

Special Review Paper: Leather Dyeing with Biodyes from Filamentous Fungi

by

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Abstract

Certain species of filamentous fungi typically produce colored substances as secondary metabolites, which can be used as dyes for industrial applications, especially for products intended to be used in direct contact with the human body such as leather goods. These natural biodyes can be an eco-friendly alternative to synthetic dyes (mainly azo dyes), since they are not originated from extractive activities of the environment and no hazardous chemicals are used while they are produced. Therefore, this biotechnological development for leather dyeing represents an important area to be explored and improved. However, this is a complex challenge due to the requirements of large-scale production with low cost and quality standardization with high stability and fastness. The aim of this study is to present a review on recent literature about improvements of leather dyeing techniques and the search of natural dyes for industrial uses emphasizing the developments related to dyes from filamentous fungi *Monascus purpureus*.

1. Introduction

The dyeing is performed in the wet-finishing step of the leather manufacturing process to impart the sensory characteristics to the final products of color shade and intensity, color penetration and uniformity.¹ The dyes are applied in drums in aqueous floats in wet leather in order to assess and fix both in the surface and interior of the tanned fiber network. Leather dyes are water soluble differently from pigments^{2,3} which are water insoluble and these are used in the finishing operations performed on crust leather manufacture together with polymeric binder substances to coat the surface of the dry leather. However, many authors do not differentiate the definitions of dyes and pigments.

Currently in the leather industry, more than 90% of leathers are dyed with azo dyes⁴ and many of these synthetic dyes extensively

used all over the world have negative impact on human health and the environment.⁵ It is estimated that about 10% of unexhausted dyes are discharged into the waste streams irrespective of the substrate involved in dyeing.⁶ It is estimated that 1 to 5% of the dye applied in the leather dyeing remains in the effluent.⁷ Furthermore, the undesirable pollution associated with colors and dyes in wastewater, especially azo and metal complexes, may result in reduced water reoxygenation capacity, acute and chronic toxicities, and difficulties in water treatment by biological methods, in addition to preventing the reuse of the water in other process steps.^{8,9,10}

Biodyes are natural dyes obtained from sustainable sources and they are important alternatives to synthetic dyes that can be potentially harmful to humans and environment¹¹ due to its better biodegradability and compatibility with the environment.^{12,13} There are several sources of natural dyes, including plant, animals and microorganisms^{14,15} and the use of environment-friendly dyes for leather dyeing is especially important for products intended to be used in direct contact with the human body. Their application must satisfy quality and productivity requirements such as high thermal stability, fastness to UV light, and low cost.¹⁶ Also, the microorganisms used for biodyes production should have non-pathogenic and non-toxic properties, be able to use a wide range of carbon and nitrogen sources for growth, produce high color yield, be stable against salt concentration, temperature, heat and in a broad pH range and be easy of separation.¹⁷ Filamentous fungi represent an incredibly rich and rather overlooked reservoir of natural products, which often show potent bioactivity and find applications in different fields, ranging from crop protection to human medicine.¹⁸ Certain species typically produce colored substances as secondary metabolites that can be used as dyes for industrial applications for various products such as leather, textiles, and food.^{19,20,21} These natural dyes are extracted from natural sources with high productivity, are produced independent of seasonal effects, and need simple quality control.²²

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Manuscript received June 6, 2018, accepted for publication June 17, 2018.

The aim of this study is to present a review of recent literature about improvements of leather dyeing techniques and the search of natural dyes for industrial uses emphasizing the developments related to dyes from filamentous fungi *Monascus purpureus*.

2. Leather Dyeing Techniques Improvements

Several studies aim to enhance the leather dyeing step, including process optimization, use of dyeing auxiliaries and environmentally friendly dyes. The use of ultrasound in leather processing enhances the chrome uptake and reduces the shrinkage in chrome tanning²³, also improves the rate of exhaustion in leather dyeing due to aid in the increasing of the apparent diffusion.^{24,25} A method of two stages dyeing in which leather were treated with the dye, fixed separately in another drum and fat liquored with reuse of 20% from the segregated dye bath, promoted a dye discharge reduction of 77 to 81% in effluent and keeping the required color intensity comparing to conventional dyeing.²⁶ The pretreatment with acid protease in dyeing process resulted in increased absorption and diffusion of dye into the leather matrix.⁶ The application of graft copolymer developed from skin trimming hydrolysate with starch and poly vinyl alcohol at the level of 10% gave improved dye exhaustion of 96% and reduced the biochemical oxygen demand and chemical oxygen demand to the level of 66.7 and 53.0%, respectively.²⁷ The utilization of nanoparticle polymer prepared from amino acids of keratin hydrolysate through copolymerization reaction enhanced dye uptake at level of 99% when 2% dye was used.²⁸

Also, different shades of color on leather can be obtained by changing the mordant used and they can be applied before, simultaneously and/or after the dyeing.²⁹ The use of mordant in dye process can enhance the shade range and improve the affinity between the dye and fibre³⁰ and the most commonly used mordants in natural dyeing are aluminum potassium sulfate, potassium dichromate, stannous chloride, ferrous sulfate and copper sulfate.^{13,31} The oxalic acid used in leather dyeing with the aqueous extracts of tea plant (*Camellia Sinensis*), turmeric (*Curcuma* sp), eucalyptus bark and walnut (genus *Juglans*) by the post mordanting method showed positive influences in the properties of leather, including color fastness to perspiration and color fastness to rubbing.³² Mixed tocopherols are abundantly available in nature and are produced from renewable sources such as soybean.³³ They are well-known antioxidants commonly used in the cosmetic and food industries. Moreover, they have been reported as potent free radical scavengers and highly protective agents for collagen fibers against UV and heat damage.³⁴ The best condition to produce black dyed leather with powder extract of gallnut and sapan wood was achieved using 5% of the natural dye with addition of 1% $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ mordant.²⁹ The dyes from urucum (*Bixa orellana* L.) and cochineal carmine (*Dactylopius coccus*)

showed good surface coating of the leather, good penetration and good dye exhaustion. During leather dyeing, the use of tocopherol in the first addition of urucum provided a more intense color in the wet white leather, but the reverse occurred for the cochineal carmine dye.³⁵ In the leather dyeing with *Bixa orellana* seeds extract, it was determined $81 \pm 1\%$ of dye exhaustion in dye bath and the leather showed fastness to dry and wet rub rating 4/5 and light fastness rating 3 on grey scale.³⁶ The natural iridoids genipin, loganin aglycone, oleuropein aglycone, E-6-O-methoxycinnamoyl scandoside methyl ester aglycone could react with hide powder at 35°C and pH 7.5 - 8.0 within 6 h at 5% of leather mass, imparting different colors such as dark blue, brown-yellow, yellow and mauve. They also improved the hydrothermal stability of hide powder.³⁷ The best leather dyeing conditions with a green fluorescent protein was 5 mM concentration of protein during 4 h and the efficiency of protein binding was estimated around 85 and 96%.³⁸ The natural beetroot dye has been found to be suitable for leather and ultrasound was beneficial in natural dyeing of leather with improved rate of exhaustion.³⁹ The natural dyes carmine cochineal (*Dactylopius coccus*, pink), turmeric (*Curcuma Longa* L., yellow), indigo carmine (*Indigofera*, blue) and annatto were encapsulated in a silica matrix via sol-gel process and injected into a polyvinyl chloride (PVC) matrix in order to enhance the stability of color characteristics after being subjected to Xenon-accelerated weathering.⁴⁰

3. Biodyes Source from Microorganisms

The production of many currently permitted natural colorants has a number of disadvantages, including the dependence on the supply of raw materials and variations of extraction methods,⁴¹ which adds high production cost and renders low resistance to light and heat.³² Many microorganisms such as bacteria, yeasts, fungi, microalgae are biodyes producing in nature and have broad of application⁴² which are more advantageous in terms of production than that extracted from vegetables or animals due to the high growth rate, independence from weather conditions, and cost effectiveness.⁴³ Biodyes of basidiomycetous fungi were used in the past to dye wool and silk, but these fungi are difficult to culture in the laboratory and large-scale industrial conditions.^{44,45} By contrast, many ascomycetous fungi provide a readily available alternative source of natural colorants that can be easily produced with high yields using optimized culture technologies,⁴⁶ without any toxins²² and there is no dependence of the seasonal supply of raw materials, thus minimizing batch-to-batch variations.⁴⁷ The production of biodyes from filamentous fungi is gaining interest owing to their use as food colorants, in cosmetics and textiles, and because of the important biological activities of these compounds,⁴⁸ such as antioxidant, anticancer, antimicrobial, and anticarcinogenic activities.^{49,50,51} Filamentous fungi are known to produce an extraordinary range

Table I
Biodyes produced by fungi and their applications.

Fungal source	Biodyes / Color	Potential use or characteristics
<i>Ashbyagossypi</i> <i>Penicillium oxalicum</i>	Riboflavin (Yellow) Arpink Red - anthraquinone	Food additives. ⁴⁷
<i>Epicoccum nigrum</i> <i>Penicillium flavigenum</i>	Orevactaene (orange) dihydrotrichodimerol (yellow)	Colored compounds with antioxidant potential for industrial applications. ⁴⁸
<i>Fusarium oxysporum</i>	Pinkish-purple Anthraquinone	Wool dyeing. ¹²
<i>Penicillium</i> spp.	Red	Food and beverage. ⁵³
<i>Monascus purpureus</i> <i>Isaria farinosa</i> <i>Emericella nidulans</i> <i>Fusarium verticillioides</i> <i>P. purpurogenum</i>	Red Pink Reddish - Brown Red Yellow	Cotton fabric and Leather dyeing. ^{16,54,55}
<i>P. minioluteum</i>	Red	Leather dyeing. ²⁰
<i>Monascus</i> spp.	Monascorubrin (Orange), Rubropunctatin (Orange), Monascin (Yellow), Ankaflavin (Yellow), Monascusones (Yellow)	Food coloring. ⁵⁶
<i>Penicillium persicinum</i> <i>Penicillium fagi</i>	Reddish-pink Greenish-blue	Pigments not yet characterized. ⁴¹
<i>Paecilomyces sinclairii</i>	Red at pH 3–4 Violet at pH 5–9 Pink at pH 10–12	Pigments not yet characterized. ⁵⁷
<i>Aspergillus ruber</i> <i>Penicillium melinii</i>	Physcion (Yellow) Atrovetin (Yellow)	Food additives. ⁵⁸
<i>Blakeslea trispora</i>	B-carotene and Lycopene (Red)	Food additives. ^{59,60}
<i>Phaffia rhodozyma</i>	Astaxanthin	Food additives. ⁶¹
<i>Dermocybe sanguinea</i>	Anthraquinones: Emodin (brownish-orange); dermocycin (wine-red)	Dyeing of polyester and polyamide fabrics. ⁶²
<i>Talaromyces albobiverticillius</i> 30548	Red colorant	Food industry. ²²

of pigments that include several chemical classes such as carotenoids, melanins, azaphilones, anthraquinones, flavins, phenazines, quinones, violacein and indigo.⁵² The Table I shows some biodyes produced by fungi and their applications.

4. Industrial Use of Fungal Secondary Metabolites

Fungi are known to produce a wide variety of secondary metabolites that can be divided into several classes, namely, terpenes and terpenoids, polyketides, non-ribosomal peptides,

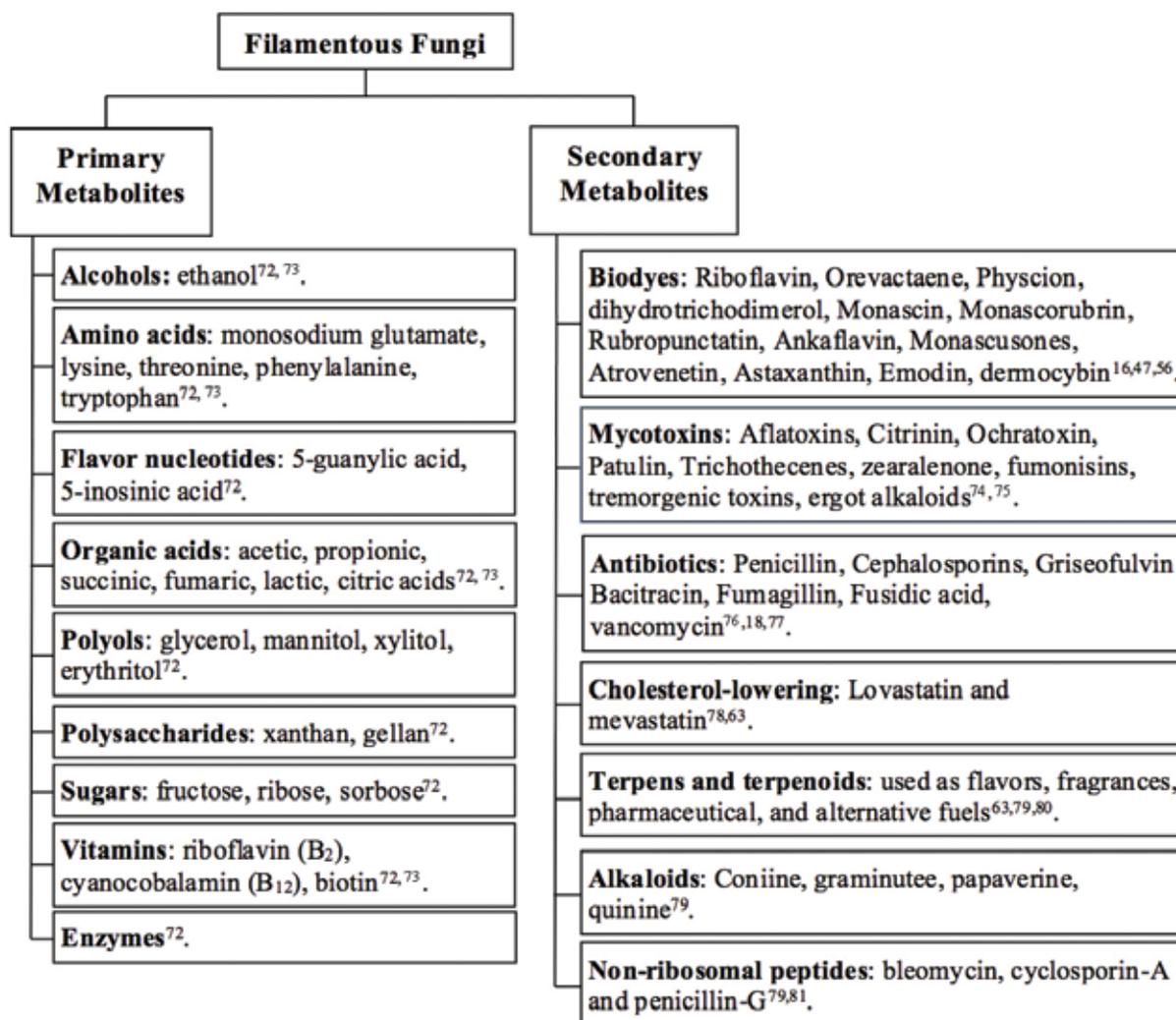


Figure 1. Some important compounds obtained from filamentous fungi metabolites.

and alkaloids. Polyketides are compounds with a high degree of structural diversity and various known biological activities which are considered the most abundant and valuable class of fungal secondary metabolites,⁶³ including antibiotics, biodyes and mycotoxins.

Natural dyes are known for their use in coloring of food,^{46,51} dyeing of leather,^{19,32,35,55} fabrics,⁶⁴ as well as natural protein fibers like wool, silk and cotton as major areas of application since pre-historic times.³¹ Pigments such as β -carotene, Arpink Red, Riboflavin lycopene and *Monascus* pigments are used in food industry and other pigments like Anthocyanin, Prodigiosin and Violacein are widely used in the pharmaceutical industry to treat diseases.⁴² The approval of fungal carotenoids as food colorants by the European Union has strengthened the prospects for fungal cell factories for the production of polyketide pigments.⁴⁵ *Monascus* biodyes have been widely applied to food products as natural colorants and preservative in China and Japan.⁶⁵ Moreover, these colorants possess a range of biological activities

such as antimutagenic, anticancer, antimicrobial, antioxidant, and potential anti-obesity activities.^{57,66,67} They are used as alternative medicine to improve blood circulation, on the treatment of dengue infection, helps on regulation of cholesterol levels and treatment of diabetes mellitus.⁶⁸ The search for new sources of biodyes has increased, mainly because of the toxic effects caused by synthetic dyes used in food, pharmaceutical, textile, and cosmetic industries. Fungi provide a readily available alternative source of natural colorants.¹¹ In nature, light is one of most crucial environmental signals for developmental and physiological processes in various organisms, including filamentous fungi.⁶⁹ The highest production of biomass and dyes (both extracellular and intracellular) from the fungi *Monascus purpureus*, *Isaria farinosa*, *Emericella nidulans*, *Fusarium verticillioides* and *Penicillium purpurogenum* were, respectively, 2.51 g/L of dry mass, 36.75UA/g of substrate and 18.27UA/g of substrate when incubated in total darkness, followed by the use of red, blue, white, green, and yellow light.⁷⁰ Incubation in total darkness increased red biodye production from 14.5 OD/g dry

substrate to 22 OD/g dry substrate. In contrast, growth of the fungus under direct illumination resulted in total suppression of biodyes production.⁷¹ In Fig. 1 is shown some important compound obtained from cultivation of filamentous fungi.

5. Mycotoxin Production

Some fungal strains can secrete toxic compounds during the biodyes production, which limit their application. Therefore, some methods should be explored to avoid or control the production of mycotoxin such as optimizing the production process, addition of nutrients, pH control, screening for mycotoxin free strains, and genetic regulation.

Mycotoxins are natural compounds produced as secondary fungi metabolites of low molecular weight,⁸² usually less than 1000 Daltons,⁸³ with no biochemical significance for the fungal development.⁸⁴ They have wide ranging of biological effects and some of them include potent poisons that are known to inhibit protein synthesis⁸⁵ and can cause several diseases to humans.⁷⁵ The production of mycotoxins due to fungal colonization and contamination can occur in crop plant in the field, at harvest, during post-harvest operations or when the crop product is in storage.⁸⁶ The mycotoxins with the greatest agro-economic and public health impact are aflatoxins (AFs), ochratoxin A (OTA), patulin (PAT), trichothecenes (deoxynivalenol DON, nivalenol NIV, HT-2 toxin, T-2 toxin), zearalenone (ZEN), fumonisins (FUM), tremorgenic toxins and ergot alkaloids,^{87,88} mainly produced by *Aspergillus* (AFs, OTA, PAT), *Penicillium* (OTA and PAT) and *Fusarium* (DON, NIV, HT-2, T-2, ZEN) genera.⁷⁴

Mycotoxins enter the human food chain by consuming contaminated food commodities such as cereals, nuts, dried fruits, coffee, cocoa, spices, oil seeds, and some derived products such as beer, wine, and fruit juices and also by consuming products from animal feed with contaminated source.⁸⁹ Over 500 mycotoxins are currently known and about 20 classes of them are known to cause health concerns in both humans and animals.⁹⁰ The most important fungal genera producing mycotoxins that are found in food products are *Aspergillus*, *Fusarium*, *Alternaria* and *Penicillium*.⁹¹

Fusarium mycotoxins can be categorized into four major groups: *Trichothecenes*, *zearalenone*, *fumonisin*s, and *enniatins*.⁹² Aflatoxins are the most toxic and potent hepatocarcinogenic natural compounds ever characterized,⁹³ wherein Aflatoxin B1 is the most toxic and has been classified as a Group I carcinogen by the International Agency for Research on Cancer.⁹⁴ T-2 toxin is a highly toxic mycotoxin produced mainly by *Fusarium sporotrichoides* and has been reported to have toxic effects on reproductive system of adult male animals.⁹⁵ Ochratoxin A possesses nephrotoxic, immunosuppressive, teratogenic, and carcinogenic properties.²¹

Some strains of *Monascus* can produce citrinin, a nephrotoxic metabolite, which was previously found mainly in some mold species of the genera *Aspergillus* and *Penicillium*.⁹⁶

The Table II lists some known mycotoxin-producing fungi and the type of contaminated materials.

6. Biodyes Production from Filamentous Fungi

Filamentous fungi secrete diverse classes of pigments as secondary metabolites, including carotenoids, melanins, flavins, phenazines and quinones and more specifically monascins, violacein, phycocyanin, or indigo.¹⁰⁵ Several factors influence the production of pigments by filamentous fungi, such as type of cultivation,¹⁰⁶ substrate,^{11,107} pH of the medium,^{108, 109} carbon, nitrogen and mineral sources,^{110,111} effect of light,^{69,70,71} temperature of incubation,⁵⁶ moisture content, and aeration rate,⁴² and mycelia morphometry.^{112,113}

The filamentous fungi produce pigments by solid state or submerged cultivation processes, wherein the productivity and rate of utilization of several nutrients differ in each case. Although many authors refer to these processes as fermentation, the correct term of production in aerobiosis system is cultivation.¹¹ In solid state cultivation, the culture media is in solid form with low moisture content and the substrates are utilized very slowly and steadily, so the same substrate can be used for longer periods. On the other hand, the submerged cultivation is carried out in liquid culture media with high water content and the substrates are utilized quite rapidly, hence need to be constantly replaced/supplemented with nutrients.⁵⁸ The operational parameters as temperature, pH, dissolved oxygen, heat transfer, and homogenization of the culture medium are easier to control in the submerged cultivation than in solid state procedure.¹¹⁴ Submerged cultivation technology was found to be faster and more appropriate for human use, while solid-state cultivation seems to be most appropriate for the production of pharmaceutically active animal feed supplements.¹¹⁵ Generally, dye production in industrial scale by filamentous fungi has been carried out using submerged cultivation,¹¹¹ wherein the solid-state cultivation is advantageous when there is no need to separate the produced biomolecules from the solid matrix.¹¹⁴

The production of microbial pigments by utilizing natural inputs, agro-industrial residues and by-products of industry as low-cost raw materials may not only reduce the process cost but also help in the control of waste disposal and environment pollution.¹¹ Different industries related to the agriculture sector generate a lot of waste in the form of peels, seeds, whey, waste liquid, molasses, bagasse, fruit pulp, pea pod powder, corn steep liquor, bran, which are rich in nutrient components (carbohydrates, proteins, fibers, minerals, vitamins, etc.) that

Table II
Relation of some known mycotoxin-producing fungi with the type of contaminated substrate.

Mycotoxin	Fungi source	Food commodity
Alternariol; altertoxin II	<i>Alternaria alternata</i>	Cereal grains, tomato, animal feeds. ⁸⁹
T-2 Toxin	<i>Fusarium sporotrichoides</i>	Corn, feeds, hay. ⁹⁵
Deoxynivalenol	<i>Fusarium graminearum</i> and <i>F. culmorum</i>	wheat and wheat-based foods. ⁹⁷
Ochratoxin A	<i>Aspergillus carbonarius</i> , <i>A. niger</i> , <i>A. ochraceus</i> , <i>Penicillium verrucosum</i>	Cereals, cheese, Grapes and grape-derived products; Coffee. ^{98,99}
Aflatoxins B1, B2, G1, G2	<i>Aspergillus flavus</i> , <i>A. parasiticus</i> , <i>A. nomius</i>	Rice, oil seeds, nuts, dried fruit, spices, and beans; Coffee. ^{94,99}
Aflatoxins B1; ochratoxin A	<i>Aspergillus sp.</i> and <i>Penicillium sp.</i>	Dry-cured meat products. ^{100,101,75}
Aflatoxins M1	Metabolite of Aflatoxins B1	Milk. ⁸³
Citrinin	<i>Monascus spp.</i> , and some <i>Aspergillus spp.</i> and <i>Penicillium spp.</i>	Beans, fruits, herbs, spices, barley, corn, rice, walnuts. ¹⁰²
Fumonisin Moniliformin	<i>Fusarium moniliforme</i>	Coffee; Corn, sorghum. ⁹⁹
Fumonisin B1, B2, B3	<i>Fusarium verticillioides</i> and <i>F. proliferatum</i>	Maize, sorghum. ⁸³
Patulin (PT)	<i>Penicillium expansum</i> , <i>P. patulum</i> , <i>P. urticae</i> , <i>Aspergillus clavatus</i>	Apple, beans, wheat. ^{83,86}
Cyclochlorotine	<i>Penicillium islandicum</i>	Rice. ⁸⁶
Roquefortine C; Mycophenolic acid	<i>Penicillium roqueforti</i>	Cheese. ¹⁰³
Deoxynivalenol; fumonisin B2	<i>Fusarium graminearum</i> and <i>Fusarium culmorum</i>	Beers, cereals and spices. ^{74,104}
Zearalenone	<i>Fusarium graminearum</i> and <i>Fusarium culmorum</i>	Cereals, maize, wheat, barley. ⁸³

can be considered as potential carbon, nitrogen and mineral sources for microbial production of pigments. The potential of microbial pigments including independence from weather conditions, fast growth and cost effectiveness can be further enhanced by growing on low-cost substrates.⁵⁸ The recovered nutrients from bakery waste in the form of sugars and amino acids can be valorized as feedstocks in fungal bioconversion processes of *M. purpureus*.¹¹⁰ *Monascus purpureus* red pigments were produced in submerged cultivations employing sugarcane bagasse as carbon source in combination with various nitrogen sources, in which the best pigment yields was achieved with peptone and soy protein isolate (organic nitrogen sources) while the use of NH₄Cl (inorganic nitrogen source) had not supported

red pigment production.¹¹¹ Rice water could be an effective substrate for the production of pigments by *M. purpureus* without any external nitrogen source.¹¹⁶ The whole sorghum grain is a potential alternative to rice for the solid-state fermentation of *M. purpureus*.¹⁰⁷

7. *Monascus* Biodyes

Research efforts have been focused on submerged cultivation of *Monascus* in order to increase the biodye yield and to overcome the problems of space, scale up and process control required in solid culture and thus to compensate its economic viability.^{109, 111}

Some studies aim to select and identify strains of filamentous fungi as potential pigment producers using agro-industrial wastes as growth substrate.^{11, 106} The greatest production of red pigments from *Monascus ruber* CCT 3802 using glycerin (by-product from the biodiesel industry) and glucose as substrates was 8.28 UA₅₁₀ with a productivity of 0.13 UA₅₁₀.h⁻¹ and 2.15 g.L⁻¹ of biomass. The nitrogen source and the pH had great effects favoring the production of red pigments and *Monascus* sp. growth.¹¹⁷ It was proved that pH is an extremely important factor for obtaining orange (precursor) or red pigment from *Monascus ruber*, since the isoelectric point of NH₃⁺ sources is critical to converting orange pigment into red pigment. Higher radial growth rate was obtained when *M. ruber* was growing at pH 4.0 and the production of yellow pigment was obtained at low pH (2.0-3.0), while the production of orange and red pigments were in the pH range from 3.0 to 4.0 and above of 5.0, respectively.¹⁰⁸ The red pigment produced by *M. ruber* is not water-soluble and is unstable at extreme pH values (2 and 14) and when exposed to heat and light. These intracellular lipophilic colorants can react with amine groups of proteins, amino acids, nucleic acids, and amino sugars, leading to the formation of extracellular water-soluble pigments. The use of glutamic acid as both a nitrogen source and reactive entity to stimulate the extracellular accumulation of pigments has improved the efficiency of dye production.¹¹⁸ The maximum red pigment yield produced by *Monascus purpureus* MTCC 369 using rice water based medium as substrate with pH of 4.0 and without any ammonium nitrate as nitrogen source after 12 days of incubation period was 20.44 UA₅₀₀/mg dry fungal biomass.¹¹⁶ The higher red pigment production by *Monascus purpureus* MTCC 410 was 14.50 UA₅₀₀ and it was significantly influenced by initial pH of medium, glucose and tryptone concentration.¹⁰⁹ The higher yield of red pigment from *Monascus purpureus* NRRL 1992 using cheese whey, soy protein, and potato dextrose as substrates were 3.70 UA₅₀₉, 20.95 UA₅₁₄, 17.15 UA₅₀₀, respectively.¹¹

Chemical modification of natural compounds could be an interesting field of study as it could appreciably facilitate the synthesis of dye molecules.¹² Several natural dyes are fixed to the fibers aided by mordants, which are usually salts of inorganic elements such as chromium, iron, copper, and aluminum, or organic compounds like tannins. They have the ability to form complexes with the coloring matter, opening fiber structure and allowing the diffusion of the dye into it. The use of mordants^{54,32} and application of antioxidant agents such as tocopherol,^{34,35} as well as the development of new sources of dyes extracted from fungi are promising techniques to enable the application of natural dyes in food, textile, and leather industries.^{53,119}

Many studies have been carried out to minimize the production of citrinin in *Monascus* cultivation such as the form of conduction, the addition of nitrogen compounds the cultivation medium, concentration of dissolved oxygen, pH, and genetic alteration and manipulation.^{114,66} The cultivation of *Monascus ruber*

using culture media supplemented with the amino acids glycine, tyrosine, arginine, serine, or histidine as sole nitrogen sources favored the production of red pigments, and restricted the synthesis of the citrinin. And also, the histidine was found to be the most valuable amino acid as it resulted in the highest production of red dyes and almost completely eliminated the formation of mycotoxin.¹²⁰ An ideal nitrogen source can be selected to control the low final pH and then produce citrinin-free *Monascus pigments*.¹²¹ The production of the mycotoxin citrinin by *Monascus aurantiacus* was successfully eliminated through genetic engineering using ctnB-disrupted strains,¹²² as well as the ctnE gene disruptant reduced citrinin production in 96% and the mutant strain produced 40% more biodyes than the wild-type.¹²³ An efficient and reliable method was proposed for citrinin removal from *Monascus* treated foods by surface active maghemite nanoparticles.¹⁰²

Conclusion

The search of novel potential natural dyes from microbiological sources such as fungal, bacterial, and cell cultures can advance new opportunities of biodyes for leather. Appropriate selection, mutation, or genetic engineering significantly improves the yield of dye or pigment production compared to wild relatives. The development of biodyes must associate both the production and the feasibility of application in the leather dyeing step, according to the required quality parameters for different articles. This is crucial because it involves multidisciplinary knowledge of microbiology/taxonomy, bioengineering, and chemical processing of leather. Several fungi are promising natural sources of biodyes that can be alternatives to synthetic dyes with carcinogenic potential or toxic effects to humans. Natural dyes that are non-toxic and non-allergenic are very important for various sensitive applications. Natural dyes offer scope for eco-friendly dyeing of fibrous materials such as textiles and leather, as well as for food coloration.

Acknowledgements

The authors thank to Conselho Nacional de Desenvolvimento Científico e Tecnológico - CNPq - Edict Universal 14/2013 for the financial support and Coordenação de Aperfeiçoamento de Pessoal de Nível Superior - CAPES for providing financial support for this research.

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Life Lines

Prof. Ph.D. Mariliz Gutterres, Chemical Engineer (1984), M Sc. Engineer (Brazil, 1996), Doctor in Chemistry (2001). Has worked as production manager of a tannery from 1985 to 1989. She is professor at the Chemical Engineering Department at the Federal University of Rio Grande do Sul - UFRGS, since 1990, teaching graduate and post-graduate courses. She is academic advisor of undergraduates, masters, PhD, post-doctor students and technical researchers. Dr. Gutterres is head of the Laboratory for Leather and Environment Studies (LACOURO) at UFRGS and has been Coordinator of the Chemical Engineering Course, Coordinator of the Posgraduate Program on Chemical Engineering and Coordinator of the Research Committee of the Engineering School at UFRGS. She is author of papers, publications and lectures in journals, congresses and conferences. She also participates in several committees and in the management of leather industry associations. Dr. Gutterres received the IULTCS Merit Award 2017 for the Excellence in the Leather Industry.

Prof. Ph.D. Adriano Brandelli, Chemist (1990), Doctor in Chemical Sciences (1994). He has been a professor at the Department of Food Science at the Federal University of Rio Grande do Sul - UFRGS, since 1996, teaching undergraduate and post-graduate courses, advising master and doctorate students and coordinating research projects on bioconversion of agro-industrial byproducts. Dr. Brandelli is head of the Laboratory of Biochemistry and Applied Microbiology at UFRGS and author of papers, publications and lectures in journals, congresses and conferences. He also participates in several committees and as associate editor for Microbiology journals.

M Sc. Wagner Fernando Fuck, graduated in Chemical Engineering at Pontifícia Universidade Católica do Rio Grande do Sul (Brazil, 2006), M Sc. Engineer (2008) and doctorate student in Chemical Engineering at Federal University of Rio Grande do Sul. He has professional experience on issues related to restricted substances analyses in leather and research, development and innovation of leather chemical. He is also author of papers and publications in journals, congresses and conferences.
