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Extraction, Characterization and Application of Natural Dyes from the Fresh Rind of Index Colour 5 Mangosteen (*Garcinia mangostana* L.)

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***Abstract*—**This study was to explore and utilize the fresh rind of mangosteen *Index Colour 5* as an upcoming raw material for the production of natural dyes. Rind from the fresh mangosteen *Index Colour 5* was utilized to extract the dyes. The established extracts were experimented on silk fabrics via three types of mordanting and dyeing procedures; pre-mordanting, simultaneous mordanting and post-mordanting. As a result, the applications of the freeze-drying methodology and mechanizable equipment have helped to produce excellent range of natural colours. Silk fabric treated simultaneously with mordanting and dyeing with extract dye *Index Colour 5* produced a brilliant shade of the red colour and the colour from this index is also discovered sensitive to light and washing during the fastness tests. The preliminary evaluation and instrumentation analysis allowed us to examine whether the application of different mordanting and dyeing procedures with the same extract samples and concentrations affected the colours and shades of the fabric samples.

***Keywords*—**Natural dye, Freeze-drying, Garcinia mangostana Linn, Mordanting.

I. Introduction

As people become more conscious of the environmental effects brought about by the use of synthetic dyes in textile products, dye compounds extracted from natural resources; mainly plants are becoming important alternatives to be used in the textile industry [1]. Natural dyes have been found biodegradable, non-toxic and generally have higher compatibility with the environment as compared to the synthetic dyes [2]. Hence, varieties of plant origins which focuses on the regeneration of waste and environment-friendly materials was recognized and applied in many textile products [3]. Colours that are achieved are mostly in pastel shades and commonly considered as soothing and pleasing to the eyes [4].

Mangosteen, tumeric, *rambutan* skins and mango seeds are among the plant origin materials utilized in the manufacture of natural dyes in Malaysia [5]. There are many other types of plants that could yield dye were discovered growing throughout the year in Malaysia. The suitable climate here characterized with its annual rainfall that exceeds the rate of evaporation and temperature has created suitable soil formations for these plants to grow. The heavy and scattered rainfall of 2500 mm a year has resulted in the abundant growth of various types of plant species. Nevertheless, being located at 23.5ºN and 23.5ºS of the equator, this country experiences two seasons; the hot, dry season and the wet, monsoon season. The dry season in the northern states of Peninsular Malaysia takes place from November to February, while the wet season in the east coast areas is from November to December. This climatic condition is an added advantage for the locals to utilize fully the resources that are made available [6].

The purpose of this study was to explore and utilize the fresh rind of mangosteens as an upcoming raw material for the production of natural dyes. Only the rind of the mangosteen is used to extract natural dye. The discarded rind contains rich *anthocyanins* that produce the impressive red colour extraction. The red pigment can also be associated with the benefits of reducing health hazards, lowering toxicity levels and avoiding allergic reactions [7]. Such waste can be recycled and reused to earn its place in our textile industry.

II. Materials and Method

A. Material

There are six colour scales indexes developed by Malaysian Agriculture Research Development Institute (MARDI) to determine the level of ripeness of the fruit. The material of this study is based on selected mangosteens from *Index Colour 5* as the fruit from this index is considered the most suitable to be adopted for the research [8]. The rinds contained a high level of major and minor anthocyanin pigments which are Cyanidin-3-sophoroside and cyanidin-3-glucoside [9].

Mangosteen with *Index Colour of 5*, from Pahang was selected due to the fact that this region is well known for producing abundant and good quality of mangosteens. With adequate annual rainfall that promotes fertile soil for healthy growing, mangosteen produced from the area are of excellent quality.

The selection of mangosteens was based on three important criteria: 1) the quality (physical characteristics); 2) the quantity of the similar quality of fruits and the time and; 3) condition of the fruits from when they were originally collected until they are brought to the research area. The good quality fruit, in large quantities as shown in Fig. 1; must be considered to ensure that only the consistent amounts of fresh fruits are used for each experiment. The amount of time for the mangosteens to be transported to the research area from the time they are picked from the orchard or plantation is also an important concern to ensure the high level of anthocyanin content of the fruits.

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Fig. 1 Selected mangosteens from *Index Colour 5*

B. Extract Preparation

The rinds were cut into small pieces, weighed and then ground using a heavy duty juice extractor at a speed of 3000 rpm. The ground rinds were collected and added with mixture of distilled water (pH 6) and ascorbic acid with a concentration of 0.5%, 1.0%, 1.5% and 2.0% [10]. The processing of the fruits was expedited further by the use of a heavy duty juice extractor to grind the hard rind into small particles and anti-browning agent to preserve the colour. They were mixed with water and filtered to obtain the aqueous solutions (see Fig. 2). The samples were then placed into a plastic container and kept in a freezer at -20ºC for 6 hours [11].

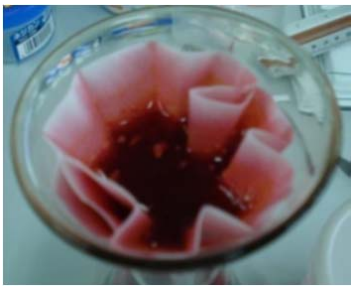

Fig. 2 A filtration process to obtain the extracts from Index Colour 5

C. Freeze-Drying

The frozen labeled samples from *Index Colour 5* were taken for freeze-drying by using a freeze dryer Heto Lab Equipment FD4. Each sample was placed one at a time into the freeze dryer at -45ºC, pressure 0.036 mB for primary drying [12]. After the completion of the primary drying process, the temperature and pressure were set at -29ºC and 0.010 mB respectively for the secondary drying.

D. Dyeing Procedure

All the silk fabric samples were treated using three different mordanting (pre-mordanting, simultaneous mordanting and post-mordanting) methods to fix the freeze-dried mangosteen extract dye onto the natural fibre, thus establishing the best range of shades for further analysis. Table I specifies the conditions for its preparation.

E. Colour Measurement and Characterization

Analysis of shades and colour of the fabric samples has been undertaken to confirm the changes that occurred due to the different methods of mordanting and dyeing procedures. The testing and analysis include the Colour Spectrophotometer Test as shown in Fig. 3; to measure the value of colours on each fabric sample and the difference in colour between samples and the Lightfastness Test and Washfastness Test to measure the resistance of the dye to fading when exposed to light and washing. The colour difference in each coordinate is presented as ΔL*, Δa* and Δb*. Whereas the difference in coordinates in the CIE L*a*b* space can be resolved by indicating the magnitude and direction of the colour difference between the two colours; 1 and 2 [13]. In assessing the change of colour in fastness testing, the procedures were carried out according to the standard methods of the ISO or International Standards Organization or the equivalent BS or British Standard for the lightfastness and ISO 3/MS 308 or the Malaysian Standard for washfastness.

TABLE I
THE FORMULATION FOR MORDANTING SILK (SAMPLES)

Preparation	Condition
Material-to-liquor	1:100
Alum (Salt Mordant)	30% of the weight of the fabric's sample
Temperature	Room temperature
Weight of the fabric's sample	0.25 gm (25 cm2)

Fig. 3 The coordinates in the CIELAB or CIE *L*a*b**

III. RESULTS AND DISCUSSIONS

The process to extract the anthocyanins from mangosteen rind *Index Colour 5* were carried out and it has been proven that anthocyanins exist almost exclusively in the rind of mangosteens, though in variations in terms of the appearance of the colours. The results showed that anthocyanins (pinkish-red) shades existed in this index's fruit.

The aqueous solution shades obtained from *Index Colour 5* were in pinkish-red shades. The addition of ascorbic acid had made the anthocyanins more stable in the aqueous solution extracts by indicating no appearance of grey or brown colour effects. More latex had accumulated from the mangosteen rind sample *Index Colour 5* after the precipitation procedures.

Visual evaluation from the colours and range of shades established, showed that variables such as mordanting and dyeing procedures, dye extract samples and concentrations had significantly affected the colour changes that occurred. The colour and shade of the silk fabric sample dyed with the extract of *Index Colour 5* which was experimented on through the pre-mordanting and dyeing method showed the deepest brown shade (see Fig. 4).

Fig. 4 Colour of a silk fabric sample dyed through pre-mordanting and dyeing procedure

Fig. 5 Colour of a silk fabric sample dyed through simultaneous mordanting and dyeing procedures

Fig. 6 Colour of a silk fabric sample dyed through post-mordanting and dyeing procedures

Fabric sample which was experimented on through simultaneous mordanting and dyeing appeared with an excellent depth and shade (see Fig. 5). Indeed, a report by Mussak and Bechtold [14] indicated that simultaneous mordanting and dyeing procedure has an advantage over the two other methods due to only one mordant and dye bath present throughout the procedure which enables the stabilization of the solution.

The shade obtained from the silk fabric sample of the post-mordanting and dyeing procedures as shown in Fig. 6, appeared in dull shade after it was completely dried. This outcome can be suggested due to the instability of the colour attachment on the fabric sample when two separate dye-bath solutions were required to conduct the procedures.

The CIE *L*a*b** measurement of values obtained from the fabric samples dyed with the extracts of *Index Colour 5* is shown in Table II. Each of these recorded values was based on the normal condition of the samples. The *L** values for samples dyed in the extract *Index Colour 5* showed that the samples D1 and D3 appeared in the coordinate of gray to white as compared to the sample D2 which appeared in darker and deeper shades. This is to confirm that there was maximum absorption of the dye under the simultaneous mordanting method for the sample D2, which was dyed with extract *Index Colour 5*.

TABLE II
THE CIE *L*A*B** VALUE OF SILK FABRIC SAMPLES DYED IN THE EXTRACTED SOLUTION OBTAINED FROM THE FRESH RIND OF THE MANGOSTEEN INDEX COLOUR 5 (*EXC*) AT THE NORMAL CONDITION (BEFORE EXPOSURE TO LIGHT)

Silk Samples	Procedures	CIE		
		*L**	*a**	*b**
Control	No Mordant	74.93	4.37	13.56
D1	Pre-mordanting and Dyeing	54.69	11.05	23.67
D2	Simultaneous mordanting and Dyeing	46.31	12.92	16.88
D3	Post-mordanting and Dyeing	64.72	10.31	17.69

The CIE *L*a*b** measurement of values obtained from the fabric samples dyed with the extracts of Index Colour 5 (*ExC*) after being exposed for 120 hours under artificial light. Samples *D1* and *D3* represent the higher and highest *L** values when dyed with Index Colour 5 (*ExC)*. Meanwhile sample *D2* showed the lower *L** value and it exhibited the deeper and darker shade of colours.

IV. CONCLUSION

The efforts to rediscover alternative red dyes from Malaysia's very own natural resources, namely from plant origins, has already been conducted. The established description of the index becomes a guideline to determine the suitability of the fruit to be utilized. The consistent nature of the extracted materials is very essential to eliminate any disparity of the resultant colour shades. The result suggested that besides the addition of supplementary material such as ascorbic acid during the experiments, the maturity level of the *Index Index Colour 5* fruits have greatly determined the shade of colours and the anthocyanin content. Within the parameters of the freeze-drying methodology, the extract obtained is produced at excellent condition.

By experimenting with silk as a textile dyeing sample, and alum as an agent to fix the colour to the fabrics, the dyeing abilities of the dyes were investigated in three stages of procedures; Pre-mordanting and dyeing, Simultaneous mordanting and dyeing and Post-mordanting and dyeing. The amount and concentration of dye extract and mordant were observed, the results achieved were visually evaluated in terms of colours and shades. The preliminary evaluation and instrumentation analysis allowed us to examine whether the application of different mordanting and dyeing procedures with different dye extract samples and concentrations affected the colours and shades of the fabric samples. All these processes will pave the way for more discoveries of new types of natural dyes which could be manufactured through systematic and well-structured experiments.

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