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ORIGINAL ARTICLE

Potential of microalgae Bio-Coke as a sustainable solid fuel alternative to coal coke

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Abstract

Energy and environmental issues have triggered the search for new sources of green energy alternatives in recent years. Biofuel production from renewable sources is widely considered one of the most sustainable alternatives for environmental and economic sustainability. Microalgae are currently being promoted as one of the most promising liquid biofuel feedstocks due to their rapid growth, high lipid production capacity, and carbon–neutral cycle. In this study, whole microalgae cells were utilized as raw material to produce solid biofuel, i.e., Bio-Coke, and this study aimed to investigate the possibility of microalgae Bio-Coke as an alternative to coal coke. The results show that Bio-Coke can be produced from microalgae in the temperature range of 80–100 °C. The apparent density is between 1.253 and 1.261 g/cm^3, comparable to the apparent density of lignocellulosic Bio-Coke. Additionally, the calorific value is higher than the calorific value of lignocellulosic Bio-Coke and within the range of the calorific value of subbituminous coal. Therefore, microalgae Bio-Coke can be utilized to replace coal coke usage in the future.

Keywords Bio-Coke · Microalgae · *Euglena gracilis* · Solid fuel · Coal coke

Introduction

Sustainable energy is required to meet the need for energy without neglecting future generations, as reflected in the Sustainable Development Goals (SDGs) program. However, despite the urge to search for more sustainable energy sources, fossil fuel remains the largest share of the energy mix, with oil contributing up to 31.2%, while coal is the second largest fuel, accounting for 27.2% of total primary energy consumption [1]. Meanwhile, the share of renewable energy rose to record highs but only accounted for 5.7% of total consumption [1]. The dirtiest of fossil fuels, coal, is responsible for 30% of global emissions and a major contributor to the air pollution that has taken a devastating toll on the health of local populations. The Glasgow climate summit (COP26) produced a new global pact committing governments to take increasingly ambitious steps to address the climate crisis. It was announced that 190 countries and organizations committed to phasing out coal power in the 2030s for Organization for Economic Co-operation and Development (OECD) countries and in the 2040s (for non-OECD). Therefore, the development of new technologies

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that can deliver fuel from renewable resources is indispensable. Among the renewable energy resources available, microalgae represent a potential source. Microalgae, which are third-generation feedstocks, show a promising future as biofuel stocks due to their features.

High lipid content in microalgae makes them suitable for energy use [2]. With their significant potential, microalgae are expected to surpass first- and second-generation feedstocks. The conversion of microalgae into liquid biofuel has received much attention. Several mechanical, chemical, and enzymatic techniques have been developed for lipid extraction. However, the cell disruption process, including lipid extraction followed by trans-esterification, is costly, produces a high volume of waste, and is considered a major bottleneck [3]. Furthermore, the oil part of algae biomass is approximately 30%, and the remaining 70% is an algae byproduct [4]. Therefore, the conversion of microalgae into solid biofuel or Bio-Coke is an alternative conversion of microalgae into biofuel in a less expensive way without residue. Solid biofuel can be a good alternative to conventional solid fossil fuels dominated by coal.

Lignocellulosic biomass has already been recognized as an alternative to solid fossil fuels, especially coal. Pelletizing and briquetting are well-known densification treatments to produce solid biofuel from lignocellulosic biomass. However, there is a need to improve their ability to burn at high temperatures for longer combustion times. Bio-Coke is a new densification method that aims to overcome these problems. Bio-Coke exhibits moderate calorific value and good mechanical strength. This method shows almost 100 percent yield on a weight basis [5, 6]. A comprehensive study showed that Bio-Coke is superior to pellets and briquettes due to its increased density and compressive strength at both room temperature (25 °C) and high temperatures (700 °C); furthermore, Bio-Coke demonstrates an ability to burn at high temperatures for a longer combustion time compared to pellets [6].

Bio-Coke produced from various lignocellulosic biomass wastes in Malaysia had apparent densities ranging from 1.255 to 1.438 g/cm^3[7, 8], while apparent densities of biomass pellets ranged between 0.8 and 1.2 g/cm^3 [9]. Another superior characteristic of the Bio-Coke is that its combustion duration was 1300–2081 s. Nevertheless, since the calorific value depends on the raw material, the calorific value of lignocellulosic Bio-Cokes is similar to that of the pellets and briquettes [6]. A study shows that the calorific values from various lignocellulosic Bio-Cokes from abundant waste in Malaysia were 16.48–19.15 MJ/kg [7].

In Japan, where Bio-Coke densification technologies were developed, Bio-Cokes have been manufactured from a wide variety of biomass wastes for various project purposes. They have been applied as fuel in boilers or waste incinerators [6, 10]. Bio-Coke is also comparable to coal coke. Bio-Coke can be mixed with coal coke in metal casting as a heat source as well as a carburizer [5]. Moreover, this Bio-Coke can be further carbonized to produce carbonized Bio-Coke in a short time [11]. This carbonized Bio-Coke is comparable to biochar, which can substitute pulverized coal applied in pulverized coal power plants [12].

However, it is widely known that the grinding process of lignocellulosic biomass prior to solid fuel production is expensive due to high energy consumption, accounting for approximately 33% of the entire electricity requirement of the process [13]. Energy consumption is typically dependent on the type of lignocellulosic biomass used. A study reported that softwoods like corn stover and switchgrass require 11.0 and 27.6 kWh/metric ton for size reduction, whereas hardwoods like pine and poplar chips require 85.4 and 118.5 kWh/metric ton, respectively [14].

Several experiments have been performed to utilize microalgae as a binding agent in solid biofuel production from lignocellulosic material. The results indicate a better strength of the pellet with the addition of up to 30% weight microalgae [15, 16]. An experiment to utilize 100% microalgae as raw material to produce fuel pellets was also conducted. The results show that algal pellets verified all standards used as fuel in biomass boilers [17]. Nevertheless, there has not been any study to apply microalgae as the raw material for Bio-Coke production.

In this study, *Euglena gracilis*, a species of microalgae, was utilized as a raw material for Bio-Coke production. *E. gracilis*, a photosynthetic protist, produces protein, unsaturated fatty acids, wax esters, a unique β-1,3-glucan called paramylon, and other valuable compounds [18]. Under anaerobic conditions, wax esters become the main storage compounds in *E. gracilis* [19]. These esters are composed of medium-chain fatty acids and fatty alcohols, which can be converted into biofuels [20]. *E. gracilis* used in this study was in the form of dry powder as received from the producer company. Microalgae biomass is generally harvested at cell sizes between 3 and 30 µm [21]. This fine powder form is advantageous because it provides a greater number of contact points, a larger exposed surface area, and a higher surface energy per unit weight regardless of the physical and chemical properties [22]. This powder form was one of the advantages of microalgae as the raw material over lignocellulosic material. It is widely known that the grinding process required to produce raw uniform material is the most expensive, time-consuming, and energy-intensive process involved in the preparation of lignocellulosic solid biofuel. All the Bio-Coke production was conducted at a loading pressure of 21.7 MPa. A high loading pressure generally resulted in the formation of a solid bridge via the diffusion of molecules between particles, increasing the density [6, 22]. The moisture contents of the raw materials were 7% ± 0.25%. Initial moisture content is a significant factor in Bio-Coke production because it influences the solid fuel's calorific value, combustion efficiency, and mechanical durability [23]. A study of solid biofuel production from major lignocellulosic biomass suggested that a low moisture content of approximately 5% to 10% resulted in denser and more durable solid biofuel than a high moisture content (15%) [24].

Previous studies of Bio-Coke production from lignocellulosic biomass suggested formation temperatures between 120 and 180 °C [7]. However, for microalgae biomass, several attempts at formation temperatures above 100 °C resulted in high backpressure in the equipment and produced cracked Bio-Coke. Hence, the formation temperatures were set between 40 and 100 °C. After the target temperature was achieved, samples were held for 6 min and 15 s. The retention times between 5 and 10 min produced Bio-Coke with better compressive strength [25]. A small 20 mm diameter molding was chosen for this study. The previous experiments indicated that for a diameter less than 20 mm, homogeneous heat permeation occurred along the sample's cross-section, resulting in the homogenous adhesive force along the region [26].

This study aims to evaluate the feasibility of *E. gracilis* in the production of Bio-Coke. The conversion of microalgae into solid Bio-Coke can be an advantage compared with wood biomass since grinding is not required, resulting in the production cost being less than Bio-Coke made by wood biomass. Furthermore, the microalgae Bio-Coke characteristics, including the apparent density, compressive strength, calorific value, and combustion characteristics, were evaluated to show the possibility of utilizing microalgae as solid fuel replacement for coal coke in the future.

Materials and methods

The Bio-Coke production process included material preparation, loading, pressurizing while simultaneously heating at the set temperature, and cooling to room temperature [11]. After finishing the production process, the product was analyzed to evaluate the Bio-Coke characteristics.

Source and preparation

E. gracilis, a species of microalgae, was used as raw material for Bio-Coke production. The dewatering and drying processes were conducted at Euglena Co. The cells are concentrated by ultrafiltration to a 50-fold concentration (PELLICON, Merck Millipore Corp., Darmstadt, Germany). After ultrafiltration, the concentrated culture liquid is centrifuged at 3500*g* for 5 min to retrieve the cells. The pelleted cells are then resuspended in deionized water to 15–20% solid content to remove the growth medium components from the collected *E. gracilis* cells. Using a spray drier (SD-1000, EYELA, Tokyo, Japan) with a constant inlet air temperature of 155 °C and an outlet air temperature of 95 °C, the concentrated suspension of *E. gracilis* is turned into a dried powder [27]. The powder is utilized directly in this study.

Components of *E. gracilis* powder are moisture contents 9.0% (w/w), crude proteins 62.8% (w/w), crude fat 13.9% (w/w), crude fiber 0.2% (w/w), crude ash 5.8% (w/w), soluble nitrogen-free substance 5.3% (w/w), and carbohydrates 3.0% (w/w).

Production of Bio-Coke

The Bio-Coke production process was conducted at temperatures ranging from 40 to 100 °C. The production process followed the method as described in U.S. Patent No. US 8211274 B2 [11]. *E. gracilis* powder was utilized as a raw material in Bio-Coke production. The initial moisture content of the dried *E. gracilis* powder was 7% ± 0.25 wt% as received from the source. 9 g of raw material was loaded into a 20 mm diameter molding barrel. After that, a pressure piston (SMP-3012B, Riken Kiki, Japan) was inserted into the molding barrel to apply a loading pressure of 21.7 MPa and simultaneously heated by an electric heating furnace (ARF-50 M, Asahi Rika, Japan) to the target temperature. The sample was held at the target temperature and constant pressure for 6 min and 15 s. Afterward, the sample was cooled to room temperature while maintaining a pressurized condition. The pressure was released after reaching room temperature, and the Bio-Coke sample could be removed from the molding barrel [11].

Apparent density measurement

Following the production of Bio-Coke, the weight, length, and diameter of the Bio-Coke produced were measured immediately before the spring-back effect occurred [28]. The apparent density of the Bio-Coke can be determined using the data collected by dividing the weight of the Bio-Coke by its volume [29].

Compressive strength test

The compressive strength test was based on the method described in previous research to measure the strength of the solid biofuel [26, 30]. It is conducted using a compression testing machine at room temperature (SHIMADZU, AG–X plus, 300 KN, Japan). The maximum compressive strength of the Bio-Coke samples was determined using Trapezium X universal testing software.

The compressive strength in simple pressure was measured by placing the Bio-Coke sample vertically in the center of the machine. Following that, the sample was loaded at a rate of 2.5×10^5 m/s. The test was finished when the sample cracked, and the maximum load force recorded at that moment was designated to determine the maximum compressive strength of Bio-Cokes [26].

Thermogravimetric analysis

To determine the weight change of the sample as a function of temperature in a controlled environment, thermogravimetric analysis (TGA) was performed using a Q50 thermogravimetric analyzer from TAInstrument, USA. The measured weight loss curve produced from the analysis gave information regarding changes in sample composition and thermal stability. Approximately 5 mg of Bio-Coke samples were placed in platinum crucibles and heated from 30 to 1000 °C at a rate of 10 °C/min using nitrogen (flow rate 60 mL/min) as the purge gas [31].

Calorific value analysis

Calorific value analysis aimed to measure the calorific value of the Bio-Coke sample. It was conducted using a bomb calorimeter (CAL2K-ECO, DDS Calorimeter, South Africa). The measurement followed the instruction manual from the manufacturer. The sample was weighed and placed into a crucible (CAL2K-4-CB). Afterward, the crucible was inserted into the external electrode's crucible holder with firing cotton (CAL2K-4-FC) touched the sample while making contact with the electrode's firing wire (CAL2K-4-FW). Afterward, the electrode assembly was inserted into the vessel body (CAL2K-ECO-4). The vessel was then filled with oxygen to a pressure of 3000 kPa. The vessel was then placed in the bomb calorimeter, and the calorific value was determined. The calorimeter measures the temperature rise of the vessel with constant mass, volume, and pressure in an isothermal environment. The temperature rise is proportional to the energy released, which will yield the specific energy as the weight is known.

Results and discussion

Bio-Coke production by E. gracilis powder

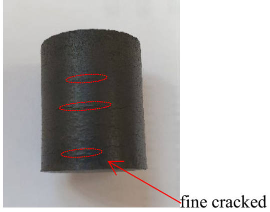
Figure 1 shows various Bio-Coke samples produced at different formation temperatures. From Fig. 1, some fine cracks were present on the surface of the Bio-Coke formed at 40 °C. Meanwhile, the surfaces of the Bio-Cokes produced at formation temperatures between 60 and 100 °C were very smooth. This is attributed to the denaturation of protein. Denaturation can be induced by heating, as the process is endothermic at elevated temperatures. Higher denaturation temperatures provide more denatured protein, resulting in the formation of new bonds and structures with other proteins, lipids, and starches, thereby aiding in enhancing binding capacity [32].

This result implied that Bio-Coke formation could occur with *E. gracilis* as the raw material despite the usual lignocellulosic biomass with lignin acting as a binder. Bio-Coke formation is achieved by applying mechanical force to the particles to create inter-particle bonding. The binding forces between particles may perform through bonding with a solid bridge between particles [33]. Lignin helps build solid bridges at elevated temperatures and plays a significant role in biomass densification. The presence of lignin in plant materials allows densification without adding binders [22]. Lignin has thermosetting properties at temperatures above 140 °C and acts as an intrinsic resin, resulting in more durable Bio-Coke. Higher lignin levels produce more durable Bio-Coke because lignin acts as the adhesive that holds particles together [34].

In this study, Bio-Coke was produced from microalgae biomass in the absence of lignin. This is attributable to the protein content in *E. gracilis,* which binds loose material together, forming the bridge and filling the void [35]. Proteins are heated during the Bio-Coke production process. Heating alters protein structure and causes the breakdown of the spatial three-dimensional structure of the protein. Next, a new covalent bond is formed, such as disulfide bridges, isopeptide bonds, and complex bonds formed in Maillard reaction products. The first step involves breaking hydrogen and van der Waal bonds, whereas the second step involves the breaking or formation of covalent bonds. Covalent bonds are the strongest chemical bond; therefore, the denaturation process during Bio-Coke production enhances the protein's ability to bind [36].

The production of Bio-Coke from microalgae may imply that one of the origins of coal might be the accumulation of photosynthetic microorganisms underground for long periods. This is in line with the theory of boghead coal originating from algae. While this theory is widely accepted, some scholars dispute it [37]. In the boghead coals, suspended matter consists chiefly of gelosic algae. Based on the similar structure, appearance, and characteristics of the yellow bodies in the boghead series, it is concluded that they originated from colonial algae [38]. The yellow bodies showed close agreement with *Botryococcus braunii* [39]. Boghead coal deposits have been discovered in Alaska, Australia, France, Scotland, South Africa, and the United States. A type of boghead coal was also reported from Illinois, although it is not a true boghead coal because it contains a significant amount of anthraxylon mixed with algal colonies, indicating a type of transitional coal towards normally banded coal [40].

Fig. 1 Images of the Bio-Coke samples

Formation Temperature	Cross-Section	Side View
40 °C		
60 °C		
80 °C		
100 °C		

Physical features of the microalgae Bio-Cokes

Following the production of the Bio-Coke, the weight, length, and diameter of the Bio-Coke produced were measured immediately. This information is used to calculate the yield and the apparent density of the Bio-Coke produced. In agreement with previous research, the conversion of 9 g *E. gracilis* dried powder into Bio-Coke resulted in an average 99.7% yield on a weight basis.

The apparent densities of the Bio-Cokes produced are listed in Table 1. One-way analysis of variance (ANOVA) showed no significant differences between the apparent density of Bio-Coke made in the temperature ranging from 60 to 100 °C. Meanwhile, the apparent densities of the Bio-Coke samples formed at 40 °C were significantly lower than the apparent density of the Bio-Coke produced in the temperature range between 60 and 100 °C.

Under this formation condition, the apparent density of microalgae Bio-Cokes is comparable to the apparent density of several lignocellulosic Bio-Cokes, which are between 1.255 and 1.438 g/cm^3 [7]. The values are still higher than the apparent densities of biomass pellets, which are 0.8–1.2 g/cm^3 [8]. High apparent density is favorable for storage handling and transportation to save space. Moreover, it indicates a high energy content per volume and provides slow-burning characteristics [41].

Compressive strength value at room temperature indicates the resistance of Bio-Coke samples stacked on top of one another to determine the potential damage to this solid fuel during storage, transportation, and handling [30]. Table 1 shows the maximum compressive strength of the Bio-Coke formed in the temperature range between 60 and 100 °C. It is observed that the maximum compressive strength of the microalgae Bio-Cokes formed at temperatures of 60–100 °C are comparable to biomass pellet and coal coke [30, 42]. However, the maximum compressive strength of the Bio-Coke formed at 60 °C is only 8.40 ± 0.56 MPa. This value is almost half of the compressive strength of the Bio-Cokes produced at 80 and 100 °C, which are 15.49 ± 0.69 MPa and 16.54 ± 0.64 MPa, respectively. This is due to the protein denaturation process. Higher formation temperatures, i.e., 80 °C and 100 °C,

Table 1 Comparison of the apparent density, compressive strength, temperature ranges, and weight loss of the Bio-Coke samples in the thermogravimetric analysis

Formation temperature (°C)	Apparent density (g/cm^3)	Compressive strength (MPa)	Thermogravimetric analysis		
			Phase	Temperature range (°C)	Weight loss (%)
40	1.2223 ± 0. 0051	n/a	First	30–140	7.40
			Second	140–500	67.55
			Third	500–910	23.14
60	1.2566 ± 0.0048*	8.40 ± 0.56	First	30–125	7.23
			Second	125–500	56.43
			Third	500–950	27.95
80	1.2534 ± 0.0022*	15.49 ± 0.69	First	30–125	6.46
			Second	125–500	56.43
			Third	500–1000	25.51
100	1.2606 ± 0.0069*	16.54 ± 0.64	First	30–120	3.30
			Second	120–500	57.08
			Third	500–1000	22.73

*$p < 0.005$ against Bio-Coke at 40 °C, $n = 3$

provide an irreversible change leading to stronger new bonds and structures [43].

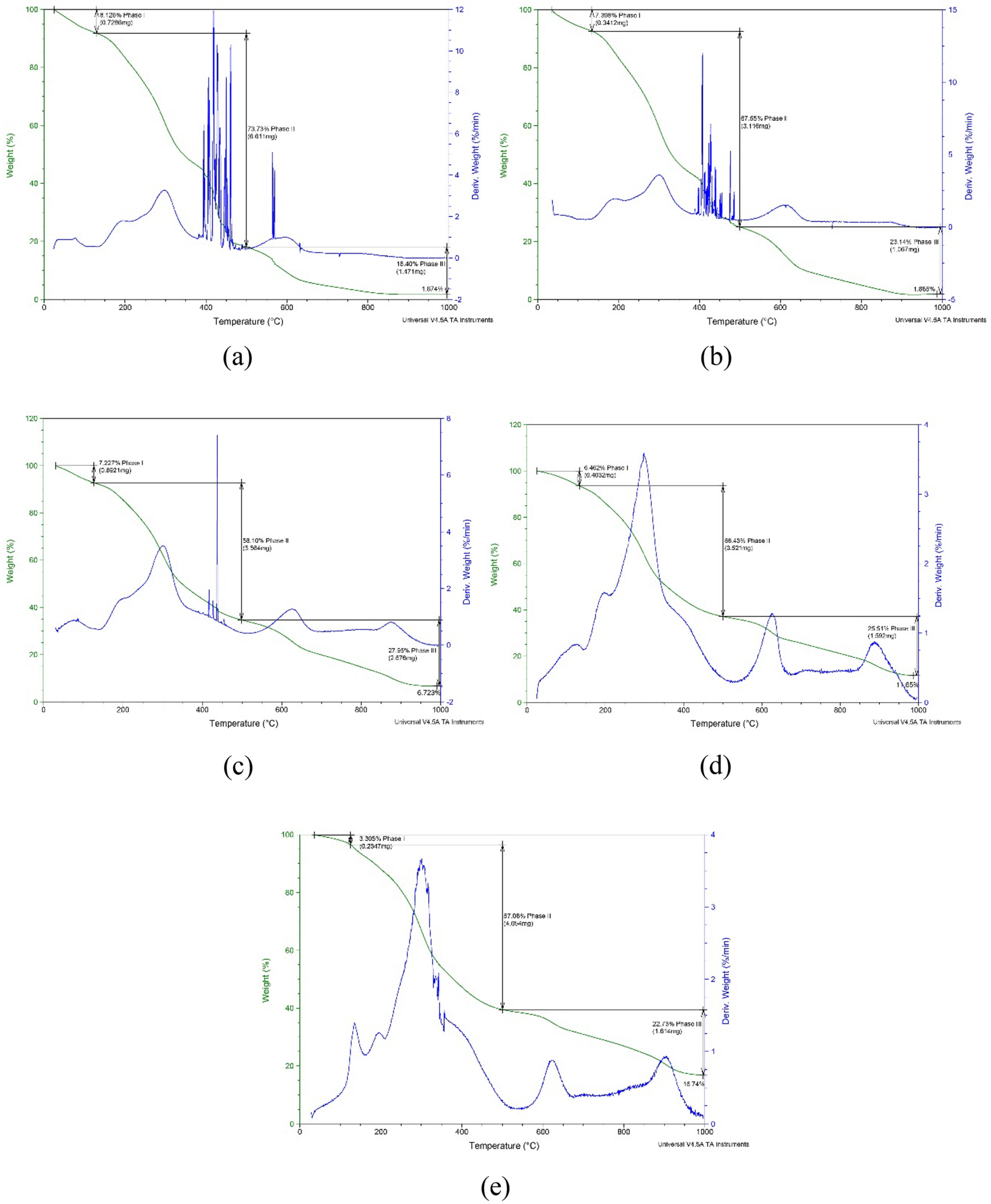
The formation temperatures of microalgae Bio-Coke were lower than the formation temperature of lignocellulosic Bio-Coke, which is 115–230 °C [11]. This is attributed to the different compositions of the materials. *E. gracilis*, as well as other species of microalgae, are mainly composed of carbohydrates, proteins, and lipids. These components are less thermally resistant than lignocellulosic biomass main components, i.e., cellulose, hemicellulose, and lignin [44]. This low formation temperature is an advantage of microalgae Bio-Coke over lignocellulosic Bio-Coke, specifically in the energy consumed for solid biofuel production, directly resulting in a lower operational cost.

Thermodynamics features of the microalgae Bio-Cokes

Figure 2 shows the TG/DTG curves of Bio-Coke made from *E. gracilis* at different temperature formations. The decomposition of microalgae Bio-Coke is divided into three main temperature zones. The first zone, which occurs until approximately 120 °C, is the reduction of moisture content and release of volatile matter with low molecular weight. The second zone, which extends to 500 °C, includes the degradation of carbohydrates and proteins. Furthermore, the third zone is due to the decomposition of carbohydrates and lipids, which is mainly associated with the breakdown of hydrocarbon chains of fatty acids [44]. A detailed observation shows that different DTG profiles were seen at 400–500 °C. The different profiles in this range were due to the denaturation process during the production of Bio-Cokes in different temperatures.

Figure 2 shows that the decomposition of *E. gracilis* powder was finished at approximately 800 °C, while the decomposition of Bio-Cokes formed at 40 °C and 60 °C was finished at approximately 850 °C and 900 °C, respectively. Meanwhile, the decomposition of the microalgae Bio-Cokes formed at 80 °C and 100 °C at 1000 °C has not yet finished, as the graphs show that the weight is still decreasing, although the rate of the decreasing weight starts to decline at 900 °C. This implied that these microalgae Bio-Cokes are thermally stable at high temperatures. This is crucial for high-temperature combustion, such as in metal casting or high-temperature gasifying and direct melting systems, which can reach a temperature of 700–1500 °C.

Denaturation is considered a two-stage process; the first involves reversible changes caused by the breakdown of hydrogen and van der Waals bonds, and the second involves irreversible changes caused by the breakdown or formation of covalent bonds such as disulfide bridges. A higher formation temperature provides a greater chance to achieve the two-stage processes that result in irreversible changes, while a lower temperature might only result in reversible changes. This process explains the DTG graph similarity from Bio-Coke formed at low temperatures, i.e., 40 °C and 60 °C, to the DTG graph from *E. gracilis* powder where many split peaks are observed, implying a fluctuating rate of decreasing weight, resulting in stiff slope in TGA curve within the temperature range of 400–450 °C. Meanwhile, higher formation temperatures, i.e., 80 °C and 100 °C, are already irreversible, leading to the formation of new bonds and structures with other available proteins, lipids, and starches, thereby changing the rate of decreasing weight profile, creating less stiff slope in TGA curve within the temperature range of 400–450 °C compared to that of low formation temperature.



(a)

(b)

(c)

(d)

(e)

Fig. 2 TG/DTG curves of **a** *E. gracilis* powder and Bio-Coke from *E. gracilis* at formation temperatures of **b** 40 °C, **c** 60 °C, **d** 80 °C, and **e** 100 °C

The calorific values of Bio-Cokes made from *E. gracilis* at formation temperatures of 80 °C and 100 °C were 22.72 ± 0.03 MJ/kg. The value was higher than that of several lignocellulosic Bio-Cokes, which were in the range of 16.48–19.15 MJ/kg, comparable to that of lignites, the lowest grade of coal [7, 43]. Meanwhile, the calorific value of algae Bio-Coke is comparable to that of subbituminous coal, which is in the range of 19–24 MJ/kg [43]. This suggests that *E. gracillis* is a potential alternative feedstock for Bio-Cokes and a prospective coal replacement.

Economic and environmental impact of Bio-Cokes application

Over the past 15 years, the percentage of coal as the world energy resource has been around 28–29% [1]. Coal is the third-highest supplier of world energy. Carbon dioxide emission from burning coal contributes about 44% of total CO_2 emissions globally [45]. Solid biofuels have been introduced as a supplement to fossil coal that helps reduce harmful coal emissions in industrial and household applications. Biomass pellets and biomass briquettes have been popularly applied as solid fuels in household heating and regional heat supply in Europe and the United States. In the industrial field, the co-firing of biomass pellets and briquettes in the industrial sector, such as power plants and large utility coal boilers, has been applied in Europe, Japan, and Korea [46].

Based on the Life Cycle Inventory modeling approach, a study in China shows that the fuel-to-heat energy consumption of wood pellets and coal was estimated to be 112.80 and 1495.11 MJ/GJ heat, respectively, indicating that switching from coal to wood pellets would result in energy savings of 1382.30 MJ per 1-GJ heat provision and reduction of 184 kg CO_2 eq/GJ [47]. Bio-Coke made from wood biomass shows superior characteristics over wood pellets and briquettes in terms of energy density and combustion time. However, the electric consumption to produce wood biomass Bio-Coke is higher than the wood pellet resulting in twice the CO_2 emission per kg Bio-Coke produced. Nevertheless, because of the high energy density, the production cost of wood biomass Bio-Coke per kg was only half the production cost of the pellet, with the selling price of Bio-Coke (0.37 $ per kg) slightly higher than the pellet (0.34 $ per kg) [48]. The production cost reflects the raw material cost, transportation, and energy consumption. Notably, wood biomass Bio-Cokes can be used in cupola furnaces as an alternative to coal coke.

The production process of wood biomass Bio-Cokes is the same as that of Bio-Cokes made from *E. gracilis*. The difference was in the first steps, whereas grinding was necessary for raw material of wood biomass Bio-Cokes. However, raw material for wood biomass Bio-Cokes was collected with a fee for disposal; hence it is considered revenue. Meanwhile, there is an estimated cost of 0.048 $ per kg for the dewatering and drying process of *E. gracilis* as the raw material for microalgae Bio-Cokes [49].

Furthermore, the energy consumption in the production process of *E. gracilis* Bio-Cokes was lower compared to wood biomass Bio-Cokes due to lower production temperature. Since the estimated heat capacity of the wood chips and *E. gracilis* are almost the same, i.e., 1.76 J/g/C [50, 51], the energy consumption for the production of *E. gracilis* Bio-Cokes at 80 °C was 0.53 times the energy consumption for the production of wood biomass at 150 °C. Table 2 compares estimated wood biomass Bio-Cokes and microalgae Bio-Cokes production costs. A further estimated 33% cost of grinding can also be deducted from the production cost of microalgae Bio-Cokes. Hence, the total production cost of the microalgae Bio-Cokes is only a bit higher than that of wood biomass Bio-Cokes but still 47% lower than the total cost for wood pellet production, resulting higher profit margin compared to wood pellet [48]. Additionally, since the calorific value of microalgae Bio-Cokes, is approximately 1.22 times higher than wood biomass Bio-Cokes, the quantity of microalgae Bio-Cokes required for the equivalent amount of energy generated is equal to 0.85 wood biomass Bio-Cokes. With the coal price fluctuating between 0.28 and 0.38 $ per kg in 2022, Bio-Coke is price competitive and prospective to replace coal and coal coke in the future.

Further study on the wood biomass Bio-Coke usage to replace coal coke indicated that 108.2 kg CO_2/GJ are produced for coal coke and 54.4 kg CO_2-eq/GJ for wood biomass Bio-Coke; therefore, a GHG reduction of 53.8 kg CO_2-eq per/GJ could be achieved by substituting wood biomass Bio-Coke for coal [48]. Based on the energy

Table 2 Comparison between estimated wood biomass Bio-Cokes and microalgae Bio-Cokes production costs

	Wood biomass Bio-Cokes	Microalgae Bio-Cokes
Cost of raw material ($/kg)	− 0.13 [48]	0.048 [49]
Cost of production ($/kg)	0.20 [48]	0.11*
Cost of grinding ($/kg)	Include in the production cost	− 0.066 [13]
Total cost ($/kg)	0.07	0.09
Total cost for the equivalent amount of energy generated	0.07	0.08

*Based on estimated energy consumption for microalgae Bio-Cokes production at 80 °C consumption per kg of wood biomass and microalgae Bio-Cokes produced, it can be assumed that the CO_2 emission is comparable. Given the amount of microalgae Bio-Cokes required to generate the same amount of energy equivalent to 0.85 wood biomass Bio-Cokes, it leads to a further decrease of 8.16 kg CO_2-eq/GJ.

Advantages and reliability of Bio-Coke production using microalgae under a tropical climate.

Microalgae are rapidly growing photosynthetic organisms that have the potential to convert 8–10% of solar energy (average sunlight irradiance) into biomass with a theoretical yield of approximately 77 g biomass/m^2/day or approximately 28 kg/m^2/year [52]. Nevertheless, under ideal growing circumstances, lab-scale sustained green microalgae productivity has never exceeded 20–40 g biomass/m^2/day [52].

Numerous studies have been conducted to investigate the production method for large quantities of microalgae biomass, including for *E. gracilis*. Since 2005, Euglena Co. has successfully developed large-scale outdoor cultivation for *E. gracilis*. The estimated theoretical maximum production of *E. gracilis* was approximately 48 g/m^2/day or 17 kg/m^2/year, although actual outdoor cultivation resulted in a maximum proliferation rate of only 25 g/m^2/day or 8.85 kg/m^2/year. This value is 2.5 times higher than Malaysia's biomass waste production per year, which reaches approximately 3.5 kg/m^2/year [53].

Microalgae use a similar mechanism to terrestrial plants, but due to their basic cell structure, high surface-to-volume-body ratio, and the fact that they are submerged in water, the mechanism is efficient at converting vast amounts of water, CO_2, and other nutrients into biomass [54]. Therefore, successful microalgal cultivation generally requires light, carbon, various macronutrients, and several micronutrients. Microalgae use light to conduct the photosynthesis process. Light, one of the essential elements in microalgae growth, may be supplied naturally or artificially. Nevertheless, a study suggested that the application of artificial light would increase the cost of production by US$ 25.3 per kilogram of dry-weight biomass [55].

Current commercial large-scale cultivation of *E. gracilis* is located at Ishigaki Island in Okinawa prefecture, Japan. The reference solar radiation intensity is approximately 18 MJ/m^2/day. It is required to discover other sites for larger-scale production of *E. gracilis*. The requirement of such land candidates includes the availability of operational land area (at least 3000 ha), climate (at least 3 h of average daily sunlight, 500 mm of average annual rainfall), and country risk (low level of crime and terrorism) [27].

Countries in tropical regions, such as Malaysia, Indonesia, and Thailand, offer benefits for microalgae cultivation. Located at the equator, the tropical zone provides the highest solar irradiances, ensuring uninterrupted and evenly distributed light necessary for photosynthesis. Tropical countries also provide high rainfall to reach the standard required by *E. gracilis*. Furthermore, tropical microalgae can grow throughout the year compared to microalgae in temperate and subtropical environments. Temperatures below 15 °C generally impede microalgae growth, while temperatures above 35 °C are lethal for some species. The majority of microalgae species are tolerant to temperatures ranging from 15 to 27 °C [54].

Therefore, the cultivation of microalgae together with Bio-Coke conversion of microalgae in tropical areas might show good advantages for solid fuel production.

Conclusion

This study implied the possibility of the future prospects of microalgae Bio-Coke made by *E. gracilis*, a microalgae species, as an alternative to coal coke. Tropical regions bring beneficial features for microalgae as they provide uninterrupted and well-distributed light to support the continuity of the photosynthesis process. Although microalgae do not contain lignin, which usually acts as a binder in Bio-Coke production, proteins in microalgae can replace this function, implying that Bio-Coke can be produced from all types of biomasses. The apparent density of this microalgae Bio-Coke is comparable to the apparent density of the lignocellulosic Bio-Coke. Apparent density is beneficial for storage and transportation as well as its physical performance in the combustion process. The combustion of the produced Bio-Coke could reach a temperature of more than 1000 °C, and the calorific value is comparable to that of subbituminous coal.

Moreover, the possibility of the formation of Bio-Coke by microalgae may imply that one of the origins of coal coke might be due to the accumulation of photosynthetic microorganisms underground for a long time because the pressure and temperature underground at 1000–2000 m could be similar to the conditions of the formation of Bio-Coke by microalgae in this study.

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Author contributions VCK, TI, and HH designed the research; VCK, SN, and NSK performed the experiments; WJY, KS, KI, and NAO prepared and provided materials, equipment, and analysis result, VCK, KS and HH wrote the manuscript; MAMY, FNMA, and NAO helped in the preparation of manuscript.

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Data availability All data generated or analysed during this study are included in this published article (and its supplementary information files).

Declarations

Conflict of interest The authors declare that they have no competing interests.

References

1. BP (2021) BP Statistical Review of World Energy 2021. https://www.undp.org/content/undp/en/home/sustainable-development-goals/goal-7-affordable-and-clean-energy.html
2. Tan WMK, Lee YK (2016) The dilemma for lipid productivity in green microalgae: importance of substrate provision in improving oil yield without sacrificing growth. Biotechnol Biofuels 9(255):1–14. https://doi.org/10.1186/s13068-016-0671-2
3. Patel A, Mikes F, Matsakas L (2018) An overview of current pretreatment methods used to improve lipid extraction from oleaginous micro-organisms. Molecules 23(7). https://doi.org/10.3390/molecules23071562
4. Alam F, Mobin S, Chowdhury H (2015) Third generation biofuel from algae. Proc Eng 105:763–768. https://doi.org/10.1016/j.proeng.2015.05.068
5. Mizuho Information and Research Institute Inc. (2015) FY 2014 Project for Promoting the Spread of Technologies to Counter Global Warming (Feasibility Study of Biocoke technology JCM project in Thailand). Mizuho Information and Research Institute, Inc.
6. Kamal Baharin NS, Cherdkeattikul S, Kanada N, Hara H, Mizuno S, Sawai T, Fuchihata M, Ida T (2022) Impact and effectiveness of Bio-Coke conversion from biomass waste as alternative source of coal coke in Southeast Asia. J Mater Cycles Waste Manag. https://doi.org/10.1007/s10163-022-01539-x
7. Kamal Baharin NS, Koesoemadinata VC, Nakamura S, Yahya WJ, Yuzir MAM, Md Akhir FN, Iwamoto K, Na O, Ida T, Hara H (2020) Conversion and characterization of Bio-Coke from abundant biomass waste in Malaysia. Renew Energ 162:1017–1025. https://doi.org/10.1016/j.renene.2020.08.083
8. Kamal Baharin NS, Koesoemadinata VC, Nakamura S, Azman NF, Muhammad Yuzir MA, Md Akhir FN, Iwamoto K, Yahya WJ, Othman Na, Ida T, Hara H (2020) Production of Bio-Coke from spent mushroom substrate for a sustainable solid fuel. Biomass Convers Biorefin. https://doi.org/10.1007/s13399-020-00844-5
9. Tumuluru JS, Sokhansanj S, Lim CJ, Bi T, Lau A, Melin S, Sowlati T, Oveisi E (2010) Quality of wood pellets produced in british columbia for export. Appl Eng Agric 26(6):1013–1020. https://doi.org/10.13031/2013.35902
10. Kakumazaki J, Kawabata H, Ota K, Ida T (2019) Demonstration of using Bio-Coke from mixed feedstock at general waste incineration facilities and cupola furnaces. J Jpn Inst Energy 98(12):340–346. https://doi.org/10.3775/jie.98.340
11. Ida T, Nakanishi A (2012) Solidified biomass and production method thereof US8211274US 8,211,274 B2
12. Tagami N, Mohd Nor F, Nakadate T, Namba K, Ida T (2016) A study of influence of the torrefaction processing on combustion properties of the powders Bio-Coke. J Smart Process 5(3):171–178. https://doi.org/10.7791/jspmee.5.171
13. Den W, Sharma VK, Lee M, Nadadur G, Varma RS (2018) Lignocellulosic biomass transformations via greener oxidative pretreatment processes: Access to energy and value-added chemicals. Front Chem 6:141. https://doi.org/10.3389/fchem.2018.00141
14. Rajendran K, Drielak E, Sudarshan Varma V, Muthusamy S, Kumar G (2017) Updates on the pretreatment of lignocellulosic feedstocks for bioenergy production—a review. Biomass Convers Biorefin 8(2):471–483. https://doi.org/10.1007/s13399-017-0269-3
15. Thapa S, Johnson DB, Liu PP, Canam T (2015) Algal biomass as a binding agent for the densification of Miscanthus. Waste Biomass Valorization 6:91–95. https://doi.org/10.1007/s12649-014-9326-3
16. Nixon EJ (2018) Co-product potential of algae biocake. Eastern Illinois University, Charleston, Illinois
17. Álvarez P, Salgueiro JL, Pérez L, Cancela A, Sánchez A, Ortiz L (2016) Total use of microalgae as feedstock for biodiesel and pellet production. Int J Environ Res 10(4):637–644
18. Wang Y, Seppanen-Laakso T, Rischer H, Wiebe MG (2018) Euglena gracilis growth and cell composition under different temperature, light and trophic conditions. PLOS One 13(4). https://doi.org/10.1371/journal.pone.0195329
19. Matsuda F, Hayashi M, Kondo A (2011) Comparative profiling analysis of central metabolites in Euglena gracilis under various cultivation conditions. Biosci Biotechnol Biochem 75(11):2253–2256. https://doi.org/10.1271/bbb.110482
20. Kawabata A, Miyatake K, Kitaoka S (1982) Effect of temperature on the contents of the two energy-reserve substances, paramylon and wax esters, in Euglena gracilis. J Protozool 29(3):421–423. https://doi.org/10.1111/j.1550-7408.1982.tb05425.x
21. Wu M, Li J, Qin H, Lei A, Zhu H, Hu Z, Wang J (2020) Pre-concentration of microalga *Euglena gracilis* by alkalescent pH treatment and flocculation mechanism of Ca3(PO4)2, Mg3(PO4)2, and derivatives. Biotechnol Biofuels 13(98). https://doi.org/10.1186/s13068-020-01734-8
22. Tumuluru JS, Wright CT, Hess JR, Kenney KL (2011) A review of biomass densification systems to develop uniform feedstock commodities for bioenergy application. Biofuel Bioprod Biorefin 5(6):683–707. https://doi.org/10.1002/bbb.324
23. Shojaeiarani J, Bajwa DS, Bajwa SG (2019) Properties of densified solid biofuels in relation to chemical composition, moisture content, and bulk density of the biomass. BioResources 14(2):4996–5015
24. Mani S, Tabil LG, Sokhansanj S (2006) Specific energy requirement for compacting corn stover. Bioresour Technol 97(12):1420–1426. https://doi.org/10.1016/j.biortech.2005.06.019
25. Ida T, Takagi R, Mizuno S, Sanchez E Jr, Namba K (2013) A study on the physical properties as affected by formation conditions of high density bio-solid fuel made from green tea. J Smart Process 2(2):83–87. https://doi.org/10.7791/jspmee.2.83
26. Mizuno S, Ida T, Fuchihata M, Namba K (2016) Effect of specimen size on ultimate compressive strength of Bio-Coke produced from green tea grounds. Mech Eng J 3(1):1–8. https://doi.org/10.1299/mej.15-00441
27. Suzuki K (2017) Large-scale cultivation of Euglena. Adv Exp Med Biol 979:285–293. https://doi.org/10.1007/978-3-319-54910-1_14

28. Frodeson S, Lindén P, Henriksson G, Berghel J (2019) Compression of biomass substances—a study on springback effects and color formation in pellet manufacture. Appl Sci 9(4302). https://doi.org/10.3390/app9204302
29. Mizuno S, Ida T, Fuchihata M, Sanchez Jr. E, Yoshikuni K (2015) Formation characteristics of Bio-Coke produced from waste agricultural biomass. ASME 2015 International Technical Conference and Exhibition on Packaging and Integration of Electronic and Photonic Microsystems San Fransisco, California, USA, International Electronic Packaging Technical Conference and Exhibtion. https://doi.org/10.1115/IPACK2015-48767
30. Brunerová A, Müller M, Šleger V, Ambarita H, Valášek P (2018) Bio-pellet fuel from oil palm empty fruit bunches (EFB): using European standards for quality testing. Sustainability 10(12). https://doi.org/10.3390/su10124443
31. Mizuno S, Ida T, Fuchihata M (2011) A study of physical properties of high-density solid biomass, Bio-Coke, with unutilized biomass. J Jpn Soc Exp Mech 11(Special Issue):SS19–SS24
32. Thomas M, van Vliet T, van der Poel AFB (1998) Physical quality of pelleted animal feed 3. Contribution of feedstuff components. Anim Feed Sci Technol 70(1–2):59–78. https://doi.org/10.1016/S0377-8401(97)00072-2
33. Kaliyan N, Morey RV (2010) Natural binders and solid bridge type binding mechanisms in briquettes and pellets made from corn stover and switchgrass. Bioresour Technol 101(3):1082–1090. https://doi.org/10.1016/j.biortech.2009.08.064
34. van Dam JEG, van den Oever MJA, Teunissen W, Keijsers ERP, Peralta AG (2004) Process for production of high density/high performance binderless boards from whole coconut husk. Ind Crops Prod 19(3):207–216. https://doi.org/10.1016/j.indcrop.2003.10.003
35. Muazu RI, Stegemann JA (2017) Biosolids and microalgae as alternative binders for biomass fuel briquetting. Fuel 194:339–347. https://doi.org/10.1016/j.fuel.2017.01.019
36. Svihus B, Zimonja O (2011) Chemical alterations with nutritional consequences due to pelleting animal feeds: a review. Anim Prod Sci 51:590–596. https://doi.org/10.1071/AN11004
37. Jeffrey EC (1910) The nature of some supposed algal coals. Proceedings of The American Academy, vol 46, vol 12. American Academy of Arts & Sciences, pp 273–290
38. Thiessen R (1925) Origin of the boghead coals. In: Mendenhall WC (ed) Shorter contributions to general geology. Washington Governer Printing Office. https://doi.org/10.3133/pp132
39. Fossil Algæ in Boghead Deposits (1937). Nature 139 (3512):340–340. https://doi.org/10.1038/139340a0
40. Kosanke RM (1951) A type of boghead coal from Illinois. Am J Sci 249(6):444–450. https://doi.org/10.2475/ajs.249.6.444
41. Tagami N, Ida T (2018) Effect of moisture content, forming temperature and structural components on Biocoke solidification from various biomass resources. Mech Eng J 5(5):1–8. https://doi.org/10.1299/mej.18-00030
42. Suopajärvi H, Dahl E, Kemppainen A, Gornostayev S, Koskela A, Fabritius T (2017) Effect of charcoal and kraft-lignin addition on coke compression strength and reactivity. Energies 10(1850). https://doi.org/10.3390/en10111850
43. Syivarulli R, Agung Pambudi N, Syamsiro M, Huat Saw L (2021) Upgrading the quality of solid fuel made from Nyamplung (*Calophyllum inophyllum*) wastes using hydrothermal carbonization treatment. Energy Eng. 118(1):189–197. https://doi.org/10.32604/ee.2020.010493
44. López-González D, Fernandez-Lopez M, Valverde JL, Sanchez-Silva L (2014) Kinetic analysis and thermal characterization of the microalgae combustion process by thermal analysis coupled to mass spectrometry. Appl Energy 114:227–237. https://doi.org/10.1016/j.apenergy.2013.09.055
45. Nunez C (2019) Fossil fuels, explained. https://www.nationalgeographic.com/environment/energy/reference/fossil-fuels/
46. Wu S-R (2017) Heating applications of bio-pellet to enhance utilization of renewable energy in the APEC region. Asia-Pacific Economic Cooperation Secretariat
47. Wang C, Chang Y, Zhang L, Pang M, Hao Y (2017) A life-cycle comparison of the energy, environmental and economic impacts of coal versus wood pellets for generating heat in China. Energy 120:374–384. https://doi.org/10.1016/j.energy.2016.11.085
48. Fuchigami Y, Hara K, Kita T, Uwasu M, Kurimoto S (2016) Analysis of effect on CO_2 emission reduction and cost estimation for the use of Bio-Coke: a case study of Osaka. Japan J Wood Sci 62(1):93–100. https://doi.org/10.1007/s10086-015-1515-6
49. Anyanwu R, Rodriguez C, Durrant A, Olabi A (2018) Optimisation of tray drier microalgae dewatering techniques using response surface methodology. Energies 11(9). https://doi.org/10.3390/en11092327
50. Wibawa D, Nasution M, Noguchi R, Ahamed T, Demura M, Watanabe M (2018) Microalgae oil production: A downstream approach to energy requirements for the Minamisoma pilot plant. Energies 11(3). https://doi.org/10.3390/en11030521
51. Thermodynamics: Heat and Enthalpy. ChemPages Netorial. https://www2.chem.wisc.edu/deptfiles/genchem/netorial/modules/thermodynamics/enthalpy/enthalpy3.htm. Accessed 6 Dec 2022
52. Melis A (2009) Solar energy conversion efficiencies in photosynthesis: minimizing the chlorophyll antennae to maximize efficiency. Plant Sci 177(4):272–280. https://doi.org/10.1016/j.plantsci.2009.06.005
53. Leong KM, Abdul Aziz N (2012) Malaysia's biomass potential. BE-Sustainable. https://issuu.com/besustainablemagazine/docs/besustainablemagazine-issue_0
54. Hossain N, Hasan MH, Mahlia TMI, Shamsuddin AH, Silitonga AS (2020) Feasibility of microalgae as feedstock for alternative fuel in Malaysia: A review. Energy Strat Rev 32:1. https://doi.org/10.1016/j.esr.2020.100536
55. Blanken W, Cuaresma M, Wijffels RH, Janssen M (2013) Cultivation of microalgae on artificial light comes at a cost. Algal Res 2(4):333–340. https://doi.org/10.1016/j.algal.2013.09.004