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CHARACTERISTICS OF SUBSTANCES EXTRACTED FROM LEAVES AND BARKS OF MATOA (POMETIA PINNATA L.) PLANTS AS RENEWABLE NATURAL PIGMENTS FOR COLORING BATIK FABRIC

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ABSTRACT

Matoa bark/leaves are abundantly available, but not yet utilized optimally. This Paper deals with characteristics of extractable substances from leaves and barks of matoa (Pometia pinnata L) plants, related to their uses for coloring batik fabrics. Bark/leaf extracts contained supposedly natural coloring-pigments (e.g., tannin, flavonoids, and chlorophyll). The main purposes of the research were mostly related to the extract characteristics by looking into color direction/orientation/appearance of leaf/bark extracts, after fixative treatment (using tunjung/Fe2[SO4]3), and extract uses in coloring batic fabrics; and by examining extracts and extract-colored fabrics, through e.g., GC-MS analysis, and color-related qualities (i.e., color-leaching/-ageing/ difference test and color visual-observation). Results revealed several significant differences occurred appreciably in characteristics between bark and leaf extracts, with respect to mainly extract yields, chemical composition (qualitatively/quantitatively), color direction/orientation/appearance, and color-leaching resistance. Judging from those already-examined characteristics, bark extracts seemed more promising as renewable natural pigments for coloring fabrics toward the successful creation of batik fabrics.

KEYWORDS: Natural Pigment, Tannin, Polyphenol Compounds, Chlorophyll.

1. INTRODUCTION

The development of small- and middle-scale industries of batik fabrics, which utilize renewable natural pigments in Indonesia, tends to increase nowadays. The trends toward natural lifestyles are viewed as safer, and the raw materials for natural pigments are abundantly available in Indonesia. Natural pigment substances, which could be obtained easily from the settlement vicinity, include animal manure, minerals, and plants that grow wild as well as cultivated. Relevantly, according to Pujilestari Titiek (2015), the pigments procured from nature or plants, either directly or indirectly, are present in their particular organs/parts, such as leaves, stems, barks, flowers, and roots, with their varying portions/contents as well as varying kinds of coloring matters. The coloring matters refer to the substances that could determine the color direction of natural pigments, which are inherently organic compounds, usually as extractable substances contained inside.

The extractable components are also present in colored woods, which include, among others, red-colored wood (Pterocarpus, Baphia, Caesalpinia pulcherima L, and Haematoxylon brasiletto), blue-colored wood (Haematoxylon campechianum), and yellow-colored wood (Indigofera tinctoria) due to containing flavonoids. Those flavonoid compounds are often found in the woods as colorless leuco compounds, and instead the flavonoid colors are generated due to the oxidation on haemotoxilin to become blue haematin and of brazilein to become red brazilein. Further, based on the contents of those compounds, wood extracts could be utilized as natural coloring-pigments (Nagoya, M. et al., 2013). Extracts from matoa woods using ethanol solvent qualitatively contained particular compounds such as flavonoids, tannins, terpenoids, and saponines. Matoa leaves, as biomass wastes from matoa trees growing in Papua, are also potentially able to produce natural coloring-pigments through the extraction process. Further, the utilization of wood wastes from nine tree species origins which grew in West Java and Riau islands could produce natural pigments with high color-leaching-resistances, categorized as good, and when used for coloring purposes, those pigments use alum ($Al2[SO4]3.18.H2O$) mordants and tunjung ($FeSO4$) fixatives (Saefudin, 2023).

Cellulose fibers from the plants, and protein fibers from animals (sheep wools and camel hairs) are potentially able to orient or direct the different colors in the application of natural pigments. In the application of cotton fabrics as well as silk fabrics, the orientation/direction of colors is affected by the material characteristics of those two fabrics. As such, the cotton fabrics could direct the colors toward stronger colors, while silk fabrics tend to direct the colors toward weaker colors. In addition, the extraction temperature could also affect the adsorption rate (as adsorbent) of natural coloring-pigments by those fibers (Satria Yudi & Seheryanto Dwi, 2016). The acidity or bacidity (pH) of the immersing solution could affect the color-ageing rates and color direction/orientation (Haerudin, 2017; Saefudin, 2023).

The use of biomass wastes generated from wood shavings as natural coloring pigments has been conducted by Haerudin & Farida (2017). In those researches the natural pigments were applied to batik fabrics, and as such, the brown color direction/orientation, as well as the color-leaching resistance, was regarded as fairly good. In this research, the dyes for coloring the fabrics were the extractable substances from barks as well as leaves of matoa (Pometia pinnata L) plants, further applied for coloring the blank white primisima fabrics toward the creation of batik fabrics. Further, in order to ensure successful results in coloring the batik fabrics using plant's biomass extracts as renewable natural pigments, it therefore neccessitates among other to know the extract characteristics. Relevantly, this research was mainly aimed (in relation to the characteristics of biomass extracts) to look-into the final color appearance as initially appeared from the leaf/bark-extracts, followed by fixative treatment, and their further uses in coloring batic fabrics; and also to examine qualities of the biomass extract as well as the colored fabrics after undergoing, e.g., the color-leaching tests. GC-MS analysis, extract yield-determination, color-ageing test, and color-differentiation tests. The more detailed related results are presented in the following.

2. RESEARCH METHODOLOGY

2.1. *Materials and Equipment*

The main raw materials used in these research activities covered, among others, biomass wastes of matoa leaves and of barks as by-products generated from wood-sawmilling industries in Bogor and Ciampea, West Java; and primisima (white-colored) fabrics as media for the coloring-application of those biomass wastes (bark/leaf extracts). Other assisting materials were, among others, fixative agents of tunjung ($FeSO4.7H2O$), alum ($AL2[SO4]3$), mordant, chemicals, teepol (a kind of soap), and distilled water.

Meanwhile, the main equipment consisted of an extractor apparatus equipped with a temperature-measuring device, stainless steel pans, gas stoves, pails, stirrers, immersing chests, strainers, beaker glasses, an appreciable universal pH meter, and a writing pen, as well as a stamping block (in preparing and creating the batic fabrics from primisima fabrics). Further, the testing instruments covered a GC-MS device, a UV-VIS spectrophotometer of Shimadzu (PC) S brand (for color difference test), and color-leaching tools, as well as color-ageing tools.

2.2. *Extraction of Matoa Wastes*

The extraction in this regard meant to remove the possible extractable compounds (including possibly natural coloring-pigments) from the biomass waste materials (i.e., matoa barks and leaves). For such, initially a representative sufficient amount of bark as well as leaf materials was prepared and then ground to small-sized solid powder. Further, as much as 1 kg of bark as well as of leaf powder (in dry-weight equivalent) was taken and then underwent the extraction separately by boiling the powder in 10 liters of water using an extraction apparatus at 80°C temperature for 3 hours. In this way, the possible natural coloring pigments would diffuse out of the solid powder, dissolving into the hot water. Afterwards, the hot water would turn into an aqueous colored liquid, and was then allowed to cool down. After cooling down, the aqueous colored liquid was strained using a cloth filter to separate the liquid from the extracted solid powder. In this way, the liquid that had passed through the filter would be obtained and then collected as liquid filtrate. The obtained liquid filtrate, supposedly containing natural pigments, further underwent evaporation under vacuum at approximately 60-70°C using a rotavapor device, which proceeded for a particular duration such that the filtrate evaporated completely to dryness.

The dry extract (from matoa bark as well as leaf powder), which adhered or stuck to the inside tube/flask of the rotavapor, was further dissolved in 100 grams (ml) of distilled water and then collected into the porcelain cup (with its already-known empty weight). The porcelain cup containing the aqueous solution of dry extract was weighed to obtain the weight of solution (Wa) **using the following formula**

Wa (g) = weight of porcelain cup (containing the solution) - weight of empty cup. [1]

After weighing, the solution of dry extract (from matoa barks as well as from matoa leaves) in the porcelain cup underwent evaporation to dryness under vacuum again at a mild temperature (60-70°C). After the vacuum-evaporation, the porcelain cup containing dry extract was weighed again to obtain the weight of dry extract (Wb) using the following formula

Wb (g) = weight of porcelain cup (containing dry extract) - weight of empty cup [2] Afterwards, the yield of dry extract (from matoa barks as well as leaves) could be calculated as follows:

Y (%) = (Wb/Wa) x 100 [3]

Where: Y = yield of dry extract (from matoa barks as well as leaves), Wa (g) = weight of solution (containing dry extract), Wb (g) = weight of dry extract (grams).

Further, the obtained dry extract (from matoa barks as well as leaves), supposedly containing natural pigments, was then ready for further application (i.e., coloring of primisima batics toward creating batik fabrics), and the subsequent testing (e.g., GC-MS analysis and color qualities / color-leaching test of pigmented fabrics).

2.3. *Data Analysis*

The data of the yield (Y) of coloring-extracts from the matoa bark as well as matoa leaves were analyzed using a completely randomized design (CRD) with the F-test, in a single factor (Ott, 1994). The factor was the origins of biomass extracts (i.e., matoa barks vs matoa leaves). The replication for each factor was replicated 5 times.

2.4. *Preparing and Creating Batik Fabrics*

The preparation/creating of batik fabrics was performed in stages. For such, initially the primisima white-colored (mori) fabrics were mordanted by immersing them in the alum ($Al_2[SO_4]_3$) solution, with the purpose to increase the affinity of fabric fibers to adsorb the dyes/pigments (i.e., natural coloring-pigments from matoa barks/leaves). The initial mordanting as such proceeded at 80°C for one hour. Afterwards, the mordanted fabrics were wetted through the immersion in the aqueous teepol/detergent emulsion (rather basic pH) to remove oil spots and other dirt on fiber surfaces. After immersion, the fabrics were washed and rinsed with clean water. Further, on one side of the fabric surface were drawn using a wax-filled pen to create particular various patterns of batik motifs.

After the drawing, the batik-patterned fabrics were immersed in an aqueous solution of matoa bark extracts as well as matoa leaf extracts (supposedly both containing natural coloring pigments) for 24 hours. Further, the pigment-colored fabrics underwent the fixation (or called final mordanting) by immersing them in the tunjung (FeSO4) fixative solution in three concentrations (i.e., 0, 10, and 20 g/L). For such, the immersion (fixation) lasted for about 5 minutes until the color appeared evenly on the fabric surface. The fixation proceded in five times or repeatedly until the basic colors of the fabrics seemed firmly fixed/adhered to the fabrics. Afterwards, the fixated fabrics were washed, dried at ambient/room temperature (under the roof), and then ended up with what was called the created batik fabrics, ready for the further testing of fabric-color qualities (e.g., GC-MS analysis, color difference, color-leaching resistance, and color ageing tests).

Colour Testing The color testing in this research was conducted on the white primisima fabrics after being colored by bark extracts as well as by leaf extracts (both as asserted before, allegedly both containing natural pigments) at various concentrations of tunjung/FeSO4 fixatives (0, 10, and 20 g/L). Also, the color testing was performed on the blank primisima fabrics, as a control (without coloring and without fixative treatment). The color testing in this research covered 3 (three) kinds of tests, i.e., color-leaching resistance, color difference, and color-ageing tests.

Color-Leaching test This test implied the resistance of colored-fabrics against the color-leaching actions by consecutively: ironing/rubbing, using AATCC 13 Crock Meter, and using Fastness Tester Meter (for-rubbing); by detergent washing, using Launder O Meter; by acidic sweat action; and by Xenon ray exposure test, using Fade O Meter-AATCC 15: 2009. The colored fabrics after the color-leaching tests and beforehand (untreated blank white primisima fabrics, as the control) were visually observed in subjective ways (e.g. the use of gray scales, color staining, and color changes/difference), using the following criteria (in classes and characteristics), with the aid of the so-called staining-scale standard, as follows: categories/classes 1 (very poor), 2 (poor), 3 (moderate/fair), 4 (good), and 5 (very good/best coloring-performance). In this research, the testing of color-leaching resistance was carried out by visually observing and assessing the possible changes in colors of the extract-colored fabrics (followed by the fixative/tunjung treatment), compared to the original color of the blank/control primisima fabrics, using the so-called staining-scale standard.

Color Difference Test In general, related to the colorimetric work, the color of a material according to the stipulation by the Commission Internationale de l'Eclairage system (CIE) could be defined in terms of three parameters, i.e., L*, a*, and b*. Those three parameters are measured using a color-measuring device (Lab Hunter, 2012). Further, from the reading on the color-gauge instrument, the color parameters/values: "L", "a", and "b". The L* parameter/value is associated with the reflection of light (brightness), which has the values of 0 / zero (black color direction) and 100 (white), revealing the color direction/orientation toward whiteness of a material. Meanwhile, the a* parameter alleges the chromatic colors of red-green mixture from 0 (green) to +100 (red) and from 0 (green) to -80 (strong green). Eventually, the b* parameter indicates the chromatic colors from 0 (blue-yellow mixture) to +80 (yellow) and from 0 (blue-yellow mixture) to -80 (yellow). In this research, the determination of the color values "L, a, and b" for each treatment level (i.e. utreated fabrics/control / without fixatives) vs colored materials, followed with consecutively 0, 10, and 20 g/L fixative concentrations/treatment was performed on 5 different location points on the surface of each of the fabric sheets (either colored/fixative-treated or uncolored/control). Further, the color difference between a standard (i.e., control) and a material of interest) In terms of a single value, ΔE was designated. Related to this research, the standard/control material was inherently blank white primissima fabrics (uncolored as well as without fixative treatment), while the material of interest implied the primissima fabrics after being colored with matoa bark/leaf extracts, followed by fixative (tunjung/FeSO4) treatment. Consequently, the ΔE discussed further would be associated with the research results. The ΔE values could be modelled mathematically (Akgul & Korkut, 2012) **using the following formula**

$$\Delta E = f(\Delta L, \Delta a, \Delta b) = ([\Delta L]2 + [\Delta a]2 + [\Delta b]2)0.5 \quad [4]$$

Where ΔE = color change value, and ΔL, Δa, and Δb = differences in consecutively L, a, and b values between the treatment levels. Further, from the obtained/calculated ΔE values, then could be determined or judged the classes of ΔE values, with the criteria as follows: classes: I ($\Delta E < 0.2$, invisible change), II ($0.2 < \Delta E < 2.0$, very small change), III ($2.0 < \Delta E < 3.0$, small changes [color changes visible by high-quality filter]), IV ($3.0 < \Delta E < 6.0$; medium [color changes visible by high-quality filter]), V($6.0 < \Delta E < 12.0$; big [distinct color changes]), and VI ($\Delta E > 12.0$, very different color).

Color Ageing Test In general, the color changes (ageing) in especially organic materials (e.g. cellulose-based fabrics) could happen due to storage, heat action, temperature of the surroundings, or exposure to light, including also the sunlight (Casey, 1980). In this research, the color ageing test was performed on the primisima fabrics (either uncolored/without fixative treatment or colored/fixative-treated), all after being stored for a particular duration (after the reversion). For such, it was by observing the K/S ratio and R values of the fabrics (either control or colored/fixated) after reversion. K refers to the light-absorption coefficient, S is light-sccattering coefficient, and R is the reflectance factor (Casey, 1980; Koch, 1995).

Visual Appearance The visual appearances of the fabrics colored with matoa bark as well as leaf extracts were observed subjectively on their surface using bare/unaided human eyes for specific matters, e.g., colors (strong /weak/direction/orientation/evenness) and other interesting/attractive visual features.

2.5. GC-MS Analysis

The GC-MS (gas chromatography – mass spectrometry) analysis used a device called the Shimadzu's PC-GC-MS-QP-2010. Further, with the GC-MS analysis, the kinds as well as proportions of possible fragmented extract compounds from the original substances in the extracts (from matoa barks as well as leaves), which might result from the high temperature imposed in the device during the GC-MS analysis. As such, in performing the GC-MS analysis (on e.g. bark and leaf extracts), high temperatures as asserted before were usually implemented in the GC-MS device/instrument, such as volatilization of the extract samples (i.e. ≥ 100°C), injection port set at high temperature (150- 170°C), separation column at 100- 160°C, and detector at 180-200°C. Accordingly, such harsh condition could cause the original chemical compounds in the examined biomass (including dry extracts from matoa barks as weel as from leaves), to partialy undergo e.g. depolymerization, degradation/fragmentation, and addition/condensation reaction, thereby leaving behind the sample partially with some fragmented extract substances and some other partly, i.e. the residual extract compounds allegedly still in intact/in original shapes (Browning, 1967; Lambert et al., 1976; Atkin, 1984).

3. RESULTS AND DISCUSSION

3.1. Yields of Extracts from Matoa Barks and Leaves

The data about the yield of biomass waste extract, supposedly containing natural-coloring pigments, from the matoa barks and leaves are presented in Table 1. On examining the CRD's F tests, it turned out that matoa bark exhibited significantly greater yield of colored extracts than matoa leaves; or in other words, the former contained more allegedly-colored extracts than the latter. The possible explanation is that the function of leaves is to serve as food-making organs, i.e., conducting photosynthesis that converts CO2 and H2O into simple sugar (glucose) as the basic photosynthetate. Further, in the leaves, the sugar undergoes assimilation and metabolism processes into the so-called primary metabolites, such as simple carbohydrates, amino acids, simple proteins, simple fats, and other simple compounds. Those primary metabolites from the leaves are subsequently transported and distributed throughout all the other plant bodies (e.g., roots, cambium, barks, parenchyma cells, and other living tissues). Meanwhile, the function of barks is also to transport the assimilates as well as the primary metabolites axially downward (to the roots) or upwards (to the apical meristem/growing buds at the stem ends) and laterally (to the cambium). In addition, inside the barks, those assimilates/primary metabolites sustain further complicated physiological processes into the so-called secondary metabolite products, such as complex sugars/fats/proteins, alkaloids, and glycosides; and consequently, the barks serve also as a storage organ for food reserves. Also, it is necessary to know that the function of bark is also to protect the inner organs/tissues (e.g., woods and cambiums) against the external extreme factors, such as sun heat, strong winds, harsh weathering, rain, and organism attacks. Consequently, the secondary metabolites produced/synthesized by the barks, besides complex foods, could also cover other intricate compounds such as polyphenols, tannins, flavonoids, tannins, sitosterol, terpenoids, phenylpropanoids, stillbenes, quinones, quinines, waxes, corks, and others, where some of them could impart specific colors as well as be toxic and provide physical barriers (Browning, 1967; Meyer et al., 1972; Sinnot & Wilson, 1975; Koch, 1995).

Table 1: Average Yield of the Extracts from the Matoa Barks and Leaves.

No	Biomass waste origins (of the extracts)	Weight of extract solution (g) 1)	Dry weight of extract (g) 1)	Yield of extract (%)1)
1	Matoa barks	10.49	0.198	2,02 2)
2	Matoa leaves	10.08	0.168	1.82 2)
Remarks: 1)Average of 5 replications; 2)significantly different, based on the CRD'a Anona (F-test)				

Judging from those facts, consequently, it is understandable that, as natural factors, the barks in general contain more of the extractable substances qualitatively as well as quantitatively than the leaves, where these phenomena likewise apply to the matoa barks/leaves (Table 1).

3.2. Results of GC-MS Analysis

GC-MS results on the dry extracts from matoa barks

The detailed chemical compounds in the bark extracts presumed as natural pigments that could be identified qualitatively/quantitatively with the GC-MS analysis, are as follows

Nickel tetracarbonyl, $Ni(CO)_4$; color: pale yellowish; solid crystal shape; quantity/portion (in total): 11.91% + 14.83% + 3.69% + 0.85% + 5.52% = 36.80%. $Ni(CO)_4$ compounds as detected by the GC-MS analysis were possiby formed from the reaction of Ni element (as one of the mineral elements in matoa barks) with the carbonyl (CO) groups that might result from the degraded hemicellulose as well as soluble / low-molecular weight (MW) lignin and other phenolic compounds in the bark extracts (Casey, 1980). In addition, CO could also be formed as carbon monoxide, which appeared from the breakage of formic acid ($HCOOH \rightarrow H_2O + CO$), while the HCOOH emerged due to the degradation of low-MW lignin and simple polyphenols (Sjostrom, 1982). Further, the CO could react with Ni metals to form colored $Ni(CO)_4$ as well. The presence of heavy metal ions (e.g., Fe^{+3}, Cu^{+2}, and Ni^{+2}) in the barks could impart colored complexes (chelations) with the bark's phenolic compounds (Smook, 2002). Consequently, the detected colored $Ni(CO)_4$ in the matoa barks could indicate the presence of phenolic compounds (e.g., gallic acids, ellagic acids, catechin, quercitin, tannin, flavonoids, lignans, phlobaphenes, quinines, and stilbenes), which contribute to the natural colors of bark extracts.

Carbon dioxide (CO_2; colorless gas, quantity/portion: 5.84%) could emerge as one of the degraded products of hemicellulose, pectic substances, and soluble/low MW lignin, as well as the low MW color-contributing simple-phenolic compounds in the matoa barks (Browning, 1967; Sjostrom, 1982). In addition, CO_2 could be generated as one product from the respiration process in the living cells inside the matoa barks (Meyer *et al.*, 1972)

Methyl alcohol/methanol (CH_3OH); colorless/clear liquid; quantity/portion: 3.39%). CH_3OH could come from the degraded hemicellulose/pectin and low-MW lignin, as well as the phenolic compounds, which contained methoxyl groups (e.g., tannins, flavonoids, plicatic acids, pinoresinol, coninderin, stillbenes, and lignans) in the matoa bark extracts. It has been known that, despite the colorless CH_3OH, those corresponding related phenolic compound origins have conjugated double bond arrangement, which are very reactive and hence could again contribute to natural coloring of matoa barks or in other words the shifting of light absorption as well as light relectance toward the visible colored electromagnetic wavelengths, i.e. 400-800 nm (Vuolo M.M., Lima V.S., Marostica Junior M.R.(2018) Bioactive Compounds: Health Benefits and Potential Applications,, pp. 33-50.Casey, 1980; Sjostrom, 1982; Bieman, 1983).

Acetic acid (CH_3COOH, colorless liquid; quantity/portion: 0.33%). Acetic acid, as detected by the GC-MS analysis, might appear from the degradation of hemicellulose and low-MW soluble lignin, as well as of the simple colored polyphenolic compounds. Consequently, the detected colorless acetic acid might also indicate the presence of colored polyphenolic compounds in the matoa bark extracts.

Acetone (CH_3-[C=O]-CH_3; colorless liquid, quantity/portion: 2.79%). The acetone and its derivatives (e.g., acetyl acetone [0.48%]; propanone [0.28%]; and propanal [0.35%]) could emerge from the degradation of phenyl-propane (C_6C_3) units, as commonly present in the low-MW lignin and particular phenolic compounds (e.g., plicatic acid, pinoresinol, and conidendrin). The lignin (especially the low-MW one), as polyphenols, was inherently pale in color, but even under the mildest treatments, it could cause the darkening of lignin color (Haygreen & Bowyer, 1999; Reus et al., 2003). Meanwhile, as described before, the phenolic compound origins (including low MW lignin), which have conjugated double bond could impart colors to the matoa barks. Accordingly, in total, the role of colorless acetones and their derivatives in contributing to the matoa-bark colors indirectly was simply the sum of 2.79% + 0.48% + 0.28% + 0.35% = 3.90%

Furan (C_4H_4O); colorless crystal/sodid; quantity/portion: 0.69%).

Furan and its derivatives could emerge from the degraded hemicelluloses/pentosans in the barks. Hemicellulose/pentosans. Hemicelluloses/pentosans were naturally white or colorless (Smook, 2002) and therefore did not contribute to the intrinsic bark colors.

Pyridine (C_5H_5N; N-containing heterocyclic compounds; yellowish colors; quantity/portion: 0.10%). The pyridine was biosynthesized as a plant metabolite, which could serve as an N-source for plant growth/development (Yanakiev S., 2020). It has been known that N-containing compounds present naturally in the plants, coupled with their conjugated C-double bond arrangements (Bulton, 2021), could contribute to their intrinsic natural colors (including the matoa-bark colors)

Indolizine (C_8H_7N; also N-containing heterocyclic compounds; bluish colors; quantity/portion: 0.43%). The indolizine was also biosynthesized as a plant metabolite, serving as well as N-source for plant growth/development. As an N-containing compound with its conjugated C-double bond arrangements (Robert & Caserio, 1975), could also impart intrinsic colors to the matoa-bark.

Formamide (CH_3NO; N-containing aliphatic compound; rather yellowish colors; quantity/portion: 0.41%). The formamide, bio-synthesized also as the plant metabolite, could serve as an N-source for the plant growth/development (Saefudin, 2023. The presence of amine (NH_2) in conjugation with the carbonyl (C=O) group rendered the formamide able to contribute natural coloring to the plant portion, e.g., matoa barks (Vuola, 2018).

Disulfide, dimethyl (DSDM; CH_3-S_2-CH_3; colorless liquid; quantity/portion: 0.08%). The presence of DSDM in plants (e.g., matoa barks) was not natural, but due to a deliberate external factor (as a pre-**plant** soil fumigant). This is because the DSDM could act as an anti-fungal action (Saefudin, 2018). Consequently, the DSDM presence in mato bark did not impart colors.

1,2-Butanediol ($C_4H_8[OH]_2$; colorless liquid; quantity/portion (in total): 4.99% + 22.00% = 26.99%). 1,2-Butanediol was inherently one of the plant's metabolite producers, where its formation was stimulated and accumulated during the plant infection (e.g., bark portions) by phytopathogenic bacteria that belong to the genera Dickeya and Pactobacterium (Ngajowa et al. 2013). Accordingly, the presence of 1,2-Butanediol, which was colorless, did not contribute to the maoa bark colors, despite its high quantity/portion (26.99%).

1,3-Cyclopentadiene ($C_5H_6O_2$; $[CH_2]_3[CO]_2$; white/colorless solid; quantity/portion: 0.40%). .

= Tetradecane ($CH_3[CH_2]_{12}CH_3$; colorless liquid, quantity/portion: 0.48%). Tetradecane was essentially a straight-chained alkane. Presumably, it was formed as an intermediate product between primary and secondary metabolites in the matoa barks, toward the further biosynthesis into the wax or fat compounds

n-Hexadecanoic acid ($C_6H_{10}O_4$; $[CH_2]_4[COOH]_2$; colorless liquid; quantity/portion: 0.13%). N-Hexadecanoic acid was allegedly formed as a secondary metabolite for further biosynthesis into fat compounds in the matoa barks.

1,2-Cyclohexanedione ($C_6H_8O_2$); colorless compound; quantity/portion: 0.32%). 1,2-Cyclohexanedione might result from the addition reaction on benzene structures, including also the phenolic groups, possibly, among others, ellagic acids and plicatic acid in the plant bodies (ETTER, M. C.1989), including mato barks. Consequently, its presence could strengthen the presence of color-bearing phenolic substances in the matoa barks.

1,3-Benzenedicarboxylic acid, bis(2-ethylhexyl ester ($C_{24}H_{38}O$; colorless solid; quantity/portion: 0.05%). 1,3-Benzenedicarboxylic acid, bis(2-ethylhexyl ester might be formed as an intermediate metabolite in plant bodies (Boulton, R. 2001) toward further biosynthesis into the so-called lignans, known as one of the color-contributing phenolic compounds in the plants, including the matoa barks.

Phenols (C_6H_5OH; colorless liquid; quantity/portion: 0.54%), and their derivatives, i.e. benzene (C_6H_6; colorless liquid; quantity/portion: 0.26%); toluene (C6H5CH3; colorless liquid; quantity/portion: 0.16%); phenol, 2 methoxy ($C_6H_4[OCH_3]_2$; colorless liquid; 0.16%); p-cresol ($CH_3C_6H_4OH$; colorless liquid; quantity/portion: 0.16%); phenol, 4-ethyl-2-methoxy- (colorless liquid; quantity/portion: 2.64%); 2-methoxy-4-vinylphenol (colorless liquid; quantity/portion: 0.84%); phenol, 2,6-dimethoxy- (colorless liquid; quantity/portion: 0.45%) and phenol, 2,6-dimethoxy-4-(2-propenyl)- (colorless liquid; quantity/portion: 0.11%). As such, those phenols and phenol derivatives could emerge, which resulted from the degradation of low-MW lignin as well as polyphenol compounds. On the other hand, it is also possible that those phenol-related compounds were formed as intermediate plant metabolites in the further biosynthesis into more complex polyphenol compounds. Regardless of those two contradictive presumptions, the phenols and their derivatives exhibited Kekule's double bond arrangement as well as conjugated double-bond system, rendering them extremely reactive in the further complex reactions, which generated color-bearing polyphenolic compounds (e.g. gallic acids, ellagic acids, catechin, quercetin, tannin, flavonoids, lignans, phlobaphenes, stilbenes, flavans, etc) and further could contribute to the natural coloring of the plant bodies (Magozwi DK et al, 2021), e.g., matoa barks.

Catechol ($C_6H_6O_2$; $C_6H_4[OH]_2$; bluish colors; quantity portions: 10.12%). The detected catechol could also appear as one of the degraded low-MW lignin and polyphenol compounds. Adversely, catechol was presumably formed as an intermediate plant metabolite, and further acted as a precursor in the subsequent biosynthesis into more complex color-bearing polyphenolic compounds in the plant bodies, including the matoa barks, into e.g., tannins, flavonoids, catechins, quercitin, lignans, stilbenes, and tropolones (Boulton, R. 2001)

Heptadecanal ($C_{17}H_{34}O$; colorless solid; quantity/portion: 0.18%). Heptadecanal was essentially a long-chain fatty aldehyde, presumably a precursor in the further synthesis into fatty acids, fats, and wax compounds in the plant body (Boulton, R. 2001), including in the matoa barks.

To summarize, the original compounds in the matoa bark extracts, after the GC-MS analysis, might contain some degraded compounds and some other (residuals) still in the intact/original compounds. Further, from those degraded compounds and those residually still in intact/original compounds, it could be deduced that three major chemical compounds brought about the natural coloring of matoa barks, i.e., polyphenolic compounds, N-containing compounds, and metal-containing compounds (nickel tetracarbonyl). The role of polyphenolic compounds was simply the sum of 5.84% + 3.39% + 3.90% + 0.33% + 0.32% + 0.05% + 5.16% + 10.12% = 29.11%. Meanwhile, the role of Ni tetracarbonyl (following the complex reaction with the color-contributing phenolic compounds) was 36.80%, and eventually the role of N-containing compounds was only 0.94%.

It seemed strongly that the role of phenolic compounds in contributing the bark extract colors was the greatest, which comprised polyphenols (gallic acids, ellagic acids, catehin, quercitin, tannin, flavonoids, lignans, quercitin, taxifolin, phlobaphenes, quinones, and stilbenes, in total as much as 28.78%) and metal-containing compounds (allegedly in complex reaction with phenolic compounds, as much as 36.80%). Meanwhile, the role of N-containing compounds was the smallest (0.94%). Eventually, all those color-contributing compounds could then be regarded as natural-coloring pigments in the matoa bark extracts.

GC-MS results on the dry extracts from matoa leaves

The detailed chemical compounds in the leaf extracts presumed as natural pigments that could be identified qualitatively/quantitatively with the GC-MS analysis (Appendix 2), are as follows

D-Alanine ($C_3H_7NO_2$; H_3C-NH-COOH; white colored; quantity/portion: 6.92%). D-alanine could allegedly be formed as one of the plant metabolites, which could serve as an N-source in the further biosynthesis into more complex, large molecular weight (MW) N-containing substances (Meyer *et al.*, 1972; Starr & Taggart, 1993), e.g., protein and in this case chlorophyll a ($C_{55}H_{72}MgN_4O_5$; exhibiting bluish green colors) as well as chlorophyll b ($C_{55}H_{70}MgN_4O_6$; with yellowish green colors) supposedly in matoa leaves. However, the detected d-Alanine (with its free methyl/CH_3 group) indirectly indicated the presence of chlorophyll a (with its free CH_3 group as well). Further, both the chlorophyll a and b had numerous conjugated double bond structures, and the presence of metal (Mg) elements in ionic bonds as well as in chelation with the nucleophilic N arrangements inside the chlorophyll could also contribute to the complex natural coloring of matoa leaf extracts (Robert & Caserio, 1975; Sinnot & Wilson, 1975; Starr & Taggart, 1993; Koch, 1995).

Sulfur dioxide (SO_2; colorless gas; quantity/portion: 4.39%). Its presence in the plant body (e.g., matoa leaves) was possibly due to the SO_2 absorption by the plants from the air/atmosphere, and then converted into anionic acids. The S elements could serve as one of the essential nutrients for plant growth. Also, the SO_2 in the plant body (e.g., matoa leaves) were metablozized into S-containing protein and possibly S-containing pigments, e.g., particular glycosides (Meyer *et al.*, 1972; Starr & Taggart, 1993).

Cyclobutylamine (C_4H_9N: $[CH_2]_3CH-NH_2$; colorless solid; quantity/portion: 5.38%). Cyclobutylamine could emerge in the matoa leaves as the degraded substances from the chlorophyll (a and b), as it is known that N-containing compounds (e.g., chlorophyll) in the plants could contribute to the natural coloring of the plant bodies (Casey, 1980; Starr & Taggart, 1993), e.g., matoa leaves.

Furan, 2-methyl- (colorless volatile liquid; quantity/portion 0.02%). Furan and its derivatives could be present in the matoa leaves, as the degradation product of hemicellulose. The hemicellulose was inherently white in color or colorless (Sjostrom, 1982). Accordingly, the presence of furans and their derivatives apparently did not contribute to the natural coloring of matoa leaf extracts.

Hydrogen isocyanate (HNCO; H-N=C=O; pale yellow to red orange colors; quantity/portion: 57.22%). The presence of hydrogen isocyanate in matoa leaves could be an intermediate plant metabolite in the further biosynthesis of complex high-MW colored N-containing compounds and possibly the chlorophyll (Starr & Taggart, 1993).

Urea ($NH_2[CO]NH_2$; white solid crystal; quantity/portion:13.01%). The urea could exist in the matoa leaves as plant metabolites, which were derived from the root uptake of N-elements in the soil. The urea could further undergo biosynthesis into protein and high-MW N-containing pigments, e.g., chlorophyll (Saefudin & Basri 2023).

Cyanuric acid ([CNOH]$_3$; $C_3H_3N_3O_3$; $C_3N_3[OH]_3$; white crystal powder; quantity/portion: 13.05%). The presence of cyanuric acid in the plant bodies (e.g., matoa leaves) with its free CH_3 group could presumably serve as a precursor in the further biosynthesis into complex high MW N-containing compounds (Starr & Taggart, 1993, e.g., protein and N-containing pigments (especially the chlorophyll a and perhaps phorphyrin)

To sum up, the original compounds in the matoa leaf extracts, after the GC-MS analysis, might also contain some degraded compounds and the residuals still in the intact/original compounds. Further, from those degraded compounds and those residually still in intact/original compounds, it could be inferred that there were two kinds of major chemicals which contributed to the natural coloring of leaf-extract colors, which comprised N-containing compounds and S-containing compounds. As such, the role of the former was 6.92% + 5.38% + 57.22% + 13.01% + 13.05% = 95.58 % (predominantly chlorophyll pigments of type a and type b); while that of the latter was much smaller (4.39%; particularly glycoside substances). Ultimately, all those color-contributing compounds could then be regarded as natural-coloring pigments in the matoa leaf extracts.

Comparison of the extractable colored compounds between matoa barks and leaves Judging from the GC-MS analysis (Appendices 1 and 2), it strongly indicates that the matoa barks in general contained more of the extractable substances (allegedly containining natural pigments) either qualitatively or quantitatively than the leaves with respect to consecutively kinds and portions of extractives, where those indications could then explain why the extractive yield from the matoa bark was greater that from matoa leaves (Table 1).

From the GC-MS analysis, it is also evident that the allegedly natural-coloring pigments in the extracts from matoa barks were dominated by the polyphenol compounds (e.g., gallic acids, ellagic acids, catechin, quercitin, tannin, flavonoids, lignans, quercetin, taxifolin, phlobaphenes, quinones, and stilbenes) and to a lesser extent by the metal-complexing compounds and the N-containing compounds, respectively. Meanwhile, conversely, the supposedly natural color compounds in the matoa-leaf extracts comprised predominantly N-containing compounds (mostly chlorophylls a & b and possibly phosphirin) and, in minor amounts, the S-containing compounds (e.g., glycosides).

3.3. *Results of Color Testing*

Color-leaching resistance In Table 2 are presented the data regarding the color-leaching tests on the colored primisima fabrics. It revealed that for the primisima fabrics colored with the bark extracts without or with tunjung/$FesO_4$ fixative (at any concentrations) exhibited consecutively the class 4-5 scale (good to very good/best) for rubbing/ironing resistance; class 3-4 (moderate to good) for acidic sweat resistance as well as for xenon ray resistance; and class 3-5 (moderate to good and verygood/best) for detergent-washing resistance. Further, for the coloring with bark extracts, the lowest color-leaching resistance against the detergent washing occurred at the colored fabric samples with the 75BTW (i.e., using 10 g/L tunjung fixative) and with the 100BTJ codes (using 20 g/L tunjung fixatives), i.e. respectively at the category/class of 3-4 and 4 scales (regarded as moderate/fair to good). Meanwhile, the washing resistance of other bark-extract samples exhibited 4-5 scales (regarded as good to very good). These results revealed that the extracts, especially from matoa barks, could be applied for coloring the primisima fabrics favorably with good criteria/category. One of the factors that could affect such high color-leaching resistance (i.e., good score) was that in the final mordanting, the alum mordant could interact positively with the tunjung fixative (especially at 20 g/L concentration), thereby serving as a more effective bridging between the natural pigments and the fabric fibers, and hence imparting even stronger affinity/bonds between the pigments and the fabrics (Suestining Diah, 2015; Saefudin, 2023). Consequently, this situation apparently contributed to better color-leaching resistance of the extract-colored fabrics (Saefudin, 2021); and as such, the fabrics appeared tobe more strongly colored, especially with bark extracts.

With respect to the color-leaching resistance of primissima fabrics colored with leaf extracts and then treated with tunjung fixatives as well as without tunjung (Table 2), it disclosed that consecutively for rubbing/ironing actions, the class/score was 3-4 (moderate/fair to good); for detergent washing as well as acid sweating actions, the class/score 2-4 (poor to good); and for xenon-ray/sunlight exposure, the class/score 2-3 (poor to moderate/fair).

Overall results disclosed that the color-leaching resistances of primisima fabrics colored with leaf extract were mostly lower (i.e., lower classes/scores), compared to those with bark extracts (Table 2). These results indicated that the tiny particles of supposedly natural pigments that originated from leaf extracts were less firmly/less strongly adhered/stuck to the fabric fibers than those from bark extracts.

This could be due to greater extract yield of bark extracts (Table 1) and to more of the natural-pigments's chemical compounds in bark extracts (qualitatively/quantitatively), compared with those of leaf extracts (as indicated from GC-MS analysis).

Table 2: The Color-Leaching Resistance of the Primisima Fabrics, After Being Colored With the Extracts From Matoa Barks as Well as From Matoa Leaves, Toward the Creation of Batik Fabrics.

Treatment No.	Biomass waste origins (of the extracts)	Color-leaching actions			
		Rubbing/ Ironing	Detergent washing	Acidic sweat	Xenon ray
		------------- Criteria / Category / Class ------------			
1.	Matoa barks	4-5	4	4	4
2.	Matoa barks	4	3-4	3	3-4
3.	Matoa barks	4	4	4	4
4.	Matoa barks	4	4-5	4	4
1	Leaf barks	3-4	4	4	3
2	Leaf barks	4	4	4	3
3	Leaf barks	3	3	3	3
4	Leaf barks	3	2	2	2-3

Remarks (for the treatment No): No 1 = control (blank primisima fabrics without any treatment / no coloring and no fixation); No 2 = primsima fabrics after being colored with natural pigments from bark as well as from leaf extracts (but without tunjung fixation or with 0% tunjung/FeSO4 fixatives); No 3 and 4 are the primisima fabrics after being colored with natural pigments (from bark/leaf extracts), with consecutively 10, and 20 g/L tunjung (FeSO4) fixatives; all No. 2, 3, and 4 were modanted with alum. No.1 = it should be colored primissima fabrics, with bark/leaf extracts, but without alum mordanting Remarks (for the criteria/category/class of color-leaching resistance): class 1 (very poor); class 2 (poor); class 3 (moderate/fair); class 4 (good); and class 5 (very-good/best).

Testing of color difference (L*,a*,b*,ΔE)

The data regarding the color difference (ΔE) for all the treatments (control or blank/control primisima fabrics vs pigment colored fabrics at 3 concentrations of tunjung/$FeSO_4$ fixative) are presented in Table 3.

Table 3: Color Parameters (L, a*, b*) and Color Difference (ΔE) for the Blank Primisima Fabrics as Well as for the Fabrics, After Being Colored With the Extracts From Matoa Barks/Leaves.*

No	Biomass waste origins (of the extracts)	Color parameters			
		L*	a*	b*	dE*ab (ΔE)
	Matoa barks				
1	A	100.48	0.03	0.25	0.00
2	B	35.73	19.84	27.20	48.39
3	C	44.16	18.12	21.62	67.74
4	D	84.16	1.38	12.99	18.45
	Matoa leaves				
1	E	44.61	18.24	16.05	16.32
2	F	28.43	13.95	19.87	27.22
3	G	30.26	6.27	8.87	21.05
4	H	17.23	1.98	13.75	17.74

Remarks: for the notations of L*, a*, b*, dE*ab (ΔE), please refer to equation [4]; A = blank white primisima fabrics/control (with no coloring and no fixation), B, C, D = primisima fabrics after being colored with bark extracts, and then fixated with tunjung (FeSO4) at consecutively 0, 10, and 20 g/L FeSO4; E = colored-primisima fabrics/control (with extract-coloring, but without alum mordant), F, G, H = primisima fabrics after being colored with leaf extracts, and then fixated with tunjung (FeSO4) at consecutively 0, 10, and 20 g/L FeSO4; F = colored primisima fabrics, mordanted with alum, but without tunjung fixatives; all B, C, D, F, G, and H-coded fabris were modanted with alum.

Referring to Table 3, the lowest L* value occurred at 17.23 (i.e., the fabrics colored with the leaf extract at 20 g/L tunjung fixatives, FeSO4.7H2O; H sample code) at 80 °C extraction temperature, previously immersed in basic detergent emulsion. Meanwhile, the highest L* value was 84.16 (i.e., the fabrics colored with bark extracts, at 20 g/L tunjung fixative, D sample code), which revealed the fabric color oriented toward the red-brown direction. The a* values exhibited positive values from 1.38 until 18.24, revealing that the fabric colored with bark extracts, as well as with leaf extracts, disclosed the color direction toward reddish brown. The b* value of all the colored fabrics (with bark as well as leaf extracts) revealed positive b* values from 8.75 to 36.09, which disclosed that the fabric colors contained a chromatic mixture of blue and color. Further, the dE*ab (ΔE) value at 17.74 (as the lowest) occurred at the fabrics colored with the leaf extracts, previously immersed in the alum mordant but without using tunjung fixative (or at 0 g/L tunjung), revealing that the fabric colors approached their color toward weak brown colors. Further, the greater the color-difference/ (ΔE; see equation [4]), then the greater would be the difference in color between the fabric standard (e.g., blank primisima fabrics) and the colored samples, e.g., the fabrics colored with the pigments, in this regard with the bark/leaf extracts, with or without the tunjung fixatives (Technical Services Department, 2008).

The variation in the resulting colors of the extract-colored fabrics (expressed quantitatively as the varying L*, a*, and b*values) after the final mordanting with alum (AL2[SO4]3) mordant combined with the fixative (tunjung/FeSO4) treatment brought about the significant color differences (as varying ΔE values). The fixation without tunjung would bring/direct the colored-fabric toward light/bright colors, while the fixation that incorporated tunjung, especially at high concentration (20 g/L), would direct the colored-fabrics toward dark/strong fabric colors. The use of tunjung fixatives allegedly affected the colors (L*, a*, b*) as well as the color difference (ΔE) of the created colored batik fabrics (Table 3). This is because during the fabric immersion in the fixative (tunjung) solution, the intricate reaction could occur between the tannin from the ZWA (allegedly in the bark/leaf extracts) and Fe+2 ions of the tunjung, resulting in a complex colored salt, the so-called ferro tannat (Saefudin, 2022). Those complex salts were formed due to the occurrence of strong covalent bonds between the metal ions (at tunjung / $FeSO_4$) and non-metal ions (in the tannin or other polyphenol compounds in the extract). Accordingly, with the use of alum as a mordanting agent, there would occur an ionic reaction between the tannin (from bark/leaf extracts) and (+) Al^{+3} ions (from the alum), as well as (+) Fe^{+2} ions (from the tunjung fixatives). These phenomena were in agreement with the opinion by Wijaya et al. (2015), where in the final mordanting, the tunjung fixatives, especially at high concentration (20 g/L), were able to create effective bridging and hence stronger bonds between the allegedly natural pigments (especially in the bark extract) and fabric fibers. In this way, therefore, it could impart higher L* values as well as consequently increase ΔE values of the colored fabrics, thereby directing the fabric colors toward stronger color, expectedly beneficial in the creation of batik fabrics successfully. In all, the ΔE values of the colored fabrics (with bark/leaf extracts) were entirely ≥ 12.0, indicating that there occurred a very distinct difference in color changes in all the colored fabrics.

Testing of color-ageing The data regarding the results of the color-ageing test (i.e., light-reflecting percentage/values and K/S ratios) are presented in Table 4. It reveals that the light-reflectance values (R) of the blank white primisima fabrics reached 108.48% (inherently 100%). This is because, according to Bieman (1983), white-colored substance (e.g., white primisima fabrics) reflects all the incoming light (100%) in the visible region (400-730 nm). Further, with 100% R value, it could explain the zero (0) value for the K/S ratio of the control/blank white primisima fabrics (see Table 4). Still related, the R-values of the the colord-primisima fabrics entirely tended to be significantly lower than those of the blank/control primisima fabrics, while the K/S ratio (in absolute values) aptly increased very appreciably. This very strongly suggests that the colored fabrics have absorbed considerably the supposedly natural pigments from bark/leaf extracts, where the colored fabrics apparently absorbed more natural pigment in bark extracts than in leaf extracts (Casey, 1980). Meanwhile, the reflectance values (R) of the fabrics colored with bark extracts were significantly greater than those colored with leaf extracts, while the K/S ratios of natural pigments between those from bark extracts and from leaf extracts seemed not significantly different from each other. For the case of such greater reflectance valueas, this also again strongly strengthened the previous indication from the results of GC-MS analysis that the matoa bark extract contained more natural coloring-pigments either qualitatively or quantitatively than the matoa leaf extracts (Appendices 1 and 2); and also the content/yield of color-contributing extracts in the barks was greater than that in the leaf extracts (Table 1).

Further, the greater R values of the primisima batik fabrics colored with bark extracts than those with leaf extracts also strongly suggested that the supposedly natural pigments in the bark extracts were able to reflect more of the incoming white-colored light, with greater intensity, compared to those withleaf extracts. It has been known that such partially reflected white light naturally contains various colors with various wavelengths (λ) as well in the λ range of visible light (400-730 nm). These phenomena implied that concurrently those fabrics (colored with bark extracts) were also absorbing the complementary colors of those various colors, also with greater intensity at various λ values, still in 400-

730 nm range, which comprise consecutively (from the lowest until highest λ) violet, blue, green, yellow, orange, red, and eventually purple colors (Bieman, 1983).

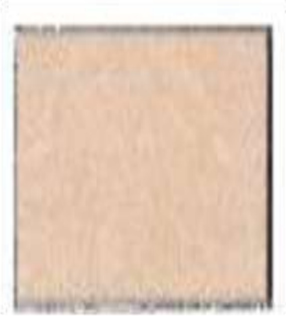
Again, this situation happened allegedly due to a greater presence of various natural coloring pigments in the bark extracts (qualitatively/quantitatively) than in leaf extracts. Further, at the batik's prmisima fabrics colored with the biomass extracts (from mato barks as well as matoa leaves), it exhibited the K/S ratio in the range of [-15.85]-[-49.18], thereby regarded as strong colors (Farida, Atika, and Saefudin, 2018).

Table 4: Results of Color-Ageing Tests on the Fabrics, After Being Coloured With the Extracts Supposedly Containing Natural Pigments, From Matoa Barks as Well as Matoa Leaves.

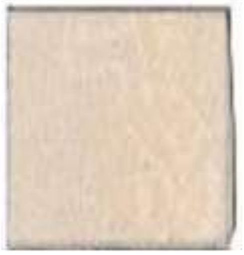
No	**Biomass waste origins (of the extracts)**	**R (%)**	**K/S ratio ¹) (of natural pigments)**
0	Blank white primisima/silk fabrics (control)	108.80	0
	Matoa barks		
1	A	12.76	-45.50
2	B	6.32	-49.18
3	C	72.13	-16.82
4	D	9.61	-46.95
	Matoa leaves		
5	E	8.46	-47.58
6	F	5.63	-49.50
7	G	50.14	-15.85
8	H	9.80	-46.89

Remarks: O = blank/untreated primisima fabrics (with no FeSO4 fixatives and no alum mordant; as control); A = primisima fabrics, after being colored with bark extracts (but without tunjung/ FeSO4 fixation and no alum mordanting); B, C, D = primisima fabrics after being colored with bark extracts, and then fixated with tunjung () at consecutively 0, 10, and 20 g/L concentration; E = primisima fabrics, after being colored with leaf extracts (but without tunjung fixation and no alum mordanting), F, G, H = primisima fabrics after being colored with leaf extracts, and then fixated with tunjung (FeSO4) at consecutively 0, 10, and 20 g/L; all B, C, D, F, G, and H-coded fabris were modanted with alum; R = reflectance percentage/values; K/S ratio = (1-R2)/R (Casey, 1980)

Visual appearance of the colored fabrics The visual appearances of the primisima fabrics, after being colored with the extracts from matoa bark as well as from matoa leaf extracts, are presented in Figure 1.



1



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3

4

Figure 1: Visual Appearances of the Primisima Fabrics Colored With Biomass Wastes of Matoa Bark Extracts, as Well as of Matoa Leaf Extracts.

Remarks

1. 1 & 3 colored with leaf extracts; 2 & 4 with bark extracts;

2. 1 & 2 and 3 & 4, each at two different concentrations of tunjung fixative
3. 1 & 2 without batik motifs; 3 & 4 with batik motifs

In Figure 1, it appears that the variation in the concentrations of tunjung fixative visually did not significantly affect the color-ageing rate, as well as the color direction/orientation/appearance of the colored primisima fabrics. On the other hand, conversely different kinds of biomass extracts (i.e. from matoa barks vs from matoa leaves) apparently affected the the color-ageing rate and color direction, where the color of fabrics after being immersed in the bark extracts tended to look more strongly colored (strong brown), while in the leaf extracts seemed to become weaker colors (weak brown toward creamy colors). This strengthens again the previous indication that such an occurrence was due to the results that the extract content in the matoa barks was greater than in the matoa leaves (Table 1); and also from the GC-MS analysis that the bark extracts very presumably contained more natural pigments either qualitatively or quantitatively than the leaf extracts. Further, without the implementation of final mordanting actions, it brought about the fabric colors which appeared weaker/more pale. Meanwhile, with final mordanting using tunjung fixative at high concentration (20 g/L), the fabrics have strong brown colors.

4. CONCLUSION AND SUGGESTIONS

The characteristics of biomass extracts from matoa barks/leaves (for their utilization as renewable natural pigments for coloring batik fabrics) were viewed from several significant aspects, comprising the extract yield, chemical composition (through GC-MS analysis), and color difference testing (using CIELab method), color-leaching resistance, as well as color-ageing tests (all performed on the resulting colored batik fabrics with those extracts, followed with fixative treatment), as follows

1. The yield of extracts (as biomass wastes) from matoa barks was significantly greater than from matoa leaves
2. The extracts from both matoa (Pometia pinnata L) barks and leaves in general could be utilized as raw materials (natural pigments) in coloring the cellulose fibers of the primissima fabris toward the creation of batik fabrics. This is because from the GC-MS analysis on those biomass's extracts, it allegedly indicates that
3. The matoa bark extracts contained the color-contributing substances (regarded as natural-coloring pigments), which comprised mostly phenolic compounds (e.g., gallic acids, ellagic acids, catechin, quercitin, tannin, flavonoids, lignans, phlobaphenes, quinones, and stilbenes) and N-containing compounds.
4. The matoa leaf extracts also contain the color-contributing substances (as natural-coloring pigments too), which comprise mostly chlorophyll (a & b) and, to a lesser extent, other N-containing compounds (e.g., porphyrin) as well as S-containing compounds (e.g., glycosides).
5. It strongly indicates that matoa barks contained more of the extractable substances (allegedly covering natural pigments) than matoa leaves qualitatively as well as quantitatively with respect to the kinds and portions.

It seemed also that the allegedly natural-coloring pigments in the extracts from matoa barks were dominated by the polyphenol compounds (e.g., gallic acids, ellagic acids, catechin, quercetin, tannin, flavonoids, lignans, quercitin, taxifolin, phlobaphenes, quinones, and sfrom tilbenes). Meanwhile, conversely, the supposedly natural color compounds in the matoa-leaf extracts comprised predominantly N-containing compounds (mostly chlorophylls a & b).

Matoa bark extracts as biomass wastes (supposedly containing natural coloring pigments) seemed better than the leaf extracts, in coloring the blank white primisima fabrics toward the creation of batik fabrics. As such, the fabrics colored with the bark extracts exhibited apparently the superior color-leaching resistance against the detergent washing with a 4-5 scale category (good to very good/best).

The values of color differences (L*, a*,b*, ΔE) brought out the color direction/orientation of the colored fabrics (allegedly with the bark extracts) toward stronger brown colors than with leaf extracts, which all significantly differed from that of the control (blank white primisima fabrics).

The value of color-ageing test results on the fabric colored with bark/leaf extracts brought about significant effects on the color-ageing rate, where the color of fabrics immersed in the bark extract solution exhibit stronger colors than that immersed in the leaf extract solution, indicating that allegedly natural pigments in bark extracts absorbed more of the complementary colors of the observed colors in the reflected incoming (visible) light. The difference in concentrations of the tunjung fixatives imparted significant effects on the rate of color-ageing at the pigment-colored fabrics, where the use of low tunjung concentration in the final alum mordanting brought the lighter/weaker colors of the fabrics (weak/pale brown colors toward creamy colors). Meanwhile, the final mordanting with tunjung fixatives also affected the fabric colors toward stronger brown colors. The use of final mordanting/fixating agents also affected the value of color difference (L*, a*,b*, ΔE), and the final mordanting action using tunjung was able to bind the L* parameters more strongly than using alum mordant. Meanwhile, the variation in 80 °C extraction temperature caused no significant effects on the color-difference values and on color-ageing rates.

To sum up, judging from those already-examined characteristics, bark extracts seemed more promising (e.g., greater extract yield, more varying chemical compounds qualitatively/quantitatively, higher ΔE values, greater color-leaching resistance, greater reflectance factor, better color-ageing results, and stronger color direction/appearances) than leaf extracts for coloring batik fabrics with satisfactory results. Therefore, scrutinizing the characteristics of biomass extracts (from matoa barks/leaves) could be beneficial, with the bark extracts allegedly deserving more emphasis, in their utilization as natural, renewable pigments for coloring the blank white primisima fabris toward the creation of batik fabrics successfully.

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