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Research Article

Natural color performance from nine species origins of lowland wood wastes

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Abstract

In the past, natural pigments derived from plant-biomass wastes had been widely used for coloring purposes (e.g. fabrics/batik), before finally getting pushed aside by the imported synthetic/inorganic pigments. Thorough attempts to revive the use of such natural pigments then deserve carrying out. Relevantly, conducting storage of liquid/wet or dry natural coloring pigments, extracted from wood wastes; and use of fixatives for batik fabrics intended to impart fabric-color variations. Wood wastes (i.e. leaves and barks) were obtained from nine plant species (*Acacia mangium* Willd, *Avicenia marina L, Caesalpinia sappan* L, *Ceriop tagal* Perr. CB. Rob, *Pelthophorum ferruginum* Benth, *Rhyzophora mucronata* Lamk, *Tectona grandis* Linn.f., *Terminalia catappa* L and *Xylocarpus granatum* KD Koenig.), prevalently growing on plain/lowland areas; and their wood portions commonly utilized by wood industries, which generate similar wastes enormously. The pigments in liquid/wet and then in dry forms were produced from those wastes. The production process of pigments adopted traditions by local batik crafters in Bogor. Variations occurring in fabric colors could be due to fixatives, i.e. lime ($CaCO_3$), ferrous sulphate/tunjung ($FeSO_4$); and various wastes. Six-month storage insignificantly changed the coloring qualities of natural coloring pigments applied to fabrics, compared to those of corresponding initially produced pigments. Leaf wastes always contained green-colored compounds, while bark wastes appeared aptly brownish in color. Color performances of bark extracts at fabrics with ferro-sulfat fixative brought about strong grey colors, and with lime fixative produced brown colors. Coloring of fabrics using leaf waste extracts with ferro-sulfat fixative aptly exhibited greenish grey; while with lime fixative became reddish brown. Testing results on the leaching resistance of pigment-colored and fixative-treated fabrics against rubbing and exposure to sunlight revealed a very good category (4-5), but against detergent, washing was very low (2-3).

More Information

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Introduction

Batik crafters located in coastal areas, particularly on northern Java island exhibited more liberal patterns, and fabric-coloring results were more varied. Non-figurative flora and fauna often become their motives. Variations occur more of the color aspects to conform to the desired markets, which are affected by foreigners; and were brought in by traders in the olden era. Initially, crafters of batik fabrics and traditional woven items from the coastal areas utilized many colors of natural pigments from particular plant species which grow profusely on plain areas, until those pigments were finally pushed aside or sustained the competition by imported synthetic coloring pigments [1]. The tradition of utilizing those natural pigments has spread widely in coastal regions in Java, Sumatera, Kalimantan and even East Nusatenggara.

The potency of natural pigments from plant biomass wastes (including wood wastes) in Indonesia could be quite immense, commensurate with the vast areas of forests and large production of wood industries. Currently, the generated wood wastes and other plant biomass wastes (such as leaves, wood pieces, tree bark, branches/twigs and sawdust) at the coastal plain areas are potentially abundant. However, efforts of putting those wastes to use are still limited, and their utilization is not yet maximally carried out, thereby rendering the wastes mostly discarded. Although local fabric crafters have recognized and identified natural pigment-sourcing plants, only a few of them utilize those plants. This inconvenient situation occurs because among others, color variation is still limited and the availability of raw materials for natural pigments is still not yet ready for use in other words it necessitates special preliminary processes to convert the pigment-containing materials to readily applicable natural pigment (usually in liquid/wet forms) for coloring of batik fabrics.

Despite some drawbacks, natural pigments could in fact afford high market potency as a superior commodity for Indonesia's products. Fans or enthusiasts of batik as well as traditional woven fabrics favor and enjoy very much products with ethical, futuristic and exclusive values in global markets [1,2]. Accordingly, batik crafters and industries should be able to read the taste trends and demands of consumers or customers. Further, intensive exploration and testing, in addition to enriching natural pigment sources, are expectedly able to add pigment color variations to batik fabrics or other woven items.

Felling and cutting down plant trees of Rhyzoporaceae and Fabaceae family, which prevalently grow in plain-area forests, could generate a lot of small-size plant portions as biomass wastes that have already been known as potentially rich sources of coloring matters, such as tannin, flavonoid, saponine, anthocyanin, and polyphenols alike [3,4]. Those compounds are quite essential as basic ingredients for the manufacture of natural pigments for batik and other woven fabrics in Indonesia. Natural pigments inherently already contained in those biomasses coulfigd appear or migrate out through heating as well as storage. The heating or storage could affect or trigger physiology or other biological processes in the biomass tissues, such as degradation, respiration, decomposition, and remediation that further bring about chemical changes in those compounds. The chemical changes further convert the complex chemical and high molecular weight compounds in biomass tissues to simple structures as well as lower molecular weight components [5].

Color variation exerted in natural pigments should be thoroughly well kept, actively maintained, or intensively attempted in order able to compete or challenge with synthetic pigments, which are usually more complete and chemically stable. Continual efforts in production technology of natural pigments from biomass wastes are worth carrying out to cope with those challenges. Production of natural pigments could be implemented to bring additional income to the fabric crafters in the vicinity of wood industries. In addition, natural pigments extracted from biomass wastes, such as leaves, wood slabs, twigs and stem barks could serve as potential alternative pigments, which are apparently more environmentally friendly and moreover renewable, than synthetic/inorganic pigments. Relevantly, the following is put forward the topic about effects of storage and fixative agents on performances of natural pigments for especially coloring batik fabrics, whereby the pigments were extracted from biomass wastes (i.e. leaves and barks) of nine old-aged plain-area tree species; and their wood portions are commonly utilized by wood industries.

It is essential to know that the tissues in the plant bodies (e.g. woods, barks, roots and leaves) of the standing trees (including also the felled/cut-down trees), no matter where the trees grow (in the mountains, plateau, lowland, etc) comprise millions of tiny-sized cells, either living or non-living/dead cells. The chemical compounds in those plant cells consist of those that make up the cell walls (e.g. cellulose, hemicellulose, and lignin) and those contained in the non-cell wall (i.e. cell lumens and wall pores). Further, the compounds contained in the non-cell walls can be among others extractives (mainly), protoplasms, and cytoplasms. The kinds of extractives could be varying, either qualitatively or quantitatively, with various plant species origins. It is the extractives that impart the plant cells (including those in living/standing trees and wood wastes) with e.g. specific smells, creating toxic effects, and exerting various attractive colors (as natural dyes/pigments). Further, the living cells inside the wood wastes (despite being already separated from their living/standing trees) remain alive and active to perform physiological processes (such as respiration, photosynthesis, metabolisms, and other enzymatic actions) as the uncertain time proceeds ahead. Accordingly, due to such, the extractives inside could undergo chemical changes (degradation, oxidation, hydrolysis, decomposition, etc), thereby causing the alteration in their extractive/pigment colors [6,7].

Methods of exploration and coloring application

Exploration of biomass wastes and storage of natural pigments

Biomass wastes as leaves and barks of *Acacia mangium* Willd*, Avicenia marina* L.*, Caesalpinia sappan* L, *Ceriop tagal* Perr. CB. Rob*, Pelthophorum ferruginum* Benth*, Rhyzophora mucronata* Lamk, *Tectona grandis* Linn.f., *Terminalia catappa* L, and *Xylocarpus granatum* KD Koenig plant species were procured and gathered from West Kalimantan; Karimun island; and Cibinong Science Centre, Cibinong, Bogor. Those seven species prevalently grow in plain areas, and their wood portions are commonly utilized by wood-processing industries. In wood processing, biomass wastes such also as leaves and bark are often generated in great amounts. The biomass wastes already selected which comprised barks 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.

Stages in the production of biomass-waste-based natural pigments began with waste sorting and dimension reduction to become small pieces 1 cm - 2 cm in size and then cleared of foreign matters to prevent the waste pieces from rotting or decaying. The selected biomass-waste pieces (e.g. leaves and barks) regarded as qualified were obtained as described before from those seven physiologically old-aged plant species. The further stage was extracting liquid-shape colored matters (or coloring-pigments) from those waste pieces, 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 °C - 80 °C 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), then could be ready directly for use (coloring the fabrics) or else stored for a particular duration.

During the storage, the screened coloring pigment solution (initially/freshly wet produced pigments) was provided with packaging; or placed in tightly closed jerry cans or plastic containers. The storage room should be such that the room was impervious to sunlight, with moderate ventilation and under normal temperature. While in the storage, a particular amount of the coloring pigment solution (wet form) could be taken apart and prepared. On the other hand, making dry natural pigments could be performed by further evaporating the already-prepared-coloring fresh pigment solution. The results of evaporation are weighted as practical dyes for trading and good durability in storage. This evaporation process could take considerable time, but afforded some advantages, whereby the obtained dry natural pigments could last longer or be more durable during storage, easily packaged and efficient for trades.

Application of natural coloring pigments

Application of coloring the fabrics by the crafters could be directly carried out, after the available sample of screened coloring-pigment solution (as previously prepared, particularly in wet form) went through several stages, as follows:

1) Bleaching or whitening of cellulose fibers of the fabrics using aqueous hydrogen peroxide (H_2O_2) solution, with a cooking system;

2) Mordanting process on the whitened fabric fibers to coat the fabrics with oxide metals (in mordanting ingredients) such that the coated fibers could adsorb or exert high affinity to the pigment colors;

3) Immersion process in soaps/detergents, whereby fibers of the mordanted fabrics were immersed in TRO (turkey red oil soap)/detergents to remove oil spots on fiber surfaces;

4) Immersion process of spot-removed fabrics in natural-coloring pigment solution (either as initially/freshly produced pigments; or as the aged pigments, after being stored for a particular duration, e.g.. days, months, etc) to impart fabric colors;

5) Fixation process, whereby the pigment-colored fabrics were treated with lime ($CaCO_3$) and tunjung ($FeSO_4$) fixatives; or combination to strengthen the bonding between the coloring-pigments and fabric fibers, such that the fabric colors would not easily fade away or leach;

6) Washing the fixative-treated fabrics with clean water to thoroughly remove residual fixatives or any other related chemicals;

7) Drying process, whereby the washed fabrics were placed in open-air places but under the roof in order that the fabrics while drying were not directly exposed to sunlight.

Application of coloring the fabrics, freshly, after the pigment solution was produced, then screening it through the filtering cloth and allowed to cool down for 24 hours. Immediately applied for coloring the fabrics, after consecutively 3-month and 6-month storages of individual pigments; and then immediately each applied for coloring fabrics;

Testing on qualities of natural-coloring pigments

Testing of the fabrics after the coloring by natural pigments/extracts was conducted at the Center for Testing and Controlling of Merchandise Qualities, under Indonesia's Ministry of Trade and Industry, Jakarta. Natural coloring pigments in wet (solution) form as well as in dry forms were prepared and then attempted in such a way optimally that they could meet three standard paradigms, which comprised quality, safety and efficacy. In addition, the coloring pigments should have their own specifications, particularly regarding color standards, color consistency and other related essential information. The nature of waste toxicity was based only on literature and the species was non-toxic based on the assessment of the user community.

Quality testing in this regard intended to examine the resistance of coloring pigments, either individually or already adhered (stuck) to the fabrics, against the destruction by microbes and by the treatment imposed on pigments as well as on pigment-colored fabrics. Another intention was to prevent or hinder enzyme actions on the active ingredients in natural pigments such that unwanted decomposition or dissociation would not occur at those ingredients. The related enzymes especially peroxidase, hydrolase and isomerase could otherwise take a significant role in the dissociation/decomposition of essential chemical compounds in natural pigments, such as phenols, polysaccharides and alkaloids.

Accordingly, it still urgently necessitated further thorough research to complete the quality requirements for those natural pigments.

Leaching tests are intended to evaluate or assess the color-leaching resistances or qualities of natural-pigment-colored materials (e.g. colored batik fabrics), further compared with the color-leaching resistance of the standards. The materials tested could involve several prototype items (e.g. batik fabrics, weaving yarns, embroidered fabrics, and other woven items) which have been colored with natural-coloring pigments (in wet/solution as well as in dry forms), with or without further fixative treatment. The testing parameters for assessing the color-leaching resistances (of the pigment-colored materials) could cover those against the allegedly leaching inducers, i.e. washing (a), rubbing/ironing (b), acid sweat (c), base sweat (d) and exposure to sunlight (e).

Results and discussions

Exploration of biomass wastes and storage of the obtained pigments

The obtained initial/fresh natural-coloring pigment solution (in liquid form), which as described before originated from plant biomass wastes (e.g. leaves and barks), then cooked/extracted in water at a 1:4 ratio (kg/liter), while partly evaporating until the residual aqueous volume became one-third, further allowed to cool down for 24 hours and ultimately passing through the filtering cloths, was after all ready for the application as coloring of the cellulose-based woven items, such as writing batik fabrics, weaving yarns, embroidered fabrics, and traditional printed batik fabrics. As such, yields of the extracts, as natural coloring pigments, from various biomass waste origins ranged about 3.0% - 3.6% (w/w, over-dry basis). Those extract yields in this research (Table 1) were considerably lower because of different solvents than the yields from the bark and leaf portions of the similar nine old-aged plant species in the previous research results. As such, those bark and root portions afforded the yields of dry extracts in the range of 11% - 28% [8].

The production process of such initial/fresh wet natural coloring pigments (in dilute extract solution form) could be continued until the pigment formed viscous (thick) extract paste or dry extract forms (Figure 1). In dry extract forms, also as described before it afforded advantages in that they could last longer or prolong their durability during storage, were easily packaged and rapidly delivered to batik fabric and other weaving yarn industries. Classical problems encountered by batik and weaving crafters are the unavailability of natural-coloring pigments which were ready for use. Accordingly, those crafters forcibly obtained the pigments directly from nature in their surroundings, concurrently while they were working; and the obtained natural pigments were frequently limited with respect to color variations as well as volumetric quantities.

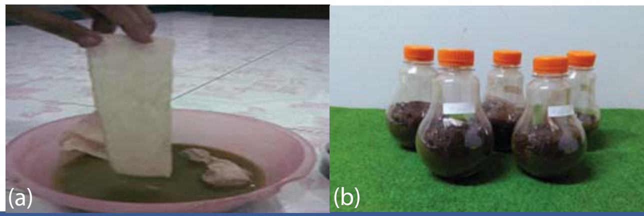

Figure 1: Production of natural coloring pigments, extracted from plant biomass wastes, in wet/liquid form or aqueous pigment solution (a); and in dry extract form (b).

Table 1: Yields of extracts, as natural-coloring pigments from biomass wastes

No	Plant species origins	Extract yields (%)*)
1	*Acacia mangium* Willd	8,3
2	*Avicenia marina* L	8,5
3	*Caesalpinia sappan* L	8,0
4	*Ceriop tagal* Perr.CB. Rob	10,6
5	*Pelthophorum ferruginum* Benth	10,5
6	*Rhyzophora mucronata* Lamk	9,6
7	*Tectona grandis* Linn.f.	9,4
8	*Terminalia catappa* L	8,6
9	*Xylacorpus granatum* KD Koenig	8,4

Remarks: *) average of 5 replications (leave + bark) ; oven dry basis (w/w)

Many ethnicities throughout the world including in Indonesia have utilized particular plant portions as natural coloring pigments. For such, they utilized particular plant portions, e.g. nila (*Indigofera tinctoria*), malam (*Pterocarpus indica*) woods, ketapang (*Terminalia catappa*) roots, teak (*Tectona grandis* L) leaves, annato (*Bixa orellana* L.) seeds, tea (*Camelia sinensis*) leaves and pinang (*Areca catechu*) fruits (Prima & Amar, 2013; Rosanah, et al. 2013 & Husodo 1999). Research results have found out that the specific essential formulae of natural-coloring pigments for coloring nature fibers were in fact previously already acquired through the information or knowledge of batik crafters and other-woven-fabric crafters. For example, red color was acquired from secang woods; yellow color from tegeran woods and mengkudu root barks; black color from acacia gunung trunk barks; brown violet color from mahoni trunk barks and weak brown color from teak leaves.

Particular portions of plant species, such as kayu tinggi (*Peltophorum pterocarpum*), soga jambal (*Pelthophorum ferruginum*), *Rhizopora apiculata, Ceriop tagal* and ketapang (*Terminalia cattappa*) were frequently utilized as mangrove-based natural coloring pigments by batik crafter community. The utilized plant portions comprised among others wood slabs, defective trunks, twigs, and barks. In fact, wood portions of those plant species were essentially required by wood-related industries for construction materials. Meanwhile, wood portions of particular plant species especially *Rhizophora apiculata* and *Ceriops tagal* were utilized as active charcoal and firewood. Other plant portions such as twigs, sawdust, leaves, roots, trunk barks and fruits were just discarded or abandoned on sites as biomass wastes. Consequently, those biomass wastes could otherwise potentially provide beneficial chances for the production of more natural pigments. A variety of biomass wastes from various tree species that grow prevalently in plain areas was considered beneficial to be utilized as natural pigments, which could afford high economic values and seemed environmentally friendly as well as renewable [2].

Referring to previous research results, there were a lot of plant biomass wastes that could serve as essential pigment sources, allegedly rich in tannin, flavonoid, anthocyanin and polyphenol compounds alike [4]. Particular wood extractives in plant tissues could impart specific colors to the plants or woods themselves. For example, flavonoids provided red, yellow, brown, or blue colors to woods; while correspondingly tannin of morin kinds imparted woods with citrus yellow colors. Roots, trunks, trunk barks and leaves of mangrove trees from *Avicennia marina*. were very rich in tannin, phenolic and flavonoid compounds [9,10]. Teak leaf wastes extracted with water released particular extracts which exhibited specific brownish-red colors [11]. Those specific brownish-red colors were indicatively attributed to the presence of anthraquinone (2-methyl anthraquinone) in the leave extracts. Extracts from teakwood sawdust contained as much as 13.54% portion as anthraquinone compounds [12].

Application of natural pigments at fabrics

Coloring of fabrics using natural pigments extracted from biomass wastes of plain-area-growing plants brought out fabric colors that were varying, depending on plant species origins and concomitantly with kinds of fixatives. Pigments extracted from leaves using ferro-sulphate fixatives aptly resulted in fabrics with greenish-grey colors; likewise using lime fixatives brought out reddish-brown-colored fabrics. Results of the production process of as many 38 kinds of liquid extracts as natural coloring pigments have been applicable for the coloring of batik fabrics, embroidered items and weaving threads. Further, application results on the coloring of batik fabrics with various natural extract pigments should be compared to each other. For example, comparing the coloring results on batik fabrics was performed between traditional ways, the one that used coloring-extract solution (wet form); and the other one that used dry coloring extract (dry form). In equal concentrations of the coloring-extract solution, the resulting color variations on fabrics remained unchanged or not so contrasted (between wet pigment form and dry pigment form origins), but strongly dominated by brown colors. Brown-colored fabric always appeared due to the predominantly tannin content in natural pigments (Figures 2,3).

Diverse variations in fabric colors could occur, depending on frequencies of fabric immersion in natural pigment/extract solution, the concentration of extract solution, and the mixture of various coloring extracts from different biomass waste origins (Table 1 and Figure 4). Pigment extracted from bakau minyak (*R. mucronata* Lamk) barks, immersion frequency: 3 times in an aqueous solution of extracts, fabric color became black-red or red. Fabric color variations were also caused by the mixture of coloring extracts from consecutively bark wastes of *C. sappan*, *R, mucronata* and *Xylocarpus granatum* KD Koenig species (Figure 5). Application of natural colors mixture at primissima fabrics was visual of waste leaves pigments. Pigment extracted from *A. mangium* bark, fabric immersion frequency: 7 times in aqueous extract, lime fixatives (left) and ferro-sulphate fixative (on the right Figure 6). The other performance of fabric colors with lime fixation and color ultimately appeared greenish brick-red of *X. Granatum, C. Tagal, and A. mangium* (Figure 7).

The performance of fabric colors that used natural pigments was also determined by the kinds of fixatives (Table 2, Figures 2). The use of lime and tunjung fixatives could exhibit weak to strong color variations on the extract-colored fabrics. The frequency of fabric immersion in coloring-extract solution could also affect how strongly the natural coloring-extracts were bonded or adhered to fabrics (Table 2, Figure 2). For example, the coloring of fabrics with allegedly tannin-predominated extracts initially imparted brown fabric colors, but after the tunjung fixative treatment coupled with many frequencies of fabric immersion in pigment (extract) solution, hence it caused blackish colors on fabrics. Conversely, when lime fixative was used, the fabric color became weak brown; while with lime fixative, it brought about yellowish-brown-colored fabrics [14]. Activities of exploration on biomass wastes and testing on the corresponding obtained pigments (waste extracts), besides enriching sources of natural pigments with their varying colors, were also able to know what plant species and which plant portions could produce natural pigments with remarkable and unique qualities. The obtained aqueous natural pigment/extract solutions (in wet form), after being screened using filtering cloths, were readily and immediately applicable for coloring the fabrics (Figures 1,2).

The storage of liquid natural coloring extracts (pigments) could provide a pigment reserve or else be directly used by the related fabric crafters. The degradation or decay of the coloring extracts (particularly in wet or liquid forms) by microbes or other microorganisms often affected the appearance of extract/pigment colors and this usually occurred due to inappropriate pigment storage. Storage is intended to prevent or inhibit the coloring pigments from the degrading/destruction action by enzymes (released by the microbes/other microorganisms) on active compounds in the pigments, thereby avoiding undesired pigment decomposition. The enzymes that took great roles in such degradation were especially oxidase, peroxidase, hydrolase and isomerase which brought about the dissociation/decomposition of pigment-active compounds such as phenol, polysaccharides and alkaloids [8,13]. The storage for three to six months strongly indicated that the qualities or coloring

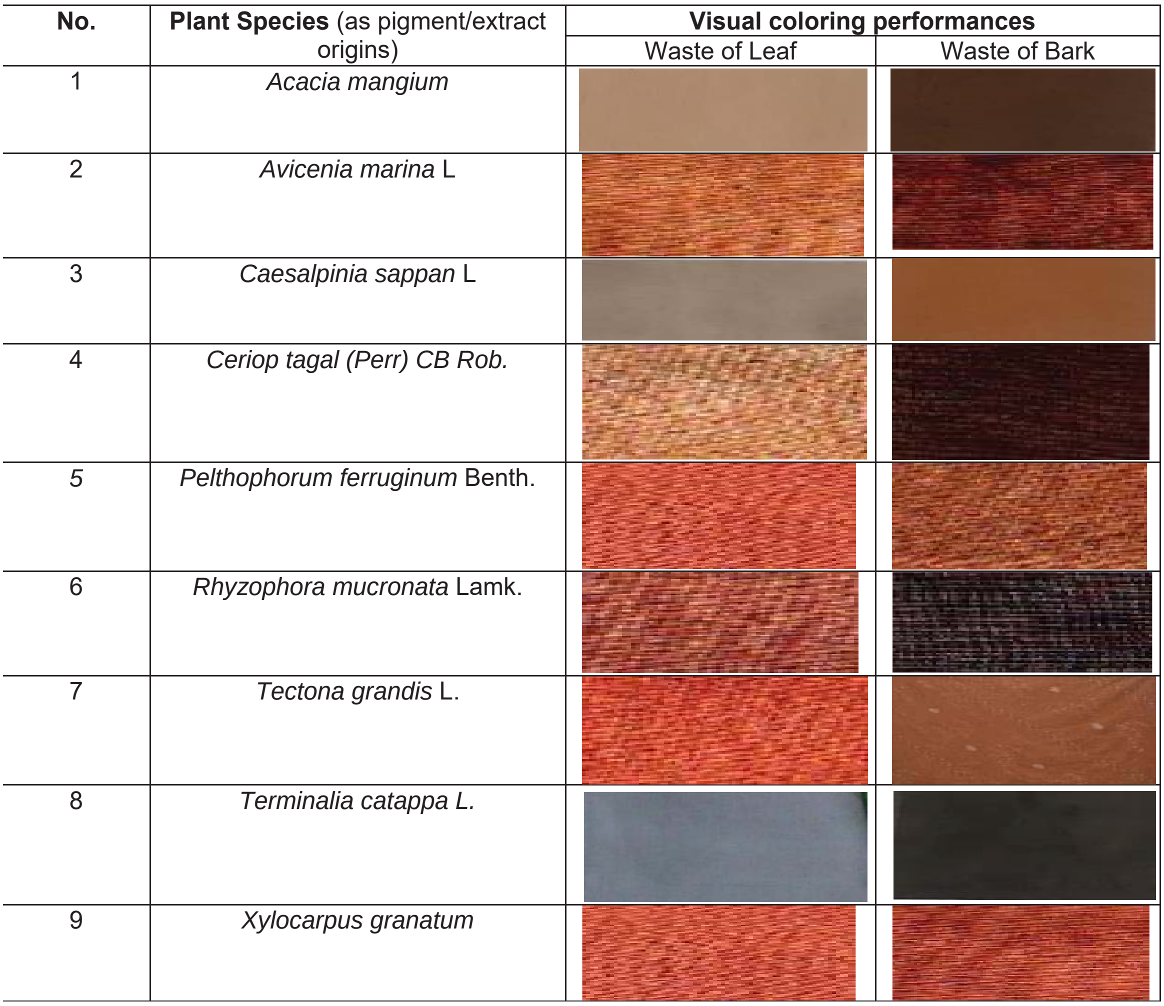

Figure 2: Color variations at fabrics due to fabric immersion in pigment (extract) solution, 10 immersion frequency and kinds of waste agents. Remarks: [1)]Initially/freshly, after the pigment solution was produced, then passing (screening) it through the filtering cloth, and further allowed to cool down for 24 hours, then immediately applied for coloring the fabrics; [2,3)] After consecutively 3-month and 6-month storages of individual pigments; and then immediately each applied for coloring fabrics;

Table 2: The color-leaching resistance of the natural coloring-pigments derived from plant-biomass wastes (i.e. leaves and barks), after 6-month storage*).

No	Color-leaching resistance against	Natural pigment-coloring and fixative-treatment A	B	C	Control
1	Rubbing/ironing	4 – 5	4 – 5	4 – 5	5
2	Exposure to sunlight	4 – 5	4 – 5	4 – 5	5
3	Detergent washing	2 – 3	2 – 3	2 – 3	5

Remarks:

*)	The fabrics were colored with natural pigment, and then treated with any of the three kinds of fixatives (A, B, or C)
A	The fabrics were colored with natural pigment, and then treated with lime ($CaCO_3$) fixative
B	The fabrics were colored with natural pigment, and then treated with tunjung/fero sulfate ($FeSO_4$) fixative
C	The fabrics were colored with natural pigment, and then treated with the mixture of lime + tunjung fixatives, in equal proportion (50:50, w/w)
Control	The fabrics were colored with natural pigment, and then treated with either lime fixative, tunjung fixative, or the mixture of lime + tunjung fixatives, after 6-month storage, but without undergoing any kind of color-leaching treatment (either lime fixative, tunjung fixative, or the mixture of lime + tunjung fixative)
Classes	class 1 (very poor); class 2 (poor); class 3 (moderate); class 4 (good); and class 5 (very good / best coloring-performance)

(A) *T. catappa* barks (B) *T. catappa* leaf

Figure 3: Coloring performance at fabrics by natural pigments extracted from different biomass waste origins (A, B); using different fixative agents (horizontally arranged, from Left side, Middle, to Right side), and two storage periods (vertically arranged, Upper, Lower).

extracts from biomass wastes (of those nine plant species origins) remained visually unchanged (at the fabrics) or practically still similar to the qualities/performance of the initially/freshly produced coloring-extracts, immediately brought out after passing through the filtering cloth; and then allowed to cool down for 24 hours (Figure 2). This occurrence strongly implied that the possible degradation/destruction by

Figure 4: Visual coloring performances at the fabrics by natural pigments extracted from leaf and bark wastes of particular plant species origins (*R. mucronata Lamk*.)
Immersion frequency: 3 times in aqueous solution of bakau minyak bark extracts, fabric color became black-red -Fixative - lime, fabric color became red; -Immersion again in solution of papaya flower extract + papaya leaf extract - Fixative tunjung, fabric color became concentrated dark brown.

Figure 5: Application of colors mixture at primissima fabricts (*C. Sappan, R. mucronata + X. granatum*) leaves, visual color, fixation lime. Fabric immersion: 7 times in aqueous of extract, and color ultimately appeared: greenish brick-red.

Figure 6: Pigment extracted from *A. mangium* bark, fabric immersion frequency: 7 times in aqueous extract, lime fixatives (left) and ferro-sulphate fixative (on the right figure). Remark: Appearance of Colors on the Pigment-imparted and Lime-fixative-treated of Weaving yarn-color of *A. Mangium*.

Figure 7: Remark:Application of colors at primissima (originally white) fabrics by natural pigments (combination of 4 types of waste origins). Application of colors at primissima (originally white) fabrics by natural pigments (extracted from *T.T. catappa, R.mucronata, A. Mangium, and C.Tagal*).

Figure 8: The final fabric staining, prepered for quality testing.

enzymes at the end of 6-month storage insignificantly affected the qualities or coloring performance of those coloring extracts (natural pigments) at the fabrics. Accordingly, the natural coloring pigments should be well maintained to meet the three standard paradigms as described before, i.e. quality, safety and efficacy. Coloring pigments should also have their own specification, particularly regarding the color standard, color consistency, and other related beneficial information.

Quality testing aimed to assess how strong or efficacious the resistance of coloring pigments was against the destruction due to microbe actions and pigment treatments. Another aim was to assess the inhibiting intensity of the extracts (pigments) against the destruction by enzymes on active compounds in the pigments, thereby avoiding undesired pigment decomposition. As described before, those enzymes covered among others oxidase, peroxidase, hydrolase and isomerase, which caused severe decomposition on particular compounds (e.g. phenol, polysaccharides and natural alkaloids). Accordingly, further in-depth research deserved carrying out to complete these standard requirements.

Testing on color-leaching quality or resistance of particular woven materials (e.g. batik fabrics) was performed after the fabrics were colored with natural pigments (with or without further fixative treatment) and then compared with the leaching-resistance standards. Standardization was essential in order not to disappoint the consumers/users and also to keep the quality of merchandise (e.g. pigment-colored fabrics) well maintained. The testing was carried out at the Center for Testing and Controlling of Merchandise Qualities, Ministry of Trade and Industry, Jakarta. The merchandise to be tested could cover several prototype items (e.g. fabrics), after being colored by aqueous coloring pigments/extract solution as well as by dry extracts and followed with fixative treatments. Also as described before, the testing parameters for assessing the color-leaching resistances (of the colored materials) could cover those against supposedly leaching inducers, i.e. washing, rubbing/ironing, acid sweat, base sweat, and exposure to sunlight (especially the final fabric staining-Figure 8). Those five criteria for color-leaching resistance of the fabrics referred to the standardized methods (Organization/ISO and Indonesia's National Standard/INS).

Exploration and research associated with the utilization of biomass wastes have not been performed a lot. In addition, the natural coloring pigments extracted from those wastes have not contributed much to the collection of national coloring pigments, particularly with regard to substitution for synthetic pigments (for coloring batik fabrics, sewing yarns, silk fabrics, and other woven items, as well). Natural pigments of whatever sources, including those from biomass wastes, were preferable by the consumers/users to synthetic pigments. Natural pigments exhibited merits or advantages, which were among others exclusive; and their visual color characteristics were very specific and ethnical, thereby creating high selling values. Natural pigment items could become regional superior potency at global markets. Developing utilization of natural pigments from biomass wastes is urgently necessitated through intensive in-depth research in order to get even better results.

There are a lot of fans or enthusiasts who favored batik crafts from countries and took great interest in the production of Indonesia's natural pigments. Utilization of particular biomass (plants) portions, such as nila (*Indigofera tinctoria*), malam (*Pterocarpus indica*) woods, ketapang (*Terminalia catappa*) roots, teak (*Tectona grandis* L) leaves, annato (*Bixa orellana* L.) seeds, tea (*Camelia sinensis*) leaves and pinang (*Areca catechu*) fruits, as renewable natural coloring pigments, has been greatly recognized in fashion world (Prima & Amar, 2013; Rosanah, et al. 2013 & Husodo, 1999). Intensive research has begun carrying out as a response to international markets. Several countries such as India, Singapore, Malaysia, and China have begun performing a transaction with consumers/customers, especially those who were very concerned with renewable and environmentally friendly merchandise.

As several examples, natural coloring pigments could be potentially extracted from biomass portions of mahoni (*Swietenia macrophylla*), acasia (*Acacia mangium*), teak (*Tectona grandis)*, ketapang (*Terminalia catappa*), kayu hitam (*Diospiros celebica*) woods, and angsana (*Pterocarpus indicus*) plant species. Actually, wood portions of those species have been especially utilized as construction materials. Meanwhile, other tree portions such as twigs, sawdust, leaves, roots, trunk or stem barks, and fruits were just discarded or abandoned enormously on sites. Based on total volume, it strongly indicated that portions of biomass wastes were considerably greater than the wood portions, which have been already utilized. Accordingly, those various biomass wastes seemed worth utilizing as natural coloring pigments which otherwise could afford high economic values [8].

Intensive research has just started carried out by particular countries that enthusiastically responded to international markets. Several countries such as India, Singapore, Malaysia and China as described before have embarked on conducting the transaction with consumers/customers, especially those who are very concerned with renewable and environmentally friendly merchandise. The use of natural coloring pigments (e.g. for fabrics) is very favored because the pigments could bring out fabric color effects that were attractive, specific and moreover could not be imitated by synthetic pigments. This fact has created meaningful attractive matters for exclusive pigment-colored items which afforded high economic values, such as batik fabrics, silk items, and woven fabrics alike. The merits or advantages of natural pigments from plant biomass wastes are that pigment production was produced in such a process that did not destroy environments or upset the delicate natural balance. According to Natasya (2015), the criteria stipulated for environmentally friendly materials or items (e.g. coloring pigments) were such that they should not cause severe problems or unwanted effects on human health. Besides in the manufacturing process, it would not generate harmful chemical compounds to the environment; also the compounds should be easily degraded or decomposed in nature. However, natural pigments for coloring the fabrics still exhibited their drawbacks or weakness, among others, the pigments still have not owned reliable color standards, are easily leachable, the pigment-manufacturing process took considerable time and moreover, the raw materials for natural pigments ready for use were still limited in quantity.

As of this occasion, production of plant-based natural coloring pigments in Indonesia was produced a lot from wild plant species origins. In fact, the trade potency able to be obtained from those plants, which grew widespread in forest areas in Indonesia; and which afforded to produce a great quantity of natural-coloring pigments could achieve high economic values, approximately worth millions of dollars; or minimally attain half of Indonesia's batik export profits. On the other hand, harvesting natural pigments directly from forests could bring about negative effects on the sustainability of particularly forest plant species. Harvesting of natural coloring pigments from biomass wastes of particular plant portions (e.g. roots and barks) was also very destructive to the sustainability of the corresponding plant source organs, moreover, should the pigment harvesting be performed through uncontrolled plant uprooting or felling down? Conservation can therefore become a reasonable choice that causes no or little damaging risks if the production of natural pigments from biomass wastes wants to proceed continually.

To recap, despite several advantages/merits afforded by natural pigments derived from plant biomass (e.g. wood wastes and other-plant wastes), however, those pigments exhibited several drawbacks (including in the processing method), e.g. the biomass wastes as biological matters with such, still able to conduct biological activities, such as respiration, metabolisms and other physiology processes, thereby possibly inducing degradation or chemical changes to the chemical components in the waste (including the dye/coloring matters) during especially the storage and hence lowering dye qualities for coloring purposes (fabrics/yarns/batik/etc); and also the location of biomass waste sources being usually scattered, thereby causing difficulties in collecting the waste raw material in sufficient quantity as well as ensuring the input supply of raw materials continuously. This inconvenient situation could render it difficult to establish industries of biomass-based natural pigments in a large-scale operation, and hence only effective for home industries with small-scale capacity.

Conclusion and recommendations

Application of natural-coloring pigments obtained or extracted through the exploration of biomass wastes (e.g. leaves, twigs, twigs and trunk barks) of the plant species origins, which prevalently grow on plain (lowland) areas, could bring out color variations on the pigment-colored fabrics. Such variations appeared visually on fabrics between red, yellow, green and brown colors. The use of tunjung fixative on pigment-colored fabrics aptly brought out strong-brown-colored fabrics. Meanwhile, the use of lime fixative tended to cause the fabrics with milky brown colors. Performance of natural coloring pigments, extracted from leaf wastes, applied to the fabrics, followed with tunjung fixation aptly brought out greenish-grey-colored fabrics; while lime fixatives tended to appear in the fabrics with reddish brown colors.

Storage of natural pigments/extracts for a 6-month period visually did not change visually the coloring performance of such pigments already applied at batik fabrics and other woven items, compared to the coloring performance of the corresponding initially/freshly produced pigments (after immediately being screened through the filtering cloth; then allowed to cool down for 24 hours and further applied at the similar fabrics/items). Testing results on coloring qualities of natural pigments (associated with color-leaching resistance of the pigment-colored fabrics) against the allegedly color-leaching inducers, i.e. washing, rubbing/ironing, and exposure to sunlight disclosed very good category (scores 4 - 5), but the coloring qualities of natural pigments from leaf wastes were very vulnerable against the washing that used detergents.

Natural coloring pigments from leaves, twigs and trunk barks of seven plant species origins did not impart many color variations on the pigment-colored fabrics, due to the implementation of just various pigments, varying storage durations of the pigments, and different kinds of fixatives. Accordingly, it still urgently necessitates a thorough in-depth exploration of natural-coloring pigments from other plant species origins that grow on a plateau (highland) areas; or from plant species of particular family or groups, which indicative can be a potential source for producing natural pigments.

Natural pigments (derived from plant biomass wastes) could afford several advantages/merits, compared to synthetic/inorganic pigments, such as being more environmentally friendly; renewable; able to make use of wood wastes and other biomass wastes from wood industry activities (and forest-logging), with might otherwise pollute and harm the environment and cause the accumulation of unused wastes/garbages/trashes; biodegradable; could afford or provide various colors; conforming to the desires/needs by consumers/users. Meanwhile, synthetic/inorganic pigments are usually produced and processed from inorganic raw materials (e.g. obtained from mineral ores), which are not renewable; and the process could be environmentally unfriendly, heavily polluting (the air and water/stream), and the residues/wastes very possibly harmful to living creatures.

However, biomass-waste-based natural pigments reveal several drawbacks. As such, those wastes could still perform biological activities, which very possibly cause chemical changes/degradation to their chemical components inside (e.g. dye/coloring matters). In addition, such wastes as the biological matter could undergo degrading/decomposing attacks by enzymes, microbes, bacteria, fungi and other destroying organisms. This situation could also lower the qualities of natural pigments for coloring purposes. Also, the locations of waste sources are usually scattered and therefore bring about difficulty in providing waste raw materials for the large-scale natural pigment-based coloring industry. Finally, careless harvesting of plant-biomass wastes (e.g. uncontrolled plant uprooting or felling down) could be very destructive as well to the sustainability of the corresponding waste-sourcing plants.

Utilization of biomass plant wastes (e.g. wood residues, leaves, and barks) into natural traditional pigments could also bring in economic gains/benefits, such as lowering the dependency on imported synthetic coloring pigments, thereby saving the country's finance and creating money/finance sources; providing job opportunity to the community, especially for those who are dealing with natural pigment collection as well as natural pigments; boosting the thriving/proliferation of fabric/batik crafters/industries; and enhancing the welfare of the related community.

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