**Research Article**
Volume 10:7, 2020
DOI: 10.37421/jtese.2020.10.425

Journal of
Textile Science & Engineering

ISSN: 2165-8064
Open Access

# A Critical Review on PLA-Algae Composite: Chemistry, Mechanical, and Thermal Properties

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## Abstract

The traditional polyester and synthetic polymer materials are imposing a boundless threat to our environment; by contrast, bio-based and biodegradable plastic material is offering a great solution to overcome this detrimental effect of conventional plastics. However, to protect our ecosystem by utilizing marine waste like algae as a polymer material is another approach and getting attention to the scientific community. This study investigates the comprehensive understanding of using biodegradable nonwoven composite made of polylactic acid (PLA) and marine waste algae. This review also finds the potential compatibility of using PLA and algae as a nonwoven composite to substitute the non-degradable synthetic polymers. Moreover, this current study explores the extent to which the adhesion between filler(algae) and matrix (PLA) in bioplastic composite can be used for the end-uses purposes, and inspects the underlying factors related to thermal and mechanical properties of PLA-algae nonwoven composite. Future research needs to quantify the adhesion (between filler and matrix) as well as advance characterizations of this potential biodegradable nonwoven composite.

**Keywords:** Biodegradable composite • Natural fiber-reinforced composite • PLA-algae composite • Filler • Matrix

## Introduction

With the attenuation of energy sources and the environmental concern due to the plastic disposal from petroleum production, the interest of biodegradable composites and renewable sources is growing up. For sustainable practice, the production and advancement of biodegradable materials are getting more attention these days [1-3]. Additionally, most of the used commercial composites such as carbon fiber, glass-reinforced composites, and polyester are fossil-based. These fossil-based composites cannot be considered as the biodegradable composites due to slow degradation rate in the environment [4-6]. Moreover, traditional biodegradable polymers have poor mechanical, thermal, and electrical properties with processing difficulties. Thus, these polymers cannot serve the actual purpose of the polymers [7-9]. Besides, to fulfill the requirements for using those polymers in different sectors like packaging and automotive industries, the properties of the composites should be improved. The mechanical properties of the traditional polyester composites could be improved by applying advanced technologies and reinforcing fillers [10]. Natural fibers [as a filler] are the most important source of biodegradable polymeric materials. As a filler in the polymeric material, cellulosic fibers such as flax, ramie, jute, sisal, and hemp are used for reinforcing the polymeric substance for many years [11]. Algae is one of the cheapest sources of filler as it is widely available throughout the world. Even though the algae are using in the sector of agriculture, food, and other cosmetic products for many years as a polymeric substance, the feasibility of using this polymer in a broader range is an utmost need [10]. The rapid proliferation of algae is causing severe water pollution. Hence, the use of this cheap and enormous sources of cellulose fiber [algae] is of great interest to researchers in the field of bio-composites [12]. There are two pivotal aspects of using algae in polymer composites. The first aspect is to reduce the production cost and the carbon emission from the polymer composites. Second, algae are used as the reinforcing fiber [as a filler] in the bio-composites [11-13]. PLA is a bio-based polymer that is derived from nature [renewable sources] such as corn and potato starch. The thermoplastic nature with biodegradable properties of this type of polymer makes it more valuable and usable in any sectors such as the packaging, automotive, and food industries. Moreover, the availability of PLA and the known production process of PLA are persuading to produce bio-composites by using PLA and other natural cellulosic fibers [14]. Hence, the characterization of PLA and algae-based composite is essential to a great extent to make it more usable [15].

Over the last three decades, the production of synthetic polymers, especially polyester polymers, have been increasing at a high rate all over the world. Every year, approximately 140 million tons of plastic polymers wastages are thrown in the environment by the United States alone [16]. Synthetic polymers, specifically the plastic which comes from the packaging industry, contributes more than 25% of disposal [17]. Furthermore, municipal waste is also made of non-degradable synthetic polyesters. Consequently, lands are filling, and water is contaminating by solid waste. Synthetic polymers are thought-about the detrimental pollutants of the environment. Thus, the development of bio-based polymeric materials is getting tremendous attention to the researchers. Henceforth, the substitution of synthetic polymers by degradable polymers such as natural fiber-based polymer composites are being developed. The composite materials, which are produced by natural polymers, are easily degradable in the environment as compared to the synthetic polymers [18]. Gautam and his co-workers studied that the blending of synthetic polymers [Poly Vinyl alcohol and nylon] with natural fibers are easily digestible by micro-organisms. Thus the rate of biodegradation of polymers has been increased [19]. However, the production of PLA has been projected 140000 tons per year globally; thus, it is predicted that the emission of greenhouse gas will be reduced, and in turn, it leads the automotive industries and manufacturing industries to the production of bio-composites in the colossal spectrum from renewable sources[14].

PLA is one of the feasible polymeric materials with its versatile applications in many sectors such as automotive, sports, packaging, and biomedical applications [20]. The favorable properties of PLA are transparent and high rigidity. However, it has some setbacks, such as brittleness because of its high degree of crystallinity, lower toughness as the elongation of PLA at break is less than 10%, and slow degradation rate than other bioplastics[due to hydrolysis of ester groups][21, 22]. Extensive research has been done to overcome the abovementioned drawbacks of PLA polymers such as the

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**Received** 25 October 2020; **Accepted** 19 November 2020; **Published** 26 November 2020

reinforcement of PLA with natural fibers such as algae, cotton, hemp, flax, etc. Natural fiber-based PLA composites have a lot of advantages due to the low processing cost and higher biodegradability rate [21,22]. Moreover, the bioplastic made of PLA and natural fibers, specifically PLA and algae, are compatible with many processing techniques such compression, injection, extrusion molding, and so on [23,24]. However, there are very few researches have been reported on PLA-algae composite, especially on chemical, mechanical and thermal properties of the PLA-algae composite. This study aims to address the comprehensive view of PLA-algae composite in terms of its fabrication techniques, thermal, mechanical properties, and applications.

# Polymer Composite

A polymer composite is a material that is made of the combination of the two or more substances or materials, and this composite possesses the different properties as compared to the original constituent materials. The approach of the composite is to bring significant change and characteristics through multi phases of the base materials [25]. The use of natural fiber-based polymer composites started in 1908. This polymer composite was made of phenol and cotton fibers. The reinforcement of plastics, specifically, glass fiber, which was reinforced with natural fibers, was brought into the market for practical use at around 1940 [26].

Moreover, the combined base materials are mixed in the composite at the microscopic level. Additionally, one of the constituent material is called reinforcing material, and another is called the matrix [27]. Synthetic polymer-based composites are used in every level of daily life, such as sporting goods, household goods, automotive, structural, and food industries.

## Natural fiber-reinforced composite polymers

Natural fiber-reinforced polymer composites consist of a polymer matrix, and this matrix is embedded with natural fibers such as jute, sisal, flax, and oil palm. Generally, polymers are categorized into thermoplastics and thermosets [28]. Thermosets polymers have properties of high strength, higher Young Modulus, and it can't back to its original state after melting. On the other hand, thermoplastics polymer such as polyethylene, polypropylene, and polyvinyl chloride are using for reinforcing composites with natural fibers to get the better properties of biodegradability [29]. The chemical structure of natural fibers [cellulose, hemicellulose, and lignin] helps to get ultimate properties while natural fiber is used as fillers or reinforcing material with other degradable or non-degradable materials [30]. Endowing some chemical properties of wool by using alkali treatment on jute has enormous potential to use as a reinforcing material with the PLA [31]. Many researchers have studied the feasibility of using natural fibers with non-biodegradable polymers and found desired properties like good degradation rate in the environment and improved mechanical properties [32,33].

Until recently, researchers started to use natural fibers as the reinforcing components in the polymer composites because of the low cost, biodegradability, and higher mechanical characteristics of the natural fibers [34]. Cellulose fibers such as algae, flax, and starch are very cheap, copious, lightweight, robust, and lenient, making them an auspicious practice as reinforcement to replace the traditional polyester materials. Hadjadj et al. found that polyurethane-cellulose composite has higher tensile strength with increased modulus than the polyurethane based nondegradable polyester composites. Therefore, the use of cellulosic fibers in the composites material can have enormous potential in reinforcing polymeric materials [35]. Since the late nineteenth century and early twentieth century, there are many biodegradable composites have been developed, which can challenge the petroleum-based composites or composites which are not biodegradable [36]. The polymers which can be derived from renewable sources like butyl group polymers are environmentally friendly. The polymers which have the butyl group ester are obtained through the fermentation of microorganisms [37]. Besides, these polymers have excellent mechanical properties with the characteristics of biodegradability in the environment. Thus, the composites which are made from those polymers with natural fibers will have a lot of applications in many fields such as the packaging industry and automotive industry. However, the applications of biodegradable composites are limited due to the mechanical performances of the composites as compared to the petroleum-based composites [38]. Many bio-based composites can get from soy protein, corn, and sunflower as shown in Figure 1 [39,40].

## PLA-algae polymer composite

The preparation of composites as shown in Figure 2 with algae is also

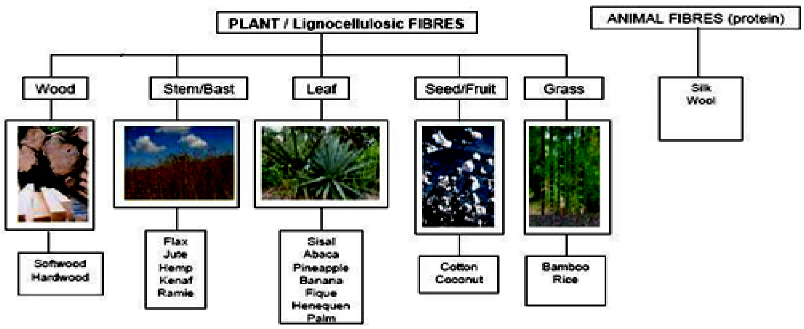


**Figure 1.** Classification of natural fibers [40].

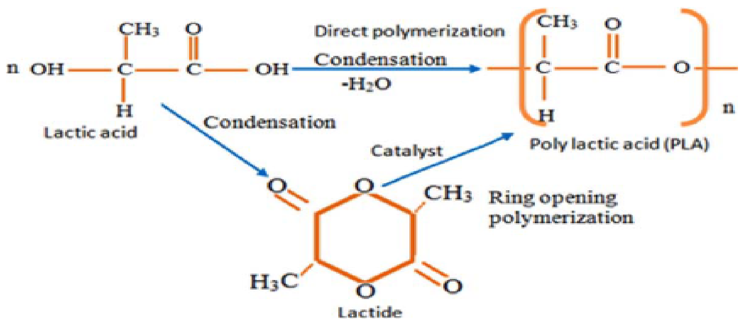


**Figure 2.** PLA polymerization [46].

another approach in terms of biodegradability and environmental pollution. Algae create water pollution, and thus, there is a shortage of fresh drinking water throughout the United States and it disturbs growing aquatic animals like fish. In the US alone, 30-48 million people get their drinking water from the reservoirs and lakes [41]. The use of algae as a sea waste has excellent potential and significance concerning environmental contamination. The use of algae as a filler in the polymer matrix helps to improve the mechanical performance of the composites. Sim et al. found that composites which are made of algae and butylene succinate has good mechanical and thermal performances. Here algae should be treated with bleaching chemicals before melt-mixing of the algae and PLA [42]. Though the production cost of composites has been increased due to treating algae with bleaching agents, using treated algae with PLA has enhanced the biodegradability [43].

The use of algae in the composite as a filler help to reduce the production price of the polymer composites. Another aspect of using algae waste is to reduce the carbon emission from petroleum resources [44]. Ferrero et al. studied bleached algae used in the polymer composites with cellulosic compounds that are easily biodegradable after disposal since there is a lack of impurities in the produced composites [45].

## Advantages of PLA-algae composite

Fully biodegradable materials have several advantages due to their low processing cost and low cost on waste management after disposal of biodegradable composites [e.g., PLA-algae composite can be disposed and degraded after incineration and landfilling] [47,48]. Moreover, biodegradable PLA-algae composite has numerous advantages regarding processing methods such as compression molding, extrusion, and injection molding, melt mixing and etc. [49]. The adhesion between brown algae and marine mussels fostered to develop glue which is known as the hydrogel composite and the adhesion strategy nourishes to produces a glue formed composite from the separation of adhesives and polymers. This approach helped researchers to develop the non-adhesive type polymers and alginate into the adhesive gel with improved properties and performance; on the other words, the development of non-adhesive gel was not viable with the traditional chemical conjugation techniques [50].

Surface modification of Cellulose Nanofiber [CN] is vital to acquire the homogeneous dispersion with hydrophobic materials due to the hydrophilic nature of the CN [51]. Surfactant modified cellulose nanocrystals in PLA showed a homogeneous dispersion. Moreover, the plasticization is brought by the surfactant on the physical and mechanical properties on PLA based composites. For instance, CN, which is treated with silane, applied to the PLA by the melt-compounding technique, PLA-CN composite has improved modulus in the glassy state of the composite and has lower degree of crystallinity of PLA-CN composite [42,51]. At the temperature of 96°C -146°C, PLA has increased its crystal fraction from 41% to 67% and the crystallization temperature was changed during the raising of temperature from 82°C to 146°C [42]. It is found from the characterization of the red algae that the fiber length and diameter are ten times shorter than other natural fibers. The crystalline cellulose has less thermal stability than the red algae fiber, which is treated with bleaching agents [42,49]. Hence, the uniform shape of algae offers to utilize these algae as reinforcing material with PLA or PHA. The algae reinforced with poly butyl succinate [PBS] showed higher fiber loadings [10,42]; thus it can improve the mechanical properties of PLA-Algae composite.

## Algae as a filler with other synthetic fibers

Lee and his co-workers used algae with synthetic polymers (polypropylene and butylene succinate) by the melt-mixing process to get better mechanical and thermal properties as shown in Figure 3 [52]. However, treating the algae before making composites with bleaching agents is somewhat costly, but this treatment has yielded good quality polymer composites as compared to polyester-based composite in terms of biodegradability [53]. The composite made of algae and polyvinyl alcohol [PVA] by film casting process has greater biodegradation rate. Besides, in this case, the particle size of the composite was smaller, and had improved mechanical properties with good cohesion between filler and matrix as shown in Table 1 [54]. The composite is made of PLA with zostera in the presence of starch produced very brittle polymer; however, this composite had a 3% higher Young Modulus than the neat PLA [55,56].

Algae is one of the major resources of an organic compound and it can convert solar energy to biomass. However, many algae species (number of known algae species is 221) are using in the many fields such as energy, food, chemical industry, and biotechnology [48]. The use of PLA-algae composite can be used in terms of industrial purposes. For example, marine micro algae contain proteins, carbohydrates, lipids, fatty acids, glycerol, pigments and specially, polyphenol groups have the antibacterial and antifungal properties [57]. Algae has polysaccharides; thus it has the potential to reinforce with different types polymers such as PLA and poly hydroxyalkanoates [PHA] to produce bioplastic considering the antifungal and antibacterial properties of the bioplastic [58].

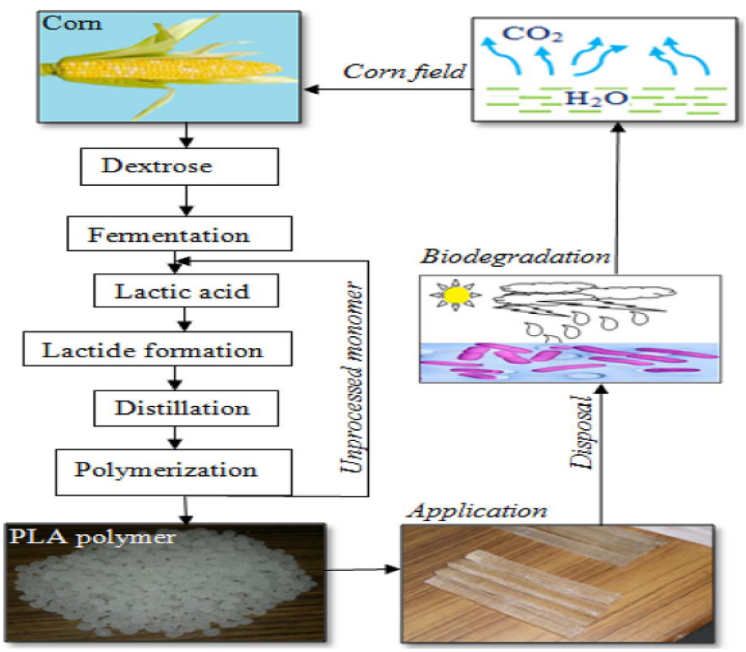


**Figure 3.** Synthesis of PLA [46].

**Table 1.** Physical Properties of some natural and synthetic fibers [56].

| Fiber | Density (g/cm³) | Elongation (%) | Young Modulus (GPa) | Tensile Strength (MPa) | Absorption (%) |
|---|---|---|---|---|---|
| E-glass | 2.5 | 2.5 | 70 | 800-1400 | - |
| Aramid | 1.4 | 3.3-3.7 | 63-70 | 2140-2250 | - |
| Carbon | 1.5 | 1.4-1.8 | 230-270 | 2860 | - |
| Flax | 1.5 | 1.2-3.2 | 27-80 | 345-1500 | 7 |
| Cotton | 1.5-1.6 | 3-10 | 5.5-12.6 | 287-800 | 8-25 |
| Sisal | 1.3-1.5 | 2-2.5 | 9.4-28 | 511-635 | 12-27 |
| Coir | 1.2 | 15-30 | 4-6 | 131-220 | - |
| Ramie | 1.5 | 2-3.8 | 44-128 | 400-938 | - |

**Table 2.** Thermal decomposition temperature of bleached red algae fiber (BRAF) reinforced PLA composite [42].

| | $T_5$ | $T_{25}$ | $T_{50}$ | $T_{75}$ | Residue % |
|---|---|---|---|---|---|
| BRAF | 84.2 | 332.1 | 353.7 | 369.5 | 11.7 |
| PLA Matrix | 303.7 | 335.8 | 350.9 | 361.6 | 0.9 |
| 30 wt.% BRFA-PLA | 299.2 | 332.4 | 343.9 | 354.3 | 4.9 |
| 40 wt.% BRFA-PLA | 298 | 333.7 | 334.4 | 354.4 | 5.5 |
| 50 wt.% BRFA-PLA | 278.7 | 331.7 | 345 | 355.9 | 6.7 |
| 60 wt.% BRFA-PLA | 276.7 | 330.5 | 343..7 | 354.2 | 7.4 |

## Application of Algae and PLA based composite in different sectors

Biodegradable composites are using in biomedical applications such as sutures, implants, and drug delivery systems. However, the use of biodegradable composites for the daily commodities is barred by its processing difficulties with poor mechanical and thermal properties [55]. PLA, polyglycolide, and other biopolymers such as starch and saccharides are manufactured from renewable sources. Blending or composites made from those polymers allow using for specific purposes [59]. Composites made from starch with synthetic or natural polymers have many applications [60]. Iannace et al. studied that composites made from polyhydroxy butyrate with PLA have good biodegradable properties. Characteristics and mechanical properties can be modified by adding other natural fibers such as algae, wood. The production cost also can be reduced in this way [11,61]. Polymer composites made from polycaprolactone with algae have improved biodegradable properties, though additional studies have not done yet on these composites. A blending of polyvinyl alcohol [PVA] with natural fibers has better performance in terms of biodegradability, but this polymer should be treated with biodegradable materials to get better degradation in the environment [62]. PLA and algae are biodegradable materials; hence they have tendency of growing microbes in the composites as the composites are favorable to absorb moistures from the environment. The use of additional antimicrobial treatment [e.g., surface modification of PLA-algae composite can be done by the phenolic compound] needs to inhibit the growing of microbes [63]. Cellulosic material like algae, and lignin comprised material like jute, both can be reinforced with PLA; have the potential to use these bioplastics in the medical textile such as baby monitoring system to reduce the chances of stroke from breathing problem [64,65].

### PLA-algae composite in tissue engineering

Tissue engineering uses the specific cells and scaffolds to heal the wounded organs of the body [66,67]. The composites are manufactured according to the predesigned scaffolds in acquiring the new tissues in the injured organ [68]. Hence in the production of scaffolds have a colossal potential to use the fibrous structure of PLA-algae composite. PLA and PLA-calcium phosphate based scaffolds are prepared by gas foaming and are used to generate bone in the damaged organ [69]. Many biomedical applications are studied, for instance, algae and PLA based scaffolds such as ulvan-PLA scaffolds are used in the drug delivery systems, wound dressings and tissue engineering [70,71]. The composite device which contains antibacterial properties made of calcium alginate hydrogel and PCL to prevent the bacterial infection in the implanted area of the organ [70]. The

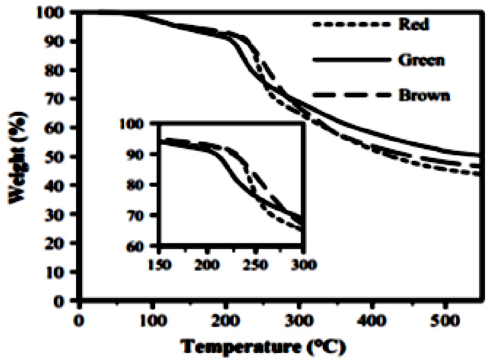


**Figure 4**. TGA (Thermogravimetric analysis) curve of red, green and brown algae flakes of 200-400 μm [73].

absorption of drug by the implanted area are warranted as a higher rate of drug absorption and thus, the potential PLA-algae composites can be a good choice for the drug delivery system and wound dressings [70,72].

### Adhesion of PLA with different types of algae

Algae as a filler (red, brown and green) with PLA matrix has been showed different types properties in terms of the concentration and size of algae in the composites. For example, the Young Modulus of PLA-algae composite which comprises algae of 40 percentage weight(for all red, green, and brown algae) has increased as compared to the Modulus of the neat PLA; though elongation at break and tensile strength has been reduced as shown in Table 2. Larger flakes of all types of algae combined with PLA showed improved mechanical properties as compare to the smaller flakes [42] (Figure 4).

### Mechanical properties of PLA-algae composite

In the PLA-algae polymer composite, PLA is acting as a matrix, and algae is used as a filler. However, Bulota et al. investigated dynamic mechanical properties in their study. They used red, brown, and green algae to produce PLA-algae composite through the melt mixing process. Basically, the size of algae flakes could effect on filler and matrix bond, thus it has influences on having better mechanical performance [73]. On the other hand, the variation of the algae by its type has a little or negligible effect on the mechanical properties of PLA-Algae composite. Though the flakes of algae were treated with the salts. The PLA-Algae made of 40% algae and 60% PLA had the better Young Modulus though the breaking strength and tensile strength of

**Table 3.** Mechanical properties of PLA based composites with different types of natural fibers.

| Fiber Type | Fiber wt. % | Tensile Strength (%) | Young Modulus (%) | Reference |
|---|---|---|---|---|
| Artificial Cellulose | 30 | 175 | 1.5 | [72] |
| Jute | 30 | 285 | - | [75] |
| Abaca | 20 | 104 | 170 | [76] |
| Flax | 30 | 121 | 202 | [77] |
| Kenaf | 70 | 286 | 492 | [78] |
| Wood flour | 30 | 92 | 196 | [79] |
| Cordenka1 | 25 | 157 | 146 | [79] |

**Table 4.** Glass transition ($T_g$) and Melting (Tm) temperature of PLA-Epoxy composite and PLA-Epoxy-algae composite [81].

| | Method | $T_g$ (°C) | $T_{cc}$ (°C) | Enthalpy (mJ) | $T_{m1}$ (°C) | Enthalpy (mJ) | $T_{m2}$ (°C) | Enthalpy (mJ) |
|---|---|---|---|---|---|---|---|---|
| PLA-EPO | | 50.62 | 102.91 | 18.86 | 145.18 | -114.25 | 154.99 | -125.39 |
| PLAEPO-Algae | Solvent casting | 52.90 | 121.66 | 23.41 | 150.19- | -94.62 | 156.04 | -100.82 |
| PLAEPO-algae | Melt blending | 56.91 | 86.69 | 136.66 | 134.12 | -142.11 | 147.96 | -150.37 |

the composite were decreased when 30% algae and 70% PLA were used to produce PLA-algae composite as shown in Table 3 [73-79].

### Thermal properties and challenge on adhesion between algae and PLA

The previous study on Polybutylene succinate (PBS)-algae composites showed that two melting peaks during the melting process of PBS-Algae composite in DSC (Differential Scanning Calorimetry)[80]. However, crystallization of PBS-Algae was declining in the DSC test and this is because algae resisted the crystallization of PBS-Algae composite [52]. Thus, the lower degree of crystallinity leads to a lower arrangement in the polymer chain of the composite. Although it is not proved that a lower degree of crystallinity leads a lower melting temperature, however; it is expected that the melting temperature of PLA-Algae composite will be lower as compared to the other natural fiber reinforced composites and will have poor thermal [melting] properties as shown in Table 4.

## Conclusion

The composite polymers which are biodegradable are economically competitive with synthetic polymers. The PLA-Algae polymer could exhibit somewhat poor tensile strength with a lower glass transition temperature. However, the biodegradability of these algae-reinforced polymer composites shows an improved biodegradation rate as it will take far less time to be degraded in the environment as compared to the polyester polymers. Moreover, the improvement of the adhesion between PLA and algae can make this composite practicable in the field of the automotive industry and packaging industry with the negligible mutilation of the environment. Further research should work on reducing moistures or water absorbency as well as improving adhesion between filler and matrix of PLA-algae composite to improve the mechanical properties of the composite. Algae-derived bioplastic which comprises PLA offers immense protection of the environment as the high yield production of algae with its tremendous physical and mechanical properties. Algae oil or agar are altered into a novel monomer for the production of polyester-based composites (e.g., PLA-algae composite), which can minimize the use of petrochemical sources and thus; reduces the carbon emission from the power plants. Moreover, the monomeric construction of algae can be exploited to produce scaffolds in the field of biomedical application. Therefore, PLA-algae scaffolds have the potential in the field of tissue engineering, wound dressing, and drug delivery system.

## Declarations

**Funding:** This research received no external funding.

**Conflicts of Interest/Competing interests:** The authors declare no conflict of interest.

**Availability of Data and material:** Data are transparent

**Code availability:** N/A

**Author Contributions:** This study was conceptualized and written by the first author. The second author contributed to prepare the figures, tables and collected literature.

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**How to cite this article:** Bulbul Ahmed and Shimo Sultana. A Critical Review on PLA-Algae Composite: Chemistry, Mechanical, and Thermal Properties. *J Textile Sci Eng* 10 (2020) doi: 10.37421/jtese.2020.10.425