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EFFECTS OF SELECTED MORDANTS ON THE APPLICATION OF NATURAL DYE EXTRACTED FROM THE PODS OF *PITHECELLOBIUM JIRINGA SEEDS*

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Abstract:

Natural dyes can be extracted from various sources such as flowers, barks, leaves, roots, wood or with other biological sources like animals, lichens or fungi. The usage of natural dyes requires application of mordant in order to increase the affinity of the colour fastness between the dye and fabric, thus enhancing the colouration of the natural dye. The colouring potential of natural dyes extracted from the pods of Pithecellobium jiringa has been studied against selected mordants such as potassium dichromate and copper (II) sulphate. Dyeing procedure for the application of natural dye onto the fabrics were done by using three different mordanting techniques which are pre-mordanting, simultaneous mordanting and post mordanting respectively. Evaluation of the colour fastness on the dye properties was assessed by evaluating the K/S ratio, on the absorbance of wash off colour strength of dye from the cotton fabric. Copper (II) sulphate showed as the best mordant towards the natural dye extracted from the pods of P. jiringa with the least value of K/S ratio 2.8×10^{-6} *that indicates low degree of dye bleeding from fabric, while the highest value for K/S ratio is* 1.08×10^{-6} *was observed with simultaneous mordanting of potassium dichromate. Thus, the present study proves that the pods dye extract of P.jiringa can be used as potential natural dye source and creates awareness on the effective usage of plant wastes in the related industries.*

Keywords: *Pithecellobium jiringa, mordanting technique, colour fastness, K/S ratio.*

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INTRODUCTION:

Dye or colorant was widely used in cosmetics, textiles, pharmaceuticals, photographic, plastics and paper industries. Dyes can be classified into two type's namely synthetic dyes and natural dyes. Synthetic dyes are basically derived from petroleum compounds, while natural dyes were obtained from natural sources such as plants, animals and minerals. Natural dye is a substance that is capable of adding or improving colour of the selected materials which was obtained from natural sources [1]. Approximately more than 10,000 different dyes and pigments were utilized in industry and there are over 7 x 10^5 tons production of synthetically-manufactured dyes annually in worldwide [2].

Natural dyes are recognized to be safe as they are non-allergic, non-toxic and are biodegraded in nature. Most of the plants used for extraction of dye are categorized as medicinal plants and therefore it was believed to impart some significant antimicrobial properties [3].

Biological oxygen demand (BOD) value produced by effluents of natural dyes is between 40-85mg/L which does not exceed the limit value of 100mg/L as supported by Central Pollution Control Board (CPCB), Government of India. The level reported for total suspended solid (TSS) and total dissolve solid (TDS) value were also does not exceed the levels as reported from many synthetic dye molecule. Unlike synthetic dyes, they are invented with good resistance to the activity of water and light thus making the dye molecules to be difficult to degrade. The contamination impact to the environment is extremely huge as the effluent containing residue of synthetic dye was hardly treated [4].

In this research, the pods of *Pithecellobium jiringa* had been chosen in order to prove its dyeing properties against the selected mordants. Traditionally, this plant can be used as natural dyes, pesticides and potential agent as antitumor and anticancer of ovary.*P. jiringa* is a plant belongs to the family Leguminosae, a traditional medicinal plant that is native to Southeast Asia [5]. It is known as "Dogfruit" in English and Malaysian recognised it as "Jering" and "Djengkol" in Indonesia. It usually grows up to 25 meter height and can be widely found in tropical countries such as Malaysia, Thailand, Bangladesh, Indonesia and Myanmyar [6]. The ethnobotanical uses of *P. jiringa* was believed to have detoxing blood activity, hypoglycemic property and its seed can be used to facilitate urination [7]. *P. jiringa* peels also proved as a natural dye in food colourings [8].

Natural dyes are known for their utilization in food colouring, natural fibres like silk, wool and cotton since prehistoric times. Historically, Mare Nostum was known as a home for the first universal dye, Tyrian purple that was produced by Phoenicians [9]. The development of natural dye had introduced other colours such as anthraquinone-based chromophores for reds, 6,6'-dibromoindigo for purple and indigo for blues. In addition, dyes for purple, red, blue and yellow can also produce diverse colours and shades such as orange and green [10].

Natural dyes have a broad classification. The first one is traditional dyes that consist most of the natural dyes potential sources that depends upon the development of metal-fiber complex to enhance the dyeing properties of the dyes to the fabric. Secondly is substantive dyes, which have ability to dye the fabric without requiring any mordant in its dyeing process. Lastly is the vat-dyes that are known to be water-insoluble that require the treatment with sodium hydrosulphite and alkali to make it soluble and stick into the fabric [11].

The usage of natural dyes helps to gain carbon credit by lessening the utilization of fossil fuel based synthetic dyes. Natural dyes are also proven safe for skin as they are anti-allergen and harmless to mankind wellbeing. Moreover, natural dyes are capable to replace synthetic dyes in children garments and food-stuff as they are believed to be moth proof and safe to be used. In spite of these advantages, natural dyes do convey a few intrinsic drawback that caused for the decline of the ancient art of dyeing textiles [12].

Uses of natural dyes will cause colours to wash out easily thus require mordants to make the colour more permanent to the fabric. Vegetables fibers like cotton and linen are not capable of holding the dyes which results in dull shades compared to wool and silk that are able to stain the bright colors. Mordant are metal salts that have potential to provide chemical bond between natural dyes and fabric thus improving the colour fastness of the dyed fabrics [13].

The mordants were broadly classified into three types namely oil mordant, metallic mordant and tannins. Example of oil mordants is castor and sesame oils that were used previously to combine with madder to produce the Turkey red colour. Metallic mordants are chromium, tin, salts of aluminum, copper, and iron. Tannin mordants can be in form of tannic acid and vegetable tannins such as oak galls, sumac or myrobolan, which are cheaper and can be found in plant parts such as leaves, bark and fruits.Tin, iron and aluminium have been selected as potential mordants that cause no harm towards the environment and human health [14].The various colours can be obtained from different mordants from the same source, i.e *Opuntia Lasiacantha* Pfeiffer in which yellow brown can be obtained from ferrous sulphate, green colour can be obtained copper sulphate, olive green can be obtained from potassium dichromate and red colour can be obtained from tannic acid mordant [15].

The usage of synthetic dyes has raised many environmental pollution and human health hazards. Therefore, interest in green technologies has restored the thought to the usage of natural dyes that are safer in health point of view as there is an increase in environment consciousness. Thus, potential natural dye have been focused on the pods extract of *P. jiringa* where by it increases its visibility in the future research community as well increases the commercial value of this plant in the dye industry.

MATERIAL AND METHODS:

Raw material collection

A matured seed (4 kg) of P. jiringa (Fig.1) was collected from Pekan Jitra market, Kedah and was authenticated at Kompleks Pertaninan Bumbumg Lima, Seberang Perai, Pulau Pinang by Mr. Suhaimi b. Hj. Din, from Plant Biosecurity Department Pulau Penang. The seeds were washed with tap water to remove the residues. The washed seeds were wiped off to remove excess moisture and broken to collect the pods for further study. The pods were shade-dried for 3 days, followed by drying in a hot air oven at 40^{O}C for 48 hours. The dried pods was subjected to grinding to obtain as coarse powder, weighed and stored in air tight container for further process.

Fig 1: matured seeds of *P. jiringa*

Dye Extraction

The pods coarse powder (300 g) was packed in soxhlet column and extracted with ethyl acetate as solvent using continuous hot extraction method at controlled temperature for 48 hours. Upon complete extraction, the excess solvent was evaporated using rotary evaporator at controlled temperature and pressure. The obtained concentrated dye extract was collected, weighed, packed in a glass container and stored in refrigerator until further usage.

Dyeing Procedure

Wash-off cotton fabric:

Pure cotton cloth pieces were washed with a mixture 2 g of detergent and 0.5 g/l sodium carbonate solution at 50^{O}C for 25 minutes. The scoured cotton fabric pieces were further subjected for rinsing with distilled water and allowed to dry at room temperature for overnight.

<u>Mordant application:</u>

Pre-mordanting technique

The fabric was treated with mordant before dying. 2 g of respective mordant, potassium dichromate and copper (II) sulphate was dissolved in 500ml distilled water. The scoured cotton sample was heated for 50 minutes at temperature monitored between 50^{O} C - 60^{O} C. The pre-mordanted cotton were allowed to dry without rinsing or washing. After the mordanted fabric have dried completely, the fabric was put in dyebath solution.

Simultaneous mordanting technique

The respective solid mordant potassium dichromate and copper (II) sulphate were added into the dye bath solution in separate beaker. The dried scoured cotton fabric was placed in the mixture and allowed to simmer at monitored temperature range 50^{O} C - 60^{O} C for 50 minutes.

Post mordanting technique

The dried scoured cotton fabric was immersed in dyebath solution for certain time and followed by complete drying. The dried and dyed fabric was treated with mordant.

2 g of respective mordant, potassium dichromate and copper (II) sulphate was dissolved in 500 ml distilled water and heated for 50minutes at temperature between 50^{O} C - 60^{O} C.

Evaluation of Colour Fastness Properties

The dyed cotton fabrics were subjected for complete drying for seven days. All the cotton dyed fabric was washed in a solution of detergent with distilled water. The wash-offs of each sample was collected and was analysed using UV-Visible spectrophotometer in order to evaluate the dye fastness and to determine the degree of dye bleeding from the fabric.

The wash-off's sample was determined by the percentage of light reflected (R) from the absorbance value (A) and percent transmitted (T) by using equation (1) mentioned below.

$$R= 100 – (A-T) \quad (1)$$

K/S value ratio can be obtained from the theory of Kubelka-Monk to define the interconnection between spectral reflectance (R) of the wash-off of the sample, its light absorption (K) and its scattering characteristic (S) using the equation (2) mentioned below.

$$K/S = (1-0.01R)^2/0.02R \quad (2)$$

The K/S ratio is an indicator of the colour strength of the wash-off in which the lower the value of this ratio indicates the lower degree of dye bleeding from the fabric.

RESULTS:

The percentage yield of the extract was found to be 5 %. A dark greenish colour was obtained upon treatment of *P. jiringa* pods extract with copper (II) sulphate as the mordant. Colour effects of copper (II) sulphate were shown in Fig. 2, 3, and 4. Post-mordanting technique with copper (II) sulphate showed the best colouration with the deepest shade of dark green colour compared to other two techniques of dyeing. Pre-mordanting technique showed moderate light green colour whereas simultaneous mordanting technique showed light green colouration. The intensity of of the colour observed in the cloth by the dye without using mordanting was less when compared to the intensity of colour obtained with the mordanting copper (II) sulphate in dye bath solution.

Fig. 2: Pre mordant with Copper sulphate

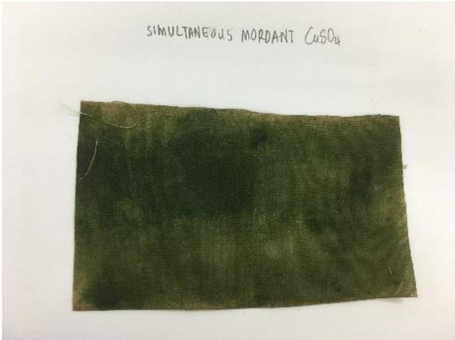

Fig.3: Simultaneous mordant with Copper sulphate

Fig. 4: Post mordant with copper sulphate

However, with potassium dichromate as mordant the colour obtained from dyeing with *P. jiringa* pods extract was greenish brown colour with different shades as can be seen in Fig. 5, 6 and 7. Based on the observation, pre-mordanting technique of copper (II) sulphate with *P. jiringa* pods extract showed the strong colouration, while simultaneous mordanting technique showed the weak colouration.

The detailed results for the *P. jiringa* pods dye extract on the application of cotton fabric using the selected mordants and different mordanting techniques was shown in Table 1 and Figure 8.

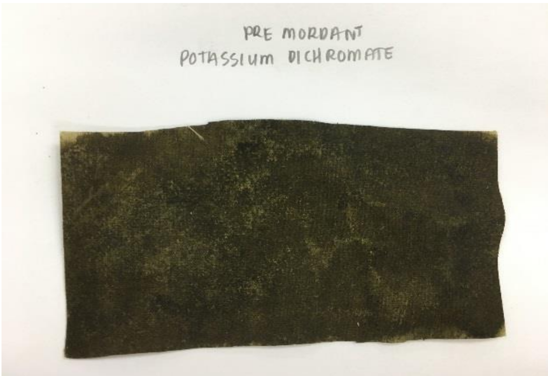

Fig 5: Pre mordant with potassium dichromate

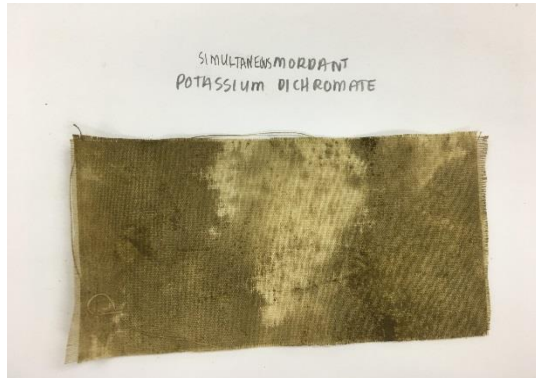

Fig 6: Simultaneous mordant with potassium dichromate

Fig 7: Post mordant with potassium dichromate

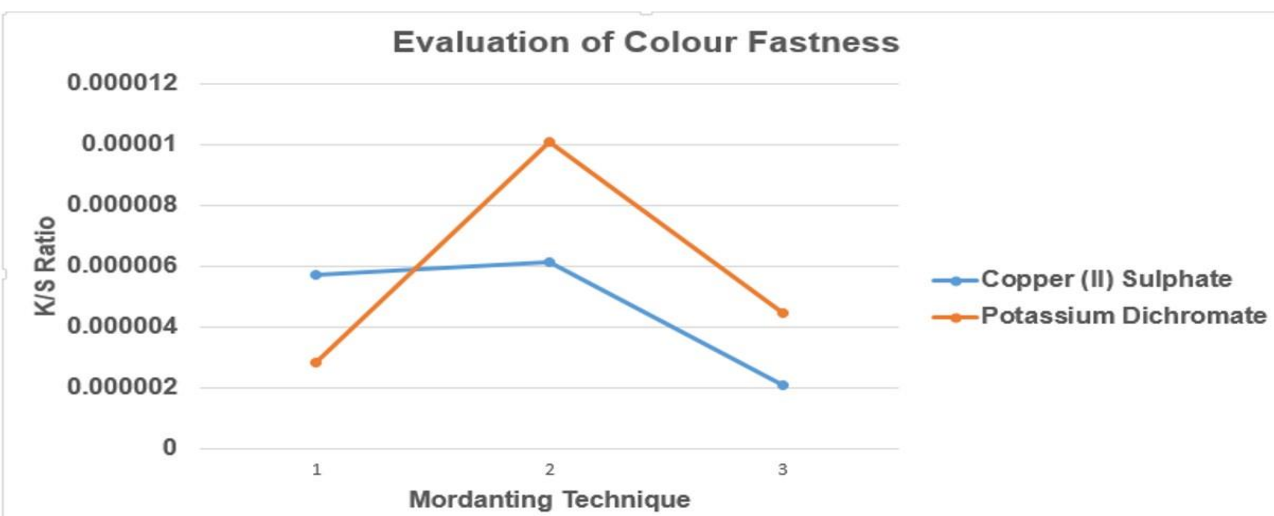

Fig. 8: Evaluation of colour fastness

Table 1: Result on the evaluation of colour fastness

Mordant	Mordanting Method	Blank (T)	Absorbance (A)	R = 100 - (A-T)	K/S Value
Copper (II) Sulphate	Pre-mordanting	0.402	0.740	99.662	0.00000573
	Simultaneous mordanting	0.402	0.829	99.573	0.00000612
	Post-mordanting	0.402	0.606	99.796	0.00000208
Potassium dichromate	Pre-mordanting	0.402	0.64	99.762	0.00000283
	Simultaneous mordanting	0.402	0.850	99.552	0.00001008
	Post-mordanting	0.402	0.70	99.702	0.00000448

DISCUSSION:

Pre-mordanting:

According to the present study, it was proved that application of *Pithecellobium jiringa* with pre mordanting technique indicates that potassium dichromate yield the best colour fastness with the K/S value 2.83 x 10^{-6}, while compared to pre-mordanting technique with copper (II) sulphate which displayed moderate colour fastness with K/S value 5.73 x 10^{-6} .

Simulatenous mordanting:

Potassium dichromate and copper (II) sulphate mordants exhibited poor colour fastness with simultaneous mordanting technique with K/S value 1.008 x 10^{-5} and 6.12 X 10^{-6} respectively. The outcome can be relate to the solubility of copper (II) sulphate and potassium dichromate in ethyl acetate solvent. Both these inorganic salts have lowest solubility in ethyl acetate.

Post mordanting:

Post-mordanting of copper (II) sulphate with *Pithecellobium jiringa* pods extract showed the best shade of colouration with K/S value of 2.08 x 10^{-6} while for potassium dichromate, the colour is quite pale with K/S value 4.48 x 10^{-6}. Thus the result indicates *Pithecellobium jiringa* pods dye extract can be best fixed by copper (II) sulphate as mordant.

Comparison of K/S value for the used mordants:

It can be observed that post-mordanting of *Pithecellobium jiringa* pods dye extract with copper (II) have the least wash off colour strength K/S value 2.08 x 10^{-6} which consequently makes it the best out of other technique and mordants. Generally, simultaneous mordanting with potassium dichromate gave the worst result as it showed the highest value (1.008 2.08 x 10^{-5}) for wash off colour fastness. The greater the value for wash off colour strength, the greater the degree of dye bleeding from the fabrics.

CONCLUSION:

P. jiringa is commonly known as “jering” in Malaysia and widely used as traditional medicinal plant in which most of plant parts have their respective uses. The cooked seeds of *P. jiringa* is commonly used as “ulam” and normally consumed with rice. However, the seeds can also be taken rawly as an antidiabetic agent and also to help purify the blood. In addition, in recent days *P. jiringa* seeds draws its application as organic insecticide due to the presence of djenkolic acid. The present study revealed the potency of *P. jiringa* pods extract to be a new source of natural dye material based on its application on pure cotton cloth with two selected mordants, copper (II) sulphate and potassium dichromate. Copper (II) sulphate showed as the best mordant towards the natural dye extracted from the pods of *P. jiringa*, when compared to potassium dichromate as the mordant. The demand in green technologies of dyeing industry has increase the interest in exploiting new source of natural dye material. Thus the study concludes the pods of *P. jiringa* can be used as a potential source of natural dye where by it increases the commercial value of this plant in the dye industry also will draw the attention of the future research community to explore more on this plant.

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