The Use of Natural Pigments to Dye Textiles Literature Review

Elise Burk

Turock School of Arts and Sciences

Keystone College

Mentored by Dr. Vicki Stanavitch

This article will focus on reviewing the recent literature information involving using natural pigments to dye textiles, specifically plant, bacterial, and fungal pigments. To begin, a quick introduction of the use of color throughout time and how synthetic dyes became the universal short-term solution. In addition, the recent motive to rediscover natural pigment uses for commercial purposes will be discussed. Then, multiple terms will be described to establish the background of the specific topics researched, including the categories of natural pigments/biopigments. Some of the most common organisms used for each category of natural pigments will also be explained as well as details of their biosynthesis. Further, this article will continue by mentioning the important factors of processing the pigments including temperature and time duration.

After the preparation and background of biopigments are established, the focus will shift toward the dyeing process and industrial application. These sections of the article will include the methods of dyeing and variables that can impact the overall output, mordant use, and color hue yielded. Not to mention, the color fastness and medical properties will also be explained. Finally, the article will end with an analysis of the data compared to commercial synthetic use, limitations of natural dyes and future research opportunities.

The literature was identified using Google Scholar, Google search engine, ScienceDirect, and Proquest databases through Keystone College's Miller Library. The search terms used were: biopigments, natural pigments, dyeing textiles, synthetic dye, microbial pigments, fungal pigments, spalting, pigment biosynthesis, environmental impacts + synthetic dye, advantages + natural pigments, advantages + biopigments, natural pigments + textile dyeing, biopigments + textile dyeing, eco-friendly textile dyeing, agricultural waste + textile dyeing, dye history.

Biopigments and natural pigments are used interchangeably in most studies, so the terms were replaced with each other when researching.

Dye Introduction + Environmental Impacts. Color has always been a substantial part of society from using different hues to express emotions to increasing social development of civilizations (Ardila-Leal et al., 2021). From the Stone Age to the Palaeolithic and Neolithic eras, all the way to the modern day, color has been advancing to create the best products for human consumption (Ardila-Leal et al., 2021). The advancement in color has gone hand in hand with the establishment of dyes. Natural pigments/biopigments or pigments derived from organic sources like plants, animals, bacteria, and fungi were solely used during early time periods until 1850's when the first synthetic dye was created, named Mauveine, out of coal tar (Ardila-Leal et al., 2021). After the development of Mauveine, synthetic dyes further transitioned into becoming the new routine due to their wide range of color variability, higher reproducibility, cost effective production, and consistent dye quality (Bechtold et al., 2005). However, despite the clear advantages of synthetic dye the long-term environmental impact has been shown to be detrimental. One of the negative environmental impacts of synthetic dyes is detailed in the article, A Brief History of Colour, the Environmental Impact of Synthetic Dyes and Removal by Using Laccases by Leidy D. Ardila-Leal, Raúl A. Poutou-Piñales, Aura M. Pedroza-Rodríguez, and Balkys E. Quevedo-Hidalgo (2021) with their research suggesting that synthetic dyes have become the largest contributor to colored wastewater. In fact, Leidy D. Ardila-Leal, Raúl A. Poutou-Piñales, Aura M. Pedroza-Rodríguez, and Balkys E. Quevedo-Hidalgo (2021) state that 20% of dye is disposed into the water and cause fluctuating pH levels, decrease in the concentration of dissolved oxygen, and increases in total dissolved solids, total nitrogen levels, total phosphorus levels, and non-biodegradable organic compounds all potentially having

carcinogenic, recalcitrant, and overall toxic effects to the ecosystem and society (Ardila-Leal et al., 2021). Not to mention, the synthesis of synthetic dyes on a commercial level relies on non-renewable resources for production and energy usage which furthers the analysis that synthetic dyes are less than favorable for continued long-term use (Gong et al., 2018; Carvalho & Santos, 2015). Henceforth, the increasing exposure to the harmful impacts of synthetic dyes has increased the demand for more eco-friendly alternatives which has led to society turning back to natural/biopigments dyes (Jha et al., 2017; Aman et al., 2022). Biopigments are seen to be non-toxic, non-polluting, and less health hazardous while also having the potential for high productivity due to rapid growth (Jha et al., 2017). They possess a biodegradable nature and are generally considered safer (Carvalho & Santos, 2015). Overall, this shows great promise for biopigments as alternative dyes.

Types of Biopigments and their Biosynthesis. There are many categories of biopigments and they are derived from animal, plant, bacteria, and fungi (Ardila-Leal et al., 2021). Each biopigment source has certain pigments that they biosynthesize, examples being melanin, carotenoids, flavins, quinones, prodigiosins, monascins, and violcein (Jha et al., 2017; Lin & Xu, 2022). The structures between melanin, carotenoids, flavins, quinones, prodigiosins, monascins, and violcein vary which leads to different hues of color and even different properties (Kramar & Kostic, 2022). These pigments are universal between the types of organisms but depend on the species in which they are produced. Melanins usually produce brown-like hues, carotenoids range from yellow to red, flavins also produce yellow tones, quinones range with a broad spectrum from blue to yellows and oranges, prodigiosins and monascins produce a red color, and violcein establish a purple hue (Lin & Xu, 2022). Biopigments are synthesized either through pathways because of outside stress stimuli in the case of bacterial, and fungal pigments

or to cause light absorbance with plants (Lin & Xu, 2022). Fungi and bacteria use pigment as forms of protection or adaptive growth while plants rely on color to bring in nutrients and to even reproduce (Lin & Xu, 2022). However, they also rely on many other factors to process the right quality and quantity of pigment.



Figure 3. Molecular structure of prodigiosin, melanin and violacein.

(Kramar & Kostic, 2022)

Factors that Impact Pigment Processing. Biopigments are produced from natural sources in a variety of different ways. For fungi and bacteria pigmentation is used as an adaptive strategy to protect the organisms from external stressors (Lin & Xu, 2022). Lan Lin and Jianping Xu detail in their article *Production of Fungal Pigments: Molecular Processes and Their Applications*, (2022) that each pigment type's purpose is stated universally as protection against substantial temperatures, detrimental milieu in a host, radiation exposure, oxidative stress, osmotic pressure, dissection, and poor nutrient supply with some such as quinones that provide extra anti-microbial and antiproliferative properties (Lin & Xu, 2022). Due to pigmentation being the protection an increase in the production process can be brought upon by all the external stressors mentioned including pH levels, oxygen levels, radiation exposure, temperature, salt levels, and lack of nutrients (Lin & Xu, 2022). However, U.V. Light and light, in general, have

also been described as factors that impact pigment production usually specifically targeting the color. In one case, *S. bambusicola* is able to produce different shades of hypocrellin pigment under different types of light even in complete darkness (Lin & Xu, 2022). In addition, plant sources also use pigmentation for survival; however, plant pigments aren't based on protection but on gaining nutrients and helping growth (Dolca, 2018). Plants use pigment as a way to bring in light and attract pollinators; this allows species to continue to grow. The pigmentation is also not collected but solely extracted from parts of the plants (Dolca, 2018). The extract requires liquid-liquid extraction so the pigment can be removed and is easier to collect (Gong et al., 2018). Although harvesting time and light absorption also impact plant sources because of plant tendencies to differ in pigments during different parts of the year (Dolca, 2018; Gong et al., 2018). While the biosynthesis of different pigments universally come with the same purpose, to keep the organism alive, the different need for other regulatory mechanisms are what impacts the adaptation for all natural organisms and the overall impact the output of pigment (Lin & Xu, 2022).

Dyeing Application and Impacting Factors. The dyeing application of biopigments is also extremely variable. As stated in the article *Colorfastness of Extracted Wood-staining Fungal Pigments on Fabrics: a new potential for textile dyes* by Eric M. Hinsch, Hsiou-Lien Chen, Genevieve Weber, and Sara C. Robinson (2015), "in order for a dye to "take" in a fabric, the dyeing process must swell the fabric fibers to allow dyeing particles in, then allow the fibers to shrink back down to entrap the dye" (p. 3) This process leaves multiple variables that can impact the result of the dye including dye bath duration, temperature, and pH of the dye bath (Kramar & Kostic, 2022; Hinsch et al., 2015). Furthermore, each pigment relies on a different structure, and because of that structure, and because of that structure they are able to dye textiles based on the

ionic interactions with fiber compounds (Kramar & Kostic, 2022). The different ionic interactions cause a difference in dyeing product compared between each substrate, the commonly tested cotton, silk, wool, and polyester fabrics (Hinsch et al., 2015; Kramar & Kostic, 2022). All pigment types waver between the different substrates; an example is cotton not performing well in multiple studies. One was concluded by Eric M. Hinsch, Hsiou-Lien Chen, Genevieve Weber, and Sara C. Robinson (2015), and the other by Jixian Gong, Fubang Wang, Yanfei Ren, Zheng Li, and Jianfei Zhang (2018) as the fiber had a low uptake percentage of the color (Hinsch et al., 2015; Gong et al., 2018). As for color impacts, mordant use and organism origin were major impacts as well in the dyeing process. Mordants allow the dye to set in the fabric and overall cause a positive effect on the results of absorption percentage, optical density, color yield, and hue quality (Dolca, 2018; Gong et al., 2018; Hinsch et al., 2015; Jha et al., 2017; Kramar & Kostic, 2022; Lin & Xu, 2022; Bechtold et al., 2005). The organism's origin, meaning plant source or strain, heavily impacts the color or ability to produce (Dolca, 2018; Lin & Xu, 2022; Kramar & Kostic, 2022). The variability of factors that impact natural pigment dyeing can be seen as a limitation but biopigments have other crucial properties that provide advantages.

Crucial Miscellaneous Properties. The advantages of using biopigments to dye textiles does not end after mentioning the eco-friendly benefits. Biopigments also provide many other crucial properties that benefit human consumption. When it comes to fungal pigments Lan Lin and Jianping Xu (2022) mention that the functionality of these natural colorants can expand into antioxidant, antimicrobial, anticancer, anti-inflammatory, and immunomodulatory properties to lead to pharmaceutical developments in the treatment of illnesses (Lin & Xu, 2022). Lin and Xu (2022) also mention that for certain species of fungal pigments radiation can cause enhanced growth in certain species that produce pigment adding another functional method of growth and

extraction (Lin & Xu, 2022). Other important properties of fungal pigments include their lack of need of water for both dyeing process or after-treatment, not being fiber specific and the short bonding time required to uptake to a substrate (Hinsch et al., 2015). Moreover, when talking about the extra benefits of microbial sources, similar to fungal pigments, Ana Kramer and Mirjana M. Kostic (2022) mentions the antioxidative, antiparasitic, anticancer, and antimicrobial properties (Kramar & Kostic, 2022). Masoud Aman Mohammadi, Hossein Ahangari, Saeed Mousazadeh, Seyede Marzeih Hosseini, and Laurent Dufosse (2022) also continue with the pharmaceutical benefits as antioxidant, anticancer, antiproliferative, antifungal, antibacterial, and antiviral properties as well (Aman et al., 2022). Not to mention, Ana Kramer and Mirjana M. Kostic (2022) also states that microbial pigments can have U.V. protection functions expanding microbial pigment use (Kramar & Kostic, 2022). In addition, plant sources also have additional benefits which raise their value industrially being that they are easily collectible and have low cost production compared to all other natural pigments (Bechtold et al., 2005). The additional benefits of natural pigments can increase the demand and motive to use as alternatives to synthetics.

Comparison to Commercial Synthetic Use. When talking about dyeing textiles with natural pigments the theoretical purpose would be to solely replace the use of synthetic pigments as an alternative to curb the negative environmental impact. Due to their capability to producing colored pigment, promising properties, and reproducibility biopigments have been described to be considered as a safer option to normal synthetic dyes (Aman et al., 2022; Dolca, 2018; Gong et al., 2018; Hinsch et al., 2015; Jha et al., 2017; Kramar & Kostic, 2022; Lin & Xu, 2022; Bechtold et al., 2005). The colors bring shades of yellow, brown, green, beige, red, and orange as well as some others. Due to the use of mordants, the dyed fabric can have great opacity and color

fastness that rival the alternative (Bechtold et al., 2005). Unfortunately, because of the limitations

of research and biopigments as a whole they are sufficient enough for production, but not necessarily as efficient as synthetic dyes have become (Bechtold et al., 2005). However, biopigments bring other additional functionalities that synthetic dyes do not which raises their value (projected up to \$2 billion by 2026) in industries like medical and food regardless of the potential limitations (Kramar & Kostic, 2022; Lin & Xu, 2022).

Limitations of Biopigments. Despite the promise and advantages towards biopigments as dyes there are also some limitations. One of the main limitations when it comes to biopigments is the lack of variety in color hues universally throughout each type. Out of all the plant, microbial, and fungal pigments extracted and used to dye textiles the colors stayed pretty universal with common themes being yellows, browns, reds, greens and black, mostly muted (Kramar & Kostic, 2022). In fact, the graphs produced by Jyoti Jha, Kumari Meenu, Pragya Sinha, and Priyaragini (2017) show two different types of natural colorants, bacterial and fungal, both creating similar hues of pigment to each other despite molecular differences in their sources (Jha et al., 2017):

Isolate	Colony cl	haracteri	stics			
no.	Color	Margin	Texture	Source	Organism	Pigment
3	Black	White	Powdery	Soil	Aspergillus spp.	Brown, Green in presence of CuSo,
4	Olivaceous green	White	Powdery	Soil	Penicillium spp.	Reddish brown

Table 1. Morphological characteristics of isolated fungi

 Table 2. Morphological characteristics of isolated bacteria

Isolate	Colon	y charact	eristics				
no.	Color	Margin	Texture	Source	Gram's	Oragnism	Pigment
1	Light orange	Smooth	mucoid	Soil	Positive	Strepto- coccus	Light yellow
2	White	Smooth	mucoid	Soil	Negative	Bacillus	Brown
5	Yellow	Smooth	mucoid	Milk	Positive	Staphylo- coccus	Yellow
6	Light green	Smooth	mucoid	Soil	Negative	Pseudo- monas	Green

The lack of different hues and shades of colors of natural colorants limits the aesthetic use of dyes for cosmetic reasons and could limit the demand of products if used industrially. In addition, another limitation seen in natural pigments is the inconsistency. As mentioned earlier there are many factors that can impact the look and function of the biopigment including substrate, temperature, pH, and even the specific strain or source the organism is derived from (Hinsch et al., 2015). The environmental factors commercially would lower the quality due to differences in colors and specificity of the origin (Hinsch et al., 2015). Subsequently, because mordants are used to enhance the color produced when dyeing the likelihood of being used industrially is very high, which could pose the issue of containing heavy metal ions and, with long-term exposure, cause health defects (Gong et al., 2018). Further, some other limitations brought upon by recent research on microbial pigments have cited the lack of technology to cause low stability, high cost of downstream processing, technological failures, low yields, and possible harmful second metabolites (toxins) being produced, all providing a disadvantage

(Aman et al., 2022). Not to mention, due to the appearance of second metabolite creation microbial pigments would have to go into lengthy toxicology testing in order to be approved for human use (Lin & Xu, 2022). As for fungal pigments another limitation they possess is that some strains lack solubility in aqueous solutions which limits the extraction options for the pigments and could increase costs as well (Lin & Xu, 2022). The final limitations seen throughout the analysis of literature are from using plant sources as they highly rely on season dependent factors such as harvesting time and growth which makes these pigments much less reliable (Dolca, 2018). Ultimately despite the advantages that come with biopigments, limitations have also been discussed and provide further questions about future research opportunities.

Perspective Applications of Biopigments. Although a generally newer field of study, natural colorants have become a trend, increasing in publications substantially in the past 10 years (Kramar & Kostic, 2022). The recent interest and expanded research have given way to a variety of new potential questions and applications when it comes to natural textile dyeing. Specifically with microbial pigments Ana Kramer and Mirjana M. Kostic (2022) describes options to further research in strengthening repeatability and durability of the achieved colors, widening the range of colors, in-depth studies on dye-fiber interactions, and exact predictive scales of industrial uses (Kramar & Kostic, 2022). Jyoti Jha, Kumari Meenu, Pragya Sinha, and Priyaragini (2017) also discussed the perspective applications of microbial pigments in textile dyeing by mentioning the crucial need for more research on upgrading the range and quality of the biopigments to completely curb the hazardous effects on the other more commonly used alternative of synthetic (Jha et al., 2017). Masoud Aman Mohammadi, Hossein Ahangari, Saeed Mousazadeh, Seyede Marzeih Hosseini, and Laurent Dufosse (2022) mentioned further opportunities such as developing improved strains, processes to optimize cultivation parameters,

the association of microbial pigments and their pharmacological properties, and using agro-waste as a low cost production option (Aman et al., 2022). This is similar to what Thomas Bechtold, Rita Mussak, Amalid Mahmud-Ali, Erika Ganglberger, and Susanne Geissler (2005) detailed in their study specifically on certain aspects that can improve agriculture waste usage as natural colorants including the optimal plant selection, optimization of extraction, deep-dive into the influence of substrate on shade and fastness, influence of types of mordants, and selection of optimum dyeing conditions (Bechtold et al., 2005). As for fungal pigments, further research ideas were presented in two studies, one by Eric M. Hinsch, Hsiou-Lien Chen, Genevieve Weber, and Sara C. Robinson (2015) encouraged the expansion into research on cellulose based fibers with the other by Lan Lin, and Jianping Xu (2022) detailing exploration into environments that can contain pigment producing fungi, and the optimization of technology to advance the field further (Hinsch et al., 2015; Lin & Xu, 2022). Cristina Carvalho and Gabriela Samos (2015) also call for the more advanced study of the biosources into expanding the range of origins of usable biopigments (Carvalho & Santos, 2015). Overall, due to the newer interest of using natural pigments to dye textile there are plenty of future perspectives able to be studied.

In conclusion, the demand for a more eco-friendly option for synthetics due to their negative health and environmental factors have given way for the alternative use of biopigments. These biopigments have been seen to be a major alternative to synthetic pigments. Biopigments have many crucial advantages to their usage, like their anti-properties, and reproducibility. Despite the potential of these dyes, they do have their limitations including limited range of color, high downstream costs, and risk of toxins, especially with the lack of technology. However, with more studies and future research natural colorants could replace man-made dyes. Bringing more eco-friendly and beneficial impacts to the ecosystem and society.

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