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ORIGINAL PAPER

Blue-Green Horizons: Redefining Alginate Bioplastics with Spirulina Dyes

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Abstract

This study introduces a novel, eco-friendly approach for the extraction and application of Spirulina (Sp.) pigments in alginate bioplastics, representing a significant advancement over traditional dyeing methods. The research explores a zero waste model process in a Fab Lab setting, involving three stages, liquid dye preparation, fibre dyeing, and bioplastic dyeing, with a focus on utilizing all by-products sustainably. In the liquid dye preparation phase, vibrant blue-green pigments were successfully extracted from Spirulina powder. The colour depth depended on the Spirulina concentration and the precipitation method used, with pH playing a critical role in achieving a range of green–blue hues. The fibre dyeing phase tested Spirulina dyes on various natural fibres, examining the impact of alum mordant pre-treatment on colour absorption and stability. Optimal dyeing results were obtained with a 1:1 ratio of Spirulina-filtered powder suspension to alum acid precipitation solution at a pH of 4. The techniques optimized were then applied to alginate bioplastics, including bio-yarns and bio-films, yielding a wide spectrum from green to blue. Bio-yarns showed better colour retention compared to bio-films, possibly due to residual alum. However, challenges in long-term colour stability and structural integrity against environmental factors like oxidation and humidity were observed. The study contributes valuable insights into the application of natural dyes in bioplastics, particularly in achieving blue and green shades with a Spirulina zero waste model. This work is significant for future sustainable material science research and emphasizes the importance of balancing aesthetic, functional, and environmental factors in circular design.

Keywords Spirulina dye · Alginate bioplastics · Design through practice · Fab Lab · Circular design

Introduction

The fast evolution of plastic material's physico-chemical properties, and relatively inexpensive production, contributed to its overused. However, it is largely non-biodegradable and ineffectively recycled, resulting in environmental complications (Wang et al. 2020). Recent studies present marine plastic pollution as ecological, economic, and health hazards. The plastic waste burdens marine life (i.e. through suffocation, intake, and absorption). During disintegration, the plastic waste creates micro- and nano-particles polluting fresh water and marine fauna (Stasiškienė et al. 2022). Moreover, polymers from plastic production are already quantified in human blood and in human placental tissue (Leslie et al. 2022; Ragusa et al. 2022). In addition, the textile industry is identified as highly chemical-intensive and a major water polluter. Modern colour compounds used in the plastic and fibre dyes are made based on petrochemicals, resulting in several risks (Kumari et al. 2013). Plastic consumption quadrupled in the past 30 years reaching 460 million tonnes in 2019. Only 9% of plastic waste was recycled, 19% incinerated, 50% goes to landfill, and 22% is

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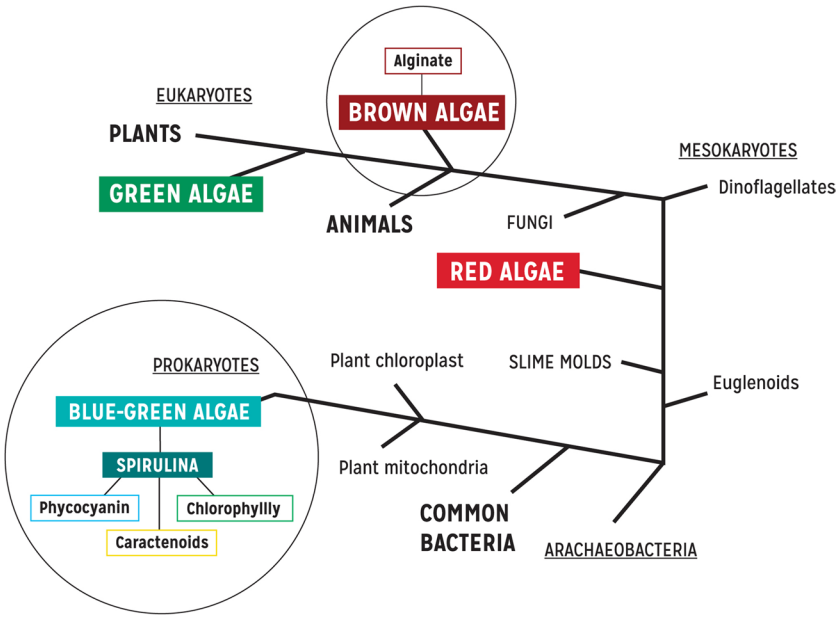

Fig. 1 Phylogenic scheme of Sp. pigments and alginate, adapted from Singh and Saxena (2015)

lost on terrestrial and aquatic environment. From this plastic waste, 40% are from packaging and 11% from textiles (OECD 2022).

Considering the Green Deal (European Commission 2021) and the Sustainable Development Goals 2030 Agenda (Brundtland Report 1987; Desa 2016), significant changes must occur in intensive pollutive industries, as textile and packaging (Sandin and Peters 2018).

Therefore, it is imperative to replace conventional plastics with bioplastics, and new design processes must be explored to reduce or transform waste (Santos 2016; Song et al. 2015).

Sustainable design requires a comprehensive approach that considers the entire product lifecycle and its environmental impact (Cooper 1994; Monteiro-De-Barros 2011). Education in sustainable practices (De los Rios and Charnley 2017; Vezzoli and Manzini 2008) is essential, with practical learning environments. A Fab Lab, originally described by Gershenfeld (2017) of the Massachusetts Institute of Technology (MIT), is a fabrication laboratory for personal fabrication: a shared space with tools and machines for digital fabrication, exploration, prototyping, and testing (Gershenfeld 2017), where teaching methods and design development are based on an interactive approach (Cabrera et al. 2018; Maxwell 2013) following the design through practice methodology (Koskinen 2011).

This research combines biomimicry and sustainable material principles, as presented by Benyus (2008) and Papanek (1985), to investigate zero waste dyeing methods using Spirulina for natural fibres and alginate-based products. The study involved creating and dyeing alginate bioplastics, specifically bio-films and bio-yarns, with Spirulina, and also documented their aging process.

Theoretical Framework

Algae Relevance for Sustainable Design

Algae presents an added value material by neutralizing greenhouse gas emissions from factories, remediating wastewater, and using CO_2 as a nutrient source (Jasmine et al. 2016).

Algae are a class of photosynthetic organisms that harvest the energy of sunlight to convert carbon dioxide to the range of organic compounds required for life, generating oxygen in the process. Remarkably, they constitute two-thirds of the earth's biomass. (McCarty and Kerna 2021).

Depending on their size, algae are named macroalgae (seaweed, i.e. benthic) or microalgae (i.e. planktonic). Macroalgae contain algin gels used for different purposes, like alginate (from brown algae), used for biomedical, food, packaging, and textiles. They are divided into three taxonomic groups, commonly known as green algae (Chlorophyta), red algae (Rhodophyta), and brown algae (Phaeophyceae) (Pereira 2015; Senturk Parreidt et al. 2018). See Fig. 1. To create sodium alginate (i.e. most common alginate salt) bio-based plastics, a hydrogel is made by mixing the sodium alginate powder with water (Gulrez et al. 2011). During this process, called gelification (i.e. creating a gel), glycerin is added as a plasticizer to improve the flexibility (Dianursanti et al. 2018). Hydrogel polymerization occurs when sodium alginate reacts with calcium chloride (solution), creating seaweed-based solid biomaterials and flexible and heat-resistant bioplastic. This process of polymerization that transforms gels

into solid forms is called gelation (van der Linden and Foegeding 2009).

Microalgae are prokaryotic (e.g. cyanobacteria: blue-green algae) or eukaryotic (e.g. Chlorophyta: green algae) photosynthetic (Kim 2015, Chapter 14). Traditionally known as algae, cyanobacteria, in modern classification, are grouped with bacteria (e.g. Kingdom Monera) due to not containing a nucleus or other membranous intracellular structures (such as animals or plants) (McCarty and Kerna 2021). The production costs of microalgae are still relatively high. Costs could be lowered through economies of scale (Ng et al. 2015) or by the use of algae as a by-product (e.g. cultivation of microalgae *Arthrospira platensis* from biological treatment of swine wastewater) (Mezzomo et al. n.d). Microalgal pigments can be water-soluble phycobiliproteins or lipid-soluble carotenoids (e.g. astaxanthin, zeaxanthin, and β-carotene) (Ghosh et al. 2015). Known for their nutritional and energy values (i.e. for biofuel) (NK Singh and Dhar 2011), the rising demand for natural pigments already contributes to the promotion of 'large-scale cultivation of microalgae for pigment production', according to Ghosh et al. (2015).

Spirulina Applications

Arthrospira platensis—Spirulina (Sp.)—is a cyanobacteria biomass (prokaryotic microalgae). Studies on Sp. refer to this microalgae as a super food and a dietary source for malnutrition (Anvar & Nowruzi 2021), presenting a valuable source of vitamins and minerals, known as the richest natural source of vitamin B12 and containing β-carotene (i.e. vitamin A), carbohydrates, and fatty acids (Khan et al. 2005; Sharoba et al. 2014; McCarty and Kerna 2021). Sp. is already used for animal feed in the aquaculture and poultry (Pabbi 2012) as well as in pharmaceuticals and cosmetics (Vieira et al. 2020) and environmental remediation (Mezzomo et al. 2010; Moradi et al. 2021).

Spirulina colours can be achieved from three different extracted pigments: phycocyanin, chlorophyll, and carotenoids (see Fig. 1). Produced by cyanobacteria, phycocyanin is used as a natural blue dye in the food and cosmetic fields (Hamid et al. 2015). Phycobiliproteins (phycocyanin) are water-soluble, while carotenoids are liposoluble. Both are potential colour alternatives to synthetic dyes in the food industry field (Ghosh et al. 2015). Sp. blue pigment, phycocyanin, is environment friendly, non-toxic, and non-carcinogenic (Pabbi 2012).

Phycocyanin is a colouring compound that precipitates with metal or alkali solution (e.g. by precipitation with potassium aluminium sulphate) and becomes a pigment powder (Durgan and Reich 1997).

Sarada et al.'s (1999) analysis of stability on Sp. pigments concluded that phycocyanin was stable over a range of pH = 5 to 7.5 and unstable beyond 40 °C (Sarada et al. 1999). Kumar et al.'s (2013) study on the optimization of Sp. pigment production showed that it is most efficient with alkaline pH (= 10.5) (Kumar et al. 2013). Nevertheless, later work on Sp. phycocyanin shows that optimal pigment extraction occurs at pH = 7.5 and recommended that it should be immediately adjusted to pH = 6.0 to 6.5 after the extraction to maintain stability (Li et al. 2020).

A review of literature and patents showed no studies specifically on Spirulina (Sp.) dyes onto natural fibres or bioplastics.

However, this review contributed significantly to the state of the art. Using databases like Google Patents and Patentscope.WIPO.int and filtering with keywords like 'Spirulina', 'Phycocyanin', 'Fibres', and 'BioPlastics', several relevant patents were identified. For instance, JPS5686958A (Kato & Shimamatsu 1981) proposed a method for obtaining non-toxic phycocyanin for human consumption using water-soluble precipitation with organic solvents. DE202017001259U1 (Rheinländer 2017) described stable liquid blue Sp. extracts without additives but did not specify acidic pH stability. Patent US20180271119A1 (Cagnac 2018) introduced an acidic pH-stable phycocyanin pigment suitable for acidic foods (pH 2–4), derived from other microalgae strains, suggesting a complex protein sequence modification not feasible for Sp. in Fab Lab settings. Lastly, patent US20190313683A1 (Sharma 2019) detailed Sp. infused gummies, focusing on the infusion of micronutrients; however, the high temperatures used in the process compromise the stability of Sp.'s blue and green colours. A recent invention, CN113698473, claims to use a thermodynamic method to improve the Sp. blue's thermal and acid stability (i.e. phycocyanin). However, this invention does not present colour and degradation studies (Shuncheng et al. 2021). Vieira et al. (2020) noted that bioactive extracts like Sp. are highly sensitive to environmental conditions, impacting their stability. DIC Corporation (1985) reports that Sp. phycocyanin is unstable below pH = 4.5.

Furthermore, the literature review highlighted several commercial brands significant to this study in the context of algae-based colouring. LinaBlue®, for instance, notes that phycocyanin pigment remains stable between pH 4.5 and 8.0, up to 60 °C (if promptly cooled) but is highly sensitive to light. Its colour stability can be enhanced with an antioxidant like sodium ascorbate (DIC Corporation 1985). Other notable brands include Spira Inc. (USA) with Electric Sky™ (blue pigment), Algen Laden (Germany) with Spirulina Blau (blue pigment), and LivingInk (USA) offering Algae Ink™ (black printing ink).

Reports on Spirulina and natural dyes were identified as relevant in terms of exploring the feasibility of extracting Sp. pigments and screen printing with Sp. (Mona et al. 2019; Hornis 2018).

As previously mentioned, no studies have been identified regarding the incorporation of Sp. dye into bioplastics.

However, there are a few studies that report on the incorporation of Sp. compounds or biomass. In 2014, it was suggested that Sp. biomass could be utilized in bioplastic development for its protein values, but discarding colours (Wang 2014). It was also demonstrated by Rajmohan and Bellmer (2019) the potential of encapsulating Sp. in alginate beads using ionic gelation. Dianursanti et al. (2018, 2019) studied the mechanical properties of Sp. in the production of bioplastic with polyvinyl alcohol and glycerol. These tested elongation and tensile strength, and only analysed the filler effects. Additionally, Głowińska (2018) found that bioplastics with Sp. and natural rubber matrices are compatible.

In terms of a cradle-to-cradle approach (Braungart and McDonough 2014), Sp. shows promise for designing bio-based or bio-compostable products, contributing positively to environmental cycles (Pandit 2019). Sp. pigments and alginate sources are described in Fig. 1.

Natural Colourants

Materials can be coloured by various processes (e.g. impression, adsorption, diffusion) with synthetic or natural colourants made from organic or inorganic sources. Organic colourants are made from plant sources (e.g. flowers, stems, leaves, bark, wood, algae), animal sources (e.g. insects, molluscs), and organisms (e.g., bacteria, myco). Inorganic colours are made from earth-based sources like minerals, rocks, salts, and metals. Depending on methods, components, and behaviour, natural colourants can be named inks, dyes, or pigments: Ink is a liquid (vehicle) solvent (e.g., water, ethanol) and a colourant applied directly onto the material. This occurs in different conditions depending on the solvent, printing materials, and process applied (e.g. wax ink, polymers, microwave, among others), drying by absorption, solvent evaporation, or precipitation (Nussinovitch 1997). Dye is a liquid colourant soluble in water that incorporates colours into fibres by a chemical reaction, through immersion bath where the fabrics are soaked (e.g. dyestuff; the dye bath), or by dispersion (Gooch 2007). The dye colours are fixed into the material by adsorption (Forman 2016). Pigment is insoluble coloured powder from organic or inorganic sources, made using metal and alkali (Durgan & Reich 1997). It is applied onto the material, using binders (e.g. guar, arabic gum) to bind the particles to the vehicle (Logan et al. 2018). In addition, mordants are chemicals used to fix colours to fibres; these also help in obtaining a vast multiplicity of shades and colours (Logan et al. 2018). Mordants contribute to colour fastness—the dye's level of colouration and properties when adsorbed by the fibres or when washed and the various reactions to light (Ran et al. 2022). Considering the textile and dyeing industries and their contribution to environmental pollution, it is imperative to adjust methods in order to provide for more sustainable patterns of production (Berradi et al. 2019; Ellen MacArthur Foundation 2021). The colouring process can be a zero waste system and use natural dyes, contributing to a significant reduction of environmental hazards (Chequer et al. 2013; Choudhury 2018). A *zero waste natural dye process*, as presented in Fig. 2, plans the materials' flow for natural dyes that can be either compostable or reused (Fabricademy 2021). See Fig. 2.

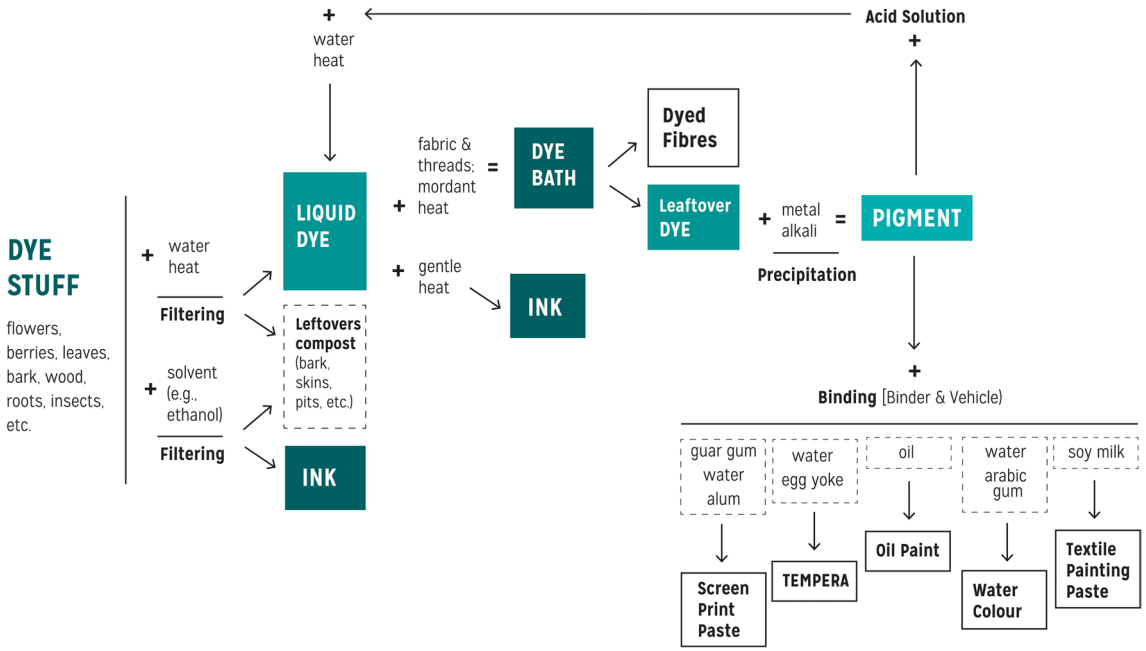

Fig. 2 Zero waste natural dye process, adapted from Cecilia Raspanti, Fabricademy (2021)

Relevance of the Study

In this work on Sp. colour and dyes, the Sp. powder (*Arthrospira platensis*) and its extracted phycocyanin pigment (i.e. commercial) were used to dye various biomaterials, vegetal fibres, and algal bio-based plastic film and yarn. Different Sp. experiments were made to achieve green and blue colours. This research study focuses on the potential of Sp. as a dyeing agent, particularly for producing blue and green hues in natural and alginate-based bioplastics. Utilizing a 'zero waste natural dye process', the study aims to optimize this approach for effectively imparting a range of colours, from green to blue, to bioplastics in various forms such as yarn and film. The study presented utilizes Fab Lab facilities, emphasizing sustainable approaches, and fosters education through practical design and the sharing of knowledge. During the literature review, studies of printing with Sp. were found (Hornis 2018; Mona et al. 2019; Rajmohan and Bellmer 2019), but none focus fibres and bioplastic dye, or any considerations on dyeing on a Fab Lab facility or through do-it-yourself (DIY) processes and their limitations.

To this end, one of the primary purposes of this research was to grasp the possibility of obtaining a natural dyeing compound (for cotton or bioplastics) in a home-made scheme or a Fab Lab setting (Cabrera et al. 2018). Using an design through practice methodology, especially in the context of Fab Lab practices (Cabrera et al. 2018; Koskinen 2011), it was created a 'process flow' for Sp. dyeing process. This flow chart presents optimized flow diagram that encapsulates all phases, processes, and materials (Haffmans et al. 2018). This study specifically investigates the effectiveness of Sp. pigments' performance, for instance, through extraction and dyeing methods on nature-based materials, utilizing a zero waste process. We conducted a series of experiments to optimize each step of the zero waste Spirulina (Sp.) dyeing process: (1) Initially, we achieved an optimized dye bath. (2) Subsequently, this was tested on commercial natural fibres, during which we optimized the fixing process and achieved a broad spectrum of colours ranging from green to blue. (3) Finally, the refined procedure was tested on homemade bioplastics, including yarn and films in various geometries. In both cases—whether using commercial natural fibres or homemade bioplastics—aging analysis was considered, evaluating how the colour held up 1 and 10 months later.

Materials and Methods

This study, building upon the foundations of algal pigment extraction (Mona et al. 2019) and the principles of zero waste natural dye processes (Fabricademy – Textile Academy 2021), introduces a novel zero waste dyeing process utilizing Spirulina (Sp.). The study is conducted in three distinct stages:

1. Liquid dye preparation: in the first step, liquid dyes are prepared using Spirulina powder aqueous suspensions. This process creates a dye bath and extract pigments. The process of extracting and using dye from Spirulina primarily involves the extraction of this pigment (phycocyanin).
2. Fibre dyeing: the second step involves using the dye bath from the first step to dye commercial natural fibres. These include cotton fabrics and reused cotton yarns, as well as seaweed yarns and threads. The goal is to achieve the desired Spirulina-specific green–blue hue (see Table 1 for details).
3. Bioplastic dyeing: in the final stage, the most effective results from the previous steps are employed to dye alginate-based bioplastics. This includes the fabrication of bio-yarns through extrusion and bio-films through moulding. Phycocyanin pigment, another algae derivative, is also utilized at this stage. The process of creating the alginate bioplastic (in film or sheet form) and bio-yarn involves specific gelification (Gulrez et al. 2011; Dianursanti et al. 2018) and gelation (van der Linden and Foegeding 2009) procedures.

Table 1 Summary of the bio-based materials tested with the Spirulina colouring compounds

Fibres	Construction	Commercial	Fabricated	From	Source
Tricoline cotton	Fabric	X		Unknown, NL	100% cotton
Re-use cotton	Yarn	X		Rosários4®, PT	100% recycled cotton
Seaweed yarn	Yarn	X		Bart & Francis, BE	100% Sud African Kelp (viscose)
Seaweed thread	Thread	X		100% Sud African Kelp (viscose)	100% Sud African Kelp (viscose)
Alginate bioplastic	Film sheet		X	Unique Products®, NL	Sodium alginate (E401) (i.e. *Fucus*, *Laminaria*, and *Macrocystis genera*)
Alginate bio-yarn	Yarn		X	Sodium alginate (E401) (i.e. *Fucus*, *Laminaria*, and *Macrocystis genera*)	Sodium alginate (E401) (i.e. *Fucus*, *Laminaria*, and *Macrocystis genera*)

Figure 3 illustrates the innovative Spirulina zero waste process flow developed in this study. It is divided into three key flow processes: liquid dye, dye bath, and leftover dye. Various fabrication techniques, such as Spirulina pigment precipitation, Spirulina dye formulation, bioplastic polymerization, and bio-yarn polymerization, are employed across these stages. Further fabrication details can be found in the supplementary information (Sect. 1. Processes).

To document the durability and stability of the materials, photographs were taken as they were prepared, after 1 month, and again after 10 months, capturing the aging process.

The Sp. experiments were made from *Arthrospira platensis* in the form of powder and phycocyanin pigment (Pp.) (i.e. the commercial blue pigment already extracted from Sp.). Sp. powder was purchased from Iswari, a Portuguese store, and phycocyanin (Sp. blue) was ordered online from the German company AlgenLaden. Household and Fab Lab materials were used: pots, blending mixers, precision scale, moulds, glass jars with lid, sprayer, colanders, and funnels; unbleached paper round coffee filter (Original 1, Melitta®, FSC certified—environmentally friendly and compostable); spoons; latex gloves; pH and temperature readers; 150 mL syringe; electric kettle; and stove. Several chemical reagents were used: calcium chloride ($CaCl_2$, 94–97% (E509), Arsegan®), glycerine; tap and demineralized water; alum powder (potassium aluminium sulphate; $KA_l(SO4)_2) > 99\%$ BTS®; and sodium carbonate ($Na_2Co_3, > 99\%$, from www.deoplosmiddelspecialist.nl).

Experiments

This section is divided according to the following three distinct stages:

1. Liquid dye preparation
2. Fibre dyeing
3. Bioplastic dyeing

In each of these stages, a series of experiments were conducted due to the extensive and complex nature of the overall experimental process. (For detailed descriptions of these experiments, please refer to the supplementary information.) The primary focus of each stage is to optimize the process for producing a wide spectrum of colours, ranging from green to blue. A significant emphasis is placed

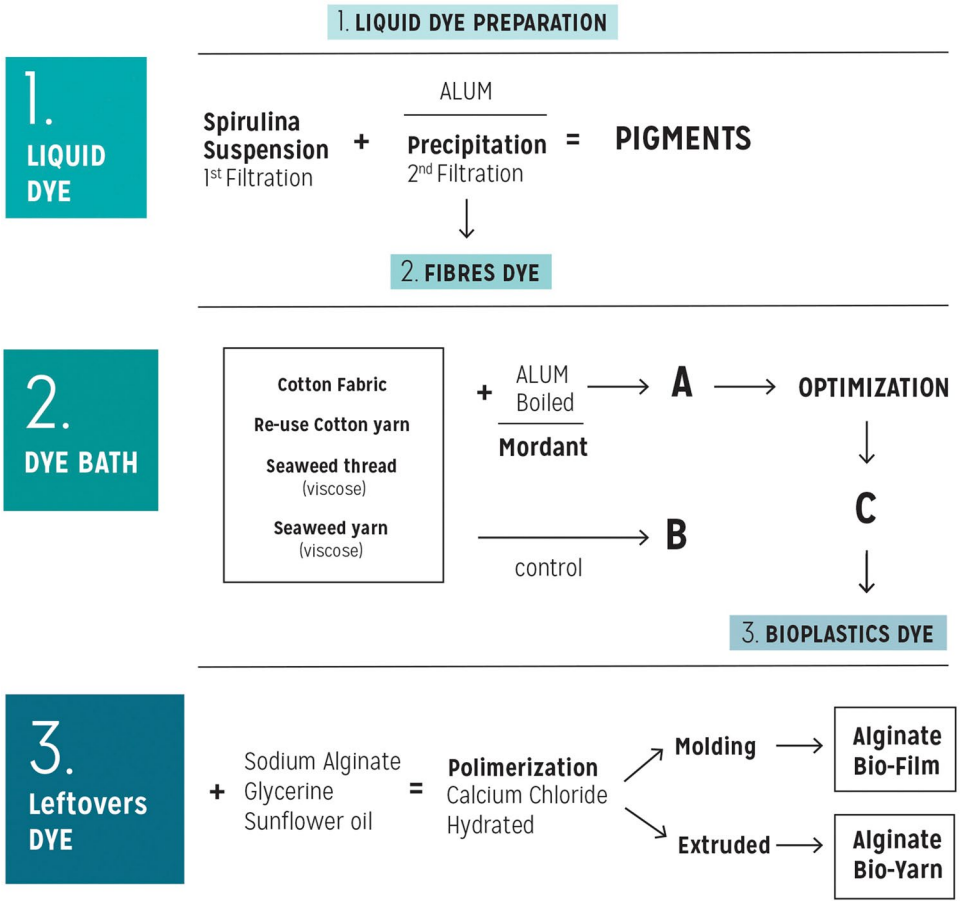

Fig. 3 Zero waste process flow—schematic diagram for fibres and bioplastic dye

on ensuring the high durability of colour fixation on the bioplastics.

Liquid Dye Preparation

The experiments conducted with Spirulina (Sp.) powder focused on extracting pigments to obtain a liquid dye—derived from an aqueous suspension of Sp. powder after filtering. The parameters of each experiment were meticulously adjusted to stabilize the liquid dye and achieve vibrant blue-green colours. This involved balancing the pH levels of the liquid dye, which have been reported as unstable at pH levels below 4.5 (DIC Corporation 1985) and stable within a pH range of 5 to 7.5 (Sarada et al. 1999). Additionally, temperature control was critical, as unstability is reported at temperatures above 40 °C. These measures ensured the stability and effectiveness of the liquid dye.

Liquid Dye Without Precipitation—Experiment 1

In a preliminary experiment conducted without precipitation, to observe direct application of a liquid dye onto the bioplastic (bio-yarn form). The liquid dye was prepared as a filtered suspension, consisting of Sp. powder with tap water (TW), at a concentration of 1.25 g/L. This mixture was stirred for 2 min at room temperature before use in bioplastic fabrication. During the bio-yarn polymerization process it was used a hydrogel that incorporated a quarter of the prepared liquid Dye. This mixture was then extruded and subsequently cured to form the final bioplastic product (consult in SI Table A1 and process in 2.1. Fibre Dye A).

Liquid Dye with Two Precipitation Solutions (Acid and Alkaline)—Experiment 2

The liquid dye was produced using a filtered aqueous Sp. suspension at a concentration of 6.25 g/L (see SI Table A1, Experiment 2). Concurrently, two solutions were prepared:

- An acidic precipitation solution was made by dissolving 10 g of $Al_2(SO_4)_3$ (alum (A.)) powder in 100 mL of distilled water (DW) at 60 °C, achieving a pH of 3.5. Once the temperature decreased to 37 °C, this solution was added to the previously prepared liquid dye.
- To enhance pigment stability, as suggested by Sarada et al. (1999), an alkaline solution was introduced. This solution comprised 5 g of sodium carbonate (Na_2CO_3) dissolved in 50 mL DW at 60 °C, resulting in a pH of 10.

Following these additions, precipitation occurred. The precipitate was then filtered out to obtain what is referred to as the dye bath (consult in SI, Table A2, Experiment 2).

Liquid Dye with One Precipitation Solution (Acid)–Experiments 3 to 10

Experiments 3 to 10 followed the initial step of using a liquid dye, as described in Experiment 2. A concentration of 25 g/L was used in all these experiments, except for Experiments 5 and 6, which had concentrations of 17 and 5 g/L, respectively.

After filtering, the liquid dye (SI, Experiments—1. Liquid Dye Preparations) was then combined with varying concentrations of an acidic $Al_2(SO_4)_3$ precipitation solution. The concentrations ranged from 25 to 83.33 g/L and pH levels were kept consistent, and the temperature was maintained between 37 and 40 °C. (Consult in SI, Table A3. Sp. Pigments Precipitation—Final formula—Experiments 3 to 10.) Post-filtration, the paper filter was left to dry, shielded from light, and observed for changes over a period of 1 month. The dye bath obtained with the conditions of Experiments 3–10 was analysed in terms of colour and transparency.

Fibre Dyeing

The Spirulina dye bath, prepared as per the methodology of Experiments 6–10 (Sect. 1.3, SI Table A3), was utilized for dyeing commercial fibres. This process involved selecting the most effective dye bath from Experiments 6–10 and applying it to natural fibres with and without mordant.

To assess the dyeing effectiveness, various collections of commercial natural fibres (detailed in Table 1) were grouped and dyed in three distinct methods:

- Fibre dye A—dyeing Sp. dye bath with mordant bath
- Fibre dye B—dyeing Sp. dye bath without mordant bath
- Fibre dye C—different Sp. sources (Sp. powder, Sp. suspension filtered (liquid dye), and commercial phycocyanin pigment) onto natural fibres with mordant bath

Fibre Dye A—Dyeing Sp. Dye Bath with Mordant Bath

Dye baths from Experiments 6 to 10 (from previous Sect. 1.3; SI-Table A1) were selected to dye the fibres. Before the dye bath, the fibres were mordanted with alum to fix the colours. Fibres were boiled in a 10% alum solution, dipped in the dye bath at 18 °C for 1 h, and left to dry at room temperature. This collection of samples is summarized in SI Experiments: 2.1. and Table A3.

Fibre Dye B—Dyeing Sp. Dye Bath Without Mordant Bath

A second collection of fibres was dipped directly into directly on the same dye baths used in the step Fibres Dye

A (i.e. dye bath) from experiments 6 to 10 (1.3 section; SI-Table A1). This group was dyed without the mordant bath, to observe the performance of the dyes directly onto the fibres. The materials were immersed in dyeing baths at 18ºC for 1 h and left to dry at room temperature to observe the performance of the dyes directly onto the fibres. The materials were immersed in dyeing baths at 18 °C for 1 h and left to dry at room temperature to observe the performance of the dyes directly onto the fibres (see SI, Table A3).

Fibre Dye C—Different Sp. Sources onto Natural Fibres with Mordant Bath

Following the previously described dyes, the collection of fibres C was done with an equal collection of fibres (as in A and B collections in Table 1). A selection of three dye baths were used to achieve different hues: Sp. liquid dye after the 1st filtration was used directly as a bath to obtain green, Sp. liquid dye bath from Experiment 8 (green–blue selected from Sect. 2.2., sample 8A based on the dye bath from 1.3—Experiment 8), and aqueous solution with commercial Pp. suspension (blue). Fibres were mordanted before (as 2.1.) and dipped for 1 h in the different dye bath solutions. See Table 2.

Bioplastic Dyeing

Alginate bioplastic films and bio-yarns were made using the dye suspensions leftover from the "Fibre Dye C—Different Sp. Sources onto Natural Fibres with Mordant Bath" section. For the hydrogel preparation, 12 g of sodium alginate (S.A.), 20 g of glycerine (Gly.), and 10 g of sunflower oil (Sf. O.) were added to 400 mL of each dye solution that was leftover (i.e. with A. remains from prior mordant fibres). This solution was mixed with an electric hand-mixer and rested during the night to release the air bubbles. The sodium alginate hydrogels were then extruded and moulded to form bio-yarns and bio-films, as here described:

Alginate Bio-yarn

For the bio-yarn extrusion, it was used a 150 mL syringe with 5 mm nozzle, filled with the bioplastics. Polymerisation occurred by extruding the syringe into a 10% C.Cl. solution with 1 L of tap water (pH = 6.5/20 °C). In order to stop the curing process and remove the residues of C.Cl., the bio-yarns were washed in T.W. and then left to dry for two days. They were then documented photographically. See Table 3.

Alginate Bio-films

Using the remaining 250 mL bioplastic (from the "Alginate Bio-yarn" section), bioplastic films were cast in Petri dishes and acrylic boards previously sprayed with 10% C.Cl. solution. The bio-film moulds were then sprayed again on top and were left to dry for 2 days and pictured. See Table 4.

Bioplastics (both bio-yarn, bio-films) and biodegradation were observed after 1 month and 10 months to assess colour fixation over time, as well as colour aging.

Results and Discussion

In this study, we introduce a novel method for extracting Spirulina pigments and applying them in the fabrication of alginate bioplastics. This method is based in the algal pigment extraction process outlined by Mona et al. (2019) and the zero waste natural dye approach described by Fabricademy—Textile Academy (2021). The traditional process typically involves two stages: the liquid dye and the dye bath. However, our novel zero waste model consists of three steps, as depicted in Fig. 3.

Table 2 Fibre dye C—dyeing different Spirulina sources onto natural fibres

Experiment		Dye Bath		Process
Nº	Description	C Sp (g/L)	C A (g/L)	methods and times
11	Sp. Liquid Dye (green) 1st Filtration	12.5	0	Liquid dye- Sp. aqueous suspension. - 1st Filtration. Fibres in Mordant bath, raised and washed in T.W. Dye bath 1 hour, raised, dried.
12	Sp. Liquid Dye (green) 1st Filtration	6.25	0	
13a	Sp. Liquid Dye 2nd Filtration (green-blue)	18.18	18.18	Dye bath - Sp. aqueous suspension+ precipitation+ 2nd Filtration Fibres in Mordant bath, raised and washed in T.W. Dye bath 1 hour, raised, dried.
13b*	Sp. Liquid Dye 2nd Filtration (Green-blue)	18.18	18.18	Dye bath - Sp. aqueous suspension+ precipitation+ 2nd Filtration Without Mordant bath . Dye bath 1 hour, raised, dried.
14	Dye Pp. (blue)	5	0	Pp. suspension. Fibres in Mordant bath, raised and washed in T.W. Dye bath 1 hour, raised, dried.
15	Dye Pp. (blue)	2.5	0	

Table 3 Alginate bio-yarn with leftover dye bath

Experiment		Leftover dye		Gelification	Cure	Process
Nº	Description	From	C (g/L)	C (g/L)	C (g/L)	Methods and times
16A	Sp. leftover dye + S.A. Bio-yarn	Dye bath 11	12.5 Sp	30 S.A 50 Gly 25 Sf. oil	100 C.Cl. (bath)	Leftover dye bath Gelification. Extrusion and cure Wash in tap water. Extend and dry
17A	Sp. leftover dye + S.A. bio-yarn	Dye bath 13a	18.18 Sp	30 S.A 50 Gly 25 Sf. oil	100 C.Cl. (bath)	Leftover dye bath Gelification. Extrusion and cure Wash in tap water. Extend and dry
18A	Pp. leftover dye + S.A. bio-yarn	Dye bath 14	5 Pp	30 S.A 50 Gly 25 Sf. oil	100 C.Cl. (bath)	Leftover dye bath Gelification. Extrusion and cure Wash in tap water. Extend and dry

Table 4 Alginate bio-films with leftover dye bath

Experiment		Leftover dye		Gelification	Cure	Process
No	Description	From	C (g/L)	C (g/L)	C (g/L)	Methods and times
16B	Sp. leftover dye + S.A. bio-yarn	Dye bath 11	12.5 Sp	30 S.A 50 Gly 25 Sf. oil	100 C.Cl. (spray)	Leftover dye bath Gelification Cast in moulds, cure, dry
17B	Sp. leftover dye + S.A. bio-yarn	Dye bath 13a	18.18 Sp	30 S.A 25 Sf. oil	l. (spray)	Leftover dye bath Gelification Cast in moulds, cure, dry
18B	Pp. leftover dye + S.A. bio-yarn	Dye bath 14	5 Pp	30 S.A 50 Gly 25 Sf. oil	100 C.Cl. (spray)	Leftover dye bath Gelification Cast in moulds, cure, dry

Figure 4 illustrates the Sp. zero waste model, summarizing the three fabrication stages of dyed bioplastics, which include yarn and films, with colours ranging from green to blue.

The first step involves preparing liquid dyes to extract pigments and collect Sp. aqueous suspensions, as shown in Figs. 3 and 4 (1. liquid dye). Following precipitation and a secondary filtration, a dye bath is obtained (Fig. 3, 2. dye bath).

In the second step, fibre dyeing (Fig. 4), we conducted tests and optimizations using the suspensions from the dye bath obtained in the first step. This process was aimed at dyeing commercial natural fabric and fibres, selecting the desired Sp. green–blue hue (Fig. 3, 2. dye bath).

The final step, bioplastic dyeing (Fig. 4), involved analysing the previous results and using the best outcomes to develop alginate bioplastics. This included fabricating bio-yarns through extrusion and bio-films by moulding (Fig. 3, 3. leftover dye). Materials were photographed as prepared, after 1 month, and after 10 months to document the aging process.

Liquid Dye Preparation

In the initial step, an optimization study was conducted to select green–blue shades. Ten liquid dyes were prepared for the extraction of Sp. pigments and Sp. liquid dye solutions. Various conditions were explored, including those without precipitation and direct use onto bioplastics, with alkaline and acid precipitations, and with only acid precipitations. The goal was to achieve and select blue-green hues. Following the filtration for pigment extraction, the remaining liquid (i.e. the dye bath) was reserved for the subsequent step of dyeing natural fibres.

Experiment 1 was a preliminary trial conducted without precipitation, where the liquid dye obtained from the first filtration was applied directly onto the bioplastic in its bio-yarn form. Initially, the aqueous suspension of Sp. powder was filtered, and the resulting liquid dye was then used in a gelation solution to create alginate bio-yarns. The yarns exhibited very faint coloration, and rapid discoloration was observed (SI, Fig. 13). Complete colour loss was noted after 3 months. As a result, this approach to bioplastic dyeing was deemed ineffective. It yielded only a faint colour, and the method was inadequate for ensuring colour durability and fixation.

On the other hand, in Experiment 2, we employed the precipitation method, where two solutions were used: an acid precipitation of potassium aluminium sulphate (A.) and an alkaline precipitation of sodium carbonate (S.C.). Following the precipitation process, the remaining liquid was filtered, yielding a liquid dye bath with a very light blue-green pigment (SI, Fig. 14). In literature, it is suggested that the best strategy for extracting Sp. pigment is

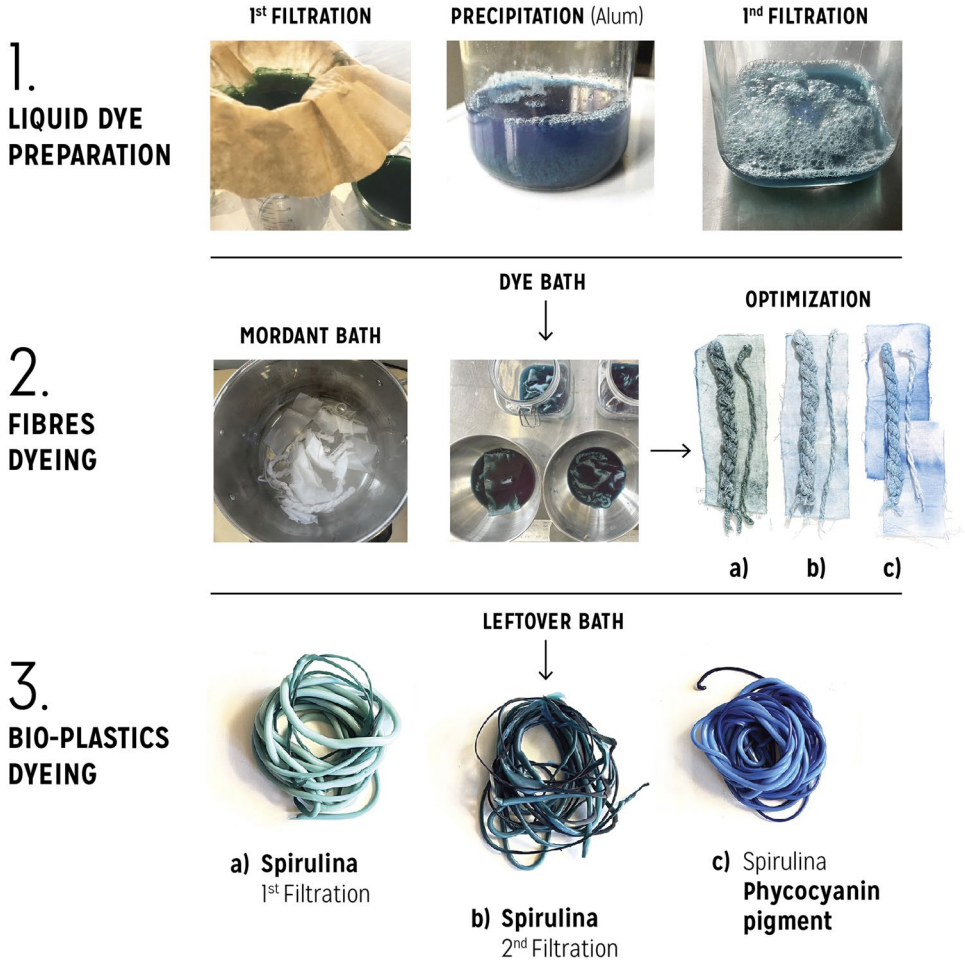

Fig. 4 Schematic of the fabrication methods and stages for fibre and bioplastic dye optimization following the zero waste process flow. (1) Liquid dye preparation—the initial stage involves containing the dye solution through different stages. (2) In fibre dye stage, various dye baths were tested to achieve a green–blue spectrum: (a) green hue obtained using the liquid dye from the first filtration, (b) an experimental optimization of the dye bath after precipitation and second filtration, and (c) use of commercial phycocyanin pigment. In the (3) bioplastic dyeing stage, the fabricated bioplastics were dyed using leftover dye baths from stages (a), (b), and (c)

at a pH close to neutral (~ pH 6) as per Li et al. (2020). Achieving this approach involved the use of two solutions, which adds an additional layer of complexity to obtaining a deeper blue-green hue. Furthermore, the dye bath exhibited relatively faint coloration.

In this context, Experiments 3 to 10 were dedicated to preparing the dye bath using various concentrations of Sp. suspensions in the liquid dye, combined with a single precipitation solution. This approach, which did not neutralize the final solution, resulted in deeper and more intense shades of blue-green pigments. Figure 5 displays the filters from these experiments (#3 to #10) showcasing the different blue-green hues obtained after drying the precipitated Sp. liquid dye solution. The interplay of varying concentrations of Sp. and A. led to a spectrum of blue-green colours, as illustrated in Fig. 6.

Our research emphasized the significant impact of pH on the extraction of pigments and their resulting vibrancy, corroborating findings from Muthu et al. (2020). In our studies, we observed that the pH of the final liquid dye fluctuated between 4 and 5. This range is particularly relevant when considering the behaviour of *Arthrospira platensis*, an organic pigment source. Muthu et al. (2020) noted that this organism exhibits moderate growth at a pH of 5 and only begins to break down when the acidity exceeds a pH of 3 (Muthu et al. 2020). This insight aligns closely with the pH range observed in our experiments, underscoring the importance of maintaining an optimal pH for effective pigment extraction and stability.

From the filtered and precipitated solutions obtained in liquid dye preparation step, we created several dye baths. These were selected based on their colour for application to different groups of commercial fibre samples (refer to Fig. 3, step 2. dye bath). Additionally, one of the leftover dye baths was chosen for dyeing bioplastics (see Fig. 3, step 3: leftover dye).

Fibre Dying

In the second step—dye bath—natural fibres were dyed using the previously collected Sp. dye bath's to optimize hues and colour fixation processes, aiming for subsequent application in alginate bioplastics. These solutions were grouped and compared across different fibre samples,

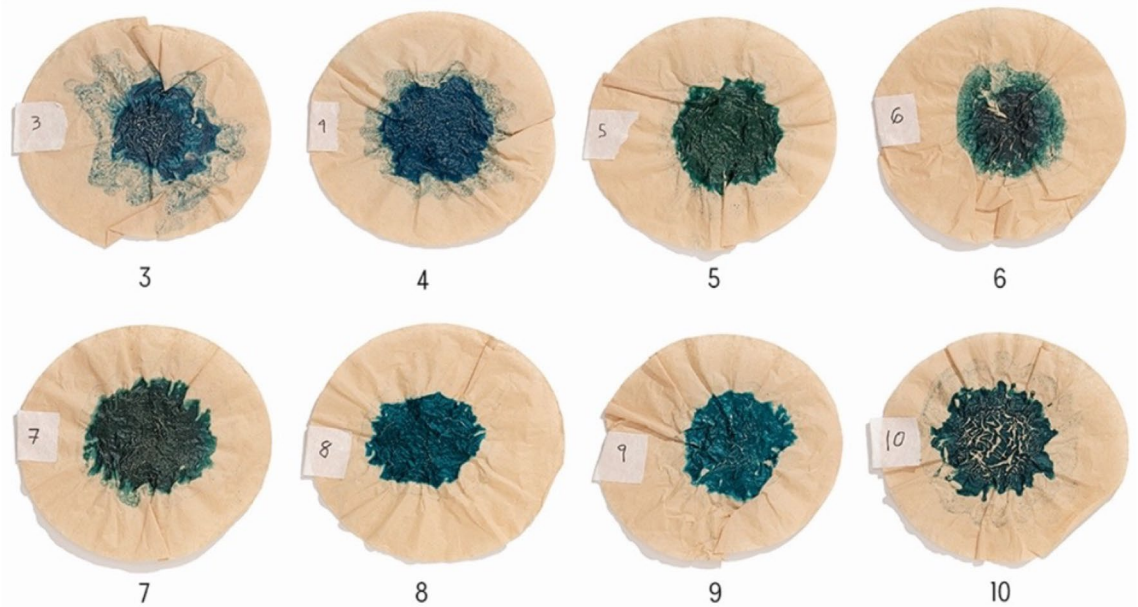

Fig. 5 Liquid dye Preparation filters - filters with pigments extracted from Liquid dye preparations solutions from Experiments 3 to 10 - aqueous suspension with different concentrations of Sp. powder and alum, conducted to a spectrum of blue-green colours liquid dyes—1-month dry, photo by Luís Silva Campos, Jan. 2022

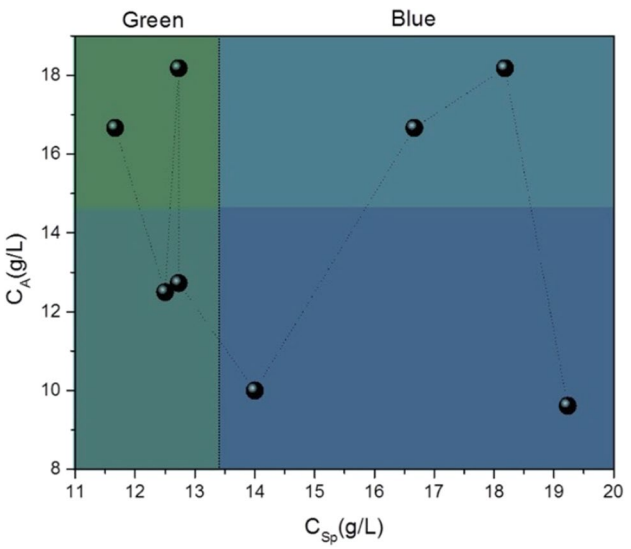

Fig. 6 Graphical Plot of Concentration of alum (C_A) as a function of Concentration of Spirulina (C_{sp}) related to experiments Liquid Dye Preparations Solutions from Experiment 3 to 10 - Sp. and A. and colours (after 24 h of rest)

including cotton fabric, re-use cotton yarn, seaweed yarn and viscose threads. One set of commercial fibres (collection A) was boiled in water with alum to mordant the fibres, while another set (collection B) served as a control group without mordanting. From the resulting dyed fibres, a blue-green hue was selected for application in a third fibre collection (C). To achieve the desired colour range of Sp., from green to blue, equal samples of fibres, both mordanted and un-mordanted, were dyed. This included using only the liquid dye without precipitation for a green hue, a selected dye bath from collection A (resulted from Experiment 8), and an Sp. phycocyanin pigment (Table 4—fibre dye C).

A comparative analysis between collections A and B (illustrated in Fig. 7A and B) showed that all fibres absorbed more dye when pre-mordanted, as expected. However, the absorption varied among materials; light cotton fabric absorbed less colour compared to reused cotton thread, which absorbed more pigment, indicating that fibre weight significantly influences the dyeing process. The final collection (fibre dye C), predominantly consisting of pre-mordanted fibres (with the exception of Experiment 13b), exhibited the most suitable hues for application in bioplastics, successfully achieving a zero waste flow, as depicted in Fig. 7C.

Fibres were photographed again after 1 month and then stored away from direct light. After 10 months, they were observed and showed no change in colour. Figure 8 displays the fibre collections A, B, and C after 1-month drying. Experiment 2.A, which involved pre-mordanting, resulted in soft hues. Experiment 2.B, conducted without mordant, produced lighter shades. Finally, Experiment 2.C, which used various Sp. sources and involved pre-mordanting with alum, resulted in deeper colour adsorption. Furthermore, after 10 months, the fibres exhibited increased fragility, showing signs of breaking and tearing at various points.

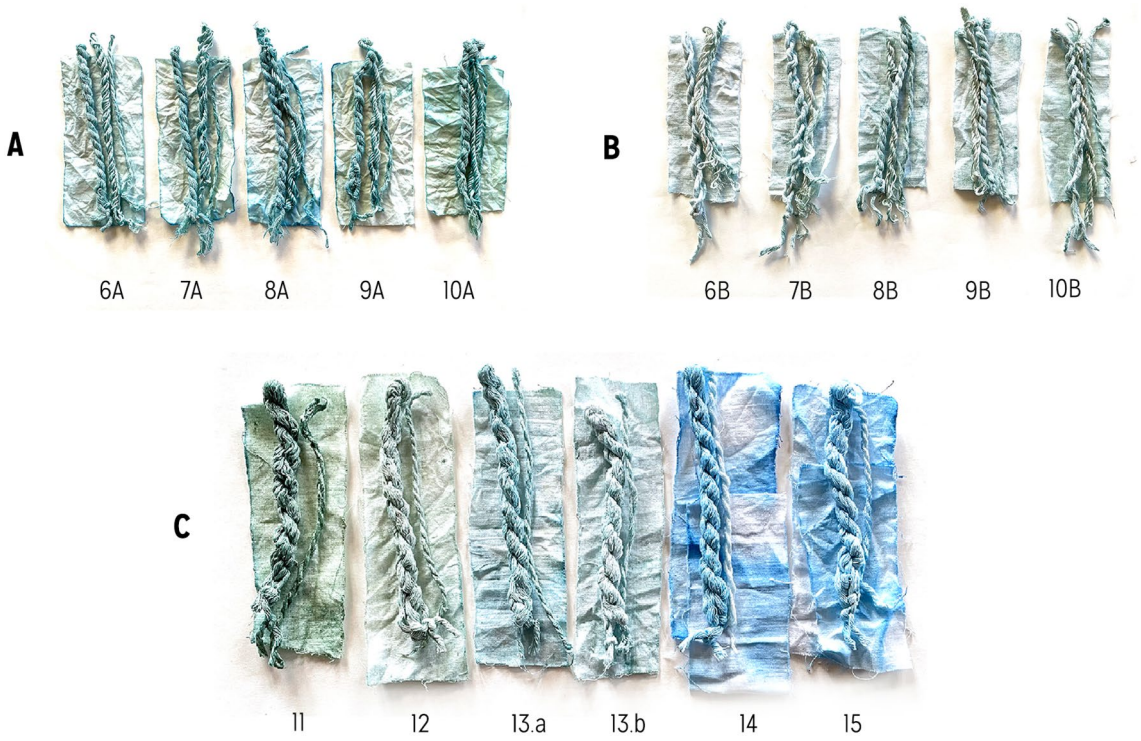

Fig. 7 Fibres as-prepared after dried: **A** fibre dye A collection of fibre dyeing Sp. dye bath with mordant bath; **B** fibre dye B collection of fibres, dyeing Sp. dye bath without mordant bath; and **C** fibre dye collection of fibres, different Sp. sources (Sp. powder, Sp. suspension filtered (liquid dye), and commercial phycocyanin pigment) onto natural fibres with mordant bath; Dec. 2021

The experiments from sample C allowed a selection of best conditions to be applied in alginate bioplastics—bio-yarn (Table 3) and bio-films (Table 4).

Bioplastic Dyeing

The optimized dye bath solutions from the previously fibre collection C (2.C.) were subsequently applied in the alginate bioplastic studies. In this third step, three distinct Sp.-coloured alginate bioplastics were fabricated. The first was coloured with liquid dye (after the first filtration of the aqueous Sp. suspension) without precipitation, aiming for a green hue (see Fig. 8C (exp. 11) and Table 2 (exp. 11)). The second utilized selected blue-green leftover bath from the previous step of fibre dyeing (see Fig. 8C (exp. 13a) and Table 2 (exp. 13a)). The final bioplastic was created with the Pp. pigment (see Fig. 8C (exp. 14) and Table 2 (exp. 14)).

Figure 9 shows alginate-based bioplastics—bio-yarns and bio-films dyed with the three distinct strategies, 1 week after, prior to complete dehydration. We can observe that the green to blue spectrum was successfully achieved in bioplastics, with intense colours. Samples 16A (yarn) and 16B (film) display the greenest colour, obtained using the first filtration bath (liquid dye) without any precipitation process. Samples 17A and 17B exhibit an intermediate green–blue colour, resulting from the optimization process following the dyeing of fibres. Finally, samples 18A and 18B showcase a deep blue hue, achieved through the direct use of commercial Pp. pigments.

The bioplastic materials were compared and analysed to study the aging process, with a focus on colour stability. During extrusion and subsequent drying, the colour appeared stable (Fig. 9). However after 1-moth, the hues remained similar but softened in shade (Fig. 10). Following a 10-month drying period, the extruded bio-yarns exhibited significant colour loss, indicative of material degradation (likely due to oxidation, humidity, and light exposure during storage, despite being kept away from direct light).

Comparatively, bio-yarns demonstrated a slower colour loss than bio-films, particularly noticeable in the leftover 13 (samples 17A and 17B in Figs. 10 and 11). This observation suggests a potential impact of alum residues in the dye bath on colour retention. Furthermore, regarding manipulation, such as knitting, the bio-yarns were easily handled immediately post-curation (approximately 1 week). It was observed that the yarns maintained their integrity, allowing for easy manipulation and knitting without difficulty (Fig. 12). However, this ease diminished

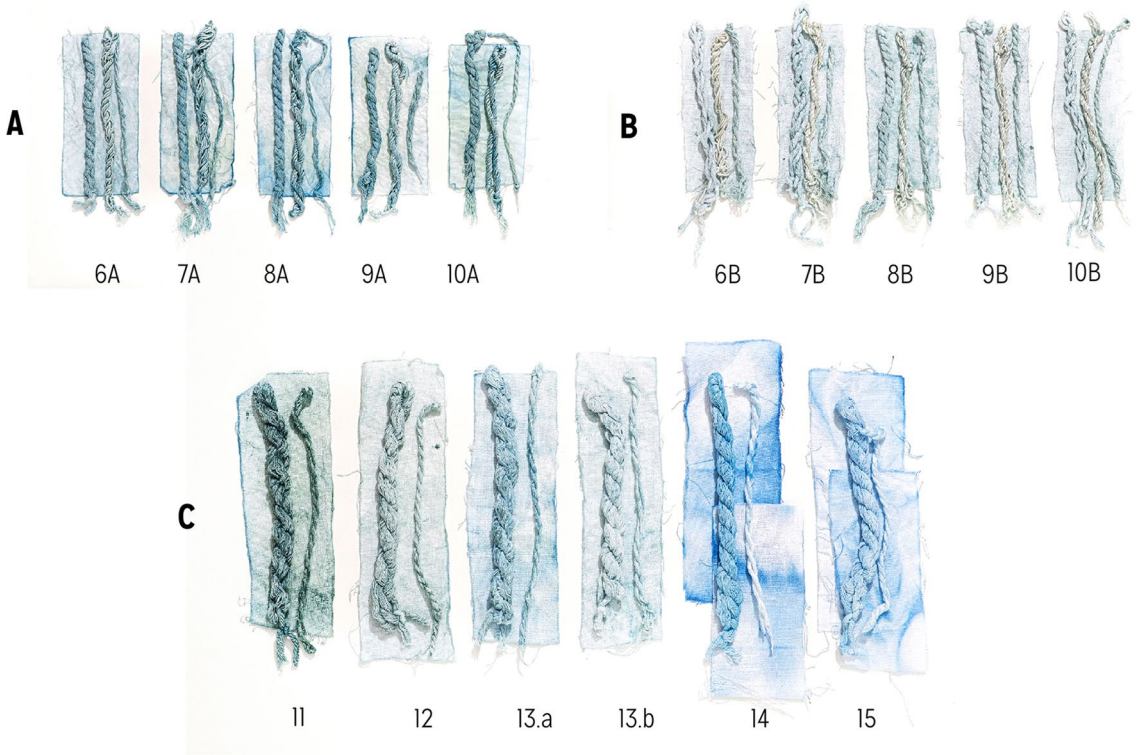

Fig. 8 Fibres after 1-month dried: **A** fibre dye A collection of fibre dyeing Sp. dye bath with mordant bath (Table 2); **B** fibre dye B collection of fibre, dyeing Sp. dye bath without mordant bath; and **C** fibre dye C collection of fibres, different Sp. sources (Sp. powder, Sp. suspension filtered (liquid dye), and commercial phycocyanin pigment) onto natural fibres with mordant bath (except 13b); photos by Luís Silva Campos, Jan. 2022

over the following months, likely due to increased susceptibility to tearing and breaking.

After 10 months, the alginate-based bio-yarns and bio-films underwent significant colour transformation. The hues became desaturated, shifting to yellow-orange shades (Figs. 11 and 12). Regarding the bio-film susceptibility for breaking, they exhibited fragility and accelerated biodegradation, which could likely be attributed to the reaction of organic compounds, such as alginate and Sp. (McCarty and Kerna 2021; Muthu et al. 2020), with glycerine grease and moisture.

Observational evidence from our study suggests that bio-yarns exhibited a longer colour retention compared to bio-films. This phenomenon indicates the possible role of alum residues in enhancing and prolonging the colour fidelity in bio-yarns. Additionally, it was noted that the bio-films, having a greater surface area exposed to air, were more prone to degradation and oxidation. This increased exposure likely accelerates the deterioration process, leading to a faster decline in both the colour and structural integrity of the bio-films.

The bio-yarns were knitted as an exploratory exercise and re-photographed. After 10 months, the fibres also presented fragility and breaking/tear points. See Fig. 12.

Conclusions

This research promotes the use of zero waste natural dyes, particularly Sp.-based greens and blues, for dyeing natural plant fibres and alginate bioplastics. This investigation successfully demonstrated that phycocyanin pigment extraction, typically conducted in industrial settings, can be effectively replicated in a Fab Lab environment using do-it-yourself methods.

Our research presents a ground-breaking method for the extraction of Spirulina (Sp.) pigments and their subsequent incorporation into alginate bioplastics, marking a notable improvement over conventional technique. Our process involves a comprehensive three-step approach: preparation of liquid dye, dyeing of fibres, and dyeing of bioplastics. This method encapsulates the zero waste model that is proposed for the case of extraction of Spirulina pigments, which effectively utilizes 3 stages—from liquid dye to dye bath and even the residual dye bath leftovers.

By optimizing the conditions in the preparation phase of liquid dye, the blue-green colours from Spirulina (Sp.) powder were expertly extracted to enhance and stabilize colour vibrancy. A spectrum of shades was produced, where the depth of colour directly correlated with the

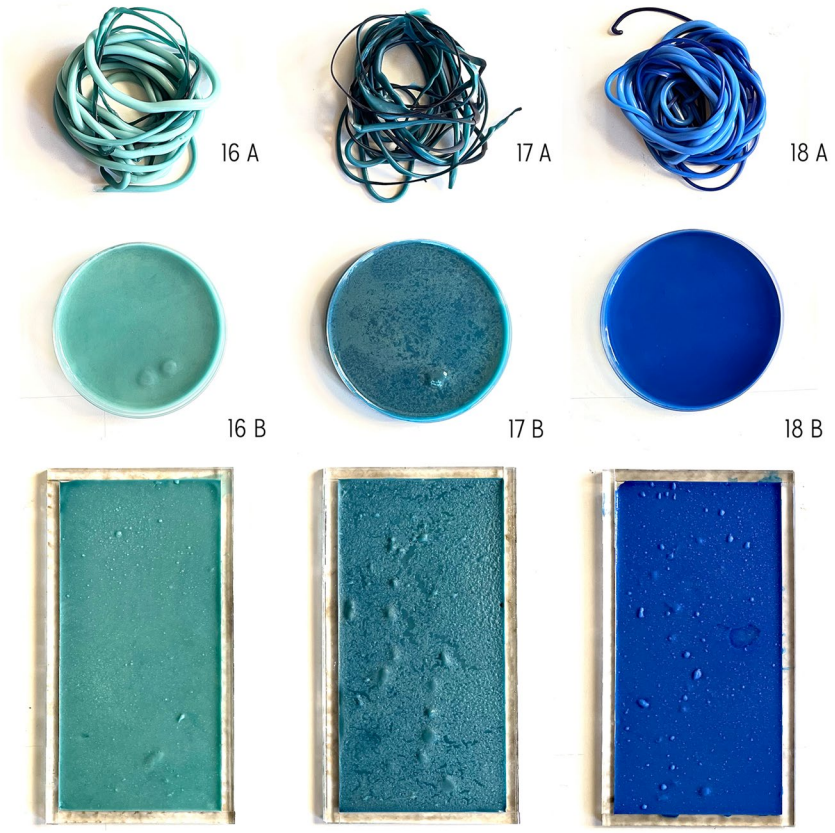

Fig. 9 Alginate-based bioplastics prior to complete dehydration: Sp. bio-yarns A and B bio-films in different geometries for samples 16A/16B, 17A/17B, and 18A/18B of Table 3 and 4, based on dye bath obtained by liquid dye first filtration, selected blue-green leftover bath from fibre dyeing, and Pp. pigment dye solution respectively; Dec. 2021

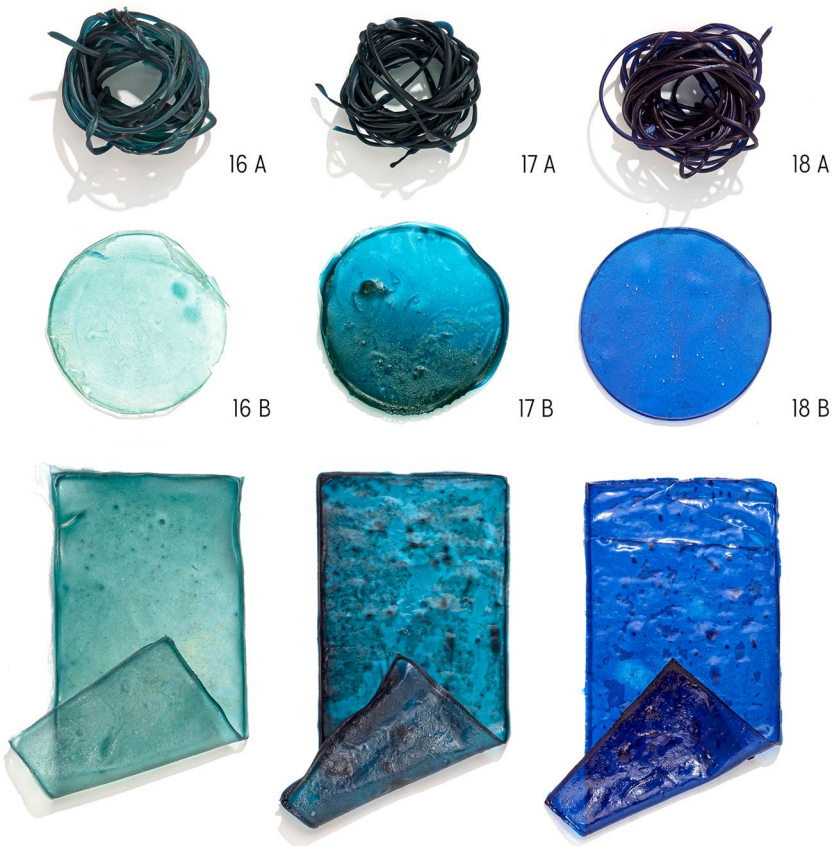

Fig. 10 Alginate-based fibres 1-month dry: Sp. bio-yarns A and B bio-films in different geometries for samples 16A/16B, 17A/17B, and 18A/18B of Table 3 and 4, based on dye bath obtained by liquid dye first filtration, selected blue-green leftover bath from fibre dyeing, and Pp. pigment dye solution respectively; photo by Luís Silva Campos, Jan. 2021

Fig. 11 Alginate-based fibres 10-month dry: Sp. bio-yarns A and B bio-films in different geometries for samples 16A/16B, 17A/17B, and 18A/18B of Table 3 and 4, based on dye bath obtained by liquid dye first filtration, selected blue-green leftover bath from fibre dyeing, and Pp. pigment dye solution respectively; photo by Luís Silva Campos, Oct. 2021

concentration of Spirulina and the specific precipitation method employed. Notably, various liquid dyes were obtained through different methods, including routes without any precipitation, and others involving precipitation, either with a combination of acidic and alkaline solutions or solely with an acidic solution. Among the various methods tested, the route involving acidic precipitation proved to be the most effective. The experiments that where conducted emphasized the crucial importance of pH in the extraction of pigments and the resulting intensity of their colours. Additionally, the same studies revealed a significant interaction between the concentrations of the liquid dye derived from Sp. powder and the acidic solution used for precipitation. This interplay was instrumental in producing a diverse spectrum of green–blue hues, highlighting the intricate balance between Sp. dye concentration and the acidic environment in determining the final colour range.

In fibre dyeing, the application of Sp. dye baths, developed via one precipitation solution, was applied to various natural fibres with and without mordant (alum) pre-treatment. It demonstrated significant colour absorption, especially in pre-mordanted fibres. Using alum as a mordant enhances colour stability and intensity. The experiment with Spirulina and alum concentration ratio 1:1, with final pH = 4, resulted in the stronger blue hue.

We observed differential colour uptake across fibre types, with a notable correlation between fibre weight and dye absorption.

In the culmination of our study, our dyeing bath techniques were successfully applied to alginate bioplastics, bio-yarn, and bio-films, achieving a range of hues from green to blue. To attain this diverse colour palette, we employed various strategies: (i) the liquid dye extracted from the initial filtration was used to achieve a green hue, (ii) the optimized results obtained from the previous stage of dyeing fibres were applied to obtain the green–blue colour, and (iii) an intense blue colour was achieved by incorporating phycocyanin (Pp) pigment solutions.

These methods collectively enabled us to explore and realize the full potential of the green–blue spectrum in our bioplastic materials.

During this bioplastic dyeing phase, it was observed that bio-yarns retained their colour longer than bio-films, hinting at the potential influence of alum residues in prolonging colour fidelity. However, over time, both bio-yarns and bio-films underwent colour desaturation and experienced physical degradation. Based on the observations, it seems that bio-yarns retained their colour longer than bio-films, hinting at the potential influence of alum residues in prolonging colour fidelity. These observations underscore the challenges faced in maintaining long-term colour stability and the structural integrity of the materials.

The study's zero waste approach, as depicted in Figs. 3 and 4, effectively utilized leftovers from each step, minimizing waste while exploring the potential of natural dyes in sustainable material applications. However, challenges

Fig. 12 Alginate-based Bio-yarns knitted: Sp. Bio-yarns from Experiments16A/17A/18A of Table 3, based on dye bath obtained by liquid dye first filtration, selected blue-green leftover bath from fibre dyeing, and Pp. pigment dye solution respectively; knitted after 10 months, photo by Luís Silva Campos, Oct. 2021

remain in ensuring long-term colour stability and physical durability in alginate bioplastics, particularly in the face of environmental factors like oxidation, humidity, and light exposure.

This research confirmed that Spirulina (Sp.) pigments are sensitive to temperature, pH, and light due to their organic composition. These findings align with cyanobacteria's natural process of energy creation through photosensitization and CO_2 capture from the atmosphere. The study underscores the value of Sp. as a practical learning tool and a sustainable raw material in design.

Overall, our findings contribute valuable insights into the sustainable use of natural dyes in bioplastics, offering a promising direction for future research in environmentally friendly material science. The study underscores the importance of continual optimization and adaptation of methodologies to balance aesthetic qualities with functional and environmental considerations.

Future Recommendations

The Spirulina-extracted pigments could be tested as ink preparations to observe the colour behaviours demonstrated in the paper. More experiments are needed on the Spirulina-dyed commercial fibres and more testing to control exposure light. Observing and evaluating colour fastness would be relevant; exposing half of the samples and covering the other half with light-protecting material could highlight colour behaviour and stability on Spirulina-dyed natural fibres. On the fabricated fibres—Sp. suspensions and phycocyanin pigment suspension with alginate bioplastic and bio-yarn—more tests with different pH solutions after dye (e.g. alkaline: vinegar, soda) are needed. Furthermore, adding antioxidation agents (e.g. sodium ascorbate), considering LinaBlue® technical recommendations (DIC Corporation 1985), could improve colour stability in alginate bio-based materials.

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Author Contribution CD performed theoretical and practical research, writing—original draft preparation and pictures.

GF performed conceptualization, validation and analysed data, writing—review, and supervision, and funding acquisition.

AA performed conceptualizzation, validation and analysed data, writing—review, supervision, and funding acquisition.

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Data Availability Main data generated or analysed during this study are included in this published article. Supporting data, Tables A1, A2, A3, and A4 and Figs. 13 and 14, are available for consultation in Supplementary information (SI).

Declarations

Competing Interests The authors declare no competing interests.

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