textiles

Article

Algae-Based Coatings for Fully Bio-Based and Colored Textile Products

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Abstract: Utilizing natural fibers for production of a fully bio-based textile is an appropriate approach to align with sustainability objectives. In case of dyed products, the used dye and further finishing agents also have to be bio-based. Algae-based materials are naturally colored and can be used as a natural dye product. The present study aims to investigate the use of algae materials as a color pigment for coating on cotton as a natural fiber material using a bio-based binder. Three different algae representing blue-green algae (BGA), unicellular microalgae (UNI), and filamentous algae (FIL) are investigated as colored additives for textile coatings. The algae are applied as powder to the coating recipe together with sodium alginate as binder component. Modification of the application is conducted by addition of calcium dichloride ($CaCl_2$) acting as a crosslinker for the natural binder system. Using these applications, strong coloration of cotton fabrics can be achieved. Scanning electron microscopy (SEM), infrared spectroscopy (FT-IR), color measurements, light fastness tests, and tests on the rubbing fastness are performed. Good values in rubbing and light fastness are reached (4–5 and 5, respectively) depending on the used algae material and the addition of calcium dichloride. Applications using the algae BGA and UNI perform better due to light fastness. In contrast, applications with the filamentous algae FIL lead to better rubbing fastness. The present study confirms that the proposed technique and formulations are appropriate for achieving fully bio-based, naturally colored textile products, offering a promising foundation for further research into and development of the use of algae for the modification and functionalization of textile materials.

Keywords: algae; textile coating; print; dyeing; light fastness; calcium dichloride; sodium alginate

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Academic Editor: Andrea Zille

Received: 9 October 2024
Revised: 27 December 2024
Accepted: 7 January 2025
Published: 8 January 2025

Citation: Blanckart, L.; Munasinghe, E.A.; Bendt, E.; Rahaman, A.; Abomohra, A.; Mahltig, B. Algae-Based Coatings for Fully Bio-Based and Colored Textile Products. *Textiles* **2025**, *5*, 3. https://doi.org/10.3390/textiles5010003

1. Introduction

For coloration of textiles and clothing, synthetic dyes are used mostly in industrial processes [1,2]. Prominent examples are the application of reactive dyes on cotton or disperse dyes on polyester fabrics [1,3–6]. The application of these synthetic dyes often leads to superior fastness properties of the dyed textiles. Synthetic metal complex dyes, such as copper phthalocyanine, are the dyes having the highest light fastness [7,8]. In addition to providing improved fastness properties, synthetic dyes are generally quite cost-effective [9]. However, their use is not viable for producing fully bio-based textile products, where natural alternatives are required for a sustainable and eco-friendly approach. In addition, microplastic particles have become a significant environmental concern due to their widespread presence in aquatic and terrestrial ecosystems [10,11]. Microalgae have been investigated for their capability to remove microplastic and even nanoplastic particles [12]. Due to growing concerns over the release of microplastic emissions from textile products during usage and washing, an increasing demand for textiles made from natural fibers can be expected for the near future [13–15]. Natural fibers like cotton decompose after release to nature and are therefore play no part in the microplastic problem [16,17]. Nevertheless, to realize a fully bio-based textile product, a holistic approach has to be used that includes the use of natural dyes and finishing agents in addition to natural fiber materials. A wide variety of natural colorants can be extracted from algae, covering a spectrum of colors. For instance, green pigments can be obtained from chlorophyll, blue from phycocyanin, red from phycoerythrin, yellow from beta-carotene, and reddish from astaxanthin [18]. In particular, microalgae-derived pigments (phycobiliproteins, chlorophylls, carotenoids) are highlighted as sustainable alternatives to synthetic dyes [18].

One of the major advantages of microalgae is their high productivity, owing to rapid growth rates and substantial pigment yields [19]. In addition, their production is sustainable, utilizing non-arable land, minimal freshwater, and possible integration with wastewater treatment. Many of those microalgae-based pigments are commercially available year-round, and not dependent on seasons. They can provide good color fastness and vibrant shades on textiles. Recent research is focused on improving extraction methods, enhancing pigment stability, and optimizing mordanting techniques to achieve better color fastness on fabrics [18]. An example for commercially available microalgae-based pigment is the product Spirulina Green Pigment offered by the German company Kremer Pigmente (Aichstetten) [20]. This company also offers an algae-based pigment named Spirulina Blue, which is based on a phycocyanin extract [21]. However, this supplying company describes both pigments as being sensitive against illumination [20,21]. The above-mentioned advances can help to establish microalgae-derived pigments as strong competitors to synthetic dyes in the textile industry. A recent study evaluated the dyeing potential of marine algae species, with a focus on optimized extraction conditions to improve pigment yield [22]. The study considered four marine algae species: *Iyengaria stellata*, *Sargassum muticum*, *Colpomenia sinuosa*, and *Laurencia obtuse.* Extraction is performed using mixtures of aqueous potassium hydroxide and acetone. Iron(II)sulfate and tannic acid are used as mordants, enhancing the color intensity. Results showed that algal pigments can produce vibrant colors with good fastness properties, positioning them as a viable alternative to synthetic dyes [22]. A broad examination of the use of bio-based dyes, including those derived from algae, for realization of sustainable textile dyeing was reviewed by Belino et al. [23]. Here, extraction methods, dyeing processes, and environmental impacts of using algal pigments are reviewed and discussed. This review considers a broad range of different bio-based dyes and refers also to different macroalgae including green algae (*Chlorophyta*), red algae (*Rhodophyta*), and brown algae (*Phaeophyceae*), which are known for their rich pigment content, such as chlorophylls, carotenoids, and phycobiliproteins [23]. Bio-based paints are developed also by use of microalgal pigments, such as phycocyanin, astaxanthin, and chlorophyll [24]. In this recent study, extraction, characterization, and application of these pigments were performed, emphasizing their environmental sustainability and commercial viability. The pigments were extracted from different microalgal species using solvent extraction methods [24]. The extracted pigments were incorporated into paint formulations and tested for their color properties, durability, and environmental impact. The described paints showed good adhesion and resistance to environmental factors such as UV radiation and moisture. Additional to their application as a colorant, further benefits are mentioned for different algae-based color components. For example, Benbelkhir et al. reviewed the potential of microalgal carotenoids as natural alternatives to synthetic dyes [25]. This study highlighted the environmental and health benefits of using natural pigments derived from microalgae, such as carotenoids, which are known for their antioxidant, anti-inflammatory, and antimicrobial properties. It emphasized the need for improvements in microalgae cultivation and extraction systems to make the carotenoid production more economical, reproducible, and eco-friendly for large-scale applications [25].

Although previous studies have extensively evaluated pigments extracted from algal biomass for textile dyeing, the use of whole algae for textile coloration remains underexplored. Evaluating whole algae as a natural colorant could open up innovative possibilities in sustainable textile manufacturing, as this approach might enhance the bio-based content of the final product while reducing processing steps involved in pigment extraction. One investigation in this field reports on the use of powder from *Spirulina platensis* for application onto textiles by using a recipe from dispersed algae powder into water, alginate, and the organic solvent glycerin [26]. However, the use of organic solvents is often not likely in industrial textile applications.

It should be clear that the use of whole algae material for coloration applications in comparison to the use of algae extracts exhibits advantages and, of course, also disadvantages. One main advantage of using the complete algae as a color pigment is clearly that no resources like energy or solvents are necessary for any extraction process. Possible disadvantages are that the whole algae pigment can contain by-components influencing the color and application process. Also, some by-components may contain possible health risks [27,28]. However, possible health risks should be avoided by selection of the algae material used for coloration applications. The potential and accidental release of algae material into a non-native environment might be considered as well [29,30]. For this, it should be ensured that only non-living algae material is applied. However, even after mentioning several disadvantages, investigating whole algae as a natural colorant could unlock innovative opportunities for sustainable textile manufacturing by increasing the bio-based content of products and simplifying processing

With this background, the present study focuses on the use of complete grinded algae in fully water-based recipes for coloration of textile fabrics from cotton. The grinded algae material is applied together with a binder system onto the textile. This study evaluates three algal species representing blue-green algae, unicellular microalgae, and filamentous algae. For this purpose, recipes with bio-based binders and crosslinkers are created. In the present study, the selected algae materials are obtained in powder form and applied as a coating additive with a binder. The filamentous algae material is further treated by the grinding process. Sodium alginate made out of seaweed algae is used as the binder in this experiment. The effect of the binder on UV exposure was analyzed first by applying only binder and binder with calcium dichloride ($CaCl_2$) as a crosslinker onto the selected fabric as follows. Calcium ions (Ca^{2+}) are divalent ions and can act as a type of crosslinker for alginate molecules [31]. This is due to the fact that the carboxyl groups (-COOH) in alginate have a strong affinity for calcium ions. When Ca^{2+} is introduced into the system, these ions bind to the carboxyl groups of adjacent alginate molecules, creating stable crosslinking. The crosslinking by Ca^{2+} ions ensures that the alginate molecules are better interconnected, leading to increased structural stability of the resulting coating [31]. For this reason, in the current investigation the recipes are also tested in terms of their performance after exposure to an aqueous solution of calcium dichloride ($CaCl_2$).

Finally, the presented technique and recipes are suitable for realizing fully bio-based colored textile products based on grinded algae material as the color component. This study has also the potential to be a starting point for future developments of algae applications for modification and functionalization of textile materials for a broad range of different applications. Nevertheless, it should be considered that several algae are used as nutrients and for food production [32–35]. For this, a certain competition in their use as nutrients versus color agents can be estimated and has to be considered in a complete view of the environmental impact. However, algae cultivation for textile applications could utilize safe wastewater sources, such as aquarium or textile wastewater, reducing the demand for food-grade resources and promoting circular economy principles [36,37].

2. Materials and Methods

2.1. Materials

For the current recipe development, three different algae groups were evaluated, namely blue-green algae (BGA, *Spirulina* sp.), unicellular microalgae (UNI, *Chlorella* sp.), and filamentous algae (FIL, identified morphologically as *Rhizoclonium* sp.). Two BGA biomass were obtained, where BGA1 and BGA2 were obtained from the company Borchers fine food GmbH (Oyten, Germany) and the company Narayana Verlag GmbH (Kandern, Germany), respectively. In addition, UNI biomass was provided by Narayana Verlag GmbH (Kandern, Germany). However, FIL biomass was gained from cultivation of algal species in WHM medium for an average of 14 days to obtain the biomass [38]. The Woods Hole Medium (WHM) is a synthetic nutrient solution containing a standardized mixture of essential macronutrients (such as nitrogen, phosphorus, and potassium) and micronutrients (including iron, zinc, copper, and manganese), supplemented with chelators and trace elements to support the growth of freshwater microalgae in laboratory and pilot studies. This WHM medium is based on H.W. Nichols (1973) [38]. After air-drying, the biomass was ground for five minutes using a RETSCH Planetary Mill PM100 with six steel balls (20 mm diameter). As a reference material, “diatomeen” was used. This material is supplied as calcinated material from diatom algae and is provided as a white powder from the company Felix Teske (München, Germany). This material is also named diatomaceous earth. Sodium alginate (Sera Print M-ALV 10, Dystar, Singapore) was used as a native low viscosity thickener for print pastes and dyeing liquors. The calcium dichloride ($CaCl_2$) used was supplied by the company Merck (Darmstadt, Germany). As textile materials, plain-woven cotton fabric with weight per area of 190 g/m^2 was used. No further pre-treatment was performed on the cotton fabric before coating application. As a further coating substrate, a polypropylene foil (PP foil) was used. This is an unstructured and transparent polypropylene foil (PP foil) acquired from the retailer Action Deutschland GmbH (Düsseldorf, Germany).

2.2. Sample Preparation

For preparation of the alginate-based binder recipe, an amount of 15 g sodium alginate was stirred in 285 g water for ten minutes. After stirring, the binder recipe was stored for 24 h in a refrigerator (Eurodal KS 120E) at 3 °C. For preparation of the final coating recipe, the alginate binder recipe was mixed with the algae powder in a ratio of 1:3. The viscosity of the alginate binder recipe was 0.739 mPa s. Prepared coating recipes were applied on two selected substrates—cotton woven fabric and polypropylene foil. Coating on cotton fabric was performed by a magnetic coating machine, type MDFR242, from Johannes Zimmer (Klagenfurt, Austria). This coating machine is a table that is usable for screen printing and coating applications, which allows a metal roll to be driven in a controlled manner over a textile fabric. In the current preparation, this metal roll was driven six times on the fabric to deliver the best regular distribution and penetration of the printing paste onto the cotton fabric. One set of coated fabrics from each algae product was treated with aqueous solution of $CaCl_2$ (2 wt-%) by manual spraying. Coating on polypropylene foil was conducted manually. For applications on cotton, the samples were oven-dried at 120 °C for three minutes. Applications on polypropylene were air-dried at room temperature. Each coated cotton sample was separated into two parts, where one set was kept in a place without sunlight exposure, while the other set was kept at the window under sunlight (summer 2024 in Germany) for 30 days.

2.3. Analytical Instruments and Methods

The data color values L*, C*, and h for coated cotton samples were determined by a Data Color Spectrophotometer (Datacolor 400) (Datacolor GmbH, Marl, Germany). The infrared spectra (IR spectra) of the prepared textile samples were recorded using a FT-IR spectrometer IRTracer-100 from Shimadzu (Kyoto, Japan) equipped with a Specac Golden Gate ATR unit. The topography of the samples was examined by scanning electron microscopy (SEM) using a TM4000Plus Tabletop microscope (Hitachi, Tokio, Japan). Magnified photographs under illumination with visible light were taken using a photomicroscope Dino-Lite Premier Digital Microscope (ANMO Electronics Corporation, Taipei, Taiwan). Viscosity measurements were conducted with a HAAKE Viscotester iQ Rheometer supplied by Thermo Scientific (Waltham, MA, USA). For rubbing and light fastness evaluation, a James H Heal crockmeter (Halifax, UK) and an Atlas Xenotest Alpha LM instrument (Linsengericht-Altenhaßlau, Germany) were used. The rubbing fastness was determined according to ISO 105-X12 on a James H Heal crockmeter . The greyscale ISO 105-A03 was used to rate the rubbing fastness (rating range from 1–5/worst to best). The light fastness was determined according to ISO 105-A02. The duration of the illumination of samples in the Xenotest was set to 50 h. The light fastness was rated from 1 = worst to 8 = best by comparison of samples against blue standard reference samples.

3. Results and Discussion

In this section, the prepared coated samples are presented and their properties are discussed. The general properties of the samples related to IR spectroscopic and electron microscopic investigations are addressed. In addition, the realized coloration properties, which are the aimed-for properties of the realized coatings, and the fastness properties of realized coloration, are evaluated.

3.1. Spectroscopic and Microscopic Evaluation

To support general spectroscopic information on the realized coating and modified textile fabrics, infrared spectroscopic investigations were performed. IR spectroscopy is a suitable tool for investigating and determining fibers, textile products, and textile-containing products such as artificial leather [39,40]. In the first reference measurements, for the untreated cotton fabric, an IR spectrum was recorded and compared with the IR spectrum of the cotton fabric after alginate application without the addition of further algae powder (Figure 1). Results show that the IR spectrum of the cotton fabric is quite similar to that of cotton reported in the literature [41]. The applied alginate does not change the IR spectrum of the cotton fabric, which can be attributed to two reasons. The first is the low amount of applied alginate material, and the second is the structural similarity of alginate with the carbohydrate basic structure and cellulose as the main carbohydrate building units of cotton fibers.

IR spectra recorded for cotton fabrics coated with different algae powder are presented in Figures 2 and 3 for the fingerprint area from 1200 cm^{-1} to 1800 cm^{-1}. In Figure 2, the IR spectra of textiles samples with coatings containing algae types UNI and BGA are presented and compared to those of uncoated cotton fabric. For comparison, the IR spectra of the pure algae powder materials are also presented. The IR spectra of the algae powders are quite similar to spectra reported in the literature [42–44]. They exhibit two prominent signals around 1640 cm^{-1} and 1540 cm^{-1}, which are related to the protein content of these algae. Protein contains multiple amide groups. The signal around 1640 cm^{-1} is assigned to the stretching vibration of the C=O bond of the amide group (Amide I vibration), and the signal around 1540 cm^{-1} is assigned to an in-plane and stretching vibration of N-H and C-N bonds of the amide group (Amide II vibration) [42].

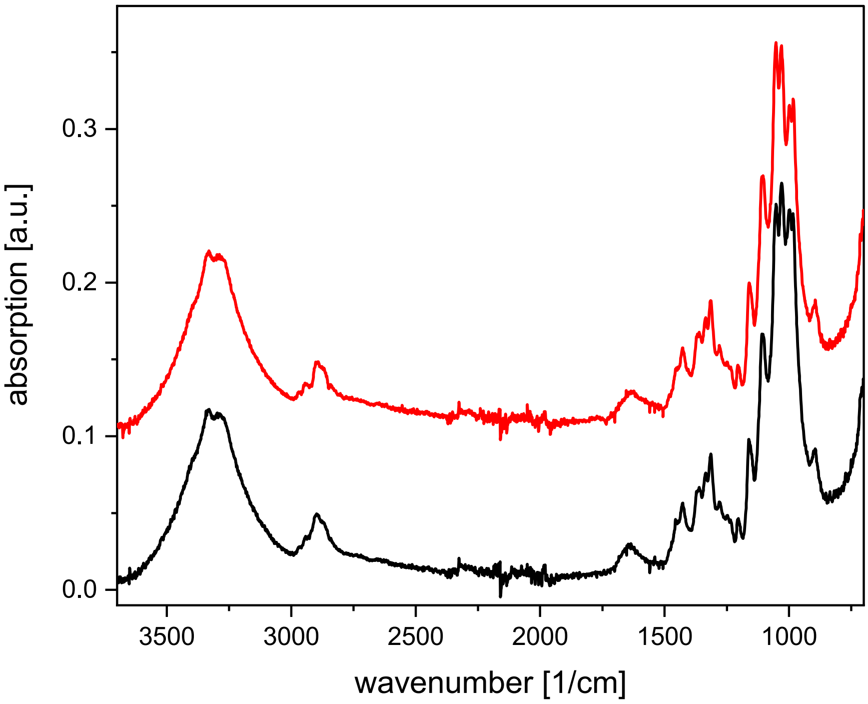


Figure 1. IR spectra of untreated cotton fabric (black) and cotton fabric after alginate application (red).

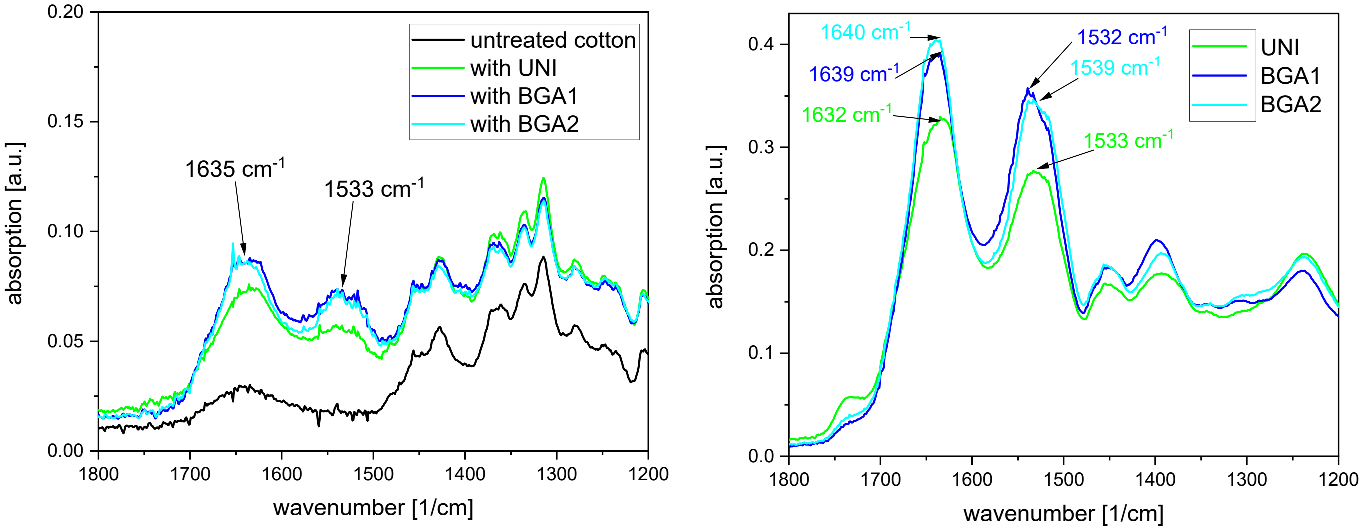


Figure 2. Comparison of textile samples and related algae powder material—left image: IR spectra of untreated cotton fabrics and those treated with recipes containing unicellular microalga (UNI, and 2 types of blue-green algae biomass (BGA); right image: IR spectra of algae powder materials.

In comparison to the IR spectrum recorded from the uncoated cotton fabric, the samples with the algae containing coatings exhibit two additional signals around 1635 cm^{-1} and 1533 cm^{-1}. These signals can be assigned to vibrations from amide groups, associated with the protein content of algal biomass [42,45–47]. Thus, the presence of the algae powders in the coatings can be detected by their content of protein, leading to IR active vibrations.

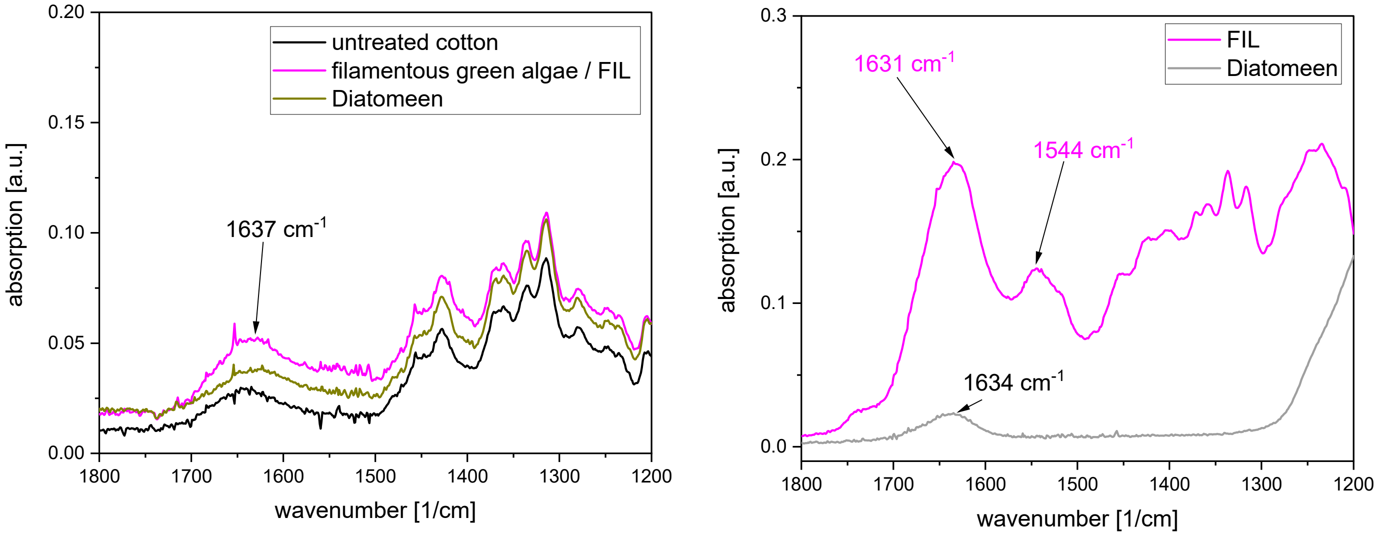


Figure 3. Comparison of textile samples and related powder material—left image: IR spectra of untreated cotton fabrics and those treated with recipes containing filamentous green algae (FIL) and white diatomeen powder; right image: IR spectra of related powder materials.

In Figure 3, the IR spectra of textiles samples with coatings containing algae type FIL and diatomeen powder are presented and compared to those of uncoated cotton fabric. For comparison, the IR spectra of the pure powder materials are also shown. For the diatomeen powder, only a weak signal at 1634 cm^{-1} is determined. This inorganic material does not have prominent signals in the IR spectral range from 1200 cm^{-1} to 1800 cm^{-1}, as is also stated in the literature for diatomaceous earth [48]. The weak signal at 1634 cm^{-1} is suggested to be caused by water molecules embedded into the inorganic structure [48]. The IR spectrum recorded from the powder material FIL exhibits two prominent signals in the IR spectral range from 1200 cm^{-1} to 1800 cm^{-1}, which are related to the protein content of this algae. The first medium signal at 1631 cm^{-1} can be assigned to C=O stretching vibrations from amide groups. The second weaker signal is recorded at 1544 cm^{-1}, and can be assigned to an in-plane and stretching vibration of N-H and C-N bonds of amide groups [42,49]. The presence of the filamentous alga FIL material on coated cotton fabric shows a single new signal with medium intensity at around 1637 cm^{-1} (Figure 3). This signal can be assigned to vibrations from amide groups, which are part of proteins as well. However, compared to UNI and BGA samples, the FIL sample exhibits a signal of lower intensity around 1637 cm^{-1} and no second signal at 1533 cm^{-1}. These differences can be explained by a lower proportion of the applied algae FIL, which would lead to a lower IR signal. In addition, FIL might contain functional groups similar to those of cotton fibers, with lower protein content compared to UNI and BGA, which requires further validation. The cotton sample with the coating containing diatomeen powder exhibits a similar IR spectrum as that of the untreated cotton fabric, which can be attributed to the inorganic material diatomeen.

SEM images of uncoated cotton fabric and the cotton fabric after simple application of the alginate are presented in Figure 4. The textile fabric structure and the typical fiber shape for cotton are clearly visible. There is almost no difference in the topography after alginate treatment. This finding suggests that the applied alginate forms a thin layer, which cannot be identified by SEM.

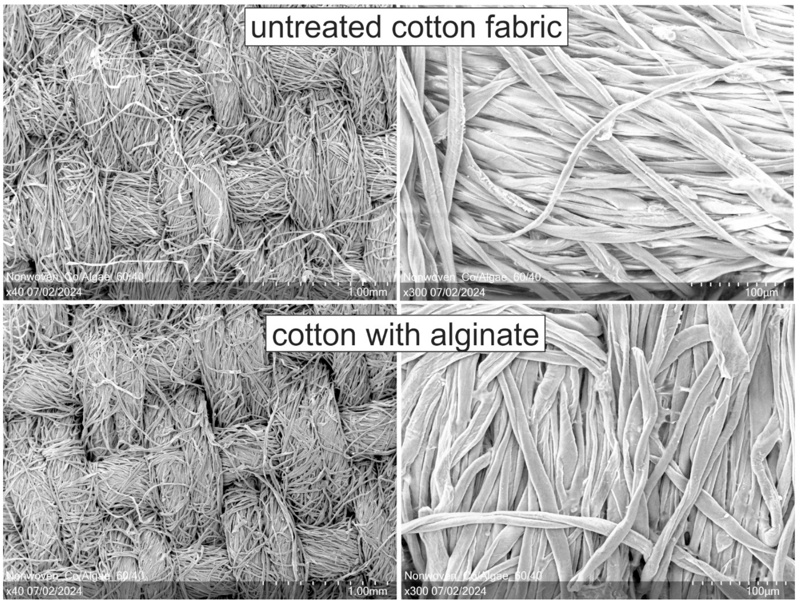


Figure 4. SEM images of reference materials without algae application, untreated cotton fabric, and cotton fabric with pure alginate application. The microscopic images are recorded in different magnifications ((**left**) lower magnification; (**right**) higher magnification).

SEM images recorded from the coatings containing the four different types of algae additives are presented in Figures 5–7. Figures 5 and 6 show those coatings on cotton fabrics in different magnifications. Figure 7 presents the coatings deposited on polypropylene (PP) foils. At lower magnification, a regular distribution of the cotton fabric by the applied coating is detected (Figure 5). For all four types of coating, the deposition on the fabric is regular and fills the space between fibers, so a flatter surface topography is obtained compared to the uncoated cotton fabric.

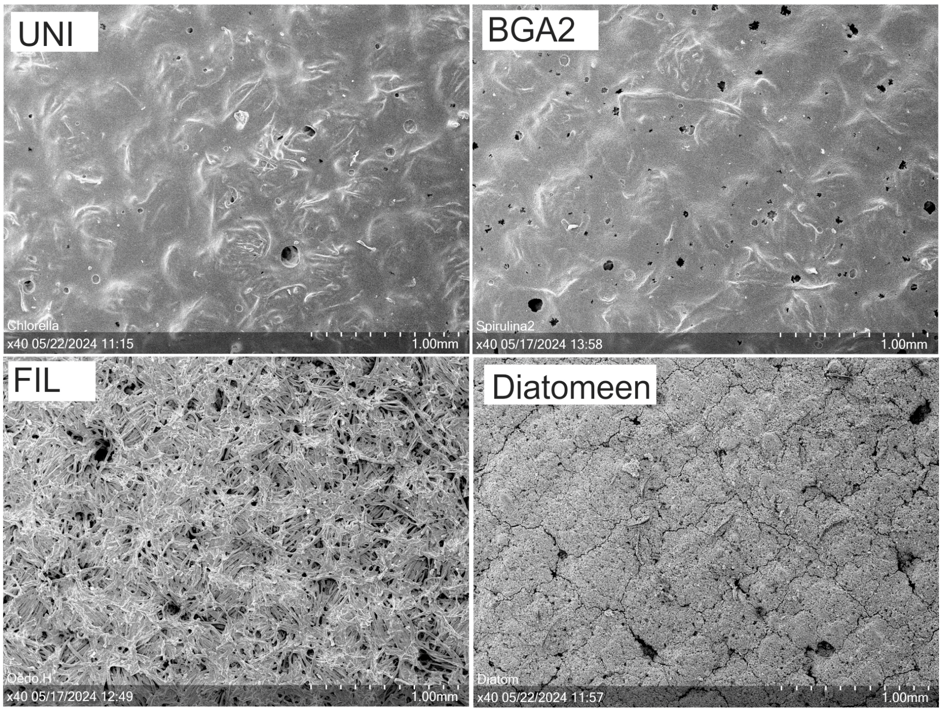


Figure 5. SEM images of cotton fabrics after application of algae containing recipes; the applied type of algae is mentioned in the single images. The SEM images are recorded at lower magnification to provide an overview image.

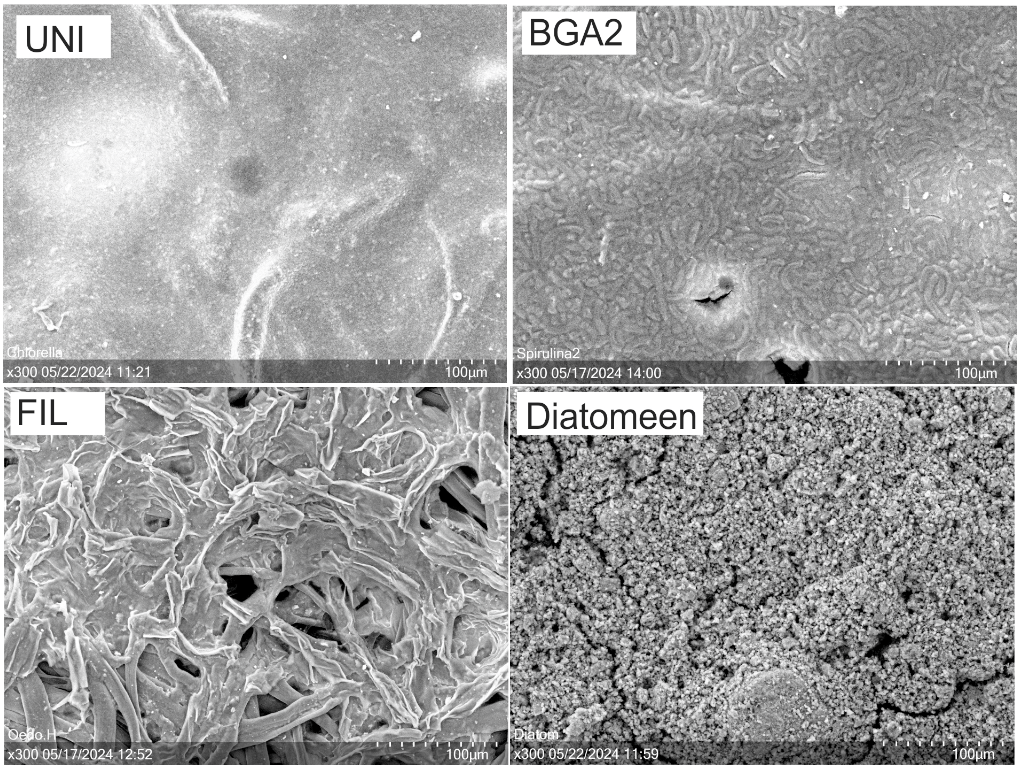


Figure 6. SEM images of cotton fabrics after application of algae-based recipes; the applied type of algae is mentioned in the single images. The SEM images are recorded in higher magnification to provide a detailed view.

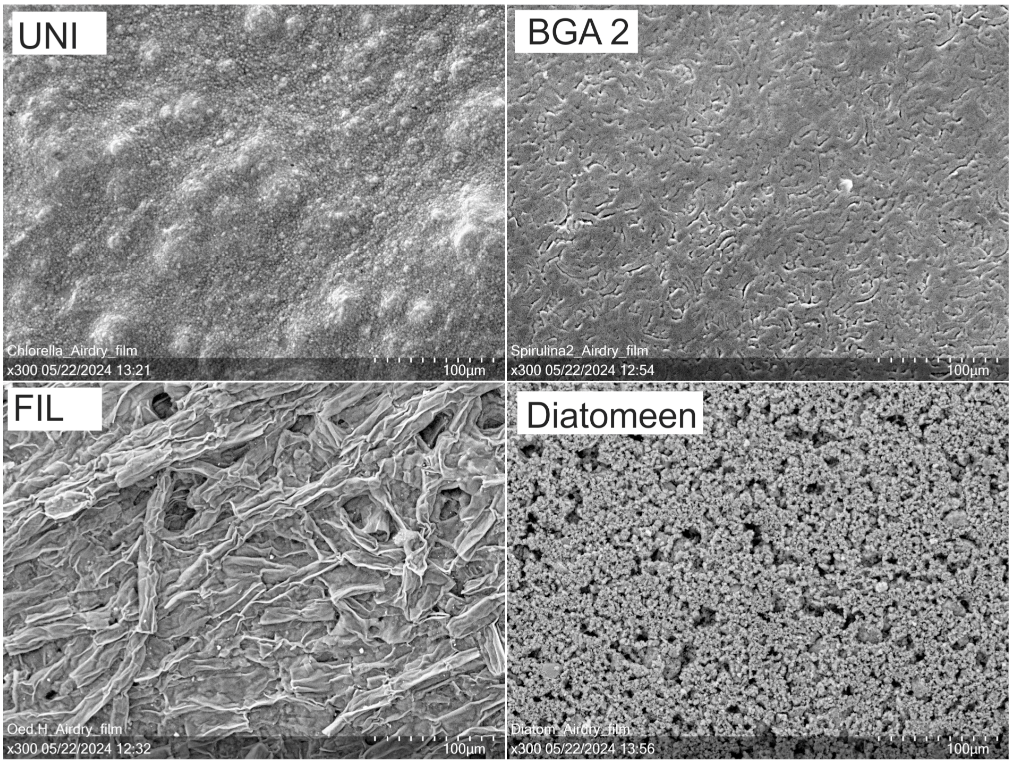


Figure 7. SEM images of algae-based recipes deposited and air-dried on polypropylene foil; the applied type of algae is mentioned in the single images. The SEM images are recorded in higher magnification to provide a detailed view.

At higher magnifications, the differences between different coating additives can be determined (Figures 6 and 7). Coatings containing UNI and BGA2 powders show less structured and quite homogenous topographies. It can be explained by the fact that these algae powders have small particles and are regularly dispersed in the binder system. This can be confirmed by the slight short filamentous structure appeared on the surface of BGA2, which is attributed to the spiral short filamentous structure of *Spirulina*. For the coating containing FIL material, a fibrous-like structure can be identified on the coated surface. Even after the performed grinding process of this filamentous alga, the fiber structure is still present and can be determined on the coated surface. For the coating with diatomeen material, a regular surface topography can also be determined, even though the fine pigmented structure of the diatomeen additive is indicated. The topographies of samples with cotton and PP foil as substrates are roughly similar (compare Figures 6 and 7). The applied coating recipe probably fills the complete interspace between the fibers of the textile substrate, so a flattened topography results, which is quite similar to the topography of an application on an already flat substrate like a PP foil.

Figure 8 presents SEM images of coated cotton fabrics after four weeks of sunlight exposure. The main structural features of the determined topography are similar to the topography of the coated samples after preparation (compare Figure 6). While the exposure to sunlight changes the coloration of algae containing samples, the influence of sunlight on the topography of the coatings cannot be determined by SEM.

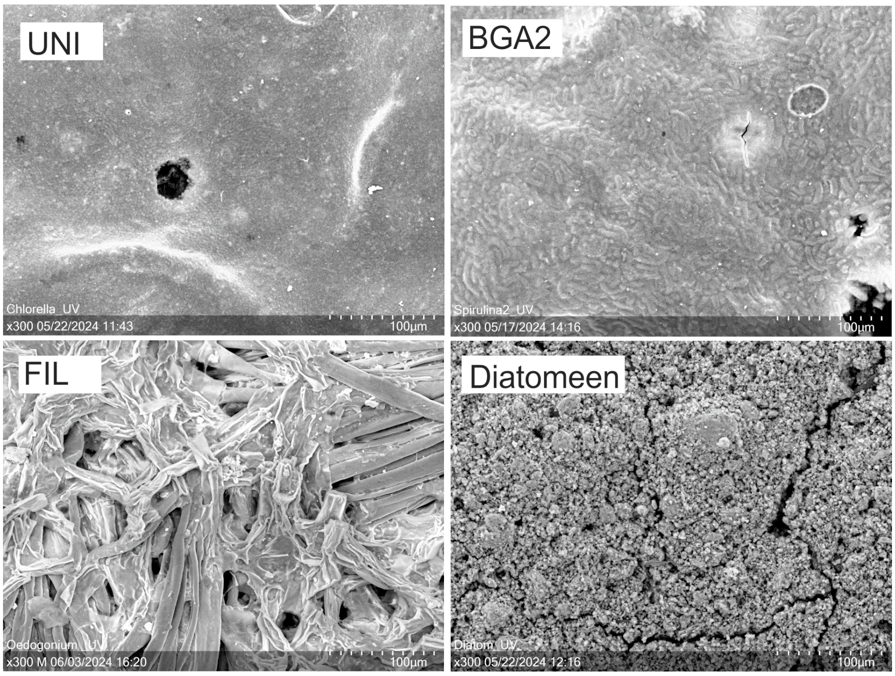


Figure 8. SEM images of coated cotton fabrics after four weeks of sunlight exposure; the applied type of algae is mentioned in the single images. The SEM images are recorded in higher magnification.

3.2. Coloration Properties

First, documentation of color properties was performed via photographs of the realized coatings applied on the cotton fabrics and the PP foils (Figure 9). By application of the algae containing the coatings, regular coloration in different green color shades is obtained. However, using diatomeen powder as an additive resulted in white to light-yellow coatings, which is in line with the coloration of this additive.

The color indices L*, C*, and h were determined for the prepared samples on cotton fabrics (Table 1). In addition, a comparison of coating application with and without the addition of calcium dichloride was performed. Additional to the information on the color properties, a first indication of properties according to light fastness can be provided. After preparation, the applied calcium dichloride shows no influence on the color of all coated samples (Table 1). With the application of the UNI and BGA additives, dark-green colorations with L* indices around 26 are obtained. In comparison, the addition of FIL resulted in lighter-green coloration with L* of around 43. The addition of the diatomeen powder leads to samples with a white appearance and the lowest coloration, with L* around 87. Based on the data color graphs and values, all samples containing algae additives, i.e., BGA, UNI, and FIL, exhibited a noticeable color change following exposure to sunlight. For some samples, this color change is more intense if calcium dichloride is present on the coated samples. By this, a negative influence of calcium dichloride on the light stability of prepared samples can be estimated. In general, after sunlight exposure, the colors of coated samples became lighter, indicating a significant alteration in their visual properties. This phenomenon can be attributed to the chromophores present in the pigment molecules responsible for the color of the algae. When exposed to sunlight, light energy can lead to photobleaching, a process where the chromophores undergo chemical changes that reduce their ability to absorb light, resulting in a loss of color [50]. This sensitivity to light exposure is particularly pronounced in organic materials, such as those derived from green algae, which often contain compounds that are susceptible to photodegradation [51]. In contrast, samples containing diatomeen additives do not show a significant change in color after exposure to sunlight. This lack of color change can be attributed to the inorganic nature of calcinated diatomeen, which does not contain light-sensitive components that would be affected by sunlight exposure [52].

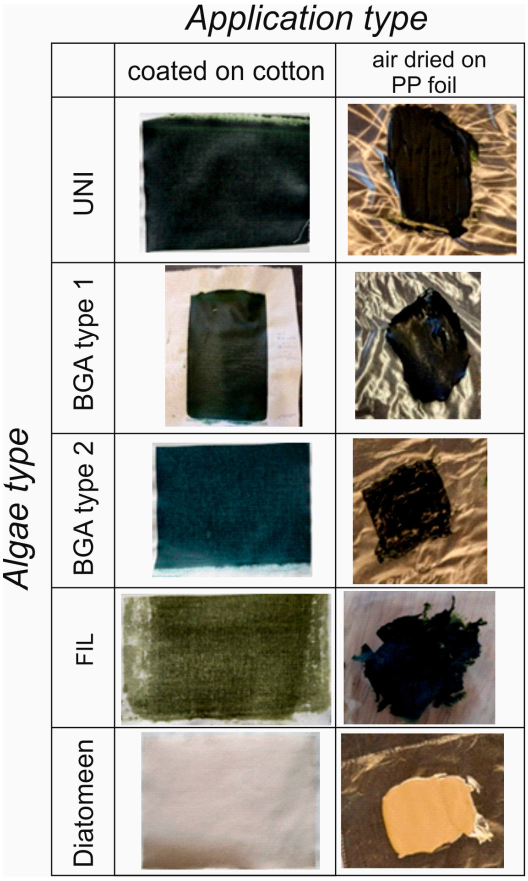


Figure 9. Photographs of prepared samples directly after application onto cotton fabrics and polypropylene foils as substrates.

Table 1. Preparations on cotton fabrics using unicellular green alga (UNI), blue-green alga (BGA), and filamentous alga (FIL), in addition to diatomeen—color indices after preparation and after four weeks of exposure to sunlight. The samples are prepared with and without the addition of calcium dichloride solution.

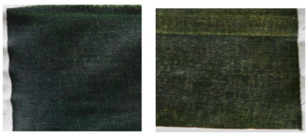
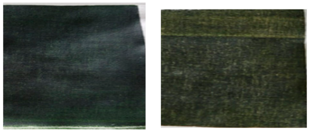
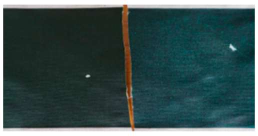
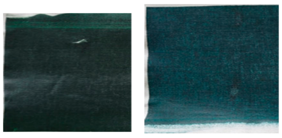
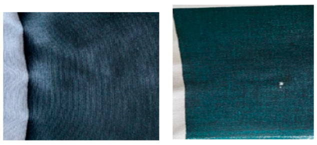
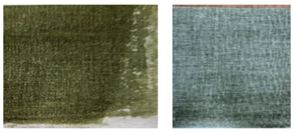
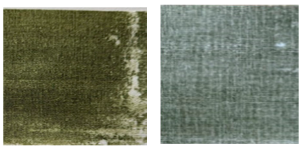
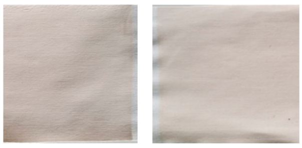
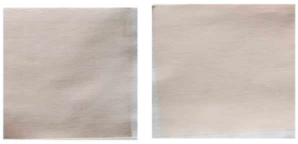
Applied Algae/Used $CaCl_2$	Photographs of Samples After Preparation and After Sunlight Exposure	Data Color Values (D65 10 Deg)		
		After Sample Preparation	After 4 Weeks Sunlight	Difference (Before and After UV Exposure)
UNI		L*—25.97 C*—4.50 h—136.90	L*—31.54 C*—7.79 h—113.38	L—21.4% C*—73.1%
UNI/$CaCl_2$		L*—25.97 C*—4.50 h—136.90	L*—30.52 C*—6.34 h—112.70	L—17.5% C*—40.8%
BGA type1		L*—26.6 C*—4.78 h—157.95	L*—31.48 C*—8.62 h—177.10	L—18.3% C*—80.3%
BGA type1/$CaCl_2$		L*—26.6 C*—4.78 h—157.95	L*—30.64 C*—7.97 h—177.29	L—15.1% C*—66.7%
BGA type2		L*—26.15 C*—4.14 h—164.67	L*—30.83 C*—8.79 h—191.14	L—17.8% C*—112.3%
BGA type2/$CaCl_2$		L*—26.15 C*—4.14 h—164.67	L*—30.83 C*—9.31 h—194.39	L—17.8% C*—124.8%
FIL		L*—42.98 C*—20.07 h—100.12	L*—59.23 C*—19.28 h—90.46	L—37.8% C*—(−3.9%)
FIL/$CaCl_2$		L*—42.98 C*—20.07 h—100.12	L*—55.04 C*—18.97 h—90.63	L—28.1% C*—(−5.4%)
Diatomeen		L*—86.80 C*—10.86 h—80.05	L*—86.35 C*—11.13 h—79.55	L—(−0.5%) C*—2.4%

Table 1. *Cont.*

Applied Algae/Used $CaCl_2$	Photographs of Samples After Preparation and After Sunlight Exposure	Data Color Values (D65 10 Deg)		
		After Sample Preparation	After 4 Weeks Sunlight	Difference (Before and After UV Exposure)
Diatomeen/$CaCl_2$		L*—86.80 C*—10.86 h—80.05	L*—86.62 C*—10.85 h—79.64	L—(−0.2%) C*—(−0.09%)

To support additional color impressions, magnified photographs of selected samples were recorded using a photomicroscope and are presented in Figure 10. These images were taken in reflection mode, and for illumination a microscope incorporating a light source was used. In this arrangement, the coated sample containing the algae material UNI appears with a dark-green coloration, changing to a lighter-green/yellow shade after sunlight exposure. The sample prepared with BGA2 appears dark blue, changing to a lighter blue after sunlight exposure. Finally, the color of the sample with FIL can be described as dull green, changing to lighter green after sunlight exposure.

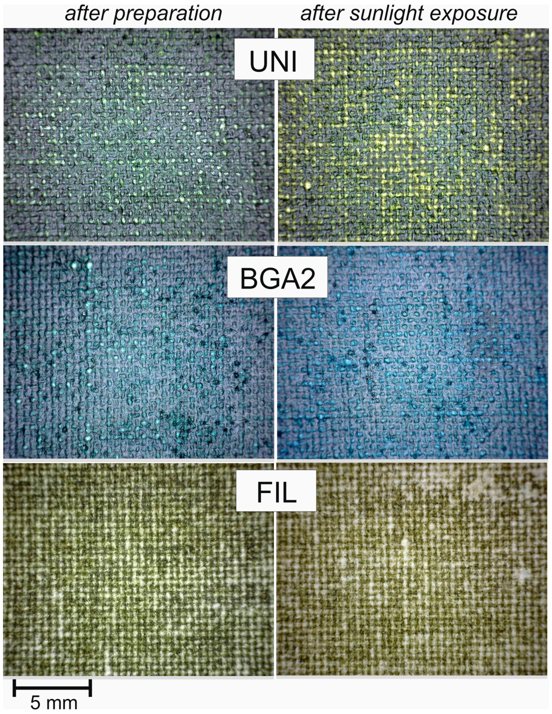


Figure 10. Magnified photographs of selected samples taken by a photomicroscope; coated samples with the algae materials UNI, BGA2, and FIL without application of calcium dichloride.

3.3. Fastness Properties

The evaluated properties of fastness to rubbing and light are presented in Tables 2 and 3. The rubbing fastness is determined for samples after coating preparation with and without the addition of calcium dichloride. After four weeks' exposure to sunlight, the rubbing fastness was determined again (Table 2). Coatings containing UNI, BGA, and diatomeen additives exhibit only a low rubbing fastness. In contrast, the coating containing FIL exhibits a grade 4 (i.e., good) rubbing fastness. This can be attributed to the fibrous structure of FIL, which supports a better interconnection to the binder system and the coated fabric, leading to a better rubbing stability. The addition of calcium dichloride shows a small positive effect on rubbing fastness, so here a crosslinking effect of alginate because of the presence of the bivalent Ca^{2+} ions may have an effect on the rubbing fastness. Surprisingly the rubbing fastness is significantly improved after exposure to sunlight. This statement is especially valid for the samples with UNI and BGA additives, showing low rubbing fastness directly after preparation. This observation can be explained by partial bleaching of the coating in the sunlight. This bleaching mainly affects color components near the coating/air interface, which is also mainly exposed for rubbing. In the case where the surface near areas of the coating is already bleached by sunlight, further applied rubbing will only remove mainly uncolored material and the color change caused by rubbing is less. For the nearly uncolored coatings with diatomeen additives, this statement is not valid and the rubbing fastness is low, even after exposure to sunlight.

Table 2. Fastness to rubbing of prepared coatings applied on cotton fabrics using unicellular green alga (UNI), blue-green alga (BGA), and filamentous alga (FIL), in addition to diatomeen.

Algae Type	Application			
	Only Algae	Algae with $CaCl_2$	Only Algae, After Sunlight Storage	Algae with $CaCl_2$, After Sunlight Storage
UNI	2	2–3	5	5
BGA, Type1	2	2	4–5	5
BGA, Type2	1–2	2	5	4–5
FIL	4	4–5	4–5	5
Diatomeen	1–2	2	1–2	1–2

Table 3. Light fastness of prepared coatings applied on cotton fabrics using unicellular green alga (UNI), blue-green alga (BGA), and filamentous alga (FIL), in addition to diatomeen.

Algae Type	Application	
	Only Algae	Algae with $CaCl_2$
UNI	5	4–5
BGA, Type1	5	4
BGA, Type2	4	3
FLI	3	3
Diatomeen	>5	>5

The prepared samples after treatment in the Xenotest are presented in Figure 11, together with the samples from the blue scale illuminated simultaneously. The related light fastness grades of prepared samples are listed in Table 3. Here, a comparison of fastness properties with and without addition of calcium dichloride is shown. For the green-colored samples with algae additives, significant light fastness in the range of 3 to 5 can be determined. The best values are achieved with additives UNI and BGA1, while the lowest light fastness is determined for the coating containing the FIL additive. This difference in light stability can be explained by the stronger coloration depth of samples with UNI and BGA additives, as samples with deeper coloration often exhibit higher light fastness.

Similarly, regarding the behavior under sunlight, the application of calcium dichloride leads to lower light fastness values in the Xenotest, even if the application of the calcium dichloride has no influence on the color of the prepared coatings. The coatings containing the diatomeen additives exhibit high light fastness because of the original bright coloration and the composition comprising a non-bleachable inorganic material.

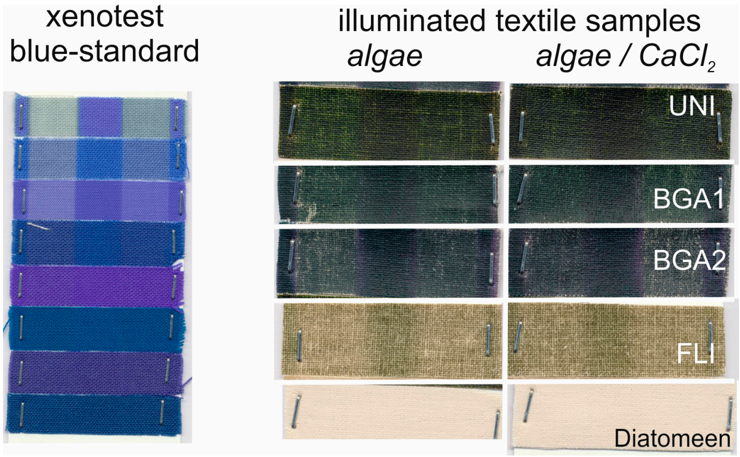


Figure 11. Photographs of samples after illumination in the Xenotest—(**left**) illuminated blue-standard; (**right**) illuminated cotton fabrics with algae containing applications.

4. Conclusions

The present study investigates the use of algae biomass as naturally colored components for possible coating of cotton textiles. This study demonstrates that algae biomass can be used as natural dyes for textiles and presents a promising alternative to synthetic dyes. These findings provide a foundation for future developments in the application of algae for textile material modification and functionalization. Despite algae-derived additives having significant potential as sustainable, eco-friendly dyes for the textile industry, future research could focus on optimizing the dyeing processes and increasing color durability, particularly through further investigation of the role of calcium dichloride as a crosslinker, which slightly improves the rubbing fastness of prepared coatings. However, a disadvantageous effect on the light fastness in the presence of calcium dichloride is also determined. For future research, evaluation and synthesizing of different crosslinkers are of great importance to determine their effectiveness in improving binding between algae-derived pigments and textile fibers. This could involve exploring alternative crosslinking agents or combining multiple additives to achieve superior color fastness. Overall, this study highlights the potential of algae as sustainable and eco-friendly dyes for the textile industry.

Author Contributions: L.B. writing draft, concept of idea; E.A.M. coating preparation, color measurements, investigation of fastness properties; E.B. concept of idea, organization of project and funding; A.R. preparation of algae, proof reading of manuscript; A.A. preparation of algae, proof reading of manuscript; B.M. concept and writing of manuscript, spectroscopic and microscopic measurements. All authors have read and agreed to the published version of the manuscript.

Funding: The presented research is realized under the funding of the research project IBÖM-08: AlgaTex—“Green algae as fiber material of the future?”—Feasibility phase for the use of filamentous freshwater algae as a potential raw material for the textile industry, funding code: 031B1358A, funded by the German Federal Ministry of Education and Research. The authors are highly grateful for this funding.

Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

Acknowledgments: The authors extend many thanks to Simone Wagner and Gudrun Lieutenant-Bister (Hochschule Niederrhein) for their help in the finishing lab and during color and fastness measurements. All product and company names mentioned in this article may be trademarks of their respected owners, even without labeling.

Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

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