

Journal of Tropical Biodiversity and Biotechnology Volume 08, Issue 01 (2023): jtbb70652 DOI: 10.22146/jtbb.70652

Research Article

The Evaluation of the Combination of Additives and Fungal Dyes to Produce Color for Textile Painting

Agung Adi Nugroho¹, Suciatmih¹*

1)Research Center for Applied Microbiology, National Research and Innovation Agency (BRIN), Cibinong Science Center, Cibinong-Bogor 16911, Indonesia

*corresponding author, email: suciatmih1958@gmail.com

Keywords:

cloth color dye fungal dye synthetic dye textile painting **Submitted:** 22 November 2021 **Accepted:** 10 October 2022 **Published:** 25 January 2023 **Editor:** Miftahul Ilmi

ABSTRACT

Fungal dyes are an eco-friendly alternative to synthetic dye. This study aims to evaluate additives to dye, using mixed fungi, to paint the picture on cloth. In the present study, the cloth was painted with mixed *Aspergillus* and *Paecilomy-ces* dye. The mixed fungi were grown on a mineral salt glucose medium. Five tests were conducted to evaluate additives to dye from mixed fungi that could be used to paint pictures on cloths to evaluate the effect of additives, a combination of additives producing tidy colors and other additives, the dye pH, mordant, and a variety of different mordants and the dye pH on color tidiness and hue. The additives used were alkali, acid, salts, glycerine, and urea. The Royal Horticultural Society (RHS) color chart was used to measure the color of filtrate and range developed on the painted color on the cloth. The results showed that the mixture of vinegar or lemon as additives and the dye pH of 3 produced tidy colors. The mordant application had a more significant effect on the color that appears than pH treatment. Colors formed on images can add variations to textile painting.

Copyright: © 2023, J. Tropical Biodiversity Biotechnology (CC BY-SA 4.0)

INTRODUCTION

The use of synthetic dyes in the textile industry is gradually receding not only due to the increase of environmental awareness for the harmful effects of either toxic degraded products or their non-biodegradable nature, but also the cause of cancer and allergic reactions (Teli et al. 2013). These problems motivate efforts to develop eco-friendly natural dyes derived from animals, microbes, and plants. Most of Indonesia's textile dyeing process use natural dyes from plants (Rini et al. 2011). Still, only few natural dyes are available in adequate quantities for use in textile dyeing (De Santis et al. 2005). Thus, it is necessary to explore the potential use of other biological sources, such as algae, bacteria, and fungi.

Dye production by microbes (including fungi) is very important to industries because microbes can proliferate, are highly productive, and are available throughout the year, regardless of the weather conditions (Méndez et al. 2011). Microbial dyes are often more soluble and stable than those obtained from animals or plants (Gunasekaran & Poorniammal 2008). Various dyes from fungi were informed, including yellow dyes by *Aspergillus sydowii* (CML 2967), two isolates of *A. aureolatus* (CML 2964 and E.4.1), and two isolates of *A. keveii* (CML 2964 and E.4.1) (Nirlane da Costa Souza et al. 2016); greyed-orange dyes by *Asper-* gillus terreus and Aspergillus sp. strain 2 (Suciatmih & Hidayat 2017); greyed-purple dyes by Paecilomyces lilacinus (Thom) Samson and Paecilomyces sp. strain 542 (Suciatmih & Yuliar 2018); and red dyes by Aspergillus sp. strain 1 (Suciatmih & Yuliar 2018) and Paecilomyces farinosus (Isaria farinosa) (Velmurugan et al. 2010a).

Previous research reported that mixed Aspergillus and Paecilomyces produced 187A greyed-purple dye and could dye cloths (Suciatmih et al. 2018; Suciatmih 2019; Suciatmih 2020), but its ability to paint pictures was untested. Using mixed fungi is intended to maintain the fungi's ability to produce dye. Except for mixed conditions, Aspergillus and Paeci*lomvces* grew in potato dextrose agar medium separately for one month. Then, each of these fungi was cultured in the production medium; each fungus was no longer able to produce dye or has lost its ability to produce dye. Bader et al. (2010) reported that in a habitat, different microorganisms may compete for substrates as well as act symbiotically. In this case, it may be a symbiosis among Aspergillus and Paecilomyces caused by synergies of their different enzymatic systems and metabolic pathways (Bader et al. 2010). Some higher activities produced by co-culture are reported. Cellulolytic fungal co-cultures were more effective in substrate saccharification, which ranged between 85~88% compared to the 62~67% saccharification shown by the monocultures (Eyini et al. 2002). The antibiotic activity produced in the co-cultures of Rhizopus peka P8 and Bacillus subtilis IFO3335 (inhibition zone of 25 mm) was higher than in each of the pure cultures (inhibition zones of 0-15 mm) (Fukuda et al. 2008).

According to Kurnia (2011), painting on the cloth called textile painting, is a technique for making motifs or decorating fabrics. The technique involves using special paints that are resistant to water and ironing. Cloth paint is generally a thick liquid with paste melted in a small amount of water. Rohandi and Listian (2015) stated that the paint component consists of additives, binders, pigments, and solvent, while Kumari and Singh (2017) informed that the main components contained in the paint are additives, pigments, solvents, and thickener.

Teli et al. (2013) reported that natural dyes with few expectations are non-substantive, hence, they must be used in conjunction with mordants. Mordant helps in binding dyes to the fiber by forming a chemical bridge between the dyes and fiber (Satyanarayana & Chandra 2013). Single dyes source added with different mordant types produced different colors and tones on the dyed cloth (Sangeetha et el. 2015; Suciatmih & Hidayat 2017; Suciatmih et al. 2019; Suciatmih 2020).

Rohandi and Listian (2015) stated that the paint quality was determined by selecting components, such as appropriate adhesives and additives. Some additives such as alkali (baking soda and soda ash), acid (cream of tartar, lemon, and vinegar), brown sugar, milk, glycerine, salts, and urea are also often added to the dyes to get the desired color. Different shades and tones were also obtained from a single dye source by application of different pH values (Wang et al. 2014; Ren et al. 2016; Suciatmih & Yuliar 2018; Suciatmih 2020).

The research objective is to evaluate the ability of various materials commonly used for dyeing cloth to help dyes from mixed fungi paint the picture on the cloth without any dye spreading beyond the boundaries of the motif of the design.

MATERIALS AND METHODS

Inoculation process

The inoculation process of mixing Aspergillus and Paecilomyces fungi

(Figure 1) was performed following previous studies (Suciatmih et al. 2018; Suciatmih 2019). Briefly, five mycelial prints were inoculated into an Erlenmeyer flask containing 200 ml of mineral salt glucose medium (Baker & Tatum 1998) with modification, and statically incubated at room temperature for one month in a dark place. Composition of the mineral salts-glucose medium is described as follows (in ppm): NaNO₃, 848; KCl, 300; MgSO₄.7H₂O, 165; NaH₂PO₄, 100; CaC1₂.2H₂O, 40; H₃BO₃, 5.7; FeSO₄.7H₂O, 5.0; ZnSO₄.7H₂O, 4.4; MnSO₄.H₂O (monohydrate), 3.1; Na₂MoO₄.2H₂O, 2.5; CuSO₄.5H₂O, 0.4; and glucose, 20,000. After the incubation period, the culture was passed through five layers of cheese-cloth, and the filtrate was centrifugated at 8500 rpm for 20 min as performed previously. The filtrate's optical density was determined spectrophotometrically (Shimadzu) at 530 nm for quantifying the dyes (Suciatmih et al. 2018; Suciatmih 2019). The dye yield was counted as OD units (UA530).



Figure 1. Mixing *Aspergillus* and *Paecilomyces* fungi in Potato Dextrose Agar medium.

Effect of additives

Primisima brand cotton cloth (4 cm \times 4 cm or 0.24 g) was first drawn with a pencil and then painted with the dye added with additives 2% and dried at room temperature. We evaluated 11 treatments of additives (Table 1). Additives applied to the dye produced tidy colors (the dye did not break and only occupied the predetermined place) on the cloths and were used for further testing.

Table 1. Treatment of additives

No	Treatment
1.	The dye 0.2 ml (pH 8) (control)
2.	The dye 0.2 ml + backing soda (pH 11) 20 µl
3.	The dye 0.2 ml + brown sugar (pH 6) 20 µl
4.	The dye 0.2 ml + cream of tartar (pH 3) 20 µl
5.	The dye 0.2 ml + glycerin (pH 4) 20 µl
6.	The dye 0.2 ml + lemon (pH 3) 20 µl
7.	The dye 0.2 ml + milk (pH 7) 20 μl
8.	The dye 0.2 ml + salt (pH 6) 20 μl
9.	The dye 0.2 ml + soda ash (pH 11) 20 μl
10.	The dye 0.2 ml + urea (pH 7) 20 μl
11.	The dye 0.2 ml + vinegar (pH 2) 20 µl

Combination effect of additives producing tidy colors and other additives

Vinegar or lemon was applied to the dye in the previous test produced a tidy color, but the hue was different from the control color. To get a tidy color and the same hue as the control color, we tested 9 treatments of a

combination of vinegar or lemon and other additives (Tables 2 and 3). The primisima cotton cloth (4 cm x 4 cm or 0.24 g), drawn with a pencil, was painted with a mixture of dye, vinegar or lemon, and other additives (2%), and then dried at room temperature. The composition of materials produced a tidy color, with the same hue as the control color, which was used for further testing.

Table 2. Treatment of combination of vinegar and other additives

No	Treatment		
1.	The dye 0.2 ml (pH 8) + vinegar (pH 2) 20 µl (control)		
2.	The dye 0.2 ml + vinegar (pH 2) 20 µl + backing soda (pH 11) 20 µl		
3.	The dye 0.2 ml + vinegar (pH 2) 20 µl + brown sugar (pH 6) 20 µl		
4.	The dye 0.2 ml + vinegar (pH 2) 20 μ l + cream of tartar (pH 3) 20 μ l		
5.	The dye 0.2 ml + vinegar (pH 2) 20 μ l + glycerin (pH 4) 20 μ l		
6.	The dye 0.2 ml + vinegar (pH 2) 20 μl + milk (pH 7) 20 μl		
7.	The dye 0.2 ml + vinegar (pH 2) 20 µl + salt (pH 6) 20 µl		
8.	The dye 0.2 ml + vinegar (pH 2) 20 µl + soda ash (pH 11) 20 µl		
9.	The dye 0.2 ml + vinegar (pH 2) 20 μl + urea (pH 7) 20 μl		

Table 3. Treatment of combination of lemon and other additives

No	Treatment
1.	The dye 0.2 ml (pH 8) + lemon (pH 3) 20 µl (control)
2.	The dye 0.2 ml + lemon (pH 3) 20µl + backing soda (pH 11) 20 µl
3.	The dye 0.2 ml + lemon (pH 3) 20 µl + brown sugar (pH 6) 20 µl
4.	The dye 0.2 ml + lemon (pH 3) 20 μ l + cream of tartar (pH 3) 20 μ l
5.	The dye 0.2 ml + lemon (pH 3) 20 μ l + glycerin (pH 4) 20 μ l
6.	The dye 0.2 ml + lemon (pH 3) 20 µl + milk (pH 7) 20 µl
7.	The dye 0.2 ml + lemon (pH 3) 20 µl + salt (pH 6) 20 µl
8.	The dye 0.2 ml + lemon (pH 3) 20 µl + soda ash (pH 11) 20 µl
9.	The dye 0.2 ml + lemon (pH 3) 20 µl + urea (pH 7) 20µl

Effect of mordant

Natural dye has a low coloring power within itself so that it requires chemicals compounds (mordants) for dye fixation into the fiber. We evaluated different mordants with 2% (alum, ferrous sulfate, and lime) painted with the mixture of the dye, vinegar, and soda ash that produced tidy colors and its hue approached the control colors; on the color's tidiness and hue. Cotton cloth (4 cm \times 4 cm or 0.24 g) was first drawn with a pencil, painted with the mixture, and dried at room temperature. There were four treatments studied (Table 4).

Table 4. Treatment of mordant

No	Treatment
1.	The dye 0.2 ml (pH 8) + vinegar (pH 2) 20 μl + soda ash (pH 11) 20 μl
	(control)
2.	The dye 0.2 ml + vinegar (pH 2) 20 μ l + soda ash (pH 11) 20 μ l + alum 20
	μl
3.	The dye 0.2 ml + vinegar (pH 2) 20 µl + soda ash (pH 11) 20 µl + lime 20
	μ
4.	The dye 0.2 ml + vinegar (pH 2) 20 μl + soda ash (pH 11) 20 μl + ferrous

Effect of the dye pH

sulfate 20 µl

Painting pH can increase or decrease the dye's ability to bind the fiber.

The differentiation effect of the dye pH on the color's tidiness and hue was evaluated. Cotton cloth $(4 \text{ cm} \times 4 \text{ cm} \text{ or } 0.24 \text{ g})$ was drawn with a pencil each was painted with the pH of the dye of 3, 7, and 11; and dried at room temperature.

Combination effect of different mordants and the dye pH

Mordant and pH can change the final color of the dyed cloth or painted picture on the cloth. We tested the combined effect of different mordants 2% (alum, lime, and ferrous sulfate) and dye pH (3, 7, and 11) on the color's tidiness and hue. Cotton cloth (4 cm × 4 cm or 0.24 g), drawn with a pencil, was painted with different mordant combinations and dye pHs, then dried at room temperature. Treatment of a combination of different mordants and pH of the fungal dye is presented in Table 5.

Table 5. Treatment of combination between different mordants and pH of the fungal dye

No	Treatment
1.	pH 3 of the dye 0.2 ml + vinegar 20 μ l + soda ash 20 μ l + alum 20 μ l
2.	pH 3 of the dye 0.2 ml + vinegar 20 μ l + soda ash 20 μ l + ferrous sulfate
	20 µl
3.	pH 3 of the dye 0.2 ml + vinegar 20 µl + soda ash 20 µl + lime 20 µl
4.	pH 7 of the dye 0.2 ml + vinegar 20 µl + soda ash 20 µl + alum 20 µl
5.	pH 7 of the dye 0.2 ml + vinegar 20 μ l + soda ash 20 μ l + ferrous sulfate
	20 µl
6.	pH 7 of the dye 0.2 ml + vinegar 20 μ l + soda ash 20 μ l + lime 20 μ l
7.	pH 11 of the dye 0.2 ml + vinegar 20 μ l + soda ash 20 μ l + alum 20 μ l
8.	pH 11 of the dye 0.2 ml + vinegar 20 μ l + soda ash 20 μ l + ferrous sulfate
	20 µl
9.	pH 11 of the dye 0.2 ml + vinegar 20 μ l + soda ash 20 μ l + lime 20 μ l

Determination of color hue

We determined the dye and the color hues after being painted with the dye from each test by matching the dye and the color hues with the Royal Horticultural Society (RHS) RH color chart, repeating each treatment twice (The Royal Horticultural Society 1966).

RESULTS AND DISCUSSION

At high glucose stress (20g/L) contained on the Baker and Tatum medium (1998) with modification, a mixed Aspergillus dan Paecilomyces produced 187A greyed-purple pigment (Figure 2) with an absorbance of 3.67 UA/L that can be used to paint the image on the cotton cloth. The control's painted cotton cloth color (only the fungal dyes) was 65B redpurple (Table 6 and Figure 3). Similar results were reported by Suciatmih et al. (2018), Suciatmih (2019) and Suciatmih (2020) that the same mixed fungi produced the same color pigment that can be used to dye cloth. The dye is included in the substantive dyes group because the dye can directly paint textile fibers or give good color when used alone (Marie et al. 2015). The fungi are thus potential sources as alternative sources for environmentally safe natural pigment production. Fungi is found to be a promising ecological source of pigments, as several fungal species are rich in stable colourants such as anthraquinone, carboxylic acids, pre-anthraquinones (Poorniammal et al. 2013). Besides adding desired colours to foods and textiles, fungal pigments have other attractive qualities such as anthelmintic activity (Sreedevi and Pradeep 2016), antidiabetic (Shi et al. 2012), anti-inflammation (Hsu et al. 2012), antimicrobial and antimutagenic (Teixeira, 2012); anti-obesity and anti-tumor (Lee et al. 2013), and antioxidants (Li et al., 2009). Sarkar et al. (2017) reported that pigments from *Aspergillus* sp. and *Penicillium* sp. found non allergic to human skin.



Figure 2. Color filtrate from mixing Aspergillus and Paecilomyces fungi.

Cloth painted with the dye added with vinegar (pH 2) or lemon (pH 3) produced tidy colors; the dye did not break and only occupied the predetermined place, while those administered with the dye alone (control) and the dye added each with other additives, such as baking soda, brown sugar, cream of tartar, glycerine, milk, salt, soda ash, and urea, produced untidy colors (Table 6 and Figure 3 & 4). The dye with a pH of 3 also produced tidy colors (Figure 5). The effect of painting pH can be attributed to the correlation between the pigment structure and cotton cloth. The anion of the pigment has complex characters, and when it is bound on the fiber at pH 2-3, with ionic forces, the ionic attraction would increase the dye-ability of the cloth (Velmurugan et al. 2010b) so that the dye only occupied the predetermined place, producing a tidy color. Meira (2016) stated that glacial acetic acid has the property of absorbing water in a pure state, thereby; making the mixture of the dye and vinegar painted on the cloth produced tidy colors. Aliño (2014) informed that acetic acid also contributes to lowering the dye bath pH. Vinegar is a mixture of glacial acetic acid and water with a ratio of 1:3. Similar results using vinegar was reported by Pliny in Adamu et al. (2013) in making a paste to paint papyrus; and Zhang et al. (2015) on the mixing of white vinegar and ink used for hand-painting silk organza, while Kumari and Singh (2017) added acetic acid in printing paste. Ramelawati et al. (2017) reported that lemon has an acid content that can bind dyes absorbed into the fabric. A similar result was reported by Sarkar (2013) using lemon juice is mixed with dye to get the desired shade for a Mithila painting of Bihar. Cream of tartar had a pH of 3 but produced untidy colors when painted on cloth with the dye. It might be that cream of tartar is containing ingredients which are capable of making the color untidy.

In contrast, cloth painted with the dye pH of 7 and 11; and other additives with pH 4–11 each produced untidy colors (Table 6 and Figures 3, 4 & 5). Velmurugan et al. (2010b) and Tayade and Adivarekar (2013) stated that the ionic interaction between the pigment and cotton cloth at high pH decreased due to the decreasing number of protonated hydroxyl groups on the cellulose leading to the electrostatic repulsion between dye and fiber. Thus, lowering the dye-ability of the cloth at the predetermined place, so the dye broken, produced an untidy color.

Table 6 and Figure 3 and 4 also present the results of color hue on the cotton cloth painted with the dye added with 20 μ l of additives (2%). The dye without additives or control (65B red-purple); and had additives added to it, such as baking soda, brown sugar, cream of tartar, glycerine, milk, salt, soda ash, and urea, each of which produced the same hue but with different intensity, namely 65A, C, and D red-purple. The dye added with vinegar or lemon each obtained 177C greyed-orange colors. The dye with a pH of 3 also produced 177C greyed-orange colors, while those with a pH of 7 and 11 each produced 65C red-purple colors (Figure 5). Even though vinegar with pH 2, lemon with pH 3 (each 177C greyedorange), or a dye with pH 3 (177C greyed-orange) could block dye or was not break when it was painted on cloth, thereby producing tidy colors. However, the resulting colors were different from the control colors (65B red-purple). Our previous study (Suciatmih & Yuliar 2018; Suciatmih 2020) reported that changing the dye pH can change the dye color of mixed Aspergillus and Paecilomyces. The dyes' ionic nature enabled the changes of the molecule structures according to the prevailing pH values and results in different colors and hues at different pH values (Mishra et al. 2012). They further stated that, when the acidic condition (vinegar with pH 2 or lemon with pH 3 or the dye with pH 3) is added to the dye, hydrogen atoms from vinegar or lemon or the dye with pH 3 stick to the dye molecules in such a way that when dyed or painted on cloth, it produced different hues.

Table 6. Effect of 0.2 μ l of additives 2% on the color tidiness and hue painted with the fungal dye.

No	Treatment	Color	Color hue
		tidiness	
1.	The dye 0.2 ml (pH 8) (control)	Untidy	65B Red-purple
2.	The dye 0.2 ml + backing soda (pH 11) 20 µl	Untidy	65A Red-purple
3.	The dye 0.2 ml + brown sugar (pH 6) 20 μ l	Untidy	65D Red-purple
4.	The dye 0.2 ml + cream of tartar (pH 3) 20 µl	Untidy	65C Red-purple
	The dye 0.2 ml + glycerin (pH 4) 20μ l	Untidy	65C Red-purple
6.	The dye 0.2 ml + lemon (pH 3) 20 μ l	Tidy	177C Greyed-orange
7.	The dye 0.2 ml + milk (pH 7) 20 μl	Untidy	65D Red-purple
	The dye 0.2 ml + salt (pH 6) 20 μ l	Untidy	65D Red-purple
9.	The dye 0.2 ml + soda ash (pH 11) 20 μ l	Untidy	65A Red-purple
10.	The dye 0.2 ml + urea (pH 7) 20 μ l	Untidy	65D Red-purple
	The dye 0.2 ml + vinegar (pH 2) 20 µl	Tidy	177C Greyed-orange

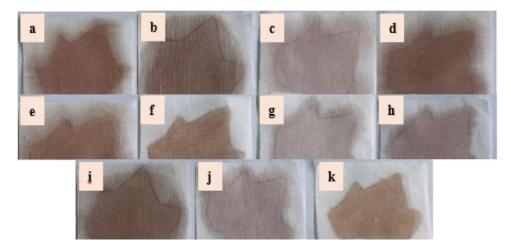


Figure 3. Color tidiness and hue painted with the dye added with additives. The dye (a), the dye + backing soda (b), the dye + brown sugar (c), the dye + cream

of tartar (d), the dye + glycerine (e), the dye + lemon (f), the dye + milk (g), the dye + salt (h), the dye + soda ash (i), the dye + urea (j), and the dye + vinegar (k).

In the previous test, vinegar or lemon applied to the dye produced the painted tidy colors, but the hue was different from the control color (the dye without additives). Therefore, it is necessary to use additives that produce tidy colors with their hue similar to the control when added to the dye. Tables 7 and 8 and Figure 6, 7, and 8 present the combination results between vinegar or lemon and other additives on hue and tidiness of the color on the cloth painted with the dye. Except for the combination of vinegar and soda ash (182C greyed-red, the color hue approaches the control, 65B red-purple) (Table 7 No 8, Figure 4.a, Figure 6.h, and Figure 8.b), the dye was only added with vinegar (177C greyed-orange) and its combination with other additives each produced the same color, namely 173D greyed-orange (the colors are different from the control, 65B red -purple) (Table 7; Figure 3.a, Figure 4.a, Figure 6, and Figure 8.a). It could be because vinegar was acid (pH 2), while soda ash was alkaline (pH 11), and when both are mixed, it will change the pH of the dye so that changing the color of the dye. The baking soda also had a pH of 11, but when combined with vinegar and painted on the cloth with the dye, it produced a different color hue (173D greyed-orange) to the control (65B red-purple). It was possible that baking soda containing ingredients which are capable of making the color hue different from the control.



Figure 4. Color tidiness and hue painted with the dye added with additives. The dye (a), the dye + vinegar (b), and the dye + lemon (c).

Table 7. Combination effect between vinegar and other additives 2% on the color tidiness and hue painted with the dye.

Treatment	Color	Color hue
	tidiness	
The dye 0.2 ml (pH 8) + vinegar (pH 2)	Tidy	177C Greyed-orange
20 μl (control)		
The dye 0.2 ml + vinegar (pH 2) 20 μ l +	Tidy	173D Greyed-orange
backing soda (pH 11) 20 μl		
The dye 0.2 ml + vinegar (pH 2) 20 μ l +	Tidy	173D Greyed-orange
brown sugar (pH 6) 20 μl		
The dye 0.2 ml + vinegar (pH 2) 20μ l +	Tidy	173D Greyed-orange
cream of tartar (pH 3) 20 µl		
The dye 0.2 ml + vinegar (pH 2) 20μ l +	Tidy	173D Greyed-orange
glycerin (pH 4) 20 µl		
The dye 0.2 ml + vinegar (pH 2) 20μ l +	Tidy	173D Greyed-orange
milk (pH 7) 20 μl		
The dye 0.2 ml + vinegar (pH 2) 20μ l +	Tidy	173D Greyed-orange
salt (pH 6) 20 µl		
- /	Tidy	182C Greyed-red
	Tidy	173D Greyed-orange
urea (pH 7) 20 μl	·	. 0
	The dye 0.2 ml (pH 8) + vinegar (pH 2) 20 μ l (control) The dye 0.2 ml + vinegar (pH 2) 20 μ l + backing soda (pH 11) 20 μ l The dye 0.2 ml + vinegar (pH 2) 20 μ l + brown sugar (pH 6) 20 μ l The dye 0.2 ml + vinegar (pH 2) 20 μ l + cream of tartar (pH 3) 20 μ l The dye 0.2 ml + vinegar (pH 2) 20 μ l + glycerin (pH 4) 20 μ l The dye 0.2 ml + vinegar (pH 2) 20 μ l + milk (pH 7) 20 μ l The dye 0.2 ml + vinegar (pH 2) 20 μ l + salt (pH 6) 20 μ l The dye 0.2 ml + vinegar (pH 2) 20 μ l + soda ash (pH 11) 20 μ l The dye 0.2 ml + vinegar (pH 2) 20 μ l +	tidinessThe dye 0.2 ml (pH 8) + vinegar (pH 2)Tidy20 μ l (control)The dye 0.2 ml + vinegar (pH 2)20 μ l + Tidybacking soda (pH 11)20 μ lThe dye 0.2 ml + vinegar (pH 2)20 μ l + TidyThe dye 0.2 ml + vinegar (pH 2)20 μ l + TidyTidycream of tartar (pH 3)20 μ lTidyThe dye 0.2 ml + vinegar (pH 2)20 μ l + Tidyglycerin (pH 4)20 μ lTidyThe dye 0.2 ml + vinegar (pH 2)20 μ l + Tidymilk (pH 7)20 μ lTidyThe dye 0.2 ml + vinegar (pH 2)20 μ l + Tidysalt (pH 6)20 μ lTidyThe dye 0.2 ml + vinegar (pH 2)20 μ l + Tidysalt (pH 6)20 μ lTidyThe dye 0.2 ml + vinegar (pH 2)20 μ l + Tidysalt (pH 6)20 μ lTidyThe dye 0.2 ml + vinegar (pH 2)20 μ l + Tidysoda ash (pH 11)20 μ lTidy

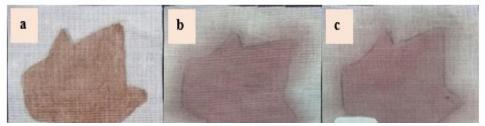


Figure 5. Color tidiness and hue painted with different the dye at pH 3 (a), pH 7 (b), and pH 11 (c).

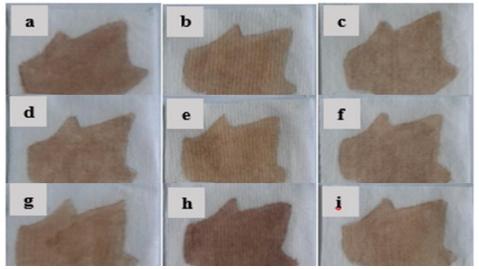


Figure 6. Color tidiness and hue painted with the dye added vinegar and other additives. The dye + vinegar (a), the dye + vinegar + backing soda (b), the dye + vinegar + brown sugar (c), the dye + vinegar + cream of tartar (d), the dye + vinegar + glycerin (e), the dye + vinegar + milk (f), the dye + vinegar + salt (g), the dye + vinegar + soda ash (h), the dye + vinegar + urea (i).

Table 8. Combination effect between lemon and other additives 2% on the color tidiness and hue painted with the dye.

No	Treatment	Color	Color hue
		tidiness	
1.	The dye 0.2 ml (pH 8) + lemon (pH 3) 20	Tidy	177C Greyed-orange
	μl (control)		
2.	The dye 0.2 ml + lemon (pH 3) 20µl +	Tidy	173D Greyed-orange
	backing soda (pH 11) 20 μl		
3.	The dye 0.2 ml + lemon (pH 3) 20 µl +	Tidy	173D Greyed-orange
	brown sugar (pH 6) 20 μl		
4.	The dye 0.2 ml + lemon (pH 3) 20 µl +	Tidy	173D Greyed-orange
	cream of tartar (pH 3) 20 µl		
5.	The dye 0.2 ml + lemon (pH 3) 20 μ l +	Tidy	173D Greyed-orange
	glycerin (pH 4) 20 µl		
6.	The dye 0.2 ml + lemon (pH 3) 20 µl +	Tidy	173D Greyed-orange
	milk (pH 7) 20 μl		
7.	The dye 0.2 ml + lemon (pH 3) 20 μl + salt	Tidy	173D Greyed-orange
	(pH 6) 20 µl		
8.	The dye 0.2 ml + lemon (pH 3) 20 μ l + so-	Tidy	177B Greyed-orange
	da ash (pH 11) 20 μl		
9.	The dye 0.2 ml + lemon (pH 3) 20 µl +	Tidy	173D Greyed-orange
	urea (pH 7) 20µl		

According to Suciatmih and Hidayat (2017), the characteristics of color shadings on the cloth depends on the mordant type used during the

dyeing process. A composition containing the dye, vinegar, and soda ash in the previous test produced the painted tidy colors that controlled the color hue. Thus, the composition is used to test different mordants (alum, ferrous sulfate, and lime) on the color tidiness and hue (Table 9 and Figure 9). Different mordants applied to the mixture of the dye, vinegar, and soda ash painted on the cotton cloth produced different color hues. The mixture, each added with mordant lime and without mordant (control), produced the same tidy and 182C greyed-red colors. When alum was added, it obtained tidy greyed-purple colors at 186 between C and B, and when ferrous sulfate was added, it produced tidy and 201 between B and A grey colors. The same results indicated that different mordants generated various colors on cotton cloth (Suciatmih & Hidayat 2017; Suciatmih 2020) and woolen yarn (Suciatmih & Yuliar 2018) each dyed with the fungal dyes. Satyanarayana and Chandra (2013) informed that mordant helps to tie the dye to the fiber by forming chemical bridges between the dye and the fiber. Pujilestari (2014) reported that the mordant serves to strengthen the color and change the dye's color according to the type of metal that binds it and locks the dye into the fiber. The mixture combined with ferrous sulfate resulted in darker color than alum and lime. Similar results showed that ferrous sulfate produced dark colors on silk Eucalyptus wool fabrics dved with leaf extract and dve (Mongkholrattanasit et al. 2011), cotton cloth and polyester wool dyed with Ficus cunia dye (Kundal et al. 2016), and cotton cloth dyed with fungal dyes (Suciatmih & Hidayat 2017; Suciatmih 2019; Suciatmih 2020).

Except for lime, when alum or ferrous sulfate applied to the mixture of the dye, vinegar, and soda ash produced colored deposits. However, when the mixture was painted on the cloth, the colored deposits were destroyed, thereby enabling the dye to bind to the cloth, but the color result is uneven. This uneven result is due to the difference in pH of the mordant solution. Lime had a pH of 11.8, while alum and ferrous sulfate had pH of 3.4 and 3.1, respectively. Alum or ferrous sulfate causes very high acidity in the dye bath, resulting in colored deposits, which cause uneven color results on the painted cloth (Figure 9 and 10). Similar results were reported by Baig (2012) at pH values lower than 5.5–6 in the bath cause most of the leucovat acid is precipitated so that at the end of dyeing, a black dispersion is produced and, therefore, very little dye is absorbed into the fabric.

Our previous study (Suciatmih & Yuliar 2018; Suciatmih 2020) obtained that a single dye source added to the application of its bath's pH value generates different colors and tones. A composition containing the dye, vinegar, and soda ash is also used to test a combination of different mordants and varieties pH of the dye (3, 7, and 11) on tidy and hue of the color on the cloth (Table 10 and Figure 10). A combination between different mordants (alum, ferrous sulfate, and lime) and pH of the dye (3 and 7) added to the mixture of vinegar and soda ash painted on the cloth each produced the same hue of color (186C greyed-purple, 201B grey, and 182D greyed-red). Except for alum (177D greyed-orange), different mordants combined with the dye pH of 11 produced the same color hue as the previous treatments. The color hue intensity was one level below them (186D greyed-purple and 201C grey). The study indicated that at pH 11, the cotton cloth could not optimally absorb the dyes. Similar results were reported by Tayade and Adivarekar (2013) on Cuminum cyminum seeds extract dye uptake decreased by cotton cloth at pH 11, and Mukherjee and Kanakarajan (2017) on dyeing cotton yarn with Aerva sanguinolenta leaves extract dye in acidic pH showed better result than alkaline pH. Tayade and Adivarekar (2013) stated that the higher pH

causes cellulose to carry more and more negative charged, de-protonated hydroxyl groups, increasing negative charge on the cellulose, leading to an electrostatic repulsion between dye and fiber, which may cause lesser dye uptake. Wang et al. in Mukherjee and Kanakarajan (2017) reported that higher pH oxidizes natural dye's conjugate structure, decreasing the dye-ability of natural dye.

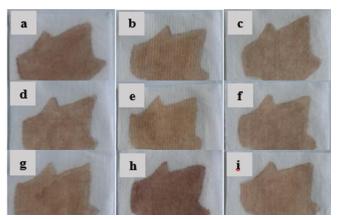


Figure 7. Color tidiness and hue painted with the dye added vinegar and other additives. The dye + lemon (a), the dye + lemon + backing soda (b), the dye + lemon + brown sugar (c), the dye + lemon + cream of tartar (d), the dye + lemon + glycerin (e), the dye + lemon + milk (f), the dye + lemon + salt (g), the dye + lemon + soda ash (h), the dye + lemon + urea (i).

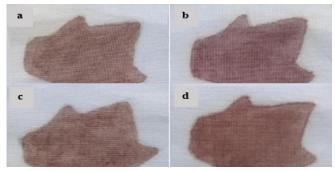


Figure 8. Color tidiness and hue painted with the dye added with additives. The dye + vinegar (a); the dye + vinegar + soda ash (b); the dye + lemon (c); and the dye + lemon + soda ash (d).

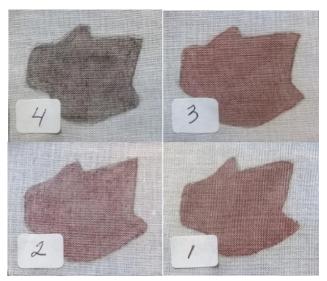


Figure 9. Color tidiness and hue painted with the mixture of the dye, vinegar, soda ash, and mordant, (1) The dye + vinegar + soda ash; (2) the dye + vinegar + soda ash + alum; (3) the dye + vinegar + soda ash + lime; and (4) the dye + vinegar + soda ash + ferrous sulfate.

Table 9. Effect of the mixture of the dye, vinegar, soda ash, and mordant on the painted color tidiness and hue.

No	Treatment	Color tidiness	Color hue
1.	The dye 0.2 ml (pH 8) + vinegar (pH 2)	Tidy	182C Greyed-red
	20 µl + soda ash (pH 11) 20 µl (control)		
2.	The dye 0.2 ml + vinegar (pH 2) 20 μ l +soda ash (pH 11) 20	Tidy	186 between Cand B Greyed-pur-
	μl + alum (pH 3) 20 μl		ple
3.	The dye 0.2 ml + vinegar (pH 2) 20 μ l +soda ash (pH 11) 20	Tidy	182C Greyed-red
	μ l + lime (pH 12) 20 μ l		
4.	The dye 0.2 ml + vinegar (pH 2) 20 μl +	Tidy	201 between Band A Grey
	soda ash (pH 11) 20 μl + ferrous sulfate(pH 3) 20 μl		

Table 10. Combination effect between different mordants and pH of the fungal dye on the painted color tidiness and hue.

No	Treatment	Color tidiness	Color hue
	$0.2 \text{ ml} + \text{vinegar } 20 \ \mu\text{l} + \text{soda ash } 20 \ \mu\text{l} + \text{alum (pH 3) } 20 \ \mu\text{l}$	Tidy	186C Greyed-purple
	$0.2 \text{ ml} + \text{vinegar } 20 \ \mu\text{l} + \text{soda ash } 20 \ \mu\text{l} + \text{ferrous sulfate (pH 3)}$	Tidy	201B Grey
 pH 3 of the dye pH 7 of the dye pH 7 of the dye 	0.2 ml + vinegar 20 μ l + soda ash 20 μ l + lime (pH 12) 20 μ l	Tidy	182D Greyed-red
	0.2 ml + vinegar 20 μ l + soda ash 20 μ l + alum (pH 3) 20 μ l	Tidy	186C Greyed-purple
	0.2 ml + vinegar 20 μ l + soda ash 20 μ l + ferrous sulfate (pH 3)	Tidy	201B Grey
7. pH 11 of the dy	$0.2 \text{ ml} + \text{vinegar } 20 \ \mu\text{l} + \text{soda ash } 20 \ \mu\text{l} + \text{lime (pH } 12) 20 \ \mu\text{l}$	Tidy	182D Greyed-red
	e $0.2 \text{ ml} + \text{vinegar } 20 \ \mu\text{l} + \text{soda ash } 20 \ \mu\text{l} + \text{alum (pH } 3) 20 \ \mu\text{l}$	Tidy	186D Greyed-purple
	e $0.2 \text{ ml} + \text{vinegar } 20 \ \mu\text{l} + \text{soda ash } 20 \ \mu\text{l} + \text{ferrous sulfate (pH } 3)$	Tidy	201C Grey
, ,	e 0.2 ml + vinegar 20 μl + soda ash 20 μl + lime (pH 12) 20 μl	Tidy	177D Greyed-orange

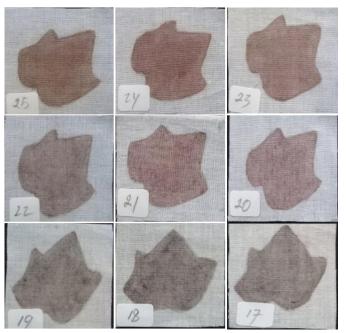


Figure 10. Color tidiness and hue painted with combination between different mordants and the dye pH (17) The dye pH 3 + vinegar + soda ash + ferrous sulfate; (18) The dye pH 7 + vinegar + soda ash + ferrous sulfate; (19) The dye pH 11 + vinegar + soda ash + ferrous sulfate; (20) The dye pH 3 + vinegar + soda ash + alum; (21) The dye pH 7 + vinegar + soda ash + alum; (22) The dye pH 11 + vinegar + soda ash + alum; (23) The dye pH 3 + vinegar + soda ash + lime; (24) The dye pH 7 + vinegar + soda ash + lime; and (25) The dye pH 11 + vinegar + soda ash + lime.

CONCLUSION

In conclusion, a mixed *Aspergillus* and *Paecilomyces* cultured on the modified medium of Baker and Tatum produced potential dyes that can be used in the paint industry. A mixture of the fungal dye, vinegar, and soda ash produced a tidy and good color hue so that it can be used to paint colors on cloth. Different mordants were applied to the mixture of the dye, vinegar, and soda ash produced varied hues of the painted color on the cloth. A combination between different mordants and pHs of the dye (3 and 7) was added to the mixture of vinegar and soda ash painted on the cloth each producing the same color hue. While except for alum, different mordants combined with the dye pH of 11 produced the same color as the treatments, but the intensity of color hue was one level below them. The present study proves the possibility of using the mixed *Aspergillus* and *Paecilomyces* as a source for the natural painting of colors on the cloth. The mixed fungi can be one of the substitute alternatives for many hazardous synthetic dyes for the painting of cloth.

AUTHORS CONTRIBUTION

Agung Adi Nugroho and Suciatmih conducted all the research and drafted manuscript.

ACKNOWLEDGMENTS

Research Center for Applied Microbiology, National Research and Innovation Agency (BRIN, formely LIPI) funded the study via Thematic Project in the Fiscal Year 2019. The authors are grateful to the Research Center for Applied Microbiology - BRIN for providing the laboratory facilities. The authors are also thankful to Ety Suryati and Nurma Nurjanah for the laboratory assistance.

CONFLICT OF INTEREST

The author stated that there is no conflict interest.

REFERENCES

- Adamu, A.K., Yakubu, M.K. & Sunmonu, O.K., 2013. Preparation of starch-reactive dyed pigment to be used in emulsion paint formulation. International Journal of Chemical, Environmental & Biological Sciences, 1(1), pp.2320-4087.
- Aliño, M.F. 2014. *Carissa carandas* fruit extract as a natural fabric dye. Journal of Nature Studies, 13(2), pp.1-12.
- Bader, J. et al., 2010. Relevance of microbial co-culture fermentations in biotechnology. *Journal of applied microbiology*, 109(2), pp.371–387. doi: 10.1111/j.1365-2672.2009.04659.x
- Baig, G.A. 2012. Effect of pH on the coloration of synthetic fibers with indigo blue. *Indian Journal of Fibre & Textile Research*, 37(3), pp. 265-72.
- Baker, R.A. & Tatum, J.H., 1998. Novel anthraquinones from stationary cultures of *Fusarium oxysporum*. Journal of Fermentation and Bioengineering, 85(4), pp.359-361.
- De Santis, D. et al., 2005. Assessment of the dyeing properties of pigments from *Monascus purpureus*. Journal of Chemical Technology & Biotechnology, 80(9), pp.1072-1079. doi: 10.1002/jctb.1285
- Eyini, M., Babitha, S. & Lee, M.W., 2002. Cellulose utilization and protein productivity of some cellulolytic fungal co-cultures. *Mycobiology*, 30(3), pp.166-169. doi: 10.4489/MYCO.2002.30.3.166

- Fukuda, T., Tsutsumi, K. & Morita, H., 2008. Antibiotic activity in coculture: Influence of *Bacillus subtilis* on the antibiotic activity of *Rhi*zopus peka. Japan Journal of Food Engineering, 9(2), pp.99-106.
- Gunasekaran, S. & Poorniammal, R., 2008. Optimization of fermentation conditions for red pigment production from *Penicillium* sp. under submerged cultivation. *African Journal of Biotechnology*, 7(12), pp.1894–1898.

https://jurnal.isbi.ac.id/index.php/atrat/article/view/384 on 29 Oct 2020.

- Hsu, Y., Lee, B. & Liao, T., 2012 Monascus fermented metabolite monascin suppresses inflammation via PPAR-γ regulation and JNK inactivation in THP-1 monocytes. Food Chem Toxicol, 50, pp.1178– 1186.
- Kumari, T. & Singh, D.R., 2017. Assessment the color fastness of developed printing paste using indalcum gum with acid dye. *IJESRT*, 6 (11), pp.97-101.
- Kundal, J., Singh S.V. & Purohit, M.C., 2016. Extraction of natural dye from *Ficus cunia* and dyeing of polyester cotton and wool fabric using different mordants, with evaluation of color fastness properties. Natural Products Chemistry & Research, 4(3), pp.214. doi: 10.4172/2329-6836.1000214
- Kurnia, N., 2011. Melukis pada Kain dan Busana. https:// hastakaryanovi.wordpress.com/2011/03/11/melukis-pada-kain/. on 29 Oct 2020.
- Lee, C., Wen, J. & Hsu, Y., 2013 Monascus-fermented yellow pigments monascin and ankaflavin showed antiobesity effect via the suppression of differentiation and lipogenesis in obese rats fed a high-fat diet. J Agric Food Chem, 61, pp.1493–1500.
- Li, D.L., Li, X.M. & Wang, B.G., 2009. Natural anthraquinone derivatives from a marine mangrove plant-derived endophytic fungus *Eurotium rubrum:* structural elucidation and DPPH radical scavenging activity. J. Microbiol. Biotechnol., 19(7), pp.675-680.
- Marie, M.M. et al., 2015. One bath cationization and dyeing of cotton fabric with brazilwood natural dye. *International Journal of Innovation and Applied Studies*, 11(4), pp.928-41.
- Meira, D.A.R., 2016. Studi komparasi hasil pewarnaan batik dengan ekstrak indigo antara yang menggunakan mordan tunjung dan mordan cuka. Semarang: State University of Semarang.
- Méndez, A. et al., 2011. Red pigment production by *Penicillium* purpurogenum GH2 is influenced by pH and temperature. Journal of Zhejiang University. Science. B, 12(12), pp.961–968. doi: 10.1631/ jzus.B1100039
- Mishra, P.K. et al., 2012. Extraction of natural dye from *Dahlia variabilis* using ultrasound. *Indian Journal of Fibre and Textile Research*, **37**, pp.83-86.
- Mongkholrattanasit, R. et al., 2011. Dyeing, fastness and UV protection properties of silk and wool fabrics dyed with *Eucalyptus* leaf extract by the exhaustion process. *Fibres and Textiles in Eastern Europe*, 19 (3), pp.94-99.
- Mukherjee, S. & Kanakarajan, S. 2017. Application of eco-friendly natural dye from Aerva sanguinolenta on cotton yarn using biomordant. Research Journal of Pharmaceutical, Biological & Chemical Sciences, 8(3), pp. 859-68.
- Nirlane da Costa Souza, P. et al., 2016. Production and chemical characterization of pigments in filamentous fungi. *Microbiology* (*Reading, England*), 162(1), pp.12–22. doi: 10.1099/mic.0.000168

- Poorniammal, R. et al., 2013. Natural dye production from *Thermomyces* sp fungi for textile application. *Indian Journal of Fibre & Textile Research*, 38, pp.276-279.
- Pujilestari, T., 2014. Pengaruh ekstraksi zat warna alam dan fiksasi terhadap ketahanan luntur warna pada kain batik katun. *Dinamika Kerajinan dan Batik*, 31(1), pp.1-9. doi: 10.22322/dkb.v31i1.1058
- Ramelawati, R., Adriani, A. & Novrita, S.Z., 2017. Pengaruh mordan tawas dan jeruk nipis (Citrus aurantifolia) terhadap hasil pencelupan ekstrak bawang merah (Allium ascalonium L.) pada bahan sutera. Padang: State University of Padang.
- Ren, Y.F. et al., 2016. Effect of dye bath pH on dyeing and functional properties of wool fabric dyed with tea extract. *Dyes and Pigments*, 134, 334-41.
- Rini, S., Sugiarti & Riswati, M.K., 2011. Pesona Warna Alami Indonesia. Kehati.
- Rohandi, T. & Listiani, W., 2015. Eksperimen cat lukis pada kertas daluang dari ekstrak warna hijau pada famili daun suji dan pandan.
- Sangeetha, K., Gomathi, R. & Bhuvaneshwari, M., 2015. Dyeing of silk fabric using lemon leaves extract with the effect of different mordants. International Journal of Innovative Research in Science, Engineering and Technology, 4(6), pp. 4692-4697.
- Sarkar, S.R., 2013. Traditional practices of natural dyes for Mithila painting of Bihar. https://www.researchgate.net/publi cation/2975646-00_Traditional_practices_of_natural_dyes_for_mithila_painting_of _Bihar#fullTextFileContent. On 29 Oct 2020.
- Sarkar, S.L. et al., 2017. Exploring textile dye from microorganisms, an eco-friendly alternative exploring. *Microbiology Research Journal International*, 18(3), pp.1-9.
- Satyanarayana, D.N.V. & Chandra, K.R., 2013. Dyeing of cotton cloth with natural dye extracted from pomegranate peel and its fastness. *International Journal of Engineering Sciences & Research Technology*, 2, pp.2664-2669.
- Shi, Y., Liao, V.H. & Pan, T., 2012. Monascin from red mold dioscorea as a novel antidiabetic and antioxidative stress agent in rats and. *Free Radic Biol Med*, 52, pp.109–117.
- Sreedevi, R. & Pradeep, B.V., 2016. Anthelmintic and antibacterial activity of red pigment

from Aspergillus terreus. RJPBCS, 7(2), pp.249-257.

- Suciatmih & Hidayat, I., 2017. Effect of different mordants on cotton cloth dyed with Aspergillus and Penicillium dyes. Aceh International Journal of Science and Technology, 6(1), pp.19-28. doi: 10.13170/ aijst.6.1.5372
- Suciatmih & Yuliar., 2018. Effect of coloring pH and mordant on fungal dyes quality using woolen yarn. *AIP Conference Proceedings*, 2002, 010001. doi: 10.1063/1.5050153
- Suciatmih, Nurfianti & Magfirani, S.V., 2018. Coloring properties assessment of dyes produced by mixed *Aspergillus* and *Paecilomyces*. IOP Conference Series: Earth and Environmental Science, 166, 012023.
- Suciatmih, Anna & Sitorus, J., 2019. Pengaruh teknik pemordanan dan bahan penguat warna terhadap warna kain oleh pewarna jamur campuran. *Prosiding Seminar Nasional Biologi*, 4, pp.846-853
- Suciatmih, 2019. Optimization of dyes production by mixed Aspergillus and Paecilomyces and its coloring on cotton cloth. IOP Conference Series: Earth and Environmental Science, 298, 012016.

- Suciatmih, 2020. Combination of culture and dyeing conditions on cloth color dyed with mixed fungi dyes. *Berkala Penelitian Hayati*, 25(2), pp.18-26. doi: 10.23869/28
- Tayade, P.B. & Adivarekar, R.V., 2013. Dyeing of cotton fabric with *Cuminum cyminum* L. as a natural dye and its comparison with synthetic dye. *Journal of The Textile Institute*, 5(2), pp. 699-706.
- Teixeira, M.F.S. et al., 2012. Amezonian biodiversity: Pigments from *Aspergillus* and *Penicillium* characterization, antimicrobial activities and their toxicities. *Curr. Trends in Biotechnol. Pharm.*, 6, pp.300-311.
- Teli, M.D., Sheikh, J. & Shastrakar, P., 2013. Exploratory investigation of chitosan as mordant for eco-friendly antibacterial printing of cotton with natural dyes. *Journal of Textiles*, 320510, pp.1-6. doi: 10.1155/2013/320510
- The Royal Horticultural Society, 1966. R.H.S. Color chart. The Royal Horticultural Society.
- Velmurugan, P. et al., 2010a. Water-soluble red pigments from *Isaria far-inosa* and structural characterization of the main colored component. *Journal of Basic Microbiology*, 50, pp.581–590.
- Velmurugan, P. et al., 2010b. Dyeing of cotton yarn with five water soluble fungal pigments

obtained from five fungi. *Fibers and Polymers*, 11(4), pp.598-605. doi: 10.1007/s12221-010-0598-5

- Wang, H.Y., Li, P. & Zhou, W.L., 2014. Dyeing of silk with anthocyanins dyes extract from *Liriope platyphylla* fruits. *Journal of Textiles*, 587497, pp.587-97. doi: 10.1155/2014/587497
- Zhang, L., Xiang, C. & Sanders, E.A., 2015. Testing Chinese ink as a natural dyestuff on silk and cotton fabrics: The foundation for a collection of wearable art. ITAA Ann. Conf. Proc. 144. https:// lib.dr.iastate.edu/itaa_proceedings/2015/posters/144 on 29 Oct 2020.