



## Naturally Extracted Inks for Digital Printing of Natural Fabrics

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### Abstract

People are getting more and more interested in the advantages of utilizing natural dyes in textiles. Synthetic dyes have been linked to toxic and allergic responses, which has led several nations to enact strict environmental laws. Natural colors are more ecologically friendly and biodegradable than synthetic dyes. Natural colors come in four different varieties: those derived from plants, animals, minerals, and microbes. Natural dyes may be used to color any natural cloth. According to a recent study, they can also be used to color synthetic materials. Natural colors are utilized in food, medicine, handicrafts, and the tanning of leather in addition to textiles. Many of the plants that give things their natural color also have medicinal uses. To be thorough, this study reviews the categorization of natural dyes and the myriad sustainability issues related to their production and use.

**Keywords:** digital printing, natural ink, natural fabrics.

### Introduction

In our daily lives, pens and inks play a significant and varied function. Our day starts with newspapers and toiletries, moves to the breakfast table, where a variety of packaged goods labeled with ink are present, including tea or coffee, bread, and butter, and gradually moves to our workplaces, whether they be schools or offices, where ink can be found on everything from books to stamps to computer prints to money. [1]

Organic or inorganic pigment or dye that has been dissolved or suspended in a solvent to create ink. Chemically speaking, it is regarded as a colloidal system of small pigment particles, either colored or uncolored, dispersed in an organic or aqueous solvent. The first inks were rumored to have been made from fruit or vegetable juices, defensive secretions from cephalopods like squid, cuttlefish, and octopus, blood from some species of shellfish, and tannin from galls, nuts, or tree bark. [2, 3]

According to some estimates, the first man-made ink initially emerged in Egypt 4,500 years ago and was composed of glue mixed with either animal or plant charcoal (lampblack). [4] The first black writing inks used before 2500 BC were suspensions

of carbon, mostly lampblack, in water stabilized with natural gums or substances like egg albumen.

Many different components are used to create modern inks. In addition to the pigment, they also include a variety of additional compounds that are generally referred to as "vehicles" in varying proportions. Examples of these include pH adjusters, humectants to stop premature drying, polymeric resins to impart binding and other properties, defoamers/antifoaming agents to control foam efficiency, wetting agents like surfactants to control surface properties, biocides to stop fungal and bacterial growth that causes fouling, and thickeners or rheology modifiers to control ink application. [5]

In other words, printing has been around for millennia in some form or another. Yet, while the fundamental functions of ornamentation and information have remained constant, printing methods and ink formulations have changed significantly. [6]

The practice of creating water-based ink-jet inks for textile printing has gained popularity in recent decades. In the 1990s, textile digital printing evolved as a means of prototyping, small-batch fabric printing for niche-market items, and the creation of promotional materials like flags and banners. [7]

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Around 5% of all textile printed materials are now produced digitally. [8] While digital textile printing is now increasing at a pace of 13% and is anticipated to achieve a growth rate of 20% in the next years, conventional textile printing is forecast to increase at a rate of 2% every year.[8-10] In Europe, textiles have been colored for a very long time using natural dyes. Vegetable, animal, and mineral extracts were used as dye sources. Madder, indigo, and saffron were the three ancient colors most often employed from about. 2500 BC onwards. [11-13]

The use of natural dyes for textile dyeing nearly completely ended with the invention of synthetic dyes. The abundance of synthetic dyes produced today satisfies consumer demand for high-quality goods at affordable prices as well as expectations for straightforward, repeatable application techniques. Natural dyes are therefore readily available, but synthetic dyes are more widely used since they are easier to make in big numbers, can be produced at a fair price, can give a diversity of colors, and can generate dyings with excellent color fastness to match today's requirements. [14, 15]

However, the production of synthetic dyes is constrained by several factors, including environmental unfriendliness due to the use of strong acids, alkalis, solvents, high temperatures, and heavy metals in dye synthesis, as well as higher costs for raw materials and energy due to the need for petroleum in the production of the raw materials as well as the generation of the energy needed for dye synthesis operations. Additionally, the environment is quite concerned about how chemical waste is disposed of. [16-31]

From the perspective of the customer, synthetic color toxicity and allergic responses are taking on more significance.

To fulfill the increasing consumer demands for health and safety, there has been a renaissance of interest in natural dyes in recent years. These dyes provide ecologically acceptable textile materials with little or low toxicity and allergic responses.

If natural "green" dyes can be manufactured for a similar cost as synthetic dyes and have similar fastness properties, then they may be a viable alternative. But, in situations where health and safety requirements are higher, such as with infant garments in light colors, they can still be utilized as an alternative to synthetic dyes. Even though natural dyes have been used extensively in traditional printing and dyeing, there are no references to natural dyes being used in digital printing.

Natural ink-jet digital printing is crucial because it not only enables the creation of textile products with a high added value that is environmentally beneficial but also enables the creation of unique environmentally friendly

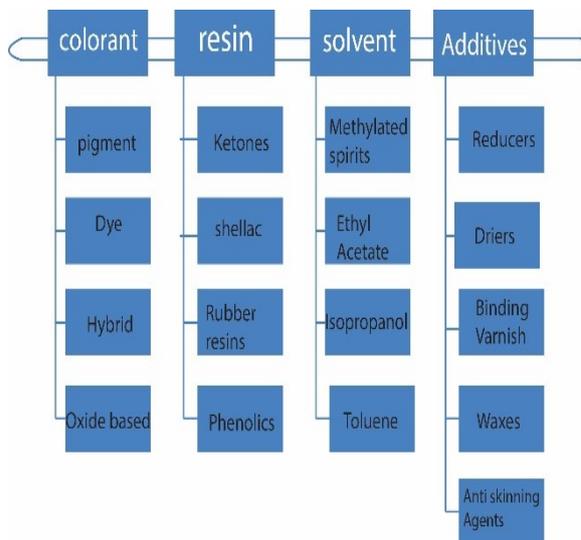
materials for the packaging, food, and cosmetics sectors. [32]

One of the fastest-growing imaging technologies is ink-jet printing, which outperforms traditional printing techniques like roller and flat-bed screen printing and offers special benefits like simplicity, lower production costs, reduced effluent waste, lower water, and energy consumption, as well as unlimited design options, enabling the creation of creative, customized finished goods. [10]

Pigments make up the majority of ink's cost and are hence its most crucial element. A pigment is a small, colorful, black, white, or fluorescent material that modifies the appearance of an object by selectively absorbing and/or scattering light.

Throughout the coloring or printing process, it remains a colloidal solution with a crystal or particle structure in ink.

To identify organic pigments in contemporary inks, people frequently utilize the Organic Pigment Identification System number. Together with structural and historical details, it reveals the pigment's color shade or hue (sequence of synthesis). The ink's color is provided by pigments, which also offer gloss, abrasion resistance, and resistance to light, heat, solvents, and other elements. Also used as special pigments are extenders and opacifiers. White pigments called opacifiers make the paint opaque, preventing visibility of the surface underneath. Translucent pigments called extenders make the hues of other pigments look less vibrant.[33]



**Fig. 1. Composition of printing links.**

**Types of pigments**

There are several varieties of pigments on the market. As they convey the ink's visual character and almost always make up the bulk of the ink's component, pigments and dyestuffs are perhaps the most important ingredients in printing ink composition. Both pigments and soluble dyes alter a substrate's reflectance to give it color. Pigments are multimolecular crystalline structures designed for an ideal particle-size distribution, hence insolubility is a crucial property. They either remain on the surface after being applied to a substrate in a vehicle or fill in any gaps left by paper or other uneven surfaces. [34]

Dyes are colored substances with a particular affinity for the substrate on which they are applied. The dye is typically applied as an aqueous solution, and a mordant may be required to increase the fastness of the fiber. There are two main categories of dyes, including:

- 1) Natural dyes
- 2) Synthetic dyes.

**Natural Dyes:** These colors are mostly made from natural materials. They are constructed with sustainable materials. [35-46]

**Synthetic Dyes:** A range of chemicals, synthetic materials, and other components are used to create these hues. It is impossible to overestimate the importance of color in textiles. Humans have been enthralled by colors from the beginning of time. Natural vegetable dyes have been a part of human life since the beginning of humanity. These hues may be found in manuscripts from the Mughal era and Egyptian mummies. The use of natural dyes has decreased since the invention of synthetic dyes in 1856. [47, 48]

**Classifications of inks**

The main contrast between a pigment and a dye is that the former may be suspended in a medium or binder while the latter is insoluble. This is due to the varied particle sizes that dyes and pigments have, which affects how they work. Think about the distinction between a pinhead-sized dye and a football-sized pigment particle (pigment). Because dyes are soluble and pigments are not, you may imagine dye particles dissolving in water whereas pigment particles need to be suspended in a binder (pigment). Although the pebbles settle to the bottom and create a suspension, the salt dissolves in the water to produce a solution.[49]

The ability to bond to a surface is another difference between pigments and dyes. While a dye can chemically bond to a surface at the molecular level and become a part of the material, a pigment needs a binder or carrier to act as the glue that is painted on the substrate and surrounds the pigment

to keep it in place. As a result, pigments are stacked on top of dyes, which form a component of the material.[49]



**Fig. 2. Classification based on structure.**

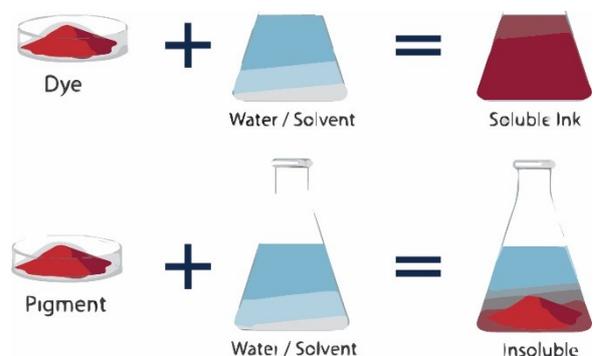


**Fig. 3. Classification based on source.**

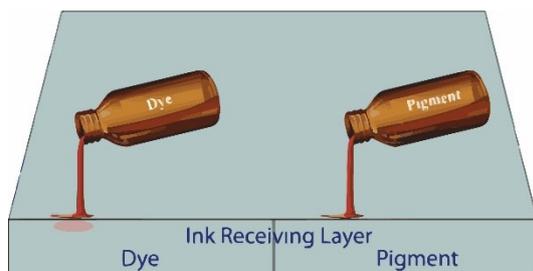


**Fig. 4. Classification based on application.**

The ability to link to a surface is another difference between pigments and dyes. While a dye may chemically bond to a surface at the molecular level and become a part of the material, a pigment needs a binder or carrier to function as the glue that is painted on the substrate and surrounds the pigment to keep it in place. As a result, pigments are stacked on top of dyes, which form a component of the material. ( Figure 5,6)[1, 50]



**Fig. 5. Difference between pigment and dye based on solubility.**



**Fig. 6. Difference between pigment and dye based on receptivity.**

These are fundamental requirements, therefore depending on the substance or dye, a dye can need a mordant to help it bond. One of the behavioral characteristics that set them apart is how quickly they fade in the presence of light. Compared to dyes, which are more prone to fading or bleaching brought on by UV radiation from the sun, pigments are more resistant to fading. [51]

Think of a pair of faded denim jeans or a patch of wallpaper where a hanging picture once was to understand why dyes fade: sunlight's ultraviolet (UV) rays may harm a dye molecule's electrical connection, causing it to lose its color. While some will need a more lasting color, other individuals will use colors, particularly for this reason, and enjoy the natural fading process. [52]

**Resins:** Resins are binders that hold the other components of the ink together to produce a film and hold the ink to the cloth.

They also help with attributes like gloss and resilience to heat, chemicals, and water. Many different types of resins are used, and most inks use more than one resin. Acrylics, Ketones, Alkyds, Maleic, Cellulose derivatives, Formaldehydes, Rubber Resins-1, and Phenolics-2 are the most popular resins. [52]

**Solvents:** When ink is put to the printing plate or cylinder and transferred to the printed surface, solvents are used to maintain the ink liquid. The solvent must now separate from the ink body to allow the image to dry and bind to the surface. Some printing processes require a solvent that evaporates quickly. Ethyl acetate, isopropanol-propyl acetate, cyclohexanone, butoxyethanol, aromatic distillates, and butyrolactone are a few of the solvents used in printing ink. [53]

**Additives:** Additives are used to alter the final properties of the formulation. such as Plasticizers that increase the printed film's flexibility, Drier, such as cobalt, manganese, or zirconium salts or soaps, that catalyze the oxidation reaction in inks that dry by oxidation, Surfactants, such as eugenol, work to moisten the pigment or the substrate better by interacting with the free radicals produced during auto-oxidation and stopping them from recombining. [54]

Antioxidant, such as eugenol, slows the initiation of oxidation polymerization by interacting

with free radicals generated during auto-oxidation and prevents them from further reacting. They act as stabilizers for the dispersion of pigment pH adjusters, biocides, and bacteriostats (usually amine compounds). [54, 55]

#### **Problems with using conventional inks:**

Toxicology may be a serious problem since it can lead to allergies such as contact dermatitis, respiratory conditions, allergic eye responses, skin rashes, and irritation of the mucous membranes and upper respiratory tract. It is neither environmentally friendly nor biodegradable and may be hazardous to both human health and the environment.

#### **Natural dyes (A sustainable option)**

In comparison to natural dyes, synthetic dyes provide a lot of benefits, such as shade uniformity and color brightness. In the present era of environmental preservation, natural colorants are once again rising in popularity and have huge economic potential. Their non-hazardousness is also becoming well understood.

We are now forced to think about safer alternatives because of the widespread use of dangerous chemicals, particularly synthetic colors, which have resulted in ecological imbalance, pollution, and the loss of natural resources. The applications for natural dyes have increased as a result of these properties.

Synthetic colors are toxic and bad for the environment and people's skin. As a result, their usage is prohibited in several European nations. This calls for the revival of natural dyes for the coloring of textiles. Natural colors are biodegradable and kind to the environment. Forestation is encouraged by plant cultivation for natural dyes, which promotes greater ecological balance. [56]

A notable maker of natural dyes is The Allergo Company. It aims to replace 1% of synthetic dyes worldwide. They anticipate that they will use 0.2% of the total available farmland. Growing raw materials, developing dyeing equipment to meet industrial needs, and transferring technology to large textile mills are some of the difficulties that have been addressed in making natural dyes an economically viable alternative to synthetic dyes. [1]

It has developed extraction processes that yield pure liquid color. The liquids are then used to create the color powder. The trash produced during the extraction process is recycled and added to the compost as additional carbon. Additionally, it has produced an eco-friendly mordant. The firm only makes five different dyes. Some of them are osage orange, cochineal, madder, catch, and indigo.

Alps Industries is a significant maker of natural dyes.

Each year, this business produces 300 tonnes of natural dye. The business has started a \$1 million research and development endeavor to create "totally eco-friendly natural dyes. (Table1) [57]

This three-year research and development project's primary objective is to produce at least six fully standardized natural colors using the following raw materials: pomegranate peel, Harada gallnuts, Catechu, Annatto, Madder, Ratan jot, and Himalayan rhubarb.

In addition, several businesses focus on specific colors like indigo, lac, cochineal, and catch. [57]

### 1) red beet

Red beets were extracted by being cut into little pieces and then blending them one at a time in ethanol and water (1:2) at 200 rpm.

The solvent absorbed the betaine dye.

1-2) Extraction of red beet:

Red beets were bought at neighborhood markets and stored at 4 C until required. The beets used in the study were washed first, and then they were grated to increase their surface area. 10 g of grated beetroot pieces were added to a 1000 mL mixture of 4:1 ethanol and water, and the mixture was mechanically stirred at 200 rpm for 2 hours at room temperature. After being filtered, the colored liquid's liquid portion received the active dye. To remove the color from the solvent, an evaporator operating at 1500 rpm and a Hoover at 50°C was utilized.

The colloiddally dispersed material was precipitated using ultracentrifugation at 13,500 rpm for 10 minutes, and it was then dried in a vacuum drying oven. A Perkin-Elmer Spectrum 100 ATR-FTIR spectrophotometer was used to identify the chemical structure of the novel colors. The dye's color character was determined by measuring the spectra with wave numbers spanning from 650 to 4000 cm, using a UV-2450 model and UV-VIS spectrophotometer. [58, 59]

**Table 1. Extraction of dyes from different sources.**

Image	source	Nature Acidic	Nature Initial	Nature Alkaline
	Butterfly pea	Red	Blue	Blue
	Allamanda	Brown	Brown	Yellow
	Pomegranate	Light orange	Dark Brown	Light Brown
	Grape	Pink	Light Purple	Green yellow
	Turmeric	Yellow	Yellow	Orange
	Red cabbage	Red	Blue	Orange-yellow
	Black bean	Red	Violet	Green yellow
	Purple potato	Pink	Pink	Green
	Red rose	Pink	Red	yellow

**Table 2. Different Methods of Extraction for Natural Dyes.**

Extraction method	Remarks
Aqueous Extraction	An ancient and traditional method. The powdered source is soaked in water, boiled, and filtered the liquid.
Ultrasonic Extraction	High power in low-frequency ultrasound waves is used to extract the dye. The powder form of the plant with a mixture of ethanol and water is placed in the ultrasonic bath and sonicated, filtering the liquid dye.
Solvent Extraction	The most effective techniques for nature dye extraction. Ethanol, methanol, acetone, petroleum ether, chloroform, and such organic solvents are used in the extraction process.
Microwave Assisted	The procedure of Microwave Assisted Extraction (MAE) involves heating solvents in contact with a sample to separate the analyst from the sample matrix into the solvent.
Alkali or Acid Extraction	We get the maximum color yield and boosts up because of the hydrolysis of glucosides.
Enzymatic Extraction and Fermentation	The advantage of the enzyme extraction approach includes softer extraction conditions and consistent physical and chemical characteristics of active components.
Supercritical Co2 Extraction	The most complicated process has benefits from both liquids and gases, high density and viscosity, lower surface tension, and increased solubility, all of which increase quickly with pressure.

**Fig. 7. Extraction of red beet.**

## 2) curcumin

is a highly popular yellow natural coloring. Curcumin may give fabrics a beautiful yellow color while also adding a variety of functionality.[60] Due to its hydrophobic nature and tiny size, curcumin has poor washing and rubbing fastness on fabrics. [61] The most common techniques for improving color fastness are mordanting, crosslinking, and molecular alterations. [61-65]

However, the color characteristics of curcumin are significantly altered as a result of the formation of coordination or covalent interactions between curcumin and chemical agents, giving the finished textile goods an unfavorable look. The decrease of free hydroxyl groups caused by the mordanting and chemical changes had a significant influence on curcumin's functioning, according to earlier research. [65, 66]

Therefore, there is a pressing need to find new techniques for improving curcumin's fastness on fabrics.

Polyphenol tannin acid is abundantly found in grains, legumes, fruits, herbs, and vegetable beverages.[67] Proteins are largely attracted to

tannin acid through a variety of hydrogen bonding interactions. [68-70]

We may add tannin acid to polyamide, silk, and wool textiles to stop curcumin from leaching out since applying tannin acid to protein films can also increase their hydrophobicity. [71] Additionally, tannic acid is a colorless chemical by nature, which may have little influence on the hue of curcumin.

## Ink preparation

Curcumin and Triton X-100 (CT), curcumin, Triton X-100, and tannin acid (CTT), and tannin acid and SDS (TS) were the three ingredients used to create the ink solutions.

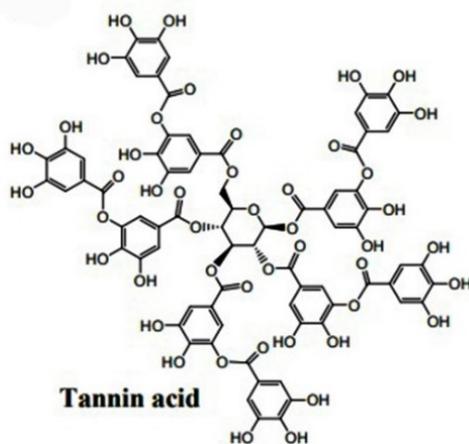
All of the ink solutions were blended using a 130 W ultrasonic processor (Cole-Parmer, USA) at 80% amplitude for 30 minutes. After that, they were filtered using a 25 mm syringe filter with a 0.45 m nylon membrane (VWR, USA), and then kept in glass bottles with a brown tint. All ink compositions' viscosity, surface tension, stability, and jet ability were evaluated.[72]

## 3)Annatto

Annatto The smallest tree in the Bixaceae family is called a bixa. The tree is distinguished by its vivid orange hue; see Figure 7 for its chemical composition.



**Fig. 8. Chemical structures of curcumin**



**Fig. 9. Chemical structures of tannin acid**

It is frequently used for items like colored butter, colorful cheese, silk, wool, and cotton. The pulp contains a lot of tannins. Under boiling conditions, the alkaline extraction method is used to remove pigment. It produces reddish-orange tones on cotton, wool, and silk.[73]

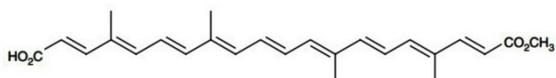
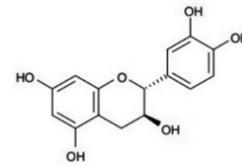


Figure10: Annatto (Bixa orellana) - CI Natural Orange 4

#### 4) Cutch (Acacia catechu)

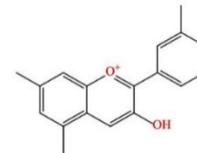
The Acacia catechu is a small to medium-sized, thorny tree that can grow to a height of 15 m. Its bark is dark grey or greyish-brown and peels off in long strips, or occasionally in narrow rectangular plates, which are brown or red on the inside. Its branches are slender, and pubescent when young, but they become glabrescent as they age, and each petiole has two curved, 8-mm prickles.[74]



**Fig.11. Cutch (Acacia catechu)**

#### 5) Pomegranate

Punica granatum rinds, which are used to bite, have a significant tannin content.[75] Additionally, a yellow teal may be used to dye cotton, wool, and silk quickly. Additionally, it is used with turmeric to increase the light speed of colored materials.[76]



**Fig.12. Pomegranate**

#### Advantages of natural dyes

Use renewable resources since they pose no health hazards and can even be used to treat diseases. There are no disposal difficulties, and the chemical reactions are rather modest. It takes a lot of ingenuity to use these colors effectively even though they are straightforward and in sync with nature. They help to maintain traditional handicrafts and provide work for rural residents, and the method of using these colors is less expensive. They have a delicate color, are calming to the eyes, and have a greater tinctorial value than other plants, which is good for the skin. Because the raw materials for dyes don't come from petroleum products and leftover materials are used to create compost, dyes are energy-efficient and biodegradable.[77, 78]

#### Challenges for natural dyes

Limitations of natural dyes that led to their death are currently being addressed. color output, availability, dyeing process complexity, and repeatability of shade. The only materials that can be dyed with natural dyes are wool, natural silk, linen, and cotton. Natural dyes are also non-standardized, have insufficient fixation and fastness, are contaminated with heavy metals and significant amounts of organic substances, lack precise application methods, and are not available in standard shade cards. They cost a lot of money and need about 500 acres of land per kilogram of dye since they can't be mass-produced. They also have low fastness attributes as a result of the natural dye's weak dye-fiber bond, the disintegration of the dye metal complex after washing, and the ionization of natural dyes after alkaline washing.[50, 58]

### Future scope of natural dyes:

To acquire an adequate shade gamut and fastness qualities, the best growing method for the essential vegetable coloring materials must be explored. For man-made fibers, natural dispersion dyes must be created; nevertheless, natural dyes can be used successfully on their own in non-toxic applications such as food, medicine, and cosmetics.

If natural dyes are given the correct attention, which is not currently happening because of their lack of expertise, they will become a more intriguing carrier. Antimicrobial, biodegradable, non-toxic, environmentally friendly, and aesthetically pleasing, which creates jobs and helps to till wasteland.[79]

These colors have beneficial medical and therapeutic effects and are safe and eco-friendly. A remarkable ecological balance is achieved by growing plants for the extraction of dyes, which supports forestation.

Numerous cancer-causing chemicals are used in the production of synthetic dyes, and the effluents that are discharged into rivers or released into the atmosphere pollute the environment.[80]

### Conclusion

Natural dyes from several plant sources have been produced recently for printing. [1]

Establishing suitable procedures for characterizing and certifying natural dyes will undoubtedly increase consumer confidence in them and benefit both producers and consumers. Small coloring units can use these colorants even though the availability of natural dyes can be increased through the aforementioned measures and the costs of cleaned dyes can be decreased with the proper certification. This is because these small coloring units lack the funds to install and run the pricy effluent treatment plants required to bring the synthetic dye effluent within the regulatory authors' set limits. If in the future, biotechnological techniques like tissue culture or genetic engineering could produce large quantities of inexpensive bacteria at any moment, dramatically increasing their availability, then the use of these colors for conventional textile manufacturing can only become viable. At this time, natural dyes can only be economically feasible for small-scale applications, but they can supplement synthesizers by providing an ecologically benign alternative and a source of income for a variety of stakeholders in the natural dye value chain. [79]

### Conflicts of interest

There are no conflicts to declare

### Funding sources

There is no fund to declare

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## الأحبار الطبيعية المستخرجة للطباعة الرقمية لبعض الأقمشة الطبيعية

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## الملخص

أصبح الناس مهتمين أكثر فأكثر بمزايا استخدام الأصباغ الطبيعية في المنسوجات. تم ربط الأصباغ الاصطناعية بالاستجابات السامة والحساسية ، مما دفع العديد من الدول إلى سن قوانين بيئية صارمة. الألوان الطبيعية أكثر صداقة للبيئة وقابلة للتحلل البيولوجي من الأصباغ الاصطناعية. تأتي الألوان الطبيعية في أربعة أنواع مختلفة: تلك المشتقة من النباتات والحيوانات والمعادن والميكروبات. يمكن استخدام الأصباغ الطبيعية لتلوين أي قطعة قماش طبيعية. وفقا لدراسة حديثة ، يمكن استخدامها أيضا لتلوين المواد الاصطناعية. تستخدم الألوان الطبيعية في الغذاء والدواء والحرف اليدوية ودباغة الجلود بالإضافة إلى المنسوجات. العديد من النباتات التي تعطي الأشياء لونها الطبيعي لها أيضا استخدامات طبية. في محاولة لتكون شاملة ، تستعرض هذه الدراسة تصنيف الأصباغ الطبيعية وقضايا الاستدامة التي لا تعد ولا تحصى المتعلقة بإنتاجها واستخدامها.

**الكلمات الدالة:** الطباعة الرقمية، الأحبار الطبيعية، الأقمشة الطبيعية.