Research Article

Biomordants and new alternatives to the sustainable natural fiber dyeings

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Abstract
The textile industry is considered a major polluter of the environment using chemical substances such as color fixatives and the production of toxic waste. There is a growing demand in the fashion market for products that are less aggressive to the environment and use of environmentally friendly processes. Considering the use of natural dyes as an alternative to synthetic dyes, and the use of natural mordants as alternatives to metal mordants, this study investigated the use of *Acacia mearnsii* sawdust as a biomordant for color fixation. Dyeing and color fixing tests were carried out on banana fiber, and the natural dyes of *Hibiscus sabdariffa*, *Allium cepa* and *Curcuma longa*. The results show the efficacy of *Acacia* as a biomordant for the tested dyes, presenting intense color and fiber softness. Thus, it is intended to contribute with new alternatives to natural dyeing through materials and sustainable processing procedures applied to natural fibers.

Keywords Banana tree fiber · Biomordant · Textile · Natural fiber · Natural dye

1 Introduction

The textile industry uses large amounts of chemicals, artificial dyes and a large volume of water in its process, being considered one of the most polluting industries in the world [1, 2]. Its process pollutes both water and the environment with toxic waste. The World Bank considers that 17 to 20% of industrial water pollution comes from textile dyeing and from the finishing treatment given to the fabric [3–5]. It is estimated that 150 L of wastewater is generated to produce one kilogram of the finished product in the textile industry and certain synthetic dyes present are not biodegradable or remain in the environment for a long period, causing pollution. Therefore, it is highly necessary to remove the dyes present from industrial effluents to avoid water pollution [6]. Synthetic dyes are made in such a way so that there is no fading away by any of the physical, chemical and biological agents [7]. It is estimated that 20% of used dyes are dumped into the environment because of their low level of affinity with the fabrics [8]. On the other hand, natural dyes come from renewable, ecological, safe, non-carcinogenic, non-allergenic and biodegradable sources [4, 9].

Currently there is a variety of applications for natural dyes such as antimicrobial, antifungal, UV protection, cosmetics, food, additives, pH indicators and other uses [10]. They are considered renewable, biodegradable, environmentally friendly and are a viable alternative for the excessive use of artificial colors, which add up to around 10 million tons per year [1, 11–13].

The most suitable and widely accepted classification system for natural dyes is based on their chemical structure, as it readily identifies dyes belonging to a particular chemical group, which has characteristic properties [5].

In the process of natural dyeing, most of the time the use of a mordant is necessary [9]. Mordants can be

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SN Applied Sciences (2019) 1:1356 | https://doi.org/10.1007/s42452-019-1384-5

Received: 13 June 2019 / Accepted: 28 September 2019 / Published online: 9 October 2019

derived from metallic salts such as sulfates (magnesium, aluminum, zinc, copper, cobalt, nickel, manganese or tin), chlorides (stannic, ferric, copper, zinc, aluminum and even neodymium or zirconium), hydroxides (calcium) and oxides (ferric or lanthanum) [14, 15]. Of the metal mordants used for natural dyes, alum and iron are environmentally safe and have not been restricted by any ecoregulation [3, 16]. Many of these mordants can cause problems with the resulting waste, soil contamination and water waste. [17]. Another problem with using these mordants is the incompatibility with the eco-friendly concept used in natural dyeing. Large-scale usage would lead to serious environmental problems [9]. In the use of these metal salts as mordants, only a small amount is fixed in the textiles and the rest is discharged as effluent, leading to contamination of terrestrial and aquatic resources [3, 18, 19].

Biomordants are reported as sustainable and ecologically correct alternatives to metal mordants, providing satisfactory dyeing and solidity properties [19]. Biomordant sources are plants with high tannin content [20] or hyperaccumulative metal plants [21].

Some biomordants, such as pomegranate peel, rosemary and thuja leaves were proposed as promising alternatives for aluminum, iron sulphate II, copper sulphate II, stannous chloride and potassium dichromate [3]. Natural polyphenols, also called tannins, are obtained from various parts of plants, such as bark, wood, fruits, fruit peels, leaves, roots and plant galls [22]. Other studies describe the presence of tannin of the banana tree pseudostem, Akpabio et al. [23] determined the physicochemical properties of the residue of the banana tree (*Musa paradisiaca*) pseudostem and banana (*Musa sapientum*). The results showed the phytochemical composition in mg/100 g of tannin for the banana and pseudostem residues of the banana tree (respectively 7.99 ± 0.26 and 6.55 ± 0.33).

Natural resources play a dominant role in the economic activities of any country and therefore, contribute substantially to the gross domestic product (GDP). In the case of the development of underdeveloped countries, this also helps economic development. There is a growing worldwide trend towards making the most of such resources through new processes and products. These, in turn, not only help in preventing environmental pollution, but also in generating jobs, particularly in the countryside and contributing to the improvement of living standards [24].

The banana belongs to the *Musaceae* family and there are approximately 300 species, of which only 20 varieties are used for consumption [25, 26]. There are over one thousand domesticated *Mus*a cultivars and their genetic diversity is high, indicating multiple origins of different wild hybrids between two major ancestral species [27].

According to the botanical systematic hierarchical classification, the banana belongs to the division of *Angiosperms*, class of *Monocotyledonous*, order *Scitamineae* and family *Musaceae*. The subgenus *Eumusa*, determines the category of edible bananas which are derived from hybrids of the wild subspecies of *Musa acuminata Colla* (genome A) and *Musa balbisiana Colla* (genome B) [28–30]. Wild bananas occur in the tropics from India to Oceania, but there is a distinction between the distribution of *M. acuminata* and *M. balbisiana* within this range [28]. The classification proposed by Cheesman [31] in 1948 for the genus *Musa*, currently accepted worldwide, is based on the basic number of chromosomes [32]. The cultivar Prata (AAB) is a variety of edible bananas, genus *Musa* [30]. In Brazil, the Vale do Ribeira region (state of São Paulo) uses the residues of pseudostem from this cultivar to produce handicrafts and textile products [33].

The residue from banana farming represents 40% of the fruit production and shows that about 13 tons of dry organic matter are generated per planted hectare when considering the pseudostem, the leaves and the peduncle of the banana tree [34].

For each ton of banana fruit harvested, about 100 kg of the fruit is discarded (that is, rotten fruit) and about 4 tons of biomass waste (e.g. leaf, pseudostem, rotten fruit, peel, peduncle, rhizome, etc.) are produced. This means that, for each banana production cycle, four times the biomass residues are also produced [35, 36].

In 2012, the Philippine Textile Research Institute concluded that banana plantations in the Philippines alone can generate more than 300,000 tons of fiber [37]. The institute, which is a pioneer in the research on banana fiber, has recently presented studies to improve fiber texture through techniques of degumming banana fiber, thus shortening processes [38].

The banana species best known for its fiber is the abaca (*Musa textiles*). Its fiber is highly important among the group of foliar fibers, while the most common banana consumed by humans is a member of the *Musa acuminata* species [39, 40].

The world production of abaca fiber in 2016 was 106,598 tons. The Philippines appear as the main dominant producer, supplying 86% of the world demand, the rest of the production being supplied by Ecuador. In 2005, the Philippines began an expansion program. By the end of 2010, a total of 48,922 hectares of new abaca plantations were established. By the end of 2010 the country had reached 167,145 hectares, capable of meeting the increasing demand for abaca fiber [41].

In India, the banana fiber has been used with a blend of cotton fiber to make fabrics, accounting for about 15.8% of all fabrics manufactured by India in the year 2015. The province of Gujarat, India, which comprises the Anand, Surat, Vadodara, Bharuch, Narmada and Kheda counties, presented in 2017 at the 8th Global Summit (Gujarat, India, 10–13 January 2017) a project to create the Gujarat Textile Eco-Park for the processing of natural and banana fibers. This covers the entire production process, with a center of excellence and training, testing labs and design studio, which will unite the chain of extraction, processing, yarn creation, and sustainable, high quality textile products. 17.6% of the total banana production in India comes from the province of Gujarat [42].

In Brazil, only about 2% of the 6.6 million tons of biomass of banana fiber produced annually by plantations are used by artisans, particularly by the coastal population in the southern states of São Paulo and Paraná. Compared to other countries, this number jumps to about 10% in countries like India, where it is used not only by artisans, but also as a source of energy and has other uses in industries. Consequently, there is an excellent opportunity to expand the use of this great source in the country [24].

The choice of *Black Acacia* as a biomordant for this study is due to its historical recognition for the quality of its bark, from which plant extracts rich in tannins and phenols are obtained. They originate the tannins, widely used in tanning, the most well-known group of products obtained from the bark. It is the main source of bark for the industry of vegetable tannin in the world [43]. *Acacia mearnsii* is one of the best species in terms of yield per tree and quality (composition and coloring) of tannin. Other species such as *A. decurrens* have approximately the same yield and *A. pycnantha* is even superior to *A. mearnsii*, but both species provide extracts of higher staining and when used for tanning, the resulting products are darker [44].

In Brazil, in plantations of *A. mearnsii* in the State of Rio Grande do Sul, at age eight, the estimated average tannin content in the bark is 27% [45], that is, of every 100 kilos of bark dried in the open air 27 kilos of tannin are obtained. The increase in the tannin content is positively correlated with the increase in age [46–48]. *Acacia mearnsii* is the third most planted forest species in Brazil, surpassed only by species of the genus *Eucalyptus* and *Pinus*. Besides its use as raw material for tannin, it provides cellulose and charcoal. It represents a significant part of the reforestation of the State of Rio Grande do Sul and is of great social importance because it is planted in small properties, thus benefiting thousands of families in the region [49].

The choice of the dyes used in this study takes into consideration their easy access in the local commerce, being easily found in street markets, emporiums and municipal markets. In addition to this is the reusing of residues, such as *Allium cepa* peels, highly rich in natural dyes, which is discarded by the food industry.

This research aims at investigating the efficacy of the use of biomordants as an alternative to metal mordants for the fixation of natural dyes in the dyeing process of banana fiber (*Musa sp.*) Cultivar Prata (AAB) for textile purposes. For this study, the sawdust of *A. mearnsii* and the dyes of *Hibiscus sabdariffa*, *Camellia sinensis*, *C. longa* and *A. cepa* were used as biomordants.

2 Experimental

2.1 Origin of banana fibers

Samples collected from the banana (*Musa* sp), Prata cultivar (AAB) pseudostem collected by women from the Banarte Association, a riverside community located in the Ribeira Valley, Miracatu municipality (SP), were used to perform the laboratory tests in the southeastern region of Brazil, between the coordinates: latitude: 24°16′53″S, longitude: 47°27′35″. The group of women artisans of the Banarte Association collects, processes and uses banana tree fibers dyed with biomordants for the making of textiles woven in manual looms and various techniques of braids [50]. Thus, a fiber supply channel has been established through this association. It is worth mentioning that the material required for analysis does not require the authorization of environmental agencies to carry out the present study, since fiber is a by-product of the banana's commercial cultivation, being the raw material of the artisans, who acquire the material from other riverside women who live in the producing sites of the region. The two types of fibers (commonly called "straws") used are, respectively, in the inner and outer part of the pseudostem. The material was transported and stored for analysis in clean polyethylene bags.

2.2 Process of artisanal processing

The basic reference to describe the process of extraction and processing of banana fiber is "The straw and the banana thread—production manual" by Maria Elisa de Paula Eduardo Garavello [50]. For the manufacturing of the straw, all the sheaths forming the pseudostem are used. They generally hold more or less 15 to 20 parts and can be manually removed one by one. After removing of the sheaths, they will be shredded into ribbons or strips to obtain the straws [51] (Fig. 1).

The types of straw obtained are [50] (Fig. 2):

- *Inner straw* It is the softest part of the sheath, that is, the inside, from which the straw is extracted (4C);
- *Lace mesh* The middle of the sheath is formed by a relatively lacy mesh layer, which is extracted by wiping the two sides of the ribbon (4B);

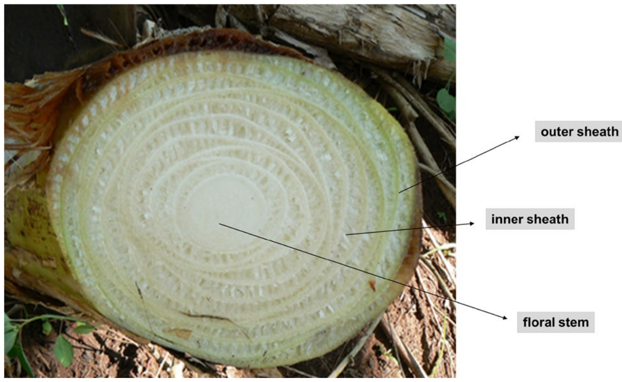

Fig. 1 Banana tree pseudostem parts (from inner to outside): floral stem, inner sheath and outer sheath. Created by the author Photo: Promusa.org [52]

- *Outer straw* It is the straw obtained from outer the layer of the sheath, extracted with the aid of a non-serrated kitchen knife, so as not to damage the fiber to be removed (4A);

In the outer and inner straws, the excess mucilage is scraped out with a knife. After this process, all the strips were washed and put on the drying rails [44].

The artisanal processing of banana fiber follows these steps (Fig. 2): collection [1], slicing [2], shredding (3 and 4), washing in water and drying [5, 6]. In this process of the community of the municipality of Miracatu (SP, Brazil), the banana straws must be malleable to be handled, that is, they cannot be too dry, and if this occur, they must be moistened again before being used in artisanal works [50].

2.3 Stages of dyeing

The fiber dyeing procedure was divided into the following steps: I. Preparation of the substrate (material to be dyed); II. Preparation and application of the biomordants; III. Preparation and application of natural dyes in the substrate (fiber); IV. Washing. In the preparation of the substrate, the banana fibers were cut into strips, weighing 25 g for each preparation, on a precision balance of 0.0001 g, and later washed with water in order to promote greater uniformity in the absorption of the biomordant and natural dyes by the fibers. Solutions with *A. mearnsii* biomordant and alum mordant were prepared for the tests performed.

2.3.1 Collection and preparation of the biomordant of *A. mearnsii* and alum mordant

The dried extracts from the barks of *Black Acacia* (*A. mearnsii*) were purchased from the company TANAC S.A. (city of Montenegro, RS, Brazil). Fifty grams of the extract were weighed and diluted in 1 L of water. They were boiled for 20 min. The obtained solution was then filtered through an analytical funnel and a paper filter and separated. The banana fibers of the cultivar Prata (AAB) were submerged in the temperature range of 45-60 °C and allowed to soak for 24 h. The potassium alum salt (KAl (SO_4) 2·12H_2O) was purchased from Emporio Terra Vitta Produtos Naturais (São

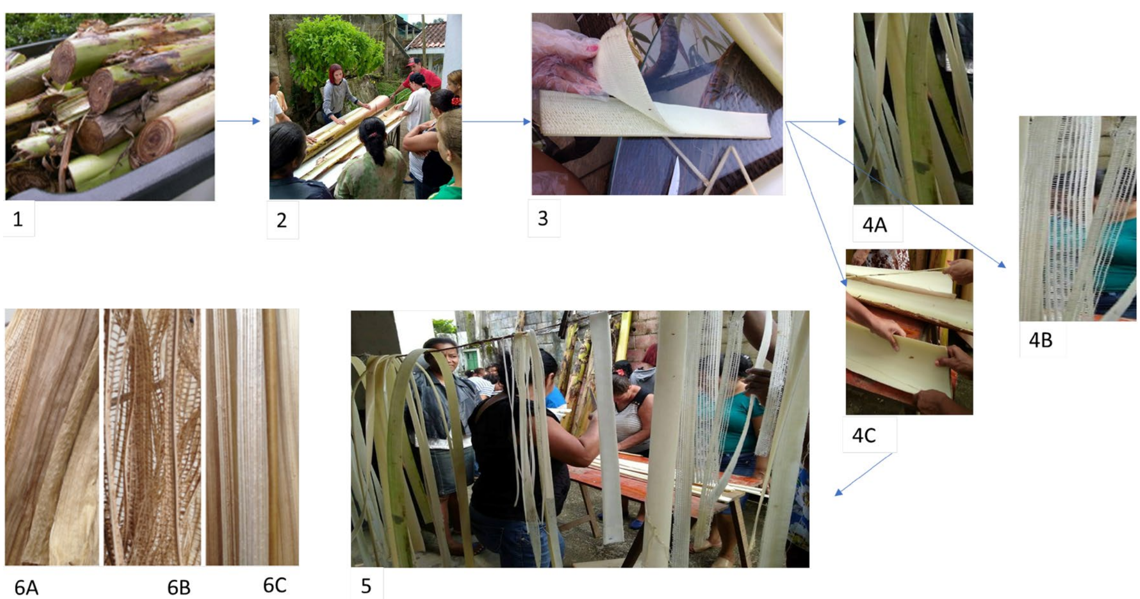

Fig. 2 Process of beneficiation of banana fiber. Steps: 1—cutting the pseudostem; 2–separation of the sheaths; 3—slicing; 4—types of straw obtained. 4A: outer straw 4B: lace and 4C: outer straw. 5—drying process; 6—dry straws. 6A: outer straw; 6B: lace; 6C: inner straw. Prepared by the author

Paulo, SP, Brazil). 20 g of the salt were weighed and diluted in 1 L of water. They were brought to boil for 20 min, cooled and stored in glass beaker.

2.3.2 Collection and preparation of dyes

The dried extracts of the leaves of *Camellia sinensis*, *H. sabdariffa* flowers, and *C. longa* root composed with corn flour were purchased from Emporio Terra Vitta Produtos Naturais (São Paulo, SP, Brazil). *A. cepa* peels were purchased from merchants in the Municipal Market of São Paulo (Brazil).

Fifty grams of each dry extract listed were weighed and diluted in 1 L of water. They were boiled for 20 min. 25 g of the *A. cepa* peels were weighed, diluted in 1 L of water and boiled for 20 min. The solutions obtained from each preparation were then filtered through analytical funnel and paper filter and separated. The banana fibers, removed from the biomordant bath with *A. mearnsii*, were submerged in these new solutions prepared in the temperature range of 45–60 °C and soaked for 12 h. After the dyeing process, the fibers were washed in running water to remove excessive unbound dye and dried at room temperature for 48 h.

A portion of the *in natura* banana fibers of the cultivar Prata (AAB) and part of the fibers with *A. mearnsii* biomordant were reserved for comparative tests between the dyed fibers. Table 1 shows a summary of the equivalence of the quantities of input used and Fig. 3 is a summary of the biomordant and dyeing process of the banana fibers (cultivar Prata AAB).

Table 1 Inputs (dyes, mordant and biomordant) used and their preparations for 1 L of water (q.s.p.)

Sample	Input	Quantity (g)
Dye	*Hibiscus sabdariffa*	50
	Camellia sinensis	50
	Curcuma longa	50
	Allium cepa	25
Biomordant	*Acacia mearnsii*	50
Mordant	*Alum*	20

3 Discussion

In the tests carried out, the employ of biomordant of *A. mearnsii* on the fiber of the banana Prata (AAB) was satisfactory, with intense brown hues, and when using the alum mordant there was a clear lightening of the fiber (Fig. 4). With respect to the nuances obtained with each natural dye, all the samples presented satisfactory results with alteration of the color of the fiber, where, with the dyeings with the extracts of *H. sabdariffa*, *A. cepa* and *C. longa* biomordanted with *A. mearnsii*, presented greater impregnation, thus obtaining colorations, respectively of intense pink (Fig. 5), mustard yellow (Fig. 6) and reddish brown (Fig. 7). Compared with the alum mordanted fibers, the *A. mearnsii* biomordant intensified all colors, while the alum mordant brightened the fibers. The samples dyed with *Camellia sinensis* obtained the less intense colors, varying in shades of light green with the two mordants used (Fig. 8). Regarding the appearance of the fibers' touch and their softness, there were no apparent changes in relation to the undyed fibers.

The main component of extracts obtained from turmeric is *curcumin*. According to Volp et al. [53], *curcumin* is a pigment that has a bioactive action, which is why it is more commonly used by the food industry, and its characteristic color is lemon yellow with shades that may vary from bright yellow to orange. In reddish pigments obtained from flowers, such as the *hibiscus*, there is the presence of substances called *anthocyanins*, among others. For Couto et al. [54] *anthocyanins* extracted from their natural condition are transformed into flavilic salts, which in turn present better adhesion on substrates that have molecules of sugars in their chemical structure.

The use of new and more ecological mordant techniques could be an option for mordants used with metal salts. [9]. Nowadays, biomordants cover a wide spectrum ranging from tannin, tannic acid, tartaric acid, plants containing metals, from bio-waste and by-products. Tannins and plants rich in metals, bio-waste and by-products act as mordants during natural coloring. Tannins make up a remarkable group of biomordants, occupying an

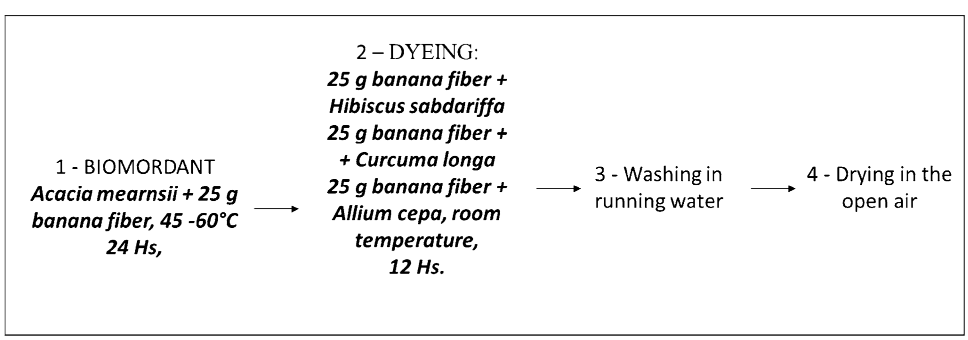

Fig. 3 Stages of the biomordant and dyeing process of banana fibers (cultivar Prata AAB)

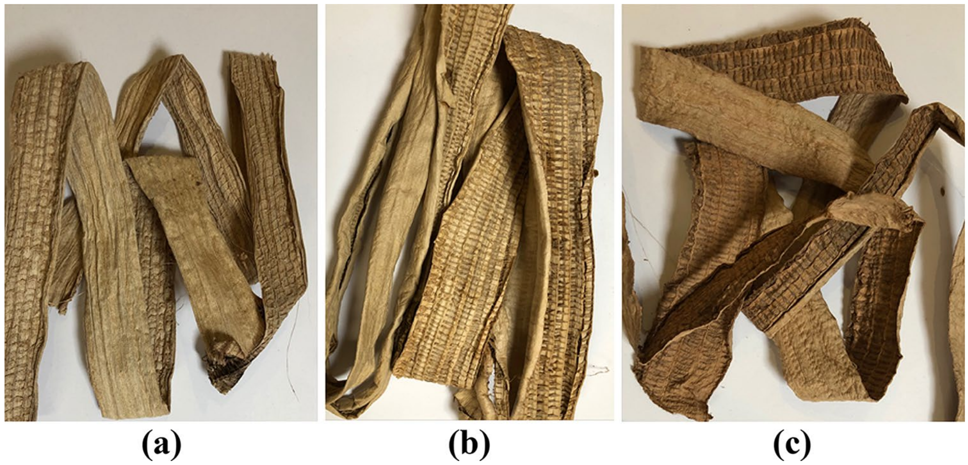

Fig. 4 Banana Prata Fiber (AAB) and mordant and biomordant processes: (a) fiber without dyeing, (b) fiber mordant with alum and extract of *A. mearnsii*; (c) fiber biomordanted with *A. mearnsii*

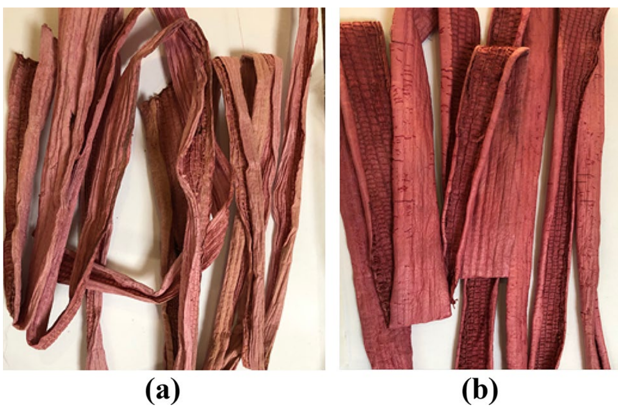

Fig. 5 Banana Prata Fiber (AAB) dyed with *H. sabdariffa*: **a** fiber mordanted with alum; **b** fiber biomordanted with *A. mearnsii*

Fig. 6 Banana Prata Fiber (AAB) dyed with *C. longa*: **a** fiber mordanted with alum; **b** fiber biomordanted with *A. mearnsii*

important place in natural dyeing and printing. Some biomordants, such as pomegranate peel (*Punica granatum* L.), rosemary (*Rosmarinus officinalis*) and thuja leaves (*Thuja orientalis* or *Platycladus orientalis*) were proposed as promising alternatives for aluminum, iron sulphate II, copper sulphate II, stannous chloride and potassium dichromate. Studies have shown their increased use of color, and color yields may compete with metal mordants and even surpass them in some cases, depending on the method and the type of mordant used [3].

Although the results of the present study are extremely promising in terms of replacing mordants with biomordants, other tests are necessary in future trials, such as: light fastness, tannin concentration and more accurate colorimetric tests.

4 Conclusions

The mordant tests performed in the present study with *A. mearnsii* sawdust showed effectiveness regarding fiber softness, color intensity and fixation of the dyes of *H. sabdariffa*, *A. cepa* and *C. longa*, indicating that they are promising as a sustainable alternative to metal mordants. The best results were obtained through the dyeing and the cold biomordant of the fibers, with a 24-h rest bath, and 12-h dyeing. Thus, it is intended to contribute with new alternatives to natural dyeing through materials and sustainable processing procedures applied to natural fibers.

Acknowledgements The authors gratefully acknowledge the funding by CAPES ("Coordenacao de Aperfeicoamento de Pessoal de Nivel Superior"—Brazilian Federal Agency for Support and Evaluation of Graduate Education).

Fig. 7 Banana Prata Fiber (AAB) dyed with *A. cepa*: **a** fiber dyed without the use of mordant; **b** Fiber mordanted with alum; **c** fiber biomordanted with *A. mearnsii*

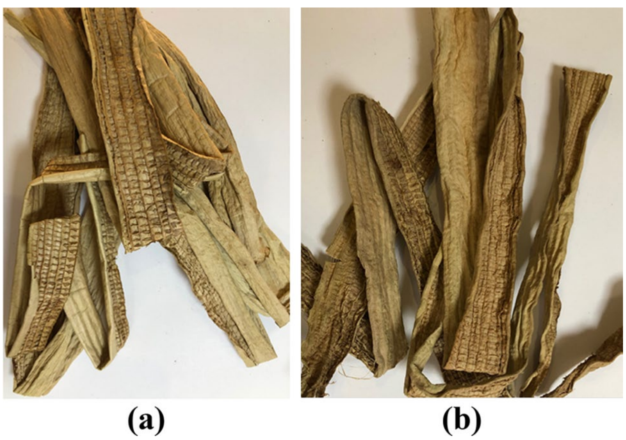

Fig. 8 Banana Prata Fiber (AAB) dyed with *Camellia sinensis*: **a** fiber mordanted with alum; **b** fiber biomordanted with *A. mearnsii*

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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