have a possible pharmaceutical application. Some of the most commonly found in anthocyanins distributed in the plants are peonidin, petunidin, pelargonidin, malvidin Cyanidin and delphinidin. In nature, Pelargonidin appears as red-coloured pigment [16] and differs from most of the anthocyanidins. Delphinidin is chemically similar to most of the anthocyanidins. It appears as a blue-reddish or purple tint in the plant. The blue hue in flowers is due to the delphinidin pigment [17]. Cyanidin is a pigment which is reddish-purple and is majorly found in berries [18] and other red-coloured vegetables such as purple corn and red sweet potato [19].

Presence of rich secondary metabolites in *Terminalia catappa* like theat of phenolic compounds, tannins, saponin, flavonoids, alkaloids, triterpenes, phytosterols and steroidal glycosides [20] has contributed to the medicinal activity of tree. Punicalagin, punicalin, quercetin, chebulagic acid, corilagin, kaempferol, tercatain, terflavin B, tergallagin, and terflavin A are the phytochemicals present in the leaf of *T. catappa* [21]. Each and every part of the tree has a medicinal property due to the presence of secondary metabolites. For example, problems related to leprosy wounds, high blood pressure, Hemorrhoids and other skin diseases can be tackled with the help of leaves [22] and has been used in India and in the Philippines as a cure for several diseases. As a remedy for dysentery, the bark of trees is used in southeast Asian countries [23] and in Taiwan Ayurveda medicine, the nut is used because of its antibacterial and aphrodisiac properties [20]. Based on the studies carried out, this tree exhibits multiple pharmacological properties like anticancer antidiabetic, anti-inflammatory, analgesic, hepatoprotective and immunomodulatory [24]. Apart from the above-mentioned therapeutic applications of *T. catappa* leaves, it is also used to improve the health of fishes against pathogens [25] by preventing fungal infections caused on fish eggs or improves the fish larval survival rate and growth performance [26]. Hence this paper reviews different extraction techniques used to extract the dye from *Terminalia catappa* fruit hull that possess different medicinal properties due to the presence of secondary phytochemicals majorly flavonoids.

## III. EXTRACTION OF DYE FROM TERMINALIA CATAPPA HULL

The extraction is a separation technique in which one or more components are separated from solid or liquid mixture using solvents. Different types of extraction processes are used to extract dyes from their natural resources using solvents such as water, methanol and ethanol majorly. Therefore, by using these extraction techniques at appropriate conditions the dye can be extracted from *Terminalia catappa* fruit hull. Initially, Indian Almond fruits can be collected from regions where these trees are available. The fruits are cleaned under tap water so as to remove the dirt and can either be peeled or cut into small pieces. Figure I depicts the picture of *Terminalia catappa* fruit [27] and figure II shows the picture of peeled / cut pieces of hull which is the raw material for any type of extraction process. (Source-photographed in department of biotechnology, Basaveshwar Engineering College, Bagalkot, Karnataka .

**Figure 1: *Terminalia catappa* fruit**

**Figure 2: Peeled hull of *T. catappa* fruit**

On obtaining the raw material, following different methods of dye extraction process can be applied to extract dye from peeled pericarp of *T. catappa* fruit.

### 3.1. ULTRASOUND ASSISTED EXTRACTION

In this method, the dried and ground samples of green walnut shell with methanol as solvent were used. This sample was placed in an El masonic S10 H ultrasonic chamber for 30 minutes with frequency 20Hz and temperature was set-up at 20-40°C. After 2 hours it rose up to 60°C this was repeated for 2-3 times and the extract was collected. The extract was filtered using Whatman filter paper, and the solution was collected. The residue was immersed back in the ultrasonic chamber and then extracted again in the same conditions [28].

### 3.2. MICROWAVE ASSISTED EXTRACTION

The equipment used for this process is microwave oven and was operated at a power of 330W. Different amounts of powdered pomegranate rind (0.1g, 0.2g 0.5g, 1g) was weighed and transferred into 100mL beakers and to this 50mL of distilled water was poured. The effect of Microwave time on the yields of colorant was examined at different time intervals (10 sec-2 min) with different pH (1.96-9.5). Extract samples were taken at different time intervals (30, 45, 60, 90, 120 and 180 minutes), filtered and dried. The optical density of the extract was measured with the help of a UV-vis spectrophotometer [29].

### 3.3. ACID METHOD OF EXTRACTION

The beetroot extract was prepared by boiling the beetroot pieces in water and HCl acid. About 1kg of beetroot was peeled, cut into small pieces and soaked overnight. Later the mixture of 1000 ml of distilled, 10 ml of concentrated HCl and beetroot was boiled for 1 hour. The extract was then filtered and collected in a fresh beaker [30].

### 3.4. ALKALINE METHOD OF EXTRACTION

The alkaline method of extraction was used to obtain beetroot extract using 1 kg of fresh beetroot.The rinsed beetroot was thoroughly peeled and then was chopped into small pieces. These pieces were soaked overnight in 1000 ml of distilled water and sodium hydroxide solution(NaOH) of different weights.(4gm, 8gm, 12.5gm, 16gm). Further this mixture was boiled for 1 hour. The extract was then filtered and collected in a fresh beaker. Natural dye extracted by alkaline medium in different concentrations like for 0.10M the yield was 53%, in 0.20M the yield was 51%, in 0.30M the yield was 54% and in 0.40M the yield was 53.5%. Clearly, it was concluded that the maximum yield was achieved in case of 0.30M NaOH solution [30].

### 3.5. AQUEOUS METHOD OF EXTRACTION

Using this method of extraction, the *T. catappa* leaves were washed thoroughly and cut into small sizes with variations of fresh and dried leaves. Weigh 500 grams of cut leaves, add it in 2 liters of water and soak for different maceration times like 2, 4, 6, 8 and 10 days. Stir the samples for every 24 hours and filter the sample into container bottles. Hence the extracted dye was analyzed by using UV-Visible spectrophotometer or Fourier transform infrared spectroscopy [31].
About 100-gram dry powder *of T. catappa* leaves were taken in 1 liter distilled water andwas allowed to soak overnight. Next day the mixture was boiled for 30 minutes and then filtered with cotton cloth and with simple filter paper to get a clear solution. The extracted dye was analyzed by using UV-Visible spectrophotometer and its pH value was estimated [32].
5gm of *Terminalia catappa* leaf and seed pericarp was weighed and 100mL of distilled water was added to it. The beaker was kept in water bath at 60 ̊C for 1 hr. The dye extract was filtered through Whatman paper grade number 1. Further, the dye extract was characterized at λmax (UV-1800, Shimadzu UV Spectrophotometer) at pH 2.0 and 9.0 the color and stability were checked [33].

### 3.6. ENZYMATIC METHOD OF EXTRACTION

The plant tissues contain starch, cellulose and pectin as binding materials, therefore enzymes such as amylase, cellulase and pectinase which are commercially available can be used to loosen the surrounding material respectively leading to the extraction of dye molecules under milder conditions [34]. Hence such enzymes can be used on the fruit hull of *T. catappa* to degrade or loosen the binding materials and a dye can be extracted out of it.

### 3.7. FERMENTATION METHOD OF EXTRACTION

In this method of extraction, the enzymes produced by the microorganisms or those present in the natural resources for assisting the extraction process are used. The freshly harvested indigo leaves and twigs were soaked in warm water by maintaining a temperature of about 32 °C and fermentation procedure was carried out for about 10-15 hours. As a result, glycoside indican present in the leaves is broken down into glucose and indoxyl by the indimulsin enzyme and indoxyl gets oxidized by air to blue-coloured insoluble indigotin which settles down at the bottom. Further, it was collected, washed, and after dehydrating excess water, it is pressed into cakes. This process can also be used for extraction of certain other colorants such as annatto [34].

### 3.8. REFLUX BOILING METHOD OF EXTRACTION

Reflex boiling method of extraction of natural dye was carried out using the peel of pomegranate in distilled water. 5 gram of powdered pomegranate peels were added in round bottom flasks at different extraction conditions and with the help of a digital pH meter, the pH of the extraction liquors was adjusted using 0.1 M NaOH or 0.1 M HCl solutions for alkaline or acidic conditions respectively. Sample extracts were taken at different time intervals (30 min, 42 min, 60 min, 78 min and 90 min) and material to liquor ratio was varied to obtain the extract [35].The extracted sample obtained can be further analyzed using UV-Visible spectrophotometer.

### 3.9. SOXHLET METHOD OF EXTRACTION

Using Soxhlet method of extraction, a natural dye is obtained from a source, marigold flower by grinding the flower to powder. About 10 grams of powder was weighed and added to the thimble of Soxhlet extractor. The ethanol (240 ml) and distilled water (160 ml) with liquor ratio of 1:40 were heated at a temperature of 95 ̊C in a round bottom flask. For 6 hours the extraction was carried continuously and the vapors so formed were passed through the tube and elevated into the condenser. The vapours obtained at the top were condensed and dripped down into the thimble. The extracted material returned into the round bottom flask and mixes with the clean solvent. Through the rotary evaporator the extracted dye was purified and the dye solution was filtered [36].

### 3.10. SUPERCRITICAL METHOD OF EXTRACTION

A simple Supercritical fluid extraction method comprises extraction and separation process. If the sample is solid, columns are filled with dried and milled samples and the pressurized supercritical solvent flows through the column and the compounds that are extractible from the solid matrix are dissolved. The compounds that are dissolved are then transported by diffusion to the separator in which the extract and solvent are separated through pressure reduction, temperature increase or by both [37]. Since *T. catappa* hull contains a phenolic compound, flavonoid which plays a significant role in plant pigmentation it can be used to extract dye.
The phenolic compound, flavonoid was extracted from *Strobilanthes crispus* leaves using the same method at varying Pressure such as 100, 150 and 200 bar at varying Temperature: 40, 50 and 60 ̊C by considering the dynamic extraction time as 0, 40, 60 and 80 minutes respectively [38].

## IV. CONCLUSION

*Terminalia catappa* fruit contains flavonoid, a phenolic compound having antimicrobial, anticancer and antioxidant properties. A dye can be extracted from it by implicating any of the discussed extraction methods. The dye extracted from Indian almond fruit can be widely used in various industries. In Food processing industries, dyes can be used to impart color to the food and make it look attractive, appealing, appetizing and provide information to the consumers regarding flavors and also can be used as food preservative agents. In textile industries, it finds its application in dyeing of clothes with or without the use of mordants and gives a cool, soothing appearance to the clothes. Dyes in pharmaceutical industries can be used to impart color to syrups or tablets and give it an aesthetic appearance. In cosmetic industries, dye can be used to impart color to the body lotions, lip cosmetic products like lip balm, lip gloss, lipstick, as a contouring product like highlighter etc. The major advantage of dye extracted from the hull of *T. catappa* is it is eco-friendly, biodegradable, renewable, and imparts soothing shade like other natural dyes. The major drawbacks of any natural dyes are these are costlier than synthetic dyes and their availability as most of the natural resources which act as raw materials are seasonal. These drawbacks can be overcome by opting proper extraction methods to reduce the cost of extraction and by choosing proper preservation techniques for preserving the raw materials and by making it more available. As a future scope, research work can be carried out for using the dye as a coloring ingredient in cosmetic products, hair color products and series of tests can be conducted to check for skin compatibility. The dye extracted from the hull of *T. catappa* can be used to minimize the usage of synthetic dyes and hence can be a substitute to artificial colors.

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