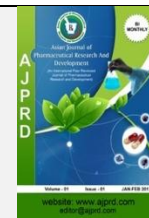


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Review Article

A Review: Natural Resources as Natural Dyes in Pharmaceutical Products

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ABSTRACT

Objectives: Several types of natural dyes have been used in pharmaceutical products such as food, beverages, cosmetics and drug additives. Natural dyes are safe and do not cause allergies or irritation to the body. The benefits of natural dyes are as a substitute for synthetic dyes which are safer for the body. The purpose of this article is to discuss natural dyes that are applied or added to pharmaceutical products or the pharmaceutical industry.

Data Sources Study Selection: The review method used is by studying the literature relevant to the research. Data sources of this article were selectively taken from Google Scholar, Pubmed, and Mendeley.

Summary of contents of the article: Pharmaceutical products always use coloring additives for consumer appeal. The addition of dyes to pharmaceutical products such as drugs will make it easier to distinguish between these types of drugs. Dyes that are safe for the body are chosen as a safe alternative in their use. Natural dyes can be derived from plants, minerals, and animals, many plants contain pigments and can be applied to pharmaceutical products such as food, beverages, cosmetics, and medicines. There are many kinds of pigments contained in plants and animals, such as flavonoids, tannins, alkaloids, indigoid, chlorophyll, and carotenoids.

Conclusion: The conclusion of this article is that natural dyes cannot only be used as a substitute for dyes that are safe for the body, but natural dyes also have pharmacological properties. The application of natural dyes to food, beverages, cosmetics and medicines does not affect the taste, aroma and texture of the product

Keywords: Beverages, Cosmetics, Food, Medicines, Natural dyes, Pharmaceutical products, Plants. and *Octyl methoxycinnamate*

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INTRODUCTION

Natural products as natural dyes can be often encountered in medicinal plants, medicinal plants are also widely used as a cure in various diseases¹. There are many sources that can be used to use natural products as natural dyes. Natural dyes are non-toxic, biodegradable and environmentally friendly². It states that the safety of color additives has been proven by testing the safety of toxicological data and is declared safe for use in the United States and elsewhere³. Natural colors come from plants, minerals and animals⁴.

Dyes are defined by the US Federal Food, Drug and Cosmetics Act (FD&C Act) as dyes, pigments, or any substance that can give color to food, medicine, or cosmetics, or to the human body. Natural dyes fall into one of the current categories of US Food and Drug Administration (FDA) regulations because they have followed FDA certification⁴. Natural dyes are currently in great demand by various groups to enhance appearances such as in food, beverages, pharmaceuticals and textiles. The old culture of using textile dyes has now begun to disappear and is replaced with natural dyes, increasing awareness of the health and dangers of synthetic dyes has shifted people to use dyes from natural sources⁵.

The purpose of this article is to discuss natural dyes in the pharmaceutical field or the pharmaceutical industry using literature study methods relevant to research, the pharmaceutical industry usually adds dyes to the manufacture of drugs as coloring agents. The purpose of coloring medicines is to increase the attractiveness of the product and help patients differentiate between drugs⁶.

Classification of natural dyes

Based on its origin, the classification of natural dyes is divided into three, namely vegetable, mineral and animal. About 500 dyes are derived from plants and all plant parts such as roots, leaves, stems, peels, flowers, fruits and seeds^{5,7}.

Some examples of natural dyes derived from plants and animals

Natural dyes can be differentiated into several pigments, namely flavonoids (flavonoids, flavanones, isoflavones, anthocyanins, anthocyanidins, and proanthocyanidins), tannins, alkaloids, indigoids, dyes contain mono or polycyclic in its structure (benzokuinone, anthraquinone and naphthakuinone), chlorophyll and carotenoids^{7,8}.

Anthocyanin

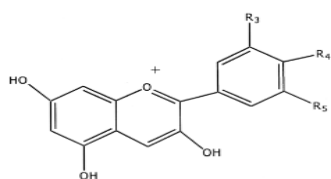


Figure 1: anthocyanin structure⁹

A single derivative of aromatic structure by addition or subtraction of hydroxyl groups or by methylation or glycosides⁹. Here are some plants that have anthocyanin pigments:

Brown seeds (purple color)

Chocolate seeds contain polyphenols, polyphenolic compounds contained in cocoa seeds are flavonoids. The purple color found in brown seeds is caused by the presence of anthocyanin pigments which are included in the pelargonidin group and are derivatives of flavonoid compounds. In this study, cocoa beans were used as natural dyes in lipsticks¹⁰.

Plantain heart

Extraction in the heart of plantain contains anthocyanin compounds which can be used as natural dyes. Not only it has the property as a natural dye, the plantain flower also has other properties, namely as an antioxidant. The results showed that the plantain heart contains anthocyanins with the dominant cyanidine derivative compound¹¹.

Red dragon fruit peels

Waste of dragon fruit peels have been used as a natural dye that is safe to use in food and beverages. In a study using red dragon fruit peel on microencapsulation, red dragon fruit peel contains anthocyanin pigments which are quite high and have hydrophilic properties. As well as safe to be

used as an alternative to natural dyes that are safe for health^{12,13}.

Purple sweet potato peels

Purple sweet potato peel has anthocyanin pigments and can be used as natural dyes. Anthocyanin stability of purple sweet potato peel extract on the influence of pH and temperature and co-pigmentation of purple sweet potato maintain the stability of anthocyanins^{14,15}. Anthocyanins are polar pigments and will dissolve well with polar solvents; these solvents can be used in anthocyanin extraction in purple sweet potatoes¹⁶.

Secang bark

Researchers have succeeded in using secang bark as a natural coloring agent for lipsticks. Secang (*Caesalpinia sappan* L) is a family of Caesalpiniaceae which has orange to dark red pigments caused by brazilin in anthocyanins¹⁷.

Rosella flowers

Rosella flowers contain anthocyanin compounds that have a natural red color, apart from being used as natural dyes, rosella flowers also have functional benefits for health and can also be used as a preservative because they contain antioxidants and antibacterial properties¹⁸. Harusnya dibawah rose flower sebelum bougainvillea flower

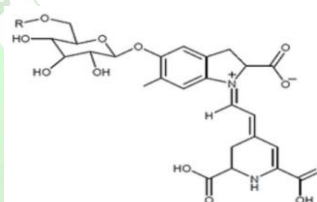


Figure 2 Betacyanin structure¹⁹

The above structure is a phenol compound that is substituted by a glycoside group in the ortho position and has a chromophore group. Existing functional groups can interact with anions that are capable of producing color changes¹⁹. Here are examples of plants that contain betacyanin:

Bougainvillea flower

Bougainvillea flower has many kinds of colors, the extraction of natural pigments in red bougainvillea flower pigment contain betacyanin. Betacyanin pigments are used as natural dyes because they have excellent coloration²⁰.

Beetroot peel

Beetroot peel has a deep red color, the red color is derived from the betacyanin pigment contained in the peel of the beetroot. Betacyanin is an antioxidant compound and belongs to the phenolic group¹⁹.

Dragon fruit and its peels

Dragon fruit and its peels contain betacyanin compounds, betacyanin is a red pigment that is widely used as a natural dye²¹.

Cactus fruit

Cactus fruit is a flowering plant that lives in the high and dry lands, cactus fruit has the family *cartacae* and contains betacyanin compounds which have a red substance²².

Carotenoids

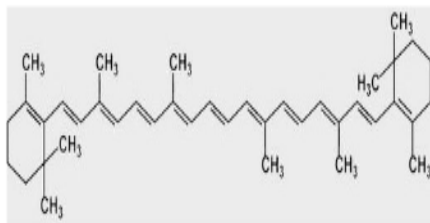


Figure 3: Structure of β -carotene²³

Well-known carotenoids are unsaturated hydrocarbons of lycopene derivatives or oxygenated lycopene derivatives (xanthophylls)²³. Beta carotene has the highest activity compared to γ -carotene and α -carotene. The following are examples of plants and animals that contain carotenoids:

Crab shells

One of the natural dyes from animals is crab shells. Crab shells have carotenoid pigments that have the function of provitamin A and antioxidants for the body²⁴.

Pumpkin

Pumpkin is one type of fruit that contains high carotenoids, because of that pumpkin is nicknamed "the king of beta-carotene". Apart from being a natural dye, pumpkin has other benefits for the body such as help preventing cancer, heart disease, diabetes mellitus, premature aging and impaired immune response²⁵.

Spinach, red capsicum, yellow capsicum, broccoli and beetroot. Of the five vegetables used as samples in the study, the five vegetables contain carotenoids and can be used as natural dyes. Below is the beta-carotene content (mg / 100g sample): spinach 5.6, red capsicum 2.4, yellow capsicum 2.4, broccoli 1.3 and beet root 1.9²⁶.

Carrots

Carrots are one of the vegetables that have the best source of β -carotene, carotene content ranges from 60-120 mg / 100 g²⁷.

Chlorophyll

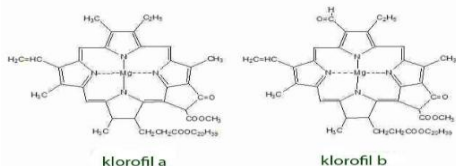


Figure 4: Chlorophyll structure²⁸

Higher plants have two types of chlorophyll, namely chlorophyll a (C₅₅H₇₂O₅N₄Mg) which is dark green and chlorophyll b (C₅₅H₇₀O₆N₄Mg) which is light green. Chlorophyll a and chlorophyll b absorb light most strongly in the red (600–700 nm) and the least in green (500–600

nm)²⁸. The following are examples of plants that contain chlorophyll:

Suji leaves

Natural dyes can be made into natural dyes that are more practical, namely by making it with prepared powder. The natural dye powder was made using suji leaf samples; suji leaves contain chlorophyll which is unstable²⁹.

Seaweed

Seaweed contains photosynthetic pigments such as chlorophyll a and c, carotene and fucoxanthin³⁰. Seaweed can be used as a natural dye, natural dyes from seaweed can be stored and would be stable for 6 days³¹.

Kale leaves

Kale leaves contain chlorophyll pigment which can be used as a natural colorant for food, but its use is less practical because kale leaves have a very thick material³².

Mango leaves

Mango is a family of *Anacardiaceae* which contains chlorophyll in its leaves and can be used as a natural dye³³.

Tannin

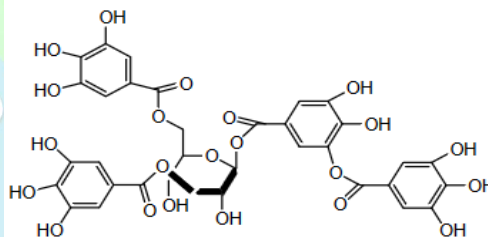


Figure 5: Structure of tannin³⁴

Tannin is a polyphenol compound with a very large molecular weight of more than 1000g / mol and can form complex compounds with protein³⁴. The following are examples of plants containing tannins:

Jamblang bark

One part of the plant containing tannins is the bark of the stem, the bark of jamblang (*Syzygiumcumini*) produces a brown color from tannin pigments³⁵.

Tamarind Seed Peel

Tamarin seed Peel which has a dark color and contains tannins in it, the tannin content is known after extracting the tamarind seed coat³⁶.

Soga

Soga (*Peltophorumpterocarpum*) is a tree that has yellow flowers and is usually used as a natural dye in clothes, using the sogu plant to enhance the color of silk threads. The part of the plant that contains tannins is the bark of sogu³⁷.

Star fruit leaves

Research on star fruit leaves identify the presence of tannin compounds, the results of these studies stated that star fruit leaves contain tannins and have other health functions such

as treating diarrhea, hemorrhoids and stopping inflammation³⁸.

Table 1: examples of some natural dyes obtained from vegetables^{4,39, 40}.

| Parts of plants | Produced by |
|------------------|--|
| Root | Turmeric, Onion, Beetroot Root |
| Bark / Branches | Purple bark, Safan wood, Sandalwood |
| Leaves | Henna, Eucalyptus, Tea, Cardamon |
| Flowers (petals) | Dahlia, hibiscus, rosella, roses |
| Fruits / seeds | Pomegranate peels, beetles, dragon fruit peels |

RESULT

DATA COLLECTION

To achieve the objectives of this review article, the authors used literature study techniques relevant to research. The way this was done was by looking for sources or literature in the form of international journals and national journals in the last 10 years (2010-2020).

The method used is the method of comparing data from one or several types of plants that is used as natural coloring additives in pharmaceutical products such as food, beverages, cosmetics and medicines.

Table: 2 Pharmaceutical, Food, And Drink Products Using Natural Dyes

| Authors | Plant | Methods | Results |
|---------------------------------|---|--|---|
| Handarini, K. ⁴¹ | Rosella flowers | Extraction of rosella flower and application of rosella flower extract into products | Addition of dye on rosella jelly on day 0 and A3 treatment showed the results of 5.8. This is because it gives a better color, namely purplish red and is much liked by many people |
| Rachmawati et al. ⁴² | Cassava leaves, kale leaves, grass jelly leaves, pandanus leaves and suji leaves. | To determine the chlorophyll content in the sample that was carried out by using visible light spectrophotometry methods at 646 and 663 wavelengths (samples were made into powder). The powder was made with variations in the concentration of 3% and 5% maltodextrin. | Natural dye powder is applied to agar food, and the result is a darker green color in the powder with 3% maltodextrin added, this is because the chlorophyll content in the powder added with 3% maltodextrin is greater than 5%. |
| Ekawati et al. ⁴³ | Dragon fruit peel | The method used was the Completely Randomized Design (CRD) method with 2 factors, where the first factor determined the concentration of dragon fruit extract and the second factor was soy milk and coconut milk. | Parameters include color, taste, aroma and overall preference level of soy milk and coconut milk. The results of the analysis of diversity showed no significant effect on color, taste and aroma, and it had overall liking levels, respectively, 5 (somewhat like), 4 (neutral) and 4 (neutral). |
| Atma, Y. ⁴⁴ | Angkak seeds | The method was carried out using a completely randomized design (CRD) with a treatment factor of adding angkak that was 5 levels and carried out 4 times so that the experimental unit was obtained 20 times. Color and organoleptic characteristics were observed. | The addition of coloring from Angkak affects the color intensity and texture of beef sausage but does not affect the taste and aroma. The greater the concentration of Angkak that was added, the red color intensity of the sausage was also higher. |

Table: 2 Cosmetic Pharmaceutical Products Using Natural Dyes

| Authors | Plant | Method | Result |
|---|--|---|--|
| 1. Marlina, L et al. ¹⁰ 2. Dwicahyani et al. ⁴⁵ 3. Yulyuswarni. ⁴⁶ | 1. Cocoa beans 2. Ruruhi fruit peels 3. Red dragon fruit peels | 1. Maceration method by using 96% ethanol and the addition of citric acid. 2. Maceration method by using 96% ethanol and 1% citric acid as solvent. 3. Maceration method by using 96% ethanol for 3 days and with the aid of a rotary evaporator to obtain a thick extract. | 1. The lipstick product has a melting point of 58-60 °C, is stable and easy to apply 2. The colors produced in this lipstick preparation are pink and dark red, with a pH of 3-4, homogeneous, sticky color (storage temperature 25 °C). 3. The color produced on the lipstick is light brown to dark brown with a semi-solid consistency that tends to be hard, the formula is not completely homogeneous, melting point of 53 °C -55.3 °C. |
| 1. Supomo et al. ⁴⁷ 2. Iqbal, Z et al. ⁴⁸ | 1. Secang wood 2. Almond shell, walnut shell, amla, soapberry | 1. Experimental method with variations in the concentration of secang wood infusion and the | 1. The resulting hair dye is blonde brown to dark brown, if exposed to sunlight the hair color will change to darker and last up to 3 |

| | | | |
|--|---|--|---|
| | peels and henna | concentration of pyrogallol and copper (II) with aquadest solvent. 2. The peel of the sample is dried and mashed to obtain a powder and then stored in a desiccator then the powder is soaked and made with 3 formulas (H1, H2, H3) | washes after washing stability test. 2. From the three formulas, the color results are pale brown in H1, medium brown in H2 and dark brown in H3. This hair dye product can strengthen hair and remove dandruff. |
| Setiawati, E & Suharyani, I. ⁴⁹ | Dayak onions | Isolated by chopping and refining using a blender and then filtering it to get a thick red dye. Before that, the optimization of cera alba is made first and then the formulation is made. | The color produced from 3 different lip gloss formulations is 20% rose, 35% brick red and 50% nude. After doing the tests on the three formulas, the results are: formula 1 "was liked" as much as 48%, formula 2 "was like" as much as 44% and formula 3 "was liked" as much as 44% |
| 1. Bindharawati, N et al. ⁵⁰ 2. Fitri, A, Z et al. ⁵¹ | 1. Rosella flower petals 2. Secang stem bark | 1. Maceration method with 96% ethanol and the addition of 3% glacial acetic acid and then evaporated. Compact powder cheek milk is made by pressing dry. The addition of isopropyl myristate as a binder with various concentrations (formula I, II and III). 2. The extraction method uses the maceration method with 70% ethanol. The data analysis technique uses primary data. | 1. Shows the results at a concentration of isopropyl myristate of 0.5% to be more brittle and easier to break. For the pH test formulas, I and II have met the requirements, and the topical test for compact powder blush has not met the requirements because the preparations cannot be attached to the skin. However, the preparation that fulfills all the tests is in formula II. 2. Based on the organoleptic test for one month there was no change in color and good stability, on the homogeneity test and on the polishing test there were no coarse grains and evenly colored, pH ranges from 6-7. |

Table: 3 Pharmaceutical Medicines Products Using Natural Dyes

| Authors | Plants | Method | Results |
|---|---------------------------------|---|--|
| Pratimasari, D & Lindawati, Y, N. ⁵² | Bunga Telang (Blue Pea flowers) | Extraction of blue pea flowers using the maceration method with ethanol solvent. Paracetamol syrup was made with 4 formulations and, there were 3 increasing concentrations at F1, F2, F3 (0.25%, 0.5%) and 1.0% in F4 added with synthetic food coloring (as a control). | The color produced in paracetamol syrup is purple which varies from light purple to dark purple, the difference in color is due to differences in concentration. The best color stability is at a concentration of 0.25% and 1% |
| Asra, R et al. ⁵³ | Dragon fruit peel | Extraction process of dragon fruit peel uses water as a solvent that has been sonicated at 50kHz for 30 minutes at 25 °C then freeze-dried for 48 hours. The extract was applied to paracetamol tablets using the direct pressing method and the color stability was evaluated over a range of 0, 1, 2 months | The betacyanin pigment produced from the peel of red dragon fruit was successfully applied to paracetamol tablets, the color produced was pink. The shelf life of the tablet for 3 months did not show a significant color change on the tablet. |

DISCUSSION

Nowadays, natural dyes have been widely used by people throughout the country, environmental concerns in the production and application of synthetic dyes have revived consumer interest in natural dyes⁵⁴. In the pharmaceutical field, there are many products that add dyes, ranging from food, beverages, cosmetics and medicines. There needs to be an education to the public, given the number of irresponsible people using dangerous dyes because they are pursuing profit⁵⁵.

The food and beverage products section have summarized some of the results of several researchers who used natural dyes, the selected natural dyes are rosella flowers, cassava

leaves, kale leaves, grass jelly leaves, pandan leaves, suji leaves, dragon fruit peels and angkak seeds. These plants produce various types of colors that are used to food, foods that use natural dyes get stable results on the product, the dyes do not affect the taste and aroma.

Cosmetic products add dyes to increase its attractiveness to customers, and enhance appearance when used, because color plays an important role in the marketing of cosmetic products⁵⁶. There are so many plants that have benefits as natural dyes. There are several cosmetic product preparations that have been tested by previous researchers such as lipsticks, hair dyes, lip gloss and blush compact powder. Researchers have succeeded in making lipsticks,

hair dyes and lip gloss with evenly colored results and can be applied well. Lipstick preparations are said to have goods readability if they provide an intensive, even and homogeneous when applied⁵⁷, as in the table of results of lipstick preparations above, however, the blush product is more fragile if the addition of 0.5% isopropyl myristate is added. It will meet the requirements for good friability if 0.75% isopropyl myristate is added, the difference in concentration will affect the brittleness of the roselle extract preparations. There are still many cosmetic preparations that use synthetic dyes, such as the Rhodamine B dye which is still found in various cosmetics. Rhodamine B is a synthetic dye which is prohibited from being used in cosmetics⁵⁸, so now people must be smart in choosing the cosmetics they use.

Dyes are used to give different appearance to pharmaceutical preparations, the types of formulations that must be colored are tablets, tablet coatings, capsules, oral fluids, toothpaste and ointments⁵⁹. Based on the results, the researchers succeeded in applying natural dyes to medicines such as tablets and paracetamol syrup with plant sources from telang (blue pea) flowers and dragon fruit peel. Adding color to tablet and syrup preparations is done to provide an attractive color so that it can increase attractiveness to consumers. The color produced in the paracetamol syrup from the telang flower extract is purple

with varying intensities, the difference in color intensity is caused by differences in the concentration of telang flower extract⁵², while the paracetamol tablets produce a pink color, pink color is produced from the pigment. betacyanin contained in dragon fruit peel. The results of the researchers indicated that betacyanin can be used as a coloring agent in pharmaceutical preparations⁵³.

Various methods can be used to get the extraction results from natural plant dyes⁶⁰, the researchers who have been summarized in the result table on average were using the maceration method. Maceration is a simple filter that is done by immersing the sample⁶¹. Natural dyes certainly have advantages and disadvantages, the advantages of natural dyes are as follows: environmentally friendly⁶², less polluting, non-toxic, non-allergic, non-carcinogenic and easy to make⁶³. As for the drawbacks of natural dyes: natural dyes require large quantities, natural dyes are more expensive if the users do not own the plants⁶⁴.

CONCLUSIONS

Multiple pharmaceutical products have switched to natural dyes from synthetic dyes, natural dyes have been chosen because they are safe for use. Besides being safe to use, natural dyes have pharmacological properties that are beneficial to the body. The use of natural dyes does not affect the taste, aroma and texture of the products.

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