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Harnessing Microalgae Biomass from Lake Taihu as a Natural Biocolorant Source for Sustainable Eco-Friendly Dyeing of Cotton with Ultrasonication

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Abstract

Most water bodies worldwide are infested with algae bloom and Lake Taihu is no exception, various techniques have been developed to harvest microalgae from Lake Taihu as part of the lake cleaning program and this results in a large algae biomass to deal with. This study made use of the algae biomass harvested as a biocolorants source for textile application and also evaluated its dyeing characteristics with mercerized and bleached cotton. The fabrics were dyed with either ultrasound or water bath method. The ultrasound method improved dye extraction yield from 17.8% for the conventional method to 26.7%, which was also enhanced to 33.2% with the addition of HCl (1 cm). The ultrasound dyeing method was effective at improving dye uptake at a reduced dyeing temperature when compared to the conventional method and also produced different shades of color after dyeing with different mordants. The dyed fabrics had good fastness properties for laundry, crocking and light. The dyed fabrics also showed a good ultraviolet protection factor. The use of algae biomass as a potential source of colorants for textile application will provide an alternate dye source that is environmentally friendly.

Keywords

Algae Biomass, Ultrasound-assisted Dyeing, Cotton Fabric, Solvent, Color Fastness

1. Introduction

There is a growing need for environmentally friendly, biodegradable, and eco-friendly products, and the textiles industry is no exception, with increased demand for natural dyes [1]. The increased interest in natural coloring sources stems from a growing understanding of the negative effects of some synthetic dyes on our health and the environment as a result of dye loss in the dyeing effluent. In order to control and minimize the harmful effluents, the use of natural dyes which are biodegradable is encouraged as well as effective effluent treatment. Some governments have banned some synthetic dyes, such as azo dyes, due to the risk they pose to humans [2]. Most natural dyes also possess additional functional finishing properties such as antifungal, UV protection, etc. Researchers are constantly exploring newer natural dye sources. Microalgae are found in freshwater, ocean, hot springs, and the arctic habitat and come in a wide variety of species [3] [4]. These microalgae reproduce quickly with the increased supply of Nitrogen and Phosphorous from waste effluents generated from both industries and homes resulting in algae blooms. There have been lots of algae blooms cases worldwide including Lake Taihu, Erie, Kasumigaura and Maracaibo [5] [6] [7] [8], which threatens the aquatic ecosystem, often forming blanket sheets depriving other aquatic animals and plants of sunlight and oxygen leading to hypoxic/anoxic conditions. Various measures have been taken to control, including massive investment in decreasing effluents discharge and effluent diversion, lake diversion programs and also harvesting these algae blankets which form during peak seasons. The disposal of these harvested algae as part of lake cleaning measures results in large lumps or sludge of algae biomass to deal with. These lumps of algae biomass could be a rich source of various bioactive compounds and colorants, the 2008 annual clean-up resulted in 0.8 to 1.6 million tons of algae mass. Lake Taihu, the lake of interest situated in China experienced algal bloom in 2007, leaving a population of over 4 million with a water crisis in Wuxi [5], a city situated on the shore of the lake. The microalgae species in Lake Taihu are Cyanophyta (blue-green algae/cyanobacteria), *Bacilliarophyta* (diatoms), Chlorophyta (green algae), Cryptophyta and Euglenophyta [9] [10]. The algae mass harvested as part of the lake cleaning process could be explored in various ways to find usage for it. Phycobilins, chlorophyll, Beta-carotenoids, fucoxanthin, phycoerythrin, and phycocyanin are pigments found in algae which find application in food coloring, pharmaceuticals, cosmetics, and paint additives [11] [12] [13]. This study would create awareness of the potential use of microalgae harvested during the river cleaning season as a potential dye source.

The ultrasound-assisted method has been used both in improving the extraction of various compounds and in the dyeing of textile materials [14] [15]. Ultrasonic cavitation has been reported for being able to cause physical and chemical effects detected in liquid or liquid/solid systems often resulting in the explosion of microbubbles generated creating pressure and temperature. The energy produced accelerates chemical reaction resulting in the reduction of chemicals, energy and time in dyeing processes. Ultrasonic treatment in dyeing results in the disintegration of agglomerated dye particles creating dispersion, degassing, strong agitation of liquid and also the swelling of substrate enhancing efficient fiber dye uptake. This treatment has been demonstrated to yield quick dye uptake at low temperature in comparison to the conventional method and also the reduction in energy consumption and wastewater pollution and has been used in various dyeing studies in the dyeing of natural dyes [16] [17]. Ultrasound treatment of leather increased chrome uptake during chrome tanning, reducing chrome dosage from 8% to 5% and shortening the processing time [18]. It is undeniable that the cost-effectiveness of ultrasound outweighs that of the traditional method [19].

The present work explores the novel use of algae biomass from Lake Taihu as a colorant source for dyeing cotton fabric and the enhancement of dyeing efficiency and fastness properties with ultrasound. The work will demonstrate and validate this new natural dye precursor as a sustainable and renewable dye source with the possibility to replace some synthetic dyes in the textile industry. The availability of algae and its fast growth rate make it a sustainable dye source with the potential to augment the increase in demand for naturally sourced dyes. The environment will benefit from these biodegradable dyes, which produce less polluted wastewater and reduce water purification demands.

2. Materials and Methods

2.1. Materials

Microalgae samples were collected from Lake Taihu in Changzhou, the samples were cleaned and dried before used. Mercerized and bleached cotton fabrics were obtained from Luolai Lifestyle Technology Co. Ltd China with the weights 125 g/m^2 and 115 g/m^2 respectively. The presence of phytochemicals in algal extract was determined using 1,1-Diphenyl-2-picrylhydrazyl (DPPH), Dragendorff's reagent, Folin-Ciocalteu reagent, Hydrochloric acid, sulfuric acid, Ascorbic acid, sodium carbonate, acetic acid, ferric chloride, ferrous sulphate, sodium hydroxide, ethanol, sodium phosphate, aluminum potassium sulfate and gallic acid in a qualitative analysis. All of the chemicals were of pure analytical grade. In the experiment, deionized water was used.

2.2. Dye Extraction

In this study, Ultrasonic assisted method was used in extracting colorant from algae sample with solvent (70/30% ethanol-water) to obtain a good yield. Alongside, thermostatted water bath (Rapid Precision Machinery) was used to extract samples for the conventional method for comparison with Ultrasonic assisted method. For the ultrasound method, extraction of algae samples was with an ultrasound instrument (Ningbo Licheng Co. Ltd., China) with ultrasound power setting of 70%. The extraction solvent's property is vital in the extraction process. Ethanol/water mixture was used as solvent for being benign and provides good recovery [20]. The extraction was carried out using a solvent mixture of 70% ethanol and 30% water (v/v) at 55˚C for 30 mins, the addition of HCl to solvent increase the yield. The ultrasonication was done in pulsed modes with a pulse length of 3 seconds and a resting interval of 2 seconds. The sample extraction using the Conventional method was at 80˚C for 60 mins. The extracted liquor was centrifuged for 5 mins at 3000 rpm in a tabletop high-speed Bioridge (TG16-WS) centrifuge. To extract ethanol and some water from the supernatant, the supernatant was concentrated under reduced pressure using a rotary evaporator (BUCHI, Labor Technik AG, Flawil, Switzerland) at 50˚C.

3. Sample Dyeing Condition

Both mercerized and bleached cotton fabrics were dyed with and without mordants and dyeing was with either ultrasound (Hechuang ultrasound KH 300DE) or thermostatted water bath shaker. Cotton fabrics were dyed after pre-mordanting treatment with the mordant aluminum potassium sulfate 6% (o.w.f) and ferrous sulphate 3% (o.w.f) at 60˚C for 30 mins and during dyeing the bath ratio of 30:1 was used at pH 7. Ultrasound dyeing (US) was at 70˚C and thermostatted water bath shaker (WB) at 90˚C for 60 mins as shown in the temperature-time dyeing diagram in **Figure 1**. The fabrics were introduced into the dye solution and the temperature rose to the desired temperatures 70˚C and 90˚C for ultrasound and water bath respectively. After dyeing the fabrics were rinsed thoroughly. Ultrasound has the ability to improve a range of chemical and physical processes, by inducing cavitation in liquid. Cavitation, which induces the creation and rupture of microbubbles, improves dye absorption by causing dye deagglomeration, agitation in the liquid, and fiber swelling, which improves fiber-dye interaction. The fabrics were dyed, rinsed, and dried at room temperature.

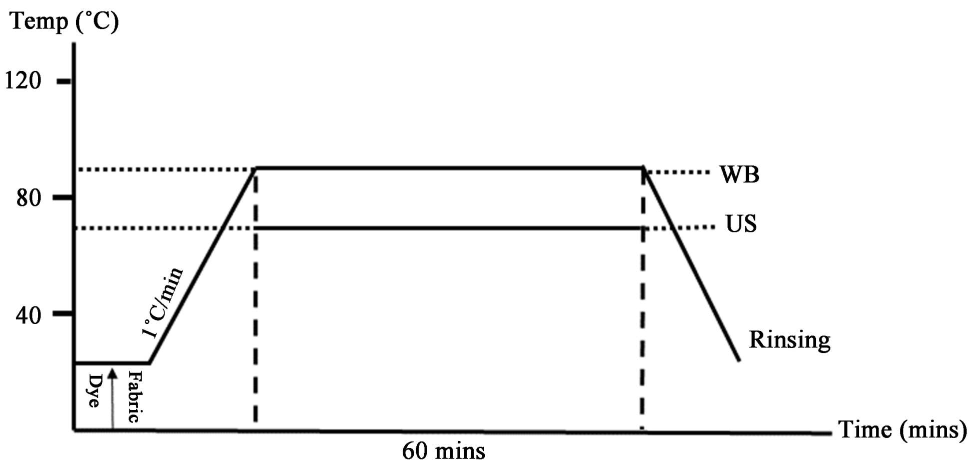

Figure 1. The temperature-time dyeing diagram.

3.1. Morphology of Algae Samples

The morphology of algae samples and extracts were observed using a HITACHI S-4800 scanning electron microscope (SEM, Japan). The samples were mounted with conductive adhesive tape, gold palladium sputter coated, and observed at a 3 kV accelerating voltage. The initial scale was magnified by 3000 times.

3.2. UV-vis Spectra and Chemical Composition of Algae Extracts

UV-Vis spectrophotometer was used to determine the absorbance spectra (V-1200, Mapada instruments). The Fourier-transform infrared spectrophotometer was used to evaluate the spectrum of algae samples (Nicolet iS10, USA). With the Attenuated Total Reflection (ATR) technique, the extract was measured from 4000 to 500 cm^{-1} at a resolution of 4 cm^{-1}.

The qualitative analysis of extracts of algae were assayed for the presence of alkaloids, tannins, saponins, glycosides, phenols, flavonoids, Quinones, protein, Coumarins, Carbohydrates, Phytosterols and terpenoids. The presence of phytochemicals was indicated by a (+) and the absence of phytochemicals was indicated by a (−).

3.3. Thermogravimetric Analysis of (TGA) Algae Extract

TGA of extract sample was conducted in a nitrogen atmosphere with a flow rate of 60 ml/min on a thermogravimetric analyzer TGA/SDTA 851e (Netzsch sta 449F5, Germany). From 30˚C to 790˚C, the samples were heated at a rate of 20˚C/min.

3.4. Colorfastness Properties of Dyed Fabrics

The color evaluation method and characteristics values (L^*, a^*, b^*, c^*, h^o and *K/S)* were reported for fabric samples. AATCC Test Method 61-2009 was used to determine colorfastness to laundry where multi-fiber fabrics (including acetate, cotton, nylon, polyester, acrylic and wool) were stitched unto dyed cotton fabrics and washed at 40˚C. The color change of dyed fabrics and the amount of dye stains on multi-fiber fabrics were determined. The AATCC Test Method 8-2007 was used to determine colorfastness to crock and AATCC Test Method 16-2004 was used to determine colorfastness to light.

The Ultraviolet protection factor (UPF) was evaluated according to the Australian/New Zealand Standard AS/NZS 4399:1996 using Cary 50 UV/Vis spectrophotometer. UPF indicates the amount of Ultraviolet (UV) protection provided the skin by a fabric. Higher UPF values mean better protection. UPF values are graded as follows: the range 15 - 24 denotes "good protection", 25 - 39 are referred to as "very good protection" and 40 - 50 and above are classified under "excellent protection" [21].

4. Results and Discussion

SEM investigation and extraction method impact on yield

The Scanning Electron Microscopic images in **Figure 2** shows the differences in morphology of algae samples before and after extraction with ultrasound. SEM was used to investigate the impact of the extraction method on the structure

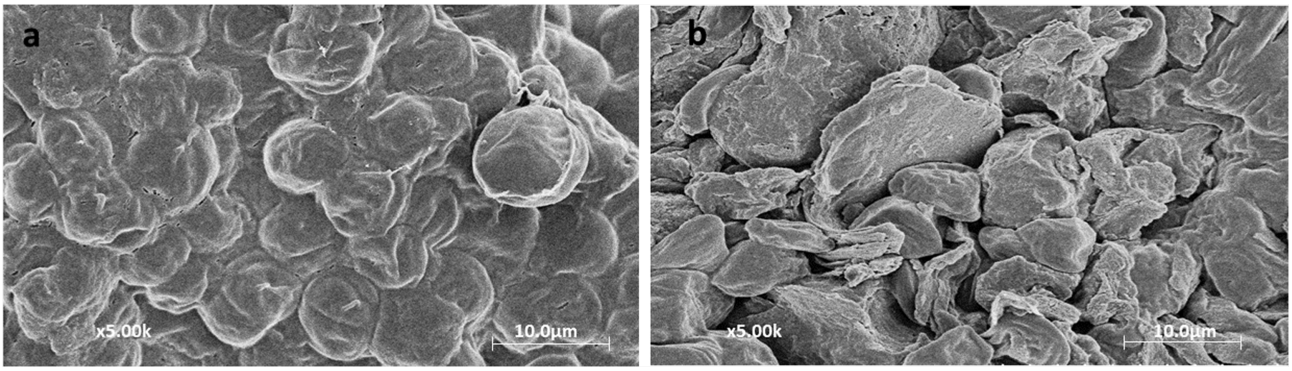

Figure 2. Scanning Electron Microscopic images of (a) Dried untreated algae biomass (b) Algae biomass after ultrasound treatment.

of the algae cell, **Figure 2(a)** shows the microscopic structure of algae before extraction, the cell surface which appears intact, unaltered and almost smooth and in **Figure 2(b)** after undergoing ultrasound treatment, the condition of the cell surface showed damages to the cell. The damage on the algal cell enabled solvent penetration and subsequent increased in yield of biocolorants when compared to the water bath method which does not show such damages to the surface of the algae cells [22]. Garcia-Vaquero *et al.*, (2020) reported similar impact of ultrasound on cell [23]. The ultrasound method increased the colorant yield with 70%/30% ethanol-water as solvent, with a yield of 26.7%, which was improved to 33.2% with the addition of 1 ml HCl to extraction. The ultrasound method had a 24.3% improvement on yield with HCl addition. The conventional method had a yield of 17.8%. The ultrasound method enhanced colorant yield at a reduced temperature and time. The mixed solvent of water-ethanol could extract polar and non-polar compounds.

The qualitative analysis reported the presences of some secondary metabolites such as phenols, flavonoids, tannins, alkaloids, terpenoids, steroids, carbohydrates, phytosterols, coumarins glycosides, proteins and quinones and these phytochemical compounds are known to have antioxidant potential and can help in dealing with several ailments [24] [25].

Effects of dyeing methods on dyed fabrics

Figure 3 and **Table 1** respectively show the mercerized and bleached cotton fabrics dyed with algae extract with different dyeing method and their characteristics values. For all the dyeing methods used, mercerized cotton had better dye yield and was improved when combined with the ultrasound dyeing method. Ultrasound dyeing yielded better results at a low temperature compared to the water bath at higher temperature to achieve close results. The use of ultrasound in natural textiles dyeing have the benefits of: low temperature dyeing which is energy saving, reduction of process time, is environmentally friendly using less chemicals, it reduces fabric deterioration during dyeing, and is cost effective and has been proposed for textile dyeing over the conventional method for mass production [21] [26]. **Figure 4** shows the improvement in dye uptake with ultrasound, which makes it an efficient and effective when compared to the

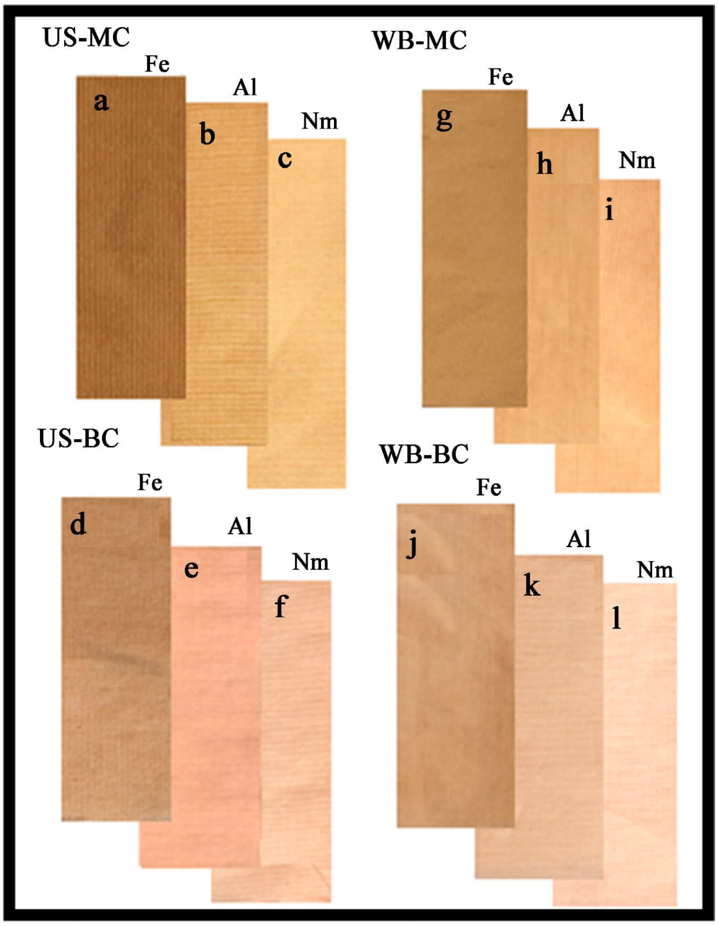

Figure 3. Mercerized cotton (MC) and bleached cotton (BC) fabrics dyed with Ultrasound (US) and water bath (WB) dyeing methods with algae extracts. (Nm) represents non mordant dyeing.

Table 1. The color characteristic values for cotton fabrics dyed with algae extracts.

The dyed cotton fabrics	Color characteristic values				
	*L**	*a**	*b**	*c**	*h*°
Figure 3(a)	67.74	13.48	24.29	27.77	61.0
Figure 3(b)	73.35	13.78	24.38	28.0	60.5
Figure 3(c)	76.51	15.36	23.78	28.3	57.1
Figure 3(d)	75.87	13.67	21.34	25.34	57.4
Figure 3(e)	77.5	14.38	18.29	23.26	51.8
Figure 3(f)	78.6	14.91	19.05	24.19	52.0
Figure 3(g)	74.86	14.14	27.12	30.58	62.5
Figure 3(h)	79.0	13.69	20.38	24.55	56.1
Figure 3(i)	78.27	15.22	24.23	28.61	57.9
Figure 3(j)	77.80	13.27	21.23	25.03	58.0
Figure 3(k)	79.85	13.29	17.18	21.72	52.3
Figure 3(l)	80.35	13.7	17.3	22.06	51.6

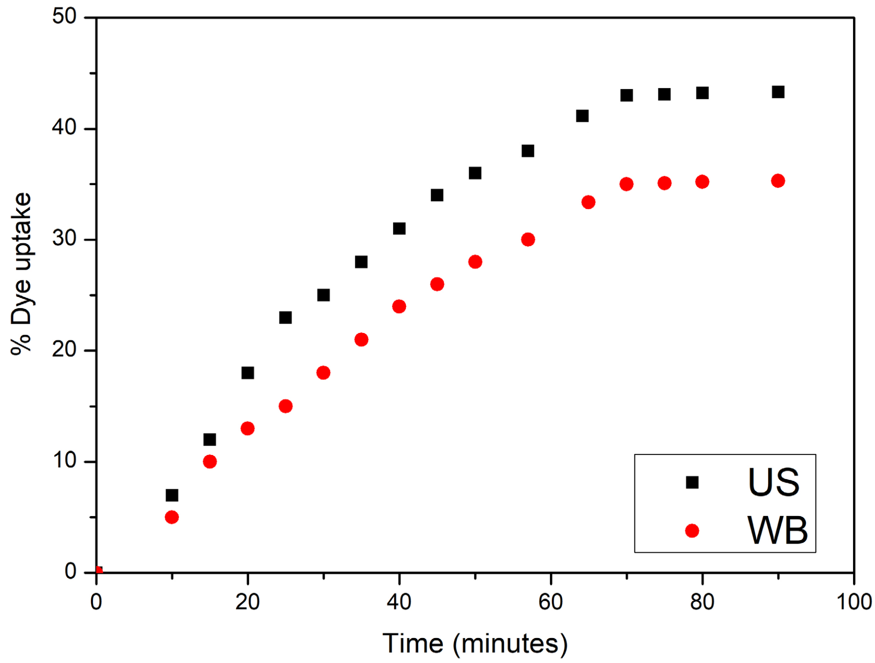

Figure 4. Results of dye uptake.

conventional method, as shown, the color strength of dyed fabric increased with time for both dyeing methods, however, there was pronounced increase for ultrasound. The use of alum and ferrous sulphate as mordants resulted in improved dye uptake and varying shades in comparison to direct dyed fabric due to the metal complexes formation with functional groups in dye and fabric. The pre-mordanting method impacted good fastness properties and good dye uptake and better shades when compared with the post mordanting method for all the dyeing techniques. The dyed fabrics showed different color shades from reddish brown to dark brown as shown in Figure 3, with Nm representing non-mordant dyed fabrics and alum or ferrous sulphate representing mordant dyed fabrics. The non-mordant fabric had light reddish brown, alum mordant resulted in reddish brown and ferrous sulphate had dark shade of the color, which is characteristic of ferrous sulphate and similar results were reported by wizi *et al.* (2018) [15]. With ferrous sulphate producing darker shades indicating a shift to towards the green co-ordinates in red-yellow zone of CIE lab color space, alum produced light shade. Light fastness of dyed fabrics is also influenced by the mordant and fiber substrate. Fabric dyeing with ultrasound produced darker shades for both mercerized and bleached fabrics, as well as a reduction in the L* value for all ultrasound treated fabrics. The mordant type and mordanting technique, dye-mordant interaction, dyeing method, and dye source can influence the color strength of dyed fabrics [27]. Figure 5 shows the color strength of dyed MC and BC. Dyeing involves a solid-liquid process, where dye molecules move from the liquid phase to the surface of fabric and a subsequent diffusion into the fabric. The ultrasound vibration influences increased dye mobility, break dye agglomeration and dye diffusion in fabric at shorter time through cavitation effect.

The ultrasound dyeing method reduces dyeing temperature, consumes less energy and reduces effluent load for treatment plants. Higher temperature ultrasound dyeing with some natural dyes has been reported to reduce dye uptake resulting from reduction in dye molecules stability [28]. The extract had less affinity for cotton, hence the need for mordant which forms complexes with dye to improve uptake and fastness properties. MC also had better dye uptake from the mercerization process which improves it dye uptake than BC. The Mercerization treatment of cotton fabric impacted changes in the surface morphology and structure which improved dye affinity, hence better dye uptake in MC than BC in both US and WB dyeing methods.

Fastness properties of cotton dyed fabrics with algae extracts

Table 2 shows the colorfastness rating for mercerized and bleached cotton

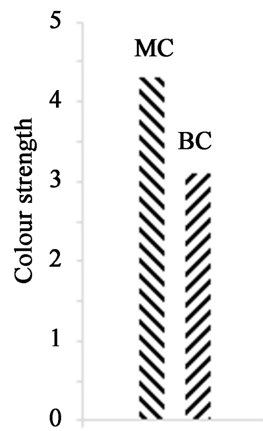

Figure 5. Color strength of mercerized and bleached dyed fabrics.

Table 2. Colorfastness assessment of cotton fabrics dyed with Algae extracts.

Dyed cotton fabrics	Colorfastness to laundry							Colorfastness to crock		Colorfastness to light
	Staining colorfastness						Color change	Dry	wet	
	Acetate	Cotton	Nylon	Polyester	Acrylic	Wool				
Figure 3(a)	5	5	4	5	5	4 - 5	4 - 5	5	5	4
Figure 3(b)	5	4 - 5	4 - 5	5	5	4 - 5	4	5	5	3 - 4
Figure 3(c)	5	3 - 4	4 - 5	5	5	5	3 - 4	5	4 - 5	3 - 4
Figure 3(d)	5	5	4	5	5	4 - 5	4	5	5	4
Figure 3(e)	5	4 - 5	4 - 5	5	5	4 - 5	4	5	5	3 - 4
Figure 3(f)	5	4	4 - 5	5	5	5	4	5	4 - 5	3 - 4
Figure 3(g)	5	5	4	5	5	4 - 5	4 - 5	5	5	3 - 4
Figure 3(h)	5	4 - 5	4 - 5	5	5	4 - 5	3 - 4	5	4 - 5	3 - 4
Figure 3(i)	5	4	4 - 5	5	5	5	3 - 4	4 - 5	4 - 5	3
Figure 3(j)	5	4 - 5	4	5	5	4 - 5	3	5	5	3 - 4
Figure 3(k)	5	4	4 - 5	5	5	4 - 5	3	5	4 - 5	3 - 4
Figure 3(l)	5	4	4 - 5	5	5	5	3	4 - 5	4 - 5	3

fabrics dyed using ultrasound and water bath method. Fabrics were dyed either directly or pre-mordanted with either Al^{3+} or Fe^{2+} prior. All the dyed fabrics showed good colorfastness to laundry, crock, and light. The algae extract indicated good fastness property to light in comparison to other natural dyes reported with lightfastness rating of less than 2 [29], the good fastness will ensure protection from the damaging effect of UV radiation. The mordant and substrate are key in determining the light fastness property of dyed fabric. Ferrous sulphate dyed fabrics are known for less fading compared to alum which might result from ferrous sulphate negative catalytic effects on the photochemical degradation of the dye, the study by Cristea *et al.*, (2006), reported that fading could be influenced by the dye's chromophore and auxochrome, mordant and fiber [29]. Mercerized cotton exhibited better dye uptake and color colorfastness properties than bleached because of its improved nature through the mercerization process, and for bleached cotton more crystalline regions contributed to low dye uptake. The mordant Fe^{2+} had substantially less fading in comparison to Al^{3+} possibly due to Fe^{2+} negative catalytic effect on the photochemical destruction of the dye [29]. The unmordanted dyed fabrics also had fairly good fastness property.

Extended exposure periods to ultraviolet radiation have detrimental effects on the skin, hence the need for UV-protected textiles materials to protect humans from hazardous radiations [30]. According to the Australia/New Zealand standard AS/NZS 4399:1996, a fabric has very good and excellent UV protection (UPF) if the UPF value is 25 - 39 and 40 - 50 and above respectively. The dyed fabrics had very good to excellent UPF results for both US and WB as shown in **Figure 6**, the study by Mongkholrattanasit *et al.*, (2011) reported very good to excellent UPF results in their study using a naturally sourced dye [31].

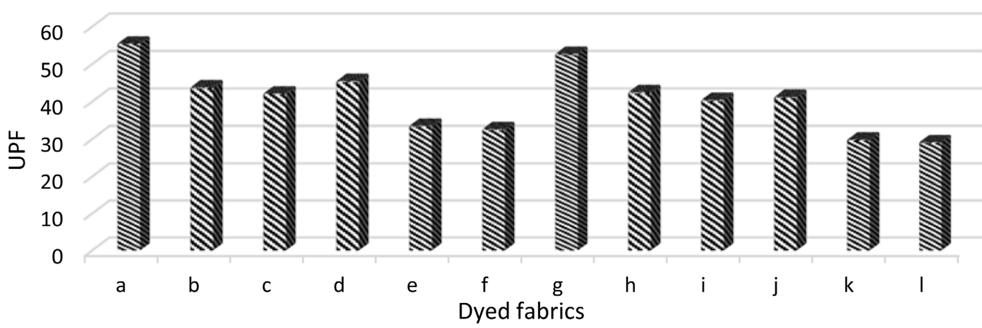

Figure 6. The UPF values for the dyed fabrics.

FTIR investigation algae extract

Fourier Transform Infrared spectra is a widely accepted spectroscopic method of assessing microalgal biomass composition and the method exhibits wavelength resolution that is significantly better than that of wavelength dispersive element like grating [32]. **Figure 7** shows the Fourier Transform Infrared spectra of algae extract. The band at 3400 cm^{-1} indicates N-H stretch, a secondary amine [33] whereas band 2920 cm^{-1} and 2850 cm^{-1} are respectively linked with the anti-symmetric and symmetric C-H stretching vibrations of methylene

(CH_2) group [33] [34] [35]. The band 1640 cm^{-1} (amide I) and 1540 cm^{-1} (amide II) indicates protein C=O stretching vibrations and N–H bending vibrations, respectively, and the absorption strength correlates to protein content [36]. The band at 1400 cm^{-1} represents the anti-symmetric bend of CH_3 group, the band 1060 cm^{-1} indicates C-O stretch and the band at 600 cm^{-1} represents O-H wag, which are characteristics of alcohols, carboxylic acids and fatty acid [37] [38].

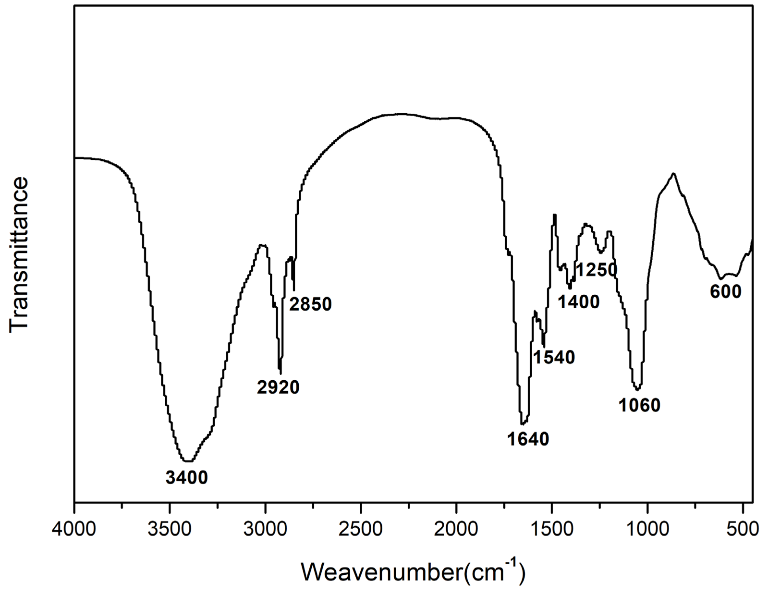

Figure 7. FTIR spectra of algae extract.

The thermal stabilities of algae extract

The TGA and DTG curves for the ultrasound-assisted algae extract are shown in **Figure 8**. At 800˚C, the weight residue percent was high. The weight loss in

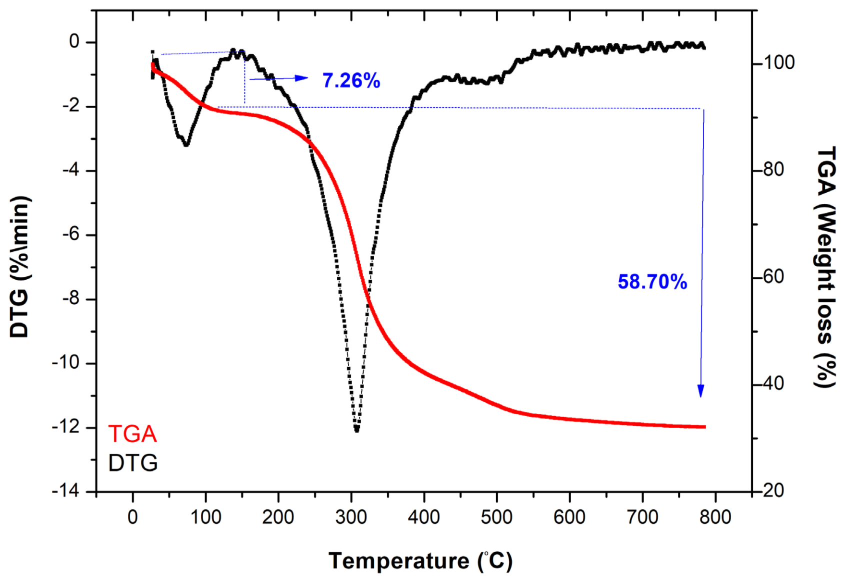

Figure 8. Thermogravimetric analysis (TGA) and Differential thermogravimetric (DTG) curves of algae extract.

the extract occurred in three stages, with the evaporation of water in the sample causing the weight loss below 100˚C. As the temperature rose from 100˚C to 250˚C, the weight reduced with the highest weight loss at 7.26%. Temperature above 250˚C resulted in a fast weight loss due to the thermal decomposition of organic compounds for the second and third decomposition. Textile products are generally dyed and finished at temperatures below 150˚C, some studies undertook high temperature dyeing; Luo *et al.* (2014) dyed at 130˚C [39], Nakpathaom *et al.* (2019) improved dye uptake at high temperature dyeing 80˚C - 130˚C with natural dyes [40], the extract's good thermal stability makes it appropriate for dyeing processes on a various textile materials [40].

In conclusion, the ultrasound dyeing method improved both dye uptake and extraction at lower temperature which is environmentally friendly in comparison to the water bath method and the mercerized fabric had better fastness properties than bleached fabric due to its enhanced treatment. Microalgae biomass has the potential to be used as a source of coloring in textiles.

5. Conclusion

The findings of this study showed the algae biomass harvested from cleaning Lake Taihu as a source of biocolorants for textile application and also contains a variety of phytochemical compounds with antioxidant properties. The ultrasound extraction method with 70/30% ethanol-water as solvent had a colorant yield of 26.7%, which was improved to 33.2% with the addition of 1 ml HCl to the extraction, indicating an increase of 24.3%, whereas the conventional method had a yield of 17.8%. With the cavitation effect, the ultrasound dyeing method further improved dyeing efficiency. The dyed fabrics also showed good fastness properties, and ultrasound-assisted dyeing method was indicated as an effective dyeing method. The study showed microalgae from Taihu as a sustainable dye source and the ultrasound dyeing approach as an effective method for dyeing textiles with functional finishing properties. Algae biomass could become a sustainable natural dye source with potential application in the textile, food coloration, and pharmaceutical industry.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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