A Comparison of Energy Consumption in Hydrothermal Liquefaction and Pyrolysis of Microalgae

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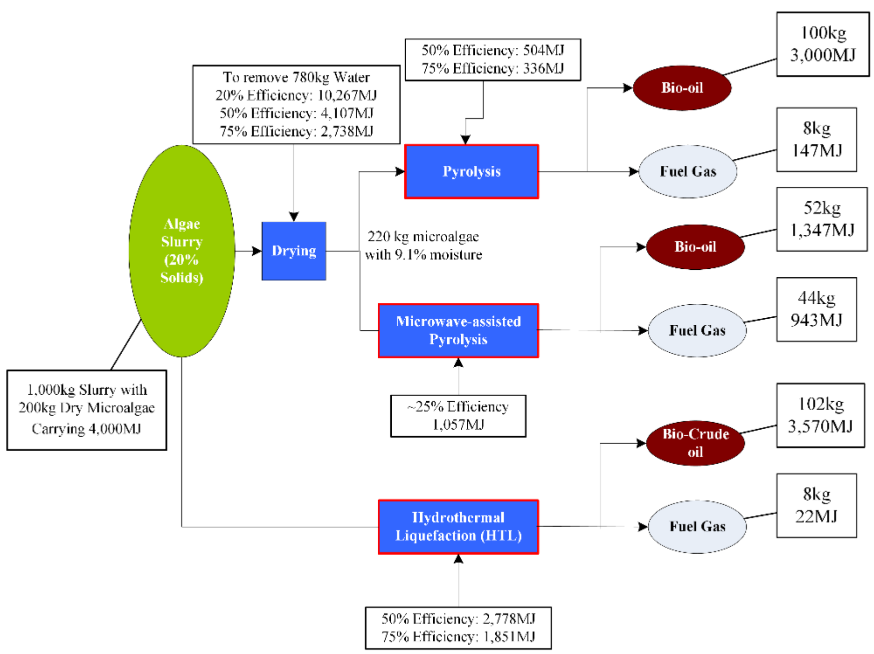

The energy requirements for converting one tonne (1,000 kg) of *Chlorella* slurry of 20 wt% solids via fast pyrolysis, microwave-assisted pyrolysis (MAP), and hydrothermal liquefaction (HTL) were compared. Drying microalgae prior to pyrolysis by using a spray drying process with a 50% energy efficiency required an energy input of 4,107 MJ, which is higher than the energy content (4,000 MJ) of raw microalgae. The energy inputs to conduct fast pyrolysis, MAP, and HTL reactions were 504 MJ (50% efficient), 1,057 MJ (~25% efficient), and 2,776 MJ (50% efficient), respectively. The overall energy requirement of fast pyrolysis is theoretically about 1.6 times more than that of HTL. The energy recovery ratios for fast pyrolysis, MAP, and HTL of microalgae were 78.7%, 57.2%, and 89.8%, respectively. From the energy balance point of view, hydrothermal liquefaction is superior, and it achieved a higher energy recovery with a less energy cost. To improve the pyrolysis process, developing drying devices powered by renewable energies, optimizing the pyrolysis process (specifically microwave-assisted), and improving the energy efficiency of equipment are options.

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Introduction

Thermochemical conversion of microalgae can be divided into pyrolysis of dry algae and hydrothermal liquefaction (HTL) of algal slurries [1]. Usually, the microalgal culture has a very dilute concentration of 0.1-1% dry solids. Currently, the proposed harvesting process is using a series of mechanical unit operations to dewater the microalgae media to a level of ~20% dry solids, which is considered as a less energy intensive processing option than completely drying microalgae for pyrolysis purpose. Drying is one of most dominant costs for algae harvest and may account for 30% of the total product costs, and the power consumption was equivalent to 15.8% of the energy of the recovered hydrocarbon [2]. Because of this energy consumption barrier, pyrolysis is considered as a kind of hopeless technologies for microalgae and only limited to laboratory investigations [3]. Meanwhile, researchers also recognized the advantages of the pyrolysis of microalgae (such as higher quality of pyrolytic bio-oil than that of cellulosic biomass) [4] and the merits of pyrolysis technology (such as lower capital cost than HTL) [5, 6].

This paper provides a simple comparison between the energy consumptions in pyrolysis of microalgae and hydrothermal liquefaction of microalgae. The purpose is not to provide a complete evaluation to these conversion technologies, but to give an idea how the energy consumption impacted the conversion processes of microalgae, and what would be the possible solutions.

Methodology

Microalgae

The composition analysis and properties of *Chlorella sp.* are summarized in Table 1. An engineered *Chlorella sp.* was assumed to be grown autotrophically, and had following components: 25% fatty acids, 50% protein, 15% polysaccharide, and 10% ash. For calculation, one tonne (1,000 kg) of *Chlorella* slurry at 20°C with 20 wt% solids and 80 wt% water (*i.e.* 200 kg dry algal cells and 800 kg water) was selected as the baseline. Cell concentration of 20 wt% has been used in multiple technical reports published by US national laboratories [7, 8]. This kind of algal slurries can be obtained via a series of dewatering unit operations such as settling, dissolved air flotation, and centrifugation. The energy content of microalgae is ~20 MJ/kg, so this microalgal slurry carried 4,000 MJ.

Table 1. Composition analysis and properties of *C. vulgaris* [9-12]

Protein (wt%)	34-58.1	**Specific heat (kJ/kg·K)***	1.57
Polysaccharide (wt%)	9.42-15.5	**Molecular weight (g/mol)***	360
Lipid (wt%)	1.04-15.6	**HHV (MJ/kg)**	19.3-21.2
C (wt%)	44.5-50.2	**Volatile matter (wt%)**	51.8-75.2
H (wt%)	6.2-7.2	**Fixed carbon (wt%)**	9-32.1
N (wt%)	6.4-10.9	**Ash (wt%)**	9.6-11.4
O (wt%)	24.6-40.7		

* [13]

Fast pyrolysis and microwave-assisted pyrolysis processes

Prior to pyrolysis, the microalgal slurry (1,000 kg) was dried with a spray dryer to 220 kg with a 9.1% moisture. Spray drying could generate *Chlorella* powders consisted of globular particles with a diameter of approximately 50-80 μm (*i.e.* 0.05-0.08 mm, approximately 270- 200 mesh) [14], which is fine enough for fast pyrolysis. Fast pyrolysis of microalgal powders were conducted in a fluidized bed reactor at 500°C with a heating rate of 600 °C/s. Pyrolytic product yields were assumed to be following: the bio-oil yield was 50 wt%, the yield of water solubles was 15 wt%, gaseous products counted for 4 wt%, and the biochar yield was 28 wt%. The gaseous products consisted of 22.2 vol% H_2, 34.9 vol% CH_4, 38.6 vol% CO_2, and 4.3 vol% C_2H_6 [11].

For microwave-assisted pyrolysis, microalgae could be air-dried by using solar dryers (Figure 1), because microwave pyrolysis doesn't require the finely ground feed [15, 16]. Microwave-assisted pyrolysis was assumed to be conducted in a pilot scale system, which could process large chunks of dry microalgae [17]. Pyrolytic product yields were assumed to be following: the bio-oil yield is 26 wt%, the yield of water solubles was 24 wt%, gaseous products counted for 22 wt%, and the biochar yield is 28 wt% [10].

A B

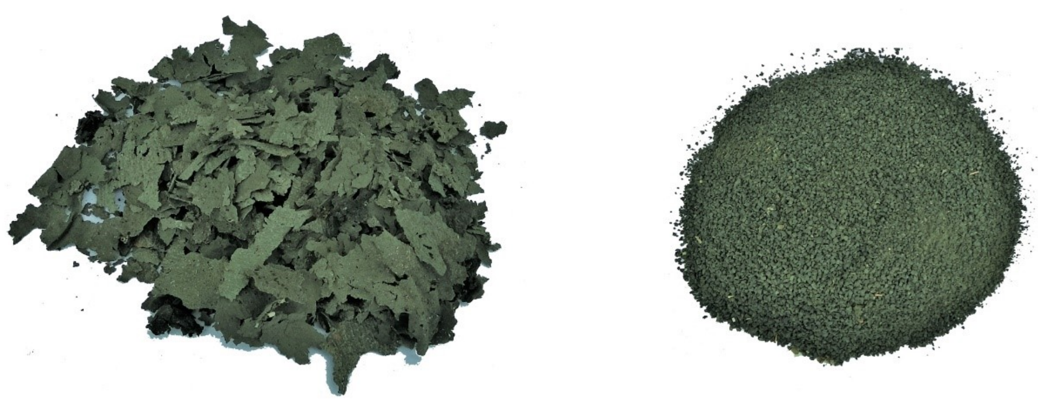

Figure 1. Naturally dried microalgae (A) and ground microalgae (B)

Hydrothermal liquefaction (HTL)

The microalgal slurry of 20 wt% solids was pumped to the HTL reactor, and hydrothermally treated in subcritical water at 2,500-3,000 psia and 350°C. The HTL process yielded 4 wt% gases, 51 wt% bio-crude oil, and 43 wt% aqueous organics and ash [5]. The non-condensable gases had following composition: 42 vol% CO_2, 50 vol% NH_3, 7 vol% CH_4, and 1 vol% ethane [18]. The non-condensable gases were mixed with natural gas and sent to a steam boiler for power generation. The predominately organic liquid phase is sent to catalytic upgrading, and the predominately aqueous phase is sent to wastewater cleanup for carbon recovery. Solids that can be removed by filtration might be recycled back to the algae ponds as nutrients [14]. The conditions and product yields for pyrolysis and HTL processes used in this study are summarized in Table 2.

Table 2. Conditions and product yields of pyrolysis and hydrothermal liquefaction of microalgae

	Fast Pyrolysis (500°C)	Microwave-assisted Pyrolysis	HTL (350°C)
Reaction temperature	500°C	-	350°C
Pressure	Atmospheric pressure	Atmospheric pressure	2500-3000 psia
Bio-oil (wt%)	50	26	51
Water (wt%)	15	24	-
Biochar yield (wt%)	28	28	-
Gaseous products (wt%)	4	22	4

Calculation

Specific heat of microalgae

According to a scientific report that studied the thermo-chemical properties of six species of microalgae, the specific heat (c_p) of microalgae was determined as 1.2 - 2 kJ/kg·K [13]. Meanwhile, to calculate the specific heat of microalga from its composition, following assumptions were applied: ash is SiO_2 with a specific heat of 733 J/kg·K or 0.175 cal/g·°C, the specific heat of polysaccharides is same as that of glucose (0.3 cal/g·°C), the specific heat of fatty acids is same as that of stearic acid (0.55 cal/g·°C), and the specific heat of protein is same as that of quinolone (0.352 cal/g·°C). Thus, the specific heat of *Chlorella sp.* was determined via Eqn. 1 as 0.376 cal/g·°C or 1.57 kJ/kg·K.

Specific heat of microalga (c_p, microalgae)

$$=10\%\times0.175+25\%\times0.55+50\%\times0.352+15\%\times0.3=0.376\ \mathrm{cal/g{\cdot}°C} \qquad \textit{Eqn. 1}$$

Energy for thermal drying of microalgal slurry

The feedstock for pyrolysis is typically quoted at <10 wt% moisture and requires thermal drying. To thermally dry one tonne of microalgal slurry (20°C) to 9.1% moisture, 780 kg water needs to be evaporated at 100°C. Water has a specific heat of 4.187 kJ/kg·K and latent heat (at 100°C) of 2256.9 kJ/kg [19].

Energy required for water evaporation:

$$=780\times4.187\mathrm{x}(100\text{-}20)+780\times2256.9=2{,}022\mathrm{MJ} \qquad \textit{Eqn. 2}$$

To evaporate 780 kg water from 1 tonne algal slurry, it will require at least 2,021,650 kJ, which is approximately 2,022 MJ or 562 kWh. This energy consumption is about 18.6 days of electricity usage of an American household [20]. Because the whole slurry shall be heated by the thermal dryer, the energy input for heating up rest water and microalgae can be calculated via following equations:

Energy required for heating 20 kg water to 100°C:

$$=20\times4.187\mathrm{x}(100\text{-}20)=6{,}699.2\mathrm{kJ}=1.86\mathrm{kWh} \qquad \textit{Eqn. 3}$$

Energy required for heating 200 kg microalgae to 100°C:

$$=200\times1.57\mathrm{x}(100\text{-}20)=25{,}120\mathrm{kJ}=6.98\mathrm{kWh} \qquad \textit{Eqn. 4}$$

The total energy for thermal drying of 1,000 kg microalgal slurry shall be equal to the sum of equations 2 through 4.

The total energy for thermal drying of 1,000 kg microalgal slurry:

$$=2{,}021{,}650.8\mathrm{kJ}+6{,}699.2\mathrm{kJ}+25{,}120\mathrm{kJ}=2{,}053\mathrm{MJ}=570\mathrm{kWh} \qquad \textit{Eqn. 5}$$

However, the overall thermal efficiency of spray dryers is only 20-50% [21]. Hence, if a dryer with 50% efficiency was used for drying the microalgal slurry, the total energy input for the drying process is 4,107 MJ or 1140 kWh. If the thermal efficiency can be improved to 75% [22], the energy requirement reduced to 2,737,960 kJ (2738 MJ) or 760 kWh.

Energy required for fast pyrolysis of microalgae

It's reported that the energy required to achieve thermal conversion (*i.e.* pyrolysis) of six different microalgae at 500°C was found to be approximately 1 MJ/kg [13]. Because only dry microalgal samples were used in their study, the energy required to evaporate moisture must be considered too.

Energy required for evaporation of 20 kg water:

$$=20\times4.187\times(100\text{-}20)+20\times2256.9=51{,}837\text{kJ}=23.6\text{kWh} \qquad \textit{Eqn. 6}$$

Energy required for pyrolyzing 200 kg microalgae:

$$=200\times1\text{MJ/kg}=200\text{MJ}=200{,}000\text{kJ}=55.6\text{kWh} \qquad \textit{Eqn. 7}$$

Total energy required for pyrolysis of 220 kg microalgae

$$=51{,}837\text{kJ}+200{,}000\text{kJ}=251{,}837\text{kJ}=252\text{MJ}=70\text{kWh} \qquad \textit{Eqn. 8}$$

If a pyrolyzer with 50% energy efficiency was used, the total energy input for the pyrolysis of microalgae rose to:

$$=251{,}837\text{ kJ}\div50\%=503{,}674\text{kJ}=504\text{MJ}=140\text{kWh} \qquad \textit{Eqn. 9}$$

Energy output from fast pyrolysis products

Pyrolyzing 200 kg dry microalgae yielded 100 kg bio-oil, 30 kg water, 8 kg gases, and 56 kg biochar. The microalgal bio-oil was assumed to have a higher heating value of 30 MJ/kg, so the energy output from the bio-oil is 3,000 MJ (3,000,000 kJ = 833 kWh). According to the composition of the gaseous products (22.2 vol% H_2, 34.9 vol% CH_4, 38.6 vol% CO_2, and 4.3 vol% C_2H_6), the gas phase had an average molecular weight:

$$\text{MW} = 2 \times 0.222 + 16 \times 0.349 + 44 \times 0.386 + 30 \times 0.043 = 24\text{ g/mol}$$
Eqn. 10

So, total gaseous products were 333.3 mol and 7,466.7 L (7.5 m^3) at normal temperature & pressure conditions, including 1.665 m^3 H_2, 2.61 m^3 CH_4, 2.89 m^3 CO_2, and 0.32 m^3 C_2H_6. The higher heating values of H_2, CH_4, and C_2H_6 are 12.769 MJ/m^3, 39.781 MJ/m^3, and 69.693 MJ/m^3 [23]. The energy output from the gases:

$$= 12.769 \times 1.665 + 39.781 \times 2.61 + 69.693 \times 0.32 = 147.4\text{MJ} = 41\text{kWh} \qquad \textit{Eqn. 11}$$

Because microalgal biochar is normally used as the soil amendment, total energy output from pyrolysis of 200 kg microalgae is 3,147.4 MJ (3,147,400 kJ or 874 kWh).

Energy required for microwave-assisted pyrolysis of microalgae

Energy requirement for microwave-assisted pyrolysis was only experimentally determined for a benchtop system that converted 30-60 g dry microalgae. Based on their results, it required 317 kJ to pyrolyze 60 g microalgae to the bio-oil with a 404 kJ energy content and gases with a 283 kJ energy content [24]. The experiments in [24] were performed in a microwave oven, which normally is less than 60% efficient [25]. If scaling up this microwave oven linearly to a system processing 200 kg microalgae with the same efficiency, the microwave-assisted pyrolysis requires an energy input of 1,056,667 kJ (1,057 MJ or 293.5 kWh), producing the bio-oil of 52 kg with a 1346,666 kJ (1347 MJ or 374 kWh) energy content and gases of 44 kg with a 943,333 kJ (943 MJ or 262 kWh) energy content.

Energy required for HTL of microalgae

One tonne (1,000 kg) of microalgal slurry was processed via HTL at 350°C. According to the steam table, the specific enthalpies of water (saturated liquid) at 20°C and

350°C (~17 MPa/2,500 psia) are 83.9 kJ/kg and 1,690 kJ/kg, respectively [26]. Energy required for heating 800 kg water from 20°C to 350°C:

$$= 800 \times (1690 - 83.9) = 1{,}284{,}880\text{kJ} = 1{,}285\text{MJ} = 357\text{kWh} \qquad \textit{Eqn. 12}$$

Energy required for heating 200 kg microalgae from 20°C to 350°C:

$$= 200 \times 1.57 \times (350 - 20) = 103{,}620\text{kJ} = 104\text{MJ} = 29\text{kWh} \qquad \textit{Eqn. 13}$$

The total energy required for heating this 1,000 kg microalgal slurry to 350°C is 1,389 MJ or 386 kWh. If an electric heater with a 50% efficiency was used for this duty, the total energy required for HTL of microalgae is 2,778 MJ (772 kWh). If a 75% thermal efficiency can be applied, the total energy required for HTL is 1,851 MJ or 514 kWh.

Energy output from HTL products

Since the yield of bio-crude oil was 51%, and thus the process yielded 102 kg bio-crude with a 35 MJ/kg heating value [27]. Total energy recovered in the bio-crude oil was 3,570 MJ.

The gaseous products (42 vol% CO_2, 50 vol% NH_3, 7 vol% CH_4, and 1 vol% ethane) had an average molecular weight:

$$\text{MW} = 44 \times 0.42 + 17 \times 0.5 + 16 \times 0.07 + 30 \times 0.01 = 28.1\text{g/mol}$$

Eqn. 14

So, total gaseous products were 285 mol and 6,377 L (6.4 m^3) at normal temperature & pressure conditions, including 2.7 m^3 CO_2, 3.2 m^3 NH_3, 0.45 m^3 CH_4, and 0.06 m^3 C_2H_6. The higher heating values of CH_4, and C_2H_6 are 39.781 MJ/m^3 and 69.693 MJ/m^3. The energy output from the combustible gases:

$$= 39.781 \times 0.45 + 69.693 \times 0.06 = 22\text{MJ} = 6\text{kWh} \qquad \textit{Eqn. 15}$$

Results and Discussion

To compare the energy consumption of different conversion technologies for microalgae, a 1,000 kg microalgal slurry was used as the baseline, and assumed to be processed with fast pyrolysis, microwave-assisted pyrolysis, and hydrothermal liquefaction processes. The energy requirements for the drying process and conversion reactors are summarized in Table 3. The energy present in original microalgae, the bio-oil or bio-crude, and gases is also summarized in Table 3.

Table 3. Breakdown of energy consumption during pyrolysis and liquefaction of microalgae (1,000 kg slurry with 20% solids at 20°C)

Energy (MJ)	Fast Pyrolysis (500°C)	Microwave-assisted Pyrolysis	HTL (350°C)
Energy in microalgae (20 MJ/kg)	4,000	4,000	4,000
Drying	4,107 [a]	4,107 [a]	N/A
Supporting conversion reaction	504 [a]	1,057 [b]	2,778 [a]
Total energy input	4611	5,164	2,778
Bio-oil	3000	1347	3570
Gas	147	943	22
Total energy in products	3147	2290	3592
Energy recovery	78.7%	57.2%	89.8%

a: 50% efficiency
b: 25% efficiency

The original 1,000 kg microalgal slurry with 200 kg dry microalgal cells carried 4,000 MJ energy. If drying this slurry to a moisture content of 9.1% by using a spray dryer with a 75% efficiency, the energy requirement for the dryer was 2,738 MJ. One advantage of spray drying for microalgae is to directly generate find powders for the need of pyrolysis. However, the spray dryers generally have 20-50% efficiency, resulting in increased energy inputs of 4,107-10,267 MJ. Obviously, the efficiency of the drying system plays a very important role. If a drying system powered by renewable energies could be introduced into this process, the pyrolysis of microalgae will be more attractive.

The energy requirements for microalgae conversion were various for different techniques. Fast pyrolysis required the lowest amount of heat, because the process was considered to be conducted under the optimal conditions. Microwave-assisted pyrolysis was scaled up from a bench-top system with a low energy efficiency, and showed an energy requirement of ~1,000 MJ for converting 200 kg dry microalgae. Because pyrolyzing 200 kg microalgae requires an energy input of 252 MJ, the actual efficiency of this microwave pyrolysis system was approximately 25%. Meanwhile, hydrothermally liquefying 1,000 kg microalgal slurry needed ~2,778 MJ (50% efficient). The energy need for HTL was less than that of drying wet microalgae, because the evaporation process was avoided and HTL reactions happened in saturated water.

The product yields of fast pyrolysis and HTL were optimal numbers, which were projected from recent experimental studies and shall be realized in the near future. Both optimized pyrolysis and HTL processes should produce ~50 wt% bio-oil or bio-crude oil with a higher heating value of 30-30 MJ/kg, which is the main energy carrier for both processes. The combustible gas yields from both processes were relatively low and less than 4 wt%. The energy recovery ratios from microalgae were 78.7% and 89.8% for fast pyrolysis and HTL, respectively. Because microalgae have a high ash content, resulting in a significant amount of ash and metals in the microalgal biochars. Normally, the microalgal biochars are considered as a good soil amendment.

The microwave-assisted pyrolysis process used for this study was not optimized, and produced large quantities of gases and less bio-oil products than fast pyrolysis or HTL. The energy recovery ratio for microwave-assisted pyrolysis was only 57.2%. Microalgae are a poor microwave absorber too, so other materials like char and activated carbon are often added to help microwave absorption [28].

From the energy balance point of view, hydrothermal liquefaction is superior, and it could achieve the higher energy-recovery ratio with a lower energy cost.

Meanwhile, the pyrolysis of microalgae might still have its chance. The major advantage of microwave-assisted pyrolysis is that it can process feedstock with a large particles size even chunks, because of the unique heating approach. If the efficiency of microwave-assisted pyrolysis can be improved to that of fast pyrolysis, and solar drying can be applied to solve the negative energy issue (as shown in Figure 2), the pyrolysis of microalgae will be more promising.

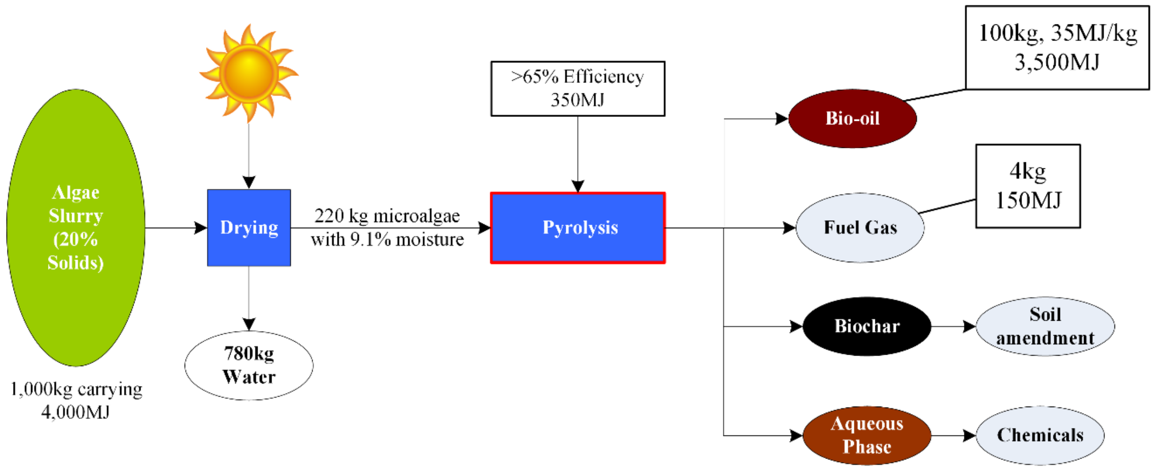

Figure 2. Proposed ideal pyrolysis process for microalgae

CONCLUSIONS

The energy requirements for converting one tonne (1,000 kg) of *Chlorella* slurry of 20 wt% solids via fast pyrolysis, microwave-assisted pyrolysis (MAP), and hydrothermal liquefaction (HTL) were compared. Drying microalgae prior to pyrolysis by using a spray drying process with 20%, 50%, and 75% energy efficiency required energy inputs of 10,267 MJ, 4,107 MJ, and 2,738 MJ, respectively. The energy inputs to conduct fast pyrolysis, MAP, and HTL reactions were 504 MJ (50% efficient), 1,057 MJ (~25% efficient), and 2,776 MJ (50% efficient), respectively. The microalgal feed contained 4,000 MJ, and the energy recovery ratios for fast pyrolysis, MAP, and HTL of microalgae were 78.7%, 57.2%, and 89.8%, respectively. From the energy balance point of view, hydrothermal liquefaction is superior, and it achieved a higher energy recovery with a less energy cost. To improve the pyrolysis process, developing drying devices powered by renewable energies, optimizing the pyrolysis process, and improving the energy efficiency of equipment are options.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this paper.

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