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Utilization Of Tannin As Renewable Natural Pigment In The Culture Of Indonesian Batik Fabrics: A Review

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ABSTRACT

Nowadays, trends have increased to replace synthetic pigments with natural pigments (e.g., tannin) due to solid consumer demands for more natural products. Sources of tannin pigments spread in particular plant parts, e.g., leaves, stems, twigs, barks, fruits, seeds, flowers, and wood wastes. Utilization of tannin pigments by the crafters of batik fabrics which covers pigment exploration, extraction, and application, has proceeded for a long time. It aims to produce batik fabrics with unique colors, suitable motives, and friendly environments. Initially, tannin-derived colors were obtained without processing the plants. Tannin's natural pigments have been developed and produced in liquid, paste, and dry forms. Natural pigment production not only increases the capacity of the selling price but should also exhibit biological activities that are environmentally friendly and beneficial for health. The abundant sources of tannin-rich wood wastes (e.g., mangrove origins) from forestry industries could replace mostly the need for synthetic pigments, which sound environmentally unfriendly. Wood waste utilization for tannin could encourage and create economic opportunities in forest villages, centers of mangrove wood industries, and small- and medium-scale batik/weaving enterprises. Coastal and inland motif batik crafters prefer the natural colors of tannin. Accordingly, exploring various natural sources of tannin is essential for fabric coloring, safe for food/beverages, and beneficial for health.

Keywords: Tannin**,** natural pigment, synthetic pigments, batik fabrics, environmentally friendly.

Introduction

Natural pigments, particularly those used for food/beverages, paper, and textiles, have attracted remarkable attention from the industry and trade world. The demand for pigments from living natural sources (e.g., plants) increases continuously. This is because those natural pigments are renewable, beneficial for human health; seem more environmentally friendly; and also due to the growing awareness of the negative impacts of possible dangerous compounds in synthetic pigments. Tannin can be defined as one type of natural polyphenolic compound commonly present in plant tissues. It is considered a secondary metabolic product that results from further biosynthesis after plant photosynthesis. Tannin can impart various colorations and be regarded as one type of renewable natural pigment. Consequently, tannin could play an essential role in the culture of weaving and crafting textile materials (including batik fabrics) in Indonesia.

The colors imparted by the tannin could vary visually as yellow patterns, brown patterns, and varying reddish appearances, which look typical and fresh. The crafters of batik fabrics and weavers of other materials could quickly obtain the tannin and other natural pigments from their home vicinities. The colors of fabrics, if dyed with tannin, will feel convenient, seem safe adapting to the individual aesthetic desires, and be able to reinforce the color fixing at the fabric fibers. The performance of tannin pigment in several plants varies with the plant species' origins, and the presence of tannin inside the plant parts often shares concurrently with other pigments, such as flavonoid, anthocyanin, chlorophyll, and carotenoid. Tannin pigments exhibit particular characteristics, among others, soluble in water (Tibor, 2007). Such characteristics are essential for many industries. For example, food/beverages, fabric weaving, and batik crafting industries have utilized tannin pigments as additives, color-reinforcing agents, and antioxidants.

The production of renewable natural pigments (including tannin) with their varying colors could be in the form of dry extract and liquid. Whatever the forms of natural pigments, attempts should be thoroughly performed to meet the standards of quality, security, and uses or benefit principles. The raw materials for pigments should be consistently available and easily assessed. The pigment products should bear their specification, especially the color standard, color consistency, and information about the ingredients and the composition of other matters added to the pigment formula. Accordingly, the testing of those pigments deserves to be carried out.

There are several plant species with their particular parts (e.g., barks, stems, leaves, and roots), which grow in our vicinities, and can synthesize and produce tannin naturally, such as pinang/betel nuts (Areca catechu) nangka/jackfruits (Artocarpus heterophylla), banana (Musa paradiciaca), bakau/mangrove (Rhizophora mucronata), pine (Pinus mercusii), and gambier (Uncaria gambir). Tannin could also be naturally produced by plant species that grow on highlands (plateau), such as tea (Camelia sinensis), as well as by plant species that grow on lowlands (plain area) or in wet environment/ecosystem, such as mangrove, for examples Rhizophora mucronata, Xylocarpus granatum, Avicenia marina, and Terminalia catappa species. Meanwhile, typical plant species that grow in coastal forests could also naturally synthesize/produce tannin, such as Rhizopora apiculata, R. macrophylla, Pterospermum ferroginum, Ceriops candolleana, etc. Further, all those mentioned plant species in the form of vegetation wastes could also be a potential source of tannin.

The vegetation wastes generated from the branch-free plant stems could reach approximately 928,934.81 m3/year (Astana, Soenarno & Endom, 2015). This amount still did not include the wood wastes from the round woods (wood logs), which were felled/cut down, in complying with the Endeavor Permit for the Utilization of Wood Forest Products at Plantation Forest (EPUWFP-PF) / Izin Usaha Pemanfaatan Hasil Hutan Kayu pada Hutan Tanaman (IUPHHK-HA), whereby the log production tended to increase annually. In 2018, the log production from plantation forests reached 40,945,378.90 m3 per year (Indonesia’s Ministry of Life Environments and Forestry / Kemen LHK, 2018). Wood wastes generated by the pulp production activities at PT TEL (Tanjung Enim Lestari) Pulp and Paper in North Sumatra were quite enormous, especially in the forms of barks from mangium woods, which could reach 216.49 tons per day or about 9% of wood logs by weight (Djarwanto et al., 2017). Further, wood wastes in wood slabs, sawdust, twigs, barks, etc., are supposedly rich in various natural pigments, such as tannin, flavonoid, anthocyanin, chlorophyll, and carotenoid. So far, those pigment-rich wood wastes are still not utilized optimally and are left abandoned or discarded. The abandoned/discarded wastes have accumulated in the forests and at nearby wood-processing sites, causing inconvenient problems in forest and factory environments.

Tannin pigments extracted from the plant parts could be produced as liquid and dry solids for the fabric coloring (e.g., the batik fabrics). For efficiency in the long-termed use of trade, the tannin pigments should be processed and produced in the form of dry solid extract. The essential activities in the coloring process of the fabrics (e.g., for batik) with the pigments, including the tannin, consist of mordanting and fixating. Mordanting aims to eliminate fat, oil, and other foreign matters that adhere to the fabric fibers. This is because such elimination could later greatly facilitate or more efficiently impart the penetration (infiltration) of pigments into the fabric's minute structures (pores or voids) during the fabric coloring. Mordanting is usually performed before the fabric coloring, and the mordants commonly used are typol, tawas/alum, tannin acid, and acetic acid/vinegar (all in an aqueous solution).

Meanwhile, the fixation is conducted after the fabric coloring, whereby the fabrics are submerged (steeped) in the coloring pigment solution (e.g., tannin). The fixation intends to strengthen or make the color of the pigmented fabrics last longer. The fixatives commonly used are inorganic salts, e.g., tawas/alum, Kapur/lime, and tunjung/ferric sulphate (Sedarlah, 2007). Further, to examine the performance of coloring pigment (e.g., tannin), the colored and fixative-treated fabrics should be tested for color-leaching or color-fading resistance against washing, rubbing, and exposure to sunlight. The testing procedures as required and conducted for such should refer to the international standard organization (ISO) and the national standard (SNI).

Relevantly, this scientific writing presents and assesses the results of exploration, extraction, and application of natural tannin pigments (e.g., tannin) obtained from various plant sources; varying plant species; motivation of batik crafters; research results; experiences acquired by the batik crafters; testing of tannin pigments implemented on various fabrics (e.g.. mori, primisima, cotton); and test on color-leaching/fading resistance of the tannin-colored fabrics against the washing, rubbing, and exposure to the sunlight. Other essential assessments include the potency of wood wastes generated from the wood-processing industries, "regarded as useless", to be further utilized in the natural renewable sources of tannin pigments and other pigments for coloring the textile (e.g., batik fabrics), which should sound environmentally friendly.

METHODOLOGY

Natural dyes containing tannins have evolved over thousands of years and have been lost. Now is the time to develop essential innovations in natural tannin pigments that are environmentally friendly. Tannin production begins by exploring biomaterials such as wood wastes from sources of tannin's natural pigments. Collection of leaf, bark, and twig waste materials from Acacia mamgium Mild., Caesalpinia sappan L, Ceriop tagal Perr. CB. Rob, Pelthophorum ferruginum Benth, Rhyzophora mucronata Lamk, Tectona grandis Linn. f., and Terminalia catappa L were sources of tannin's natural pigments (Table 1).

Stages in producing biomass-waste-based natural pigments began with waste sorting and dimension reduction to become small pieces of 1-2 cm sizes and cleared of foreign matter. The biomass wastes already selected, which comprised barks, sawdust, and leaves from those plant species but of old ages, were frequently used as sources of natural brown-coloring pigments by the traditional crafter community. The search for natural coloring pigments could also be conducted in particular places such as cutting, temporary storage, sawing, and other related processing sites in wood industries.

The further stage was extracting liquid-shape colored matters (or coloring pigments) from those materials, which could be easily fixed or bonded by the so-called primissima or white-colored cotton fabric materials. As such, those biomass waste pieces were cooked in water with the ratio of 1:4 (w/v; kg/liter) at 70-80oC temperature for a particular duration such that most of the coloring matters (natural pigments) inside were intensively moved or migrated out into the cooked water, forming aqueous pigment solution; or on evaporation, the volume of the residual solution becomes approximately one third (1/3) of the original volume. Afterward, the residual pigment solution was allowed to cool down for 24 hours and further finely screened using clean filtering cloths. The obtained filtrate (the so-called screened coloring pigment solution or wet initial/fresh pigments, which passed through the cloths) could then be ready directly for use (coloring the fabrics) or stored for a particular duration.

RESULTS AND DISCUSSION

Production and Sources of Tannin as Renewable Natural-Pigment

As a renewable natural pigment, Tannin can be obtained from almost any part of the plants, such as tree barks, roots, leaves, fruits, seeds, exudates/saps, and fruit skins, though commonly the extraction process. Currently, particular parts of the plant's considered worth tannin extraction are the so-called wood wastes generated abundantly from the tree cutting/felling in the forest and the wood sawing in the sawmilling industries. So far, the wood wastes are abandoned in the forest, accumulated in wood industry sites, or burnt, potentially polluting the environment. The wood wastes from the tree cutting/felling and the wood sawing could be regarded as indicatively rich sources of natural pigment compounds, such as Tannin, flavonoid, saponine, carotenoid, chlorophyll, and anthocyanin (Rosyida & Zulfiya, 2013; Sjostrom, 2013).

Those pigment-alleged compounds, such as Tannin, flavonoids, saponine, anthocyanin, chlorophyll, and carotenoid, besides already naturally present in the plant part origins, however, could also be formed due to the heating as well as storage processes of those plant origins as well as of the generated wood wastes. This is because chemical changes can occur during the heating/storage of the original chemical compounds in the plant origins or the generated wood wastes, such as degradation, respiration, and other physiological activities. Such changes can convert those original compounds (formerly as complex structures, high polymerization degree, and high molecular weight) to more straightforward or more degraded/depolymerized compounds with lower molecular weight, e.g., Tannin, flavonoid, and anthocyanin (Kwartiningsih et al., 2009).

The production of Tannin should be preceded by exploring the indicated pigment-bearing plant sources that might contain the

Tannin, which grows at our vicinities. Further, those indicated plants could produce Tannin and other natural pigments with high potency, regardless of the number of plant species or the various colorations. Unfortunately, only a few kinds of produced Tannin are ready for use, continuously available, and already standardized. Results of qualitative tests on identifying the presence of possible tannin compounds extracted from the fruits, leaves, and stem barks of S. alba's mangrove using the phytochemical method with $FeCl_3$ salt, revealed that the extracts positively contained the Tannin, which exhibited blackish green or blue solid colors. Meanwhile, the stem barks of S. alba's mangrove, after being dried kept, maintained the same colors as those before the drying, which were brown; and afforded dry as well as complex textures (Halimu et al., 2017; Nuraini, 2002)

Species origins of woods (or other plant parts) and kinds of cooking/extracting chemicals affected the yield of the tannin. As such, the cooking/extracting chemicals could be using water (H_2O) solvent (usually at elevated extraction temperature, ±100oC) or other polar solvents (e.g., alcohol and acetone) or aqueous alkali solution (e.g., 0.5-1.0% Na_2CO_3). Using cooking alkali chemicals (e.g., 0.5% until 1.0% Na_2CO_3 solutions) was highly influential in extracting the tannin (out of the plant parts), as it afforded more significant tannin's solid content and tannin's extract yield than those using just the cooking water liquid. The tannin pigments from the mangrove of consecutively Rhizophora mucronata Lamk., Rhizophora apiculata Blume, and Avicennia officinalis species, extracted using water solvent, exhibited soft brown colors. Meanwhile, correspondingly the mangrove of the same species, which were extracted using the cooking alkali chemicals of 0.5% and 1.0% Na_2CO_3 solutions, brought out the tannin pigments with greater viscosity compared to that extracted by the cooking water, indicating that such tannin pigments contained fewer water-soluble portions and more incredible molecular weight compounds.

It is strongly indicated that the tannin pigments with more excellent solid content and greater viscosity (e.g., containing fewer water-soluble and more excellent molecular weight compounds) contain a high proportion of free phenolic hydroxyl groups and therefore afford the tannin ability to form numerous hydrogen bonds with the OH groups at the fabric fibers (Koch, 1985). Accordingly, the tannin could be more firmly fixed or bonded to the fabric fibers, enabling more excellent pigment resistance against the washing/rubbing/leaching/fading actions and sunlight exposure. The highest solid content (7.866%) in the tannin extracts was achieved from A. officinalis woods using the cooking alkali chemicals of 1% Na_2CO_3 solutions. Meanwhile, likewise, the highest yield of tannin extracts (31.003%) was obtained from R. apiculata woods also using 1% Na_2CO_3's cooking chemical (Suhendry et al., 2017)

The total content or potency of the tannin in the whole individual plant stand species that grew in a particular area for a specific duration period could be predicted by consecutively knowing the number of any individual plant species that existed there, estimating the area where any of the particular species grew there (e.g., in ha), approximating the stand density of any particular species in that area (expressed in the number of individual plants stands per area, e.g., Σ individual plant stands / ha), measuring the average mass density of any of the plant species (e.g., in ton/m3 or kg/m3), and determining the average weight of plant mass per tree stand for each species (e.g., in kg/stand or tons/stand). Therefore, the total content or potency of tannin in a particular area could be assessed. Research results revealed that the potency of tannin extracts in the rehabilitated land located at Rembang reached 105.93 kg per ha. Further, in the third year of growth of the plants, the tannin potency became 146.36 kg per ha (Poedjirahajoe et al., 2011).

Tannin compounds, besides able to exhibit typical colors to the tannin itself or impart various colorations to the fabrics, in facts also afford significant roles in protecting the plants against the vegetation predators (herbivores) and pest attacks as well as in regulating the plant growth. The tannin presence in the young fruit causes a bitter taste to particular animals (including also to human tongue). Changes that occur in tannin compounds which are commensurate progressively with the fruit growth affect significantly the ripening process of the fruits. The tannin content in organic matters (e.g. slabs, twigs, and woods) which are further dissolved in the rain water (along side with the varying humus substances) causes the colors of flooded (stagnant) water at the swamps and peat swamps to appear blackish brown like the colors of tea water, popularly called as black water. It is also the tannin content that renders the water of this kind to taste bitter and rather astringent.

The tannin as natural pigment in the process of imparting the colors and motifs (partial colors) at Indonesia's batik fabrics could play very essential roles. The stages of fabric coloring, as described before, was preceded by submerging/soaking the fabrics in the aqueous pigment solution (including also the tannin), followed afterwards by the fixation stage. Fixation is a process whereby the pigmented-fabrics (using e.g. tannin) was treated with specific chemical salt fixatives, such as kapur/lime ($CaCO_3$), tawas/alum ($Al_2[SO_4]_3$), and tunjung/ferric sulphate ($Fe_2[SO_4]_3$). Fixation stage is very essential, because the fixatives could lock or strengthen the bonding between the pigments and fabric fibers, such that the pigmented-fabric colors would not easily fade away or leach (due to washing/rubbing/sunlight exposure). Also, the fixation can convert the color of original pigment (e.g. tannin) to other typical colors, depending on the kinds of fixative's metal ions (e.g. Ca^{+2}, Al^{+3}, or Fe^{+3}) which react with the pigments. Before conducting the fixation, an aqueous fixative solution should be prepared by dissolving approximately 50 grams of the chemical fixative (e.g. $CaCO_3$, $Al_2[SO_4]_3$, or $FeSO_4$) into one liter of water. The resulting aqueous fixative solution is allowed to settle or precipitate for 24 hours, such that there occur two layers (upper and lower). The upper/supernatant layer (of the fixative solution) which looks clear is further used in the fixation. The fixation is performed by submerging/soaking the pigmented fabrics or yarns in the clear/supernatant fixative solution for approximately 15 minutes. Afterwards, the fixative-treated pigmented-fabrics/yarns are removed and then allowed to dry. The color detection of the dried fixative-treated pigmented-fabrics/yarns could be conducted by submerging/soaking them again in the color-detecting solution for 24 hours. Thereafter, the fabrics/yarns are removed, dried again, and then undergo the color identification test. The procedures of color identification test refer to the RHS (royal horticultural society) color charts. Afterwards, color-identified pigmented-fabrics/yarns are well documented by annotating specific labels. The labels bear the information about e.g. the content of pigments in the fabrics/yarns, date/day/month/year of identification tests, etc.

Imparting the motifs in this regards implies the drawing of batik patterns on the fabrics. For such, before submerging/soaking the fabric in the aqueous pigment solution as described above, the batik crafters or artisans draw the specific patterns (batik motifs) on the upper surface of the fabrics with the wax pen (already filled with a hot melted wax). The wax while still melting could penetrate or pass through the fabric thickness, and therefore the batik motifs will trace or appear on the back (lower surface) of the fabrics. After the drawing of batik motifs all finishes, the entire upper surfaces of the fabrics (except the background or lower surface) are thickly waxed. The waxed fabrics are further submerged / steeped in the aqueous pigment solution (e.g. tannin). In this way, the background or lower surface of the fabrics will get colored (by the pigment), while the waxed upper surface is unaffected. Afterwards, the fabrics are removed from the pigment solution, and the wax (on the upper fabric surface) is scraped with a sharp tool (e.g. a knife). After the scraping, a certain background surface of the fabrics (with traces of the drawn batik motifs) remains unaffected by the coloring pigment solution, while the other background surface (without the traces of batik motifs) is adversely exposed to the pigment and then becomes colored (e.g. partially colored). The further stage is the fixation, followed by other staged activities (e.g. fabric drying and color detection), which are all performed in the same way as those described previously.

As of this occasion, the varying sources of natural pigments which are abundantly available in Indonesia unfortunately have not yet provided maximal solution. Further, despite strong suggestions and appeals that the Indonesia's batik crafters and weavers use natural pigments (instead of synthetic pigments), which are abundant, renewable, and environmentally more friendly, unfortunately the majority of domestic batik crafters/weavers still use synthetic pigments which are mostly imported and moreover environmentally unfriendly. In fact, ironically a part of the Indonesia's wealth of natural pigments has been utilized and produced as usable or ready-for-use pigments by other countries, such as India, Singapore, Malaysia, and China. Accordingly, systematic and thorough attempts deserve carrying out to reduce the dependence of Indonesia's fabric crafters/weavers on the imported synthetic pigments, among others by establishing renewable natural pigment industries. Those renewable natural pigments (with their typical colors) such as chlorophyll (green), anthocyanin (red-violet), xanthohyl (yellow), tannin (brown, reddish brown), and carotene (orange) are abundantly available or existed in the bodies of specific plants as well as of other living creatures (e.g. microorganisms)

Further, in order that the Indonesia's abundant natural pigments could be obtained and produced into ready-for-use pigments in feasible operation (technically as well as economically), then it is necessary to explore the genetic and biochemical aspects of the natural pigment's living sources (especially the plants), which otherwise can bring essential economy values. Accordingly, a breakthrough is urgently performed with respect to the effectiveness, efficiency, and rules to regulate the utilization of those natural biopigment sources. In addition, the involvement of the batik-crafting community and traditional fabric-weavers should get a priority. The intent is in order that the pigment-industry continuation and environment sustainability could be realized.

III. Variations in Colors and Motifs of Batik Fabrics

The traditions performed by the batik crafters and fabric weavers to impart the fabric colors and motifs using natural pigments are by cooking (boiling in the water) the particular parts of the plants (e.g. stem barks, roots, twigs, and plant wastes). More specifically, the traditional batik crafters and weavers boil in the water the plant parts, e.g. barks, leaves, wood slabs, sawdust, and flowers, for a particular duration. In this way afterwards the pigments (e.g. possibly containing tannin, flavonoids, saponine, chlorophyll, and other kinds) inside the plant parts will be extracted out, diffusing into the boiling water and hence causing the water colorations. When, the colorized boiling water becomes thickened or viscous enough, then into which the fabrics or yarns are immersed and subsequently undergo the coloring, with the schematic details illustrated in Figure **1**. This local wisdom/conduct is commonly implemented a lot by the community at the forest village that further becomes a guidance or reference. Further, the guidance/reference is adopted by consecutively the regional authority ; the Indonesia's state-owned forest enterprise (Perhutani) in Central and East Java; and wood-processing enterprises in Kalimantan, Sumatra, Nusa Tenggara, and Bali

Figure 1: Schematic illustration of coloring the fabrics by immersing into the boiling water, already containing the dissolved pigments (previously extracted out of the particular plant parts)

The natural colors of tannin become very essential to seek and arrange the motifs on the tannin-colored fabrics, which should be compatible with the desire by the batik/fabric crafters; and also with the prearranged preference by the users/consumers who already have ordered the batik and other woven fabrics. As such, the yellow, brown, and reddish colors can be arranged and directed based on the crafter experiences. Tannin and other natural pigments (e.g. flavonoid and anthocyanin) belong to the plant's extractives that can impart typical colors to the plants themselves as well as to the pigmented-fabrics. Depending on the species and parts of the plants, flavonoid can create yellow (Rymbai, Sharma, & Srivasta, 2011), red, brown, or blue colors (Rosyida & Zulfiya, 2013); tannin gives brown (Kasmudjo et al., 2008), brownish yellow and reddish brown (Kasmujiastuti, 2014), or citrus-like yellow colors (Rosyida & Zulfiya, 2013); and meanwhile, anthocyanin imparts blue, brown, red, orange, and violet colors to the leaves (Saefudin & Pujilestari, 2015)

Tree roots, barks, stems, and leaves at mangrove plants of Avicennia sp. are prevalently very rich in tannin, phenolic, and flavonoid compounds, where their presence is indicated from results of qualitative (or semi-quantitative) tests on those plant parts, which revealed a strongly positive category [++] to a very strongly positive category [+++] (Harbone, 1987; Hostettmann, 1991). This implies that if a part of those mangrove plants is dissolved in the water, the color imparted by the pigment that diffuses out into the water will appear very strong, i.e. blackish brown or resembling the color of tea water. The presence of tannin and flavonoid in high quantity inside the parts of mangrove plants allows the plant extracts to be used as natural pigment for coloring the fabrics.

The specific colors which appeared at the old teak trees were due to the presence of predominantly anthraquinone (2-mehtyl anthraquinone) in the teak extractives with their content quite great (reaching 13.54%), but those extractives contained tannin only a little (Basri, Balfas, & Dewi, 2013; Setyowati, 2014). Further, the yield (content) of the tannin which was extracted from Acacia mangium tree barks could reach 15-20% (Santoso, 2003). The batik fabrics which were colored with Acacia amngium's tannin were resistant against the high intensity of sunlight, and also against the pH changes (Kasmudjo et al., 2008)

In the experiment after coloring the fabrics with natural pigments, extracted from the teak (Tectona grandis) leaves, two kinds of mordants were separately used, which consisted of alum mordant and acetic acid (vinegar) mordant (Masyitoh et al., 2019). It turned out that the use of vinegar mordant rendered the resulting pigmented-fabrics to bring out brighter color motifs (i.e. 72.22%), compared to that using alum mordant, where the color motifs of the pigmented-fabrics appeared to be less brightness (55.56% and 66.67%). In another experiment (Figure 2), the use of tannins as natural pigment, which were extracted from five mangrove species (Acacia mangium, Terminalia catappa, Rhizophora apiculate, R. mucronata) and (Tectona grandis) could be implemented to create typical color motifs at the woven fabrics as well as at yarns after undergoing the mordanting and fixation (using tawas/alum, kapur/lime, as well as tunjung/ferric sulphate fixatives). This coloring process could be used to impart the basic color motifs as well as supplementing color motifs at the batik fabrics (Saefudin, 2017; Saefudin & Basri, 2021).

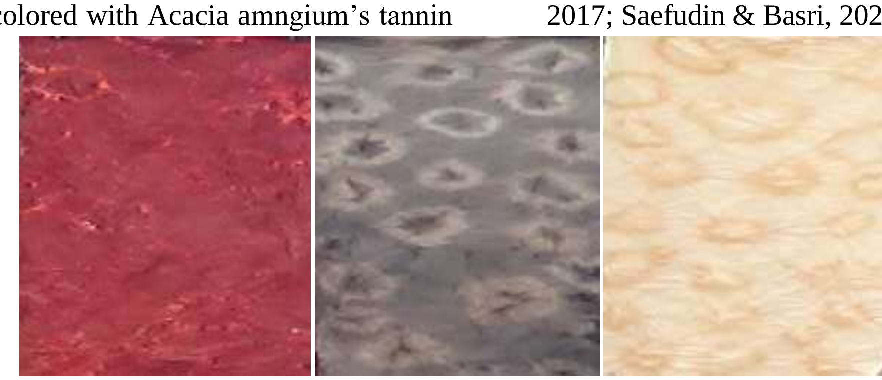

Figure 1. Natural tannin variation were extracted from mangrove species (fixator: alum, lime, and ferric sulphate)

The natural colors imparted by the tannin essentially signify the most dominant elements in the batik motifs, and also in the coloring aspects as well as in the color variations, where the two latter cases are the most important in designing and imparting the batik motifs, especially the inland motifs (Irianingsih 2017). From the research results about the fixation, it could be inferred that different fixatives which were used would bring out the pigmented-fabrics with different colors as well. This case was commensurate with the previous statement which mentioned that different colors at the fabrics could occur, despite using the same pigments, due to the use of different fixatives. The use of fixatives actually aimed to strengthen the bonding (fixing) between the pigment particles and fabric fibers.

Research experiment was already conducted in coloring the fabrics with the natural pigment, followed with the fixation treatment (e.g. using particular fixative solution). It turned out that the use of fixatives caused the color of the pigmented-fabrics not easily to undergo the leaching/fading, and moreover the fabrics were also more resistant to the rubbing/abrading actions, all compared to the leaching/fading and rubbing/abrading resistances of the fabrics without using the fixatives (Saefudin and Basri, 2022). The fixatives employed in this experiment were comprised of tawas/alum, kapur/lime, and tunjung/ferric sulphate. Those three kinds of fixatives were selected because they had been utilized a lot; and moreover the fixative prices were affordable or not too expensive. Meanwhile, the natural pigments which were used for coloring the fabrics (prior to such fixative treatment) were the tannin extracts from three mangrove species, i.e. Acacia mangium, Terminalia catappa, and Rhizophora apiculate.

The sources of natural pigments have already been tested on 73 plant species. As many as 26 species out of them s were identified as natural pigments that could impart yellowish brown, reddish brown, or strong brown color, for use as the basic color of batik fabrics (Saefudin et al., 2018; Prayitno at al., 2012, Poedjrahajoe at al., 2011; Suhendry at al., 2011). Those tannin's natural pigments could be acquired from particular parts of those 26 plant species origins, such as leaves, stems, barks, flowers, and twigs (Table 1).

Table 1. Parts of the particular plant species, regarded as sources of tannin's natural pigments for coloring the batik fabrics and other woven fabrics

No	Species of the plant	Parts of the plants	Colors imparted / appeared on the fabrics
1	Acacia mangium	Tree barks	Strong brown
2	Areca catechu	Fruits, leaves	Brown, reddish brown
3	Artocarpus heterophylla	Tree barks, woods	Yellowish brown
4	Bixa orelana	Tree barks	Blackish brown
5	Caesalpinia sappan	Tree barks	Brown, Yellowish red
6	Camelia chinensis	Leaves, tree barks	Reddish brown
7	Ceriops candolleana	Tree barks	Yellowish brown
8	Cudraina javanensis	Tree barks	Reddish brown
9	Garcinia mangostana	Tree barks, fruit skin	Brown, reddish brown
10	Morinda citrifolia	Tree barks, roots	Brown, reddish brown
11	Pterocarpus indicus	Tree barks, leaves	Red, yellowish brown

12	Koordersiodendron pinnatum	Leaves, twigs	Strong red, brown
13	Musa paradiciaca	Flowers, fruits	Brown, blackish brown
14	Peltophorum pterocarpum	Tree barks, saps/exudates	Brownish red, red-black
15	Pinus mercusii	Tree barks, exudates/saps	Reddish brown
16	Psidium guajava	Tree barks	Greenish brown
17	Rhizopora apiculata	Tree barks	Brown, blackish brown
18	Rhizopora mucronate	Tree barks, woods	Blackish brown, brown
19	Soneratia alba	Tree barks, fruits	Brown, Greenish brown
20	Syzygium conglomeratum	Tree barks	Brown
22	Swietenia mahagoni	Tree barks, fruit skins	Brown, red, yellowish brown
23	Terminalia catappa	Tree barks, leaves	Brown,-black, brownish yellow
24	Tectona grandis	Leaves, Tree barks, woods	Red, brown, blackish brown
25	Uncaria gambir	Leaves, twigs	Strong red, brown
26	.Xylocarpus granatum	Tree barks, fruits	Brownish red, brown

Sources: Jamal (2017); Basri et al. (2017); Saefudin et al. (2017); Susanto, (1980); Prayitno, et al. (2012), Poedjrahajoe et al, (2011); Suhendry et al, (2011);

The natural pigments extracted from the root parts of those particular plant species, for coloring the fabrics, could generate four different varying fabric colors due to the use of different fixatives afterwards (Table 1). As such, the pigmented-fabrics, when treated with tunjung (ferric sulphate) fixative, further appeared with dark fabric colors. In average, the fabrics treated with this tunjung fixative appeared as black until grey colors. Meanwhile, correspondingly, the pigmented-fabrics treated with tawas (alum) fixative brought out the fabric colors which were brighter than the original colors of the pigmented fabrics (i.e. without fixative treatment). Further, the pigmented-fabrics treated with kapur (lime) fixative appeared with the colors toward blackish (Irianingsih , 2017).

In other variations, the fixation treatment with tunjung/ferric sulphate fixative brought out the pigmented-fabrics with rather dark (blackish) colors; while correspondingly the treatment with kapur/lime fixative rendered the fabrics to appear toward brown colors; and ultimately the treatment with tawas/alum fixative caused the fabric colors to appear brighter than original colors of the pigmented-fabrics (i.e. without fixative treatment). The dark/blackish colors of the pigmented-fabrics (after the treatment with tunjung/ferric sulphate fixative) indicated the reaction that occurred between the tunjung/ferric sulphate fixative and the pigments (plant's extractive) apparently produced complex salts. The complex salts, being large-sized compounds, further could provide as if a bridging (intermediary) between the pigment and the fabric fibers (Brit, 1980). This situation accordingly created a strong bond (fixing) between the pigment (plant's extractive) and the fabrics, thereby eventually imparting also strong colors (dark/blackish) at the tunjung/ferric-sulphate-treated pigmented-fabrics. Meanwhile, the different colors (not dark/blackish colors, but brighter colors) of the pigmented-fabrics after the fabric treatment with lime fixative as well as with alum fixative supposedly did not produce complex salts, thereby causing the bonding between fabric fibers and the pigment (plant's extractive) to be not too strong. However, the lime fixative as well as alum fixative could afford ionic bonding with either fabric fibers or with the pigment (plant's etractive), thereby still imparting the bonds between the fabric and the pigment (plant's extractive), despite being not as strong as the bonds exerted by the complex salts (Prayitno, 2012).

Further, it can be inferred that the treatment with such different fixatives revealed that the use of tawas/alum fixative brought out the pigmented-fabrics with colors toward the brightest, followed in decreasing order by using consecutively kapur/lime fixatives (less brightness) and then tunjung/ferric sulphate fixative (the least brightness or the darkest). In another case, different kinds of fabrics (e.g. mori, cotton, cotton yarns, and wool), but colored with the same natural pigment and then also treated with the same fixatives, could bring out the pigmented-fabrics with different colors. This difference could occur because mori fabrics, cotton fabrics, and cotton yarns are made up of mostly cellulose polymers, thereby rendering the fabrics/yarns to easily adsorb the natural pigments (and also synthetic pigments). However, the wool is composed of mostly protein polymers which could have different characteristics in interacting/adsorption with the natural pigments (and possibly also with synthetic pigments), compared to those of the cellulose-based fabrics. All of these phenomena could therefore bring about the different colors as appeared at the resulting pigmented and fixative-treated fabrics. For these reasons, the fabrics composed of cellulose fibers are widely used by the batik crafters and weavers (Pujilestari, 2014)

IV. Chances and Challenges

Several parts of the plant species, from which their pigments are extracted (presumably containing the tannin) by the crafters for coloring the batik fabrics and other woven materials, are among others the leaves of nila (Indigofera tinctoria) trees and barks of soga tingi (Ceriops candolleana) trees. Other plant species with their particular plant parts supposedly rich in tannin pigments are woods of tegeran (Cudraina javanensis) trees and Camelia chinensis trees, roots of mengkudu (Morinda citrifolia) trees, barks of soga jambal (Pelthophorum ferruginum) trees and kesumba (Bixa orelana) trees, and leaves of jambu biji (Psidium guajava) trees (Susanto, 1980; Saefudin, Basri, & Santoso, 2018). Although the natural pigments (including the tannin) have been known a lot by the crafters of fabric crafters and weavers, unfortunately only a few of the fabric crafters/weavers are interested to utilize those natural pigments. The main constraints and hindrances that only a few select and utilize the natural pigments lie among others in the limitation of pigment varieties which are ready for use; lack of knowledge owned by the fabric crafters/weavers about the pigment's raw material; and insufficient technology about the pigment processings which are ready for use or easily adopted. Besides, the availability of natural pigments from the plants is very affected by the seasons, environments, and collecting-workers, thereby causing the supply of natural pigments to the fabric crafters/weavers not continuous. Despite their drawbacks, the natural pigments (especially the tannins) actually afford high market potencies as Indonesia’s superior commodity to enter into the global markets, which have attractive powers with unique, ethnical, and exclusive characteristics

The challenges for the researchers are that they should explore the sources of tannin and other natural pigments such that those pigments eventually become usable or ready for use. The creativity of the fabric crafters and weavers to seek fabric motifs which should conform to the tastes and desires by the users can become self separate problems. These classical problems also occur in the villages where the batik crafters and weavers produce the batik fabrics and other woven materials in many Indonesia’s provinces. The batik crafters and weavers should obtain the pigment’s raw materials from their plant sources, which are advisably fast growth, short ages, available in considerable quantities, and easily acquired from their home vicinities

The aspects about natural pigments (used for coloring the fabrics) that should be developed immediately in the future are especially the motif designs. This is because those motif designs are very essential to anticipate local and global markets. Kinds of laboratory researches that use experiment methods should try hard to seek the efficacious formulae for developing the unique natural pigments in the vicinity of crafter community. The process of acquiring pigments is performed through commonly the extraction by cooking or boiling the pigment sources (e.g. plants) in the water at elevated temperature (about 100^{o}C). In order that the pigment-generated colors could last long at the fabrics or could be protected from the washing/rubbing/leaching/fading actions and the sunlight exposure, the fixation treatment should be implemented (e.g. using tawas/alum, kapur/lime, and tunjung/ferric sulphate fixatives). Wood wastes generated from tree-logging/felling operation in the forests and from wood processing factories, as described before, are allegedly rich in natural pigments such as tannin, anthpcyanin, carotenoid, xanthophyl, quinone, etc. Evaluation results reveal that tannin was the most positively prospective as natural pigments for coloring purposes (e.g. fabrics) which seemed expectedly applicable for coloring purposes (e.g. batik fabrics) with satisfactory results

The tannin’s natural pigments could be obtained from the extraction of the particular parts of especially forest plants (e.g. woods, tree barks, roots, flowers, seeds, and saps/exudates). Therefore, it appears that the potency of natural pigments from the forest areas is quite huge. Unfortunately, the data and information about the forest’s pigment potency are still lacking or inadequate, and moreover have not yet been much assessed. The utilization of tannin as natural pigments from the forests could become an added value for the tannin’s plant sources and essential information for the community who love the Indonesia’s batik fabrics and other woven materials.

The vegetation-derived tannin from the particular plant species, besides being used as imparting the colors to the fabrics, has long been utilized as a tanning agent to convert the animal bones and skins (i.e. under high temperatures) into stable leathers, which are strong and elastic/flexible. This is because the tannin, as described before, which belongs to one kind of the polyphenolic compounds as high-molecular weight macromolecules contains high proportion of free phenolic hydroxyl groups which therefore affords its ability to form multiple hydrogen bonds not only with the fabric fibers but also with the hydroxyl groups and other polar groups (e.g. carboxylic) at the proteins of animal bones/skins, thereby creating also strong bonds between the tannin and the animal bones/skins. Further, in the plant leaves, it is strongly presumed that the tannin is also contained inside along side with the chlorophyll, thereby allegedly causing the color changes of the leaves themselves as well as of the tannin-colored fabrics. Further, the leaves of the same plant species could also sooner or later undergo color changes. This occurs, because the chlorophyll in the leaves absorbs the red-colored and blue-colored light from the sun ray, which hits those leaves. Such color changes are attributed to the almost continuous use of chlorophyll in the plant photosynthesis during the daylight, thereby gradually decreasing the chlorophyll capacity to absorb the sunlight and hence diminishing the intensity of chlorophyll's green colors. On the other hand, the carotene in the leaves is more stable than the chlorophyll, thereby enabling the reddish orange colors of the leaves to stay longer, although the leaf's green colors disappear (Anonim, 2006). Other factors that could affect the pigment stability are among others the presence of cation, oxygen, pH, sulphur dioxide (SO_2), proteins, and enzymes.

The aspects of tannin together with other natural pigments (e.g. chlorophyll, anthcyanin, flavonoid, etc) in the plant parts have attracted enthusiastic attention by many genetic experts. This is because the pigment presence in such might have meaningful relations with the various morphology aspects among the different plant species, which are still close relatives in the same genus. That information is very essential for the taxonomist to determine and to combine into one group the evolution lines of the plants. Further, the forming of flavonoid pigment in the plants could be induced by the blue-colored light radiation (e.g. from the sun ray) to increase the plant resistance against the ultraviolet radiation.

CONCLUSIONS AND SUGGESTIONS

Indonesia is endowed with abundant availability of natural pigments, concerning huge pigment potencies, numerous pigment varieties (e.g., tannin, flavonoids, saponine, anthocyanin, chlorophyll, and carotenoid), as well as substantially varying colors of the pigments (e.g., green, red-violet, yellow, brown, reddish brown, and orange).

Those natural pigments could be obtained from almost any part of the plants, such as tree barks, roots, leaves, fruits, seeds, exudates, and fruit skins, though commonly the extraction process.

Crafters and weavers can utilize those natural pigments for imparting colors and motives on batik fabrics or other woven materials. Also, natural pigments are beneficial, renewable, and more environmentally friendly.

Unfortunately, most of Indonesia's crafters/weavers of batik fabrics and other woven fabrics still use synthetic (inorganic) pigments, which are mostly imported and environmentally unfriendly.

One way to overcome that unfortunate situation is by establishing industries to produce natural pigments that are usable or ready for use. For such, it is necessary to explore the genetic and biochemical aspects of the natural pigment's living sources (especially the plants)

In order that the ready-use produced natural pigments could satisfy the users/consumers, especially the crafters/weavers of batik fabrics or other woven materials, among others, the imparted colors on the pigmented-fabrics/cloths should last long, e.g., proved through the test color resistance against the leaching/fading due to mainly the washing, rubbing, and sunlight exposure.

For such, the coloring of fabrics (with natural pigments) should be performed in stages incorporating the fabric mordanting, coloring, fixation, and color detection essentially (on the produced pigmented fabrics).

Tannin is the most prominent natural pigment (e.g., tannin, flavonoids, xantophyls, anthocyanins, and chlorophyll). The common potential sources of tannin are the tree barks of mangrove plants and also, allegedly, the wood wastes. The wood wastes in this regard are abundantly generated from the tree logging operation in the plantation forest (predominantly of acacia-tree species) and acacia-wood processing (pulp/paper) industries, all conducted by the pulp/paper factory.

Those acacia-related wastes are mostly left abandoned and accumulated on sites, potentially arousing environmental concerns. The wood wastes with acacia species origin are prevalently rich in tannin.

The tannin which is extracted from the plant sources (e.g., abundant wood wastes) by the mixture of hot water and alcohol/acetone; and by 0.5-1.0% alkali (Na2CO3) solution could bring out the tannin extract with favorable qualities (e.g., high yield, high solid content, high molecular weight portion, high free phenolic hydroxyl groups, and high viscosity).

The favorable-quality tannin extract is such that it can strengthen the bonding or fix between the tannin pigments and fabric fibers. In this way, the colors of tannin-pigmented fabrics (e.g., batik fabrics) could last longer and afford high resistance against washing/rubbing/leaching/fading actions and sunlight exposure.

The favorable-quality tannin can also form insoluble colloidal compounds with the proteins of the animal skins (hides), thereby converting the hides into durable leathers that are solid and elastic/flexible (as tanning agents).

Therefore, tannin as a renewable natural pigment seems very prospective to be developed in imparting the colors and motifs on the fabrics (e.g., batik fabrics). In this way, these attempts can encourage the fabric crafters/weavers to use tannin as renewable and more environmentally friendly natural pigments (instead of the synthetic/inorganic pigments, being environmentally unfriendly), and further, it could reduce the dependency (on the fabric crafters/weavers) on the imported synthetic pigments.

Meanwhile, the animal-leather industries have long utilized tannin as the tanning agent, which could be extracted from particular plant parts and many wood wastes.

Accordingly, attempts to utilize those wastes into tannin as natural pigment for coloring the fabrics (e.g., batik) and as tanning agents for leather processing, besides reducing environmental concerns, can also mean imparting added values to the wastes.

However, tannin and other renewable natural pigment also have weaknesses or drawbacks. The drawbacks comprise, among others, that the availability of pigment’s living sources (either plants or wood wastes) is often not continuous, affected by seasons, environments, the collecting workers, and their locations scattered.

This unfortunate situation deserves immediate and thorough solutions, such that renewable natural pigments (including tannin) can eventually become extensively ready for use and be utilized commercially worldwide.

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