Energy use and greenhouse gas emissions from an algae fractionation process for producing renewable diesel

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Abstract

In one approach to algal biofuel production, lipids are extracted and converted to renewable diesel and non-lipid remnants are converted to biogas, which is used for renewable heat and power to support the process. Since biofuel economics benefit from increased fuel yield, the National Renewable Energy Laboratory analysed an alternative pathway that extracts lipids and also makes ethanol from carbohydrates in the biomass. In this paper, we examine the environmental sustainability of this “fractionation pathway” through life-cycle analysis (LCA) of greenhouse gas emissions and energy use. When the feedstock productivity was 30 (18) g/m²/d, this pathway emitted 31 (36) gCO₂e / MJ of total fuel, which is less than the emissions associated

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with conventional low sulphur petroleum diesel (96 gCO₂e / MJ). The fractionation pathway performed well in this model despite the diversion of carbon to the ethanol fuel.

Keywords: algae; life-cycle analysis; greenhouse gas emissions; biofuels; renewable energy

1. Introduction

The profitability of a biofuel pathway depends, in part, upon the fuel yield from a given mass of feedstock. Yields from two algal biofuel pathways, algae lipid extraction and upgrading (ALU) and algae hydrothermal liquefaction (AHTL), benefit from feedstocks with large amounts of lipids [1-3]. Economic models typically favor AHTL because of its high fuel yield [2], but AHTL can direct significant levels of nitrogen to the biofuel intermediate, which must be removed later, during upgrading. If upgrading occurs remotely from the algal production facilities, then the nitrogen is recovered in produced waters that are unlikely to be returned to the algal farm [4]. ALU, on the other hand, keeps nitrogen on site and may have more (or complementary) options for high-value co-products. Thus, there is interest in revising the ALU process to increase its fuel yield from a given biomass feedstock.

The National Renewable Energy Laboratory (NREL) studied a process that fractionated algal biomass into carbohydrate, lipid, and protein-rich fractions that can be converted into fuels and co-products. The carbohydrate-rich stream, produced via acid hydrolysis of algal biomass, was fermented to ethanol. Lipids were extracted from the lipid rich fermentation stillage and were upgraded to renewable diesel (RD), which accounted for most of the produced fuel. The protein-rich residue left after lipid extraction was converted to biogas by anaerobic digestion (AD) and this biogas was used for co-generation of heat and power on-site. While this approach is a

41 relatively new conceptual processing pathway, early research conducted at NREL has
42 demonstrated encouraging results with high yields (>65%) for hydrolysis of algal carbohydrates
43 to monomeric sugars, and high yields (>80%) of fermentable sugars to ethanol as well as
44 recovery of lipids via a wet extraction process at roughly 80 wt% moisture content [5].

45 Davis et al. [6] performed a techno-economic analysis (TEA) of this pathway via a discounted
46 cash flow rate of return analysis that determined the minimum fuel selling price required to
47 achieve a zero net present value under a set of financial assumptions and based upon estimates of
48 capital and operating expenses [6]. That study was based upon a mass- and energy-balanced
49 Aspen Plus computer model that determined stream flows, utility demands, and material inputs,
50 which is summarized in Figure 1. The stream flows were used for equipment sizing and
51 associated costs. Details are in Davis et al. [6], but in summary, this study established that the
52 fractionation approach could achieve a selling price of \$4.35 per gallon of gasoline equivalent
53 (GGE) in 2011 dollars for a high-lipid (41 wt%) *Scenedesmus* feedstock and \$5.04/GGE for a
54 lower-lipid (27 wt%) *Scenedesmus* feedstock if projected improvements in process operating
55 conditions and conversion yields were achieved. Although Davis et al. [6] considered ethanol
56 production, the carbohydrate fraction could, in principle, be converted to other fuels or higher
57 value products.

Figure 1. Fractionation process flow diagram. Biomass production is in open ponds, harvested via settling, and dewatered via dissolved air flotation and centrifugation as described in [1, 2]. Algal biofuels are sought, in part, from the desire for fuels with fewer greenhouse gas (GHG) emissions than would be produced by conventional petroleum fuels and from the desire to reduce petroleum and fossil fuel use. Life cycle analysis (LCA) of GHG emissions and energy use computes the total GHG emissions and energy use associated with all relevant operations related to the production and use of a particular fuel [7]. Our previous LCA studies of algal biofuels [1, 2, 4, 8-11] indicated that energy use on-site is high, e.g., power demand for culture mixing, dewatering, and cell disruption. In the ALU and AHTL pathway models that we studied previously, significant fractions of the biomass were left as remnants, which were converted to renewable heat and power that supported the process. The GHG reduction and the reduction of fossil fuel consumption depended upon this renewable power.

The fractionation pathway is attractive economically, but it redirects biomass away from on-site renewable heat and power generation, so it is possible that GHG and energy use performance may be impaired. Performance is often assessed via a metric of emissions or energy use per unit of produced fuel. The question addressed by this paper is whether the fuel yield increase in the

fractionation process compensates for the decrease in on-site heat and power production and compensates for the increase in process energy demand, e.g., for ethanol distillation.

This study presents an LCA of energy use and greenhouse gas emissions for algal biofuels produced via the fractionation process as modeled by Davis et al. [6]. The LCA considered all operations in the fuel pathway, including upstream material and energy provisioning operations, cultivation, and final fuel use. See Figure 2. The purpose of this analysis is to examine the sustainability of the fractionation process when energy use and GHG emissions are used as metrics. This work extends the literature by considering the sustainability implications of redirecting carbon away from waste streams that provide process energy towards streams that produce additional fuel and high-value co-products. It must be recognized from the outset that there are uncertainties in the algae pathway at the current time because several operations have only been evaluated at small scale. Even if the present study cannot achieve final, definitive emissions and energy use numbers, the study has value as a sensitivity study that can increase our understanding of the consequences of using fixed carbon in the biomass for co-products rather than using it for energy to support the process.

Figure 2: Boundary of the system considered in the life cycle analysis. The soil amendment co-product is the digestate (solids) from the anaerobic digestion step.

2. Methods

The LCA study was carried out using the Greenhouse Gases Regulated Emissions and Energy Use in Transportation (GREET) model [12]. The analysis computes greenhouse gas (GHG) emissions as the sum of the two stages, namely the well to pump (WTP) stage and the pump to wheel (PTW) stage. The WTP stage computes the energy use and GHG emissions from feedstock and fuel production, including transportation to distribution to fueling stations. This stage includes the material and energy demands for nutrient manufacturing, growth, biomass dewatering, biomass to fuel conversion, and transportation of fuel to a pumping station. The PTW stage examines the energy use and GHG emissions associated with vehicle operation (fuel combustion). The sum of the WTP and PTW stages is the Well to Wheel (WTW) result and is the net result for the pathway.

GREET computes GHG emissions associated with methane, nitrous oxide, and carbon dioxide by considering their global warming potentials, expressed in equivalent grams of CO₂ (g CO₂e). The total GHG emissions are reported as grams of CO₂ equivalent (gCO₂e) per megajoule (MJ) of renewable diesel (RD) based on the lower heating value (LHV) of the RD.

2.1 System boundary and functional unit (RD_e)

The Aspen model of the fractionation process in [6] considered only the conversion of 20 wt% dewatered algae biomass to fuel intermediates plus upgrading the lipids to RD blendstock. The model included all recycle loops, co-generation, heat integration, and waste treatment and calculated net mass and energy inputs. The energy inputs computed by the Aspen model were

broken down into individual fuels (electricity and natural gas) for input into the GREET LCA model. Within GREET, the set of operations was expanded to include nutrient manufacturing [11], CO₂ transport by low pressure pipeline, biomass growth and dewatering (see below), transportation of fuels to bulk terminal via barge, rail and truck, transportation from the bulk terminal to the fueling station by truck, and fuel use in vehicles. These operations, plus the transportation and use of AD digestate in fields as fertilizer, define the system boundary for the present LCA analysis. Details can be found in Frank et al.[9]. The system boundary is equal to the system boundary used in our previous LCA examinations of the ALU and AHTL pathways [1,2] and is consistent with other biofuel pathways conducted with GREET. The AD digestate was assumed to be dewatered to 30 wt% solids, consistent with centrifugal dewatering, and 40% of its nitrogen content was assumed to be bioavailable [1,9]. The bioavailable nitrogen displaces fossil fuel based fertilizers when applied on land as a fertilizer, which produces a credit by displacement, but the total N in the digestate contributes to N₂O emissions. See Frank et al. [9].

Energy use and GHG emissions are reported per MJ of fuel (the functional unit in the analysis). In principle, each of the fuels could be considered separately to reflect differences in vehicle efficiencies, but most of the fuel is RD. To simplify the analysis and to avoid technical questions regarding allocation methods, the higher heating values (HHV) of the RD, ethanol, and naphtha were combined into a total fuel, which is referred to here as “RD equivalent (RD_e).” The RD_e value includes the naphtha that is produced when the algal oil is hydrotreated. The flow of RD equivalent (MJ/hr) is equal to the sum, $C_{RD} \dot{m}_{RD} / \rho_{RD} + C_E \dot{m}_E / \rho_E + C_N \dot{m}_N / \rho_N$ where C is the HHV (MJ/L), $\dot{m}$ is the mass flow rate (Kg/hr), and ρ is the density (Kg/L) and where the subscripts indicate the various fuels (RD=renewable diesel, E=ethanol, and N=naphtha). Whenever fuel mass was required in the analysis, the total HHV of the produced fuels was

divided by the energy density of the RD (energy per mass) to obtain an equivalent weight. Fuel properties are shown in Table 1.

Table 1. Fuel properties used in the analysis.

Fuel	HHV (MJ/L)	Density (Kg/L)
RD	36.4	0.768
EtOH	23.6	0.789
Naphtha	34.9	0.73

2.2 Biomass production and harvesting

Biomass is produced in open ponds, harvested via settling, and dewatered via dissolved air flotation (DAF) and centrifugation as described in [1, 2]. Those references provide details, but key parameters are in Table 2. An Excel-based growth and dewatering model was used to compute the energy consumption associated with biomass production [12]. The process represented by the Excel model (Figure 3) was identical to that which was assumed in the Davis TEA study and has been checked against an Aspen model of these steps in previous studies [1, 2]

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150 **Figure 3.** Stages in Algae growth and dewatering. See Davis et al. [1] for details. Red text
151 indicates biomass concentration.

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153 **Table 2.** Key parameters in the algae growth and dewatering model.

Parameter	Value	Source
Mixing/Circulation energy	48 kWh/ha/d	Davis et al. [1]
Settler retention efficiency	90%	Davis et al. [1]
DAF retention efficiency	90%	Davis et al. [1]
Centrifuge retention efficiency	95%	Davis et al. [1]
Overall harvesting efficiency ^a	>99%	Davis et al. [6]
Energy to pump water to site	1.23e-4 kWh/L	Davis et al. [1]
Energy to pump culture on site	2.5e-5 kWh/L	Davis et al. [1]
Energy for DAF ^b	1.33e-4 kWh /dry g	Davis et al. [2]
Centrifuge energy	1.92e-5 kWh/g afdw	Davis et al. [1]
Solids concentration from centrifuge	20%	Davis et al. [6]
Net nutrient N demand ^c	4.5e-3 g N/g afdw	Calculated

Net nutrient P demand ^c	3e-3 g P/ g afdw	Calculated
CO ₂ utilization efficiency	82%	Frank et al. [9]
^a Assumes algae in water returned to cultivation from settler, DAF, and centrifuge are viable		
^b Total dry biomass basis		
^c Net, after recycle		

The fractionation TEA study considered high-lipid *Scenedesmus* (HLSD) harvested at 6 to 9 days. Table 3 shows the biomass properties used in the Davis TEA study. Although the underlying fractionation TEA analysis assumed optimistic, future algal annual average productivity of 30 g/m²/d, we also examined a more conservative assumption of 18 g/m²/d [2].

Net nutrients were computed as the difference between the N and P in the biomass and the recovered N and P computed in the Aspen model. N and P needs were met via ammonia and diammonium phosphate. CO₂ demand was satisfied in part by recycling CO₂ generated during fermentation and from the operation of the co-generation turbine. The CO₂ demand after recycling was imported via flue gas transported in low-pressure pipelines from a central source [1, 9].

Table 3. Composition of the high lipid *Scenedesmus* strain (HLSD) conditions, excerpted from [6]. Values are dry weight percentages on an ash free basis.

Element	HLSD
C	57.4
H	8.7
N	1.6
O	31.8
S	0.2
P	0.20
Ash	2.1
Fermentable carbohydrates	38.2
Other carbohydrates	3.9

Protein	9.0
Lipids (leading to fuel)	41.2
Non-fuel lipids	2.1
Other	3.5

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169 **2.2 Biomass conversion model**

170 A summary of input and output flows was presented in Table 24 in Davis et al. [6] for the annual
171 average biomass flow for two cases. The first case (“Store case”) dries 35% of the summer
172 biomass and stores it for winter use. In the second case (“No store” case), the biomass is
173 processed as it is produced, without drying or storage. In the store case, the dried summer
174 biomass is processed in the winter together with the winter-produced biomass. The Store Case
175 helps to level the variation in processing stream flows the season to season and thus mitigates
176 oversizing the plant [2]. We analyzed both scenarios, but report only the no storage case here
177 because it seemed the more justifiable assumption, e.g., storage without refrigeration has not
178 been demonstrated in hot, summer conditions.

179 The energy use data from the Aspen model [6] were expressed on the basis of grams of
180 equivalent fuel (RD_e), as described above. Table 4 displays the material and energy use. The
181 Aspen model methane flow was used to compute fugitive emissions (2% of total, See Frank et al.
182 [9]) and the co-generation electricity calculated with the Aspen model was used to reduce the
183 demand for grid electricity.

Table 4. Material and energy inputs and outputs per equivalent gram of fuel for the fractionation conversion operation.

Material Inputs		
Biomass	2.29	g afdw/g RD_e
Hexane makeup	4.11×10^{-2}	g/g RD_e
Acid (pretreatment)	1.01×10^{-1}	g/g RD_e
Ammonia (pretreatment)	3.25×10^{-2}	g/g RD_e
DAP (ethanol fermentation)	4.66×10^{-3}	g/g RD_e
Net natural gas demand for the process	5.06×10^{-4}	kWh/g RD_e
Hydrogen	2.26×10^{-2}	g/g RD_e
Phosphoric acid (lipid purification)	1.76×10^{-3}	g/g RD_e
Silica (lipid purification)	9.50×10^{-4}	g/g RD_e
Clay (lipid purification)	1.85×10^{-3}	g/g RD_e
Outputs		
RD_e	1	g
AD digestate, dry	3.75×10^{-1}	g/g RD_e
Electricity ^a	3.12×10^{-4}	kWh/g RD_e
Gaseous CO ₂ recovered	1.57	g/g RD_e
Methane produced ^b	0.126	g/g RD_e

^aThis co-gen power reduces grid electricity use in the entire biomass to fuel pathway

^bIntermediate stream used to calculate fugitive emissions

3. Results and Discussion

Figure 4 presents the fractionation pathway direct energy consumption for each season. Direct energy demand is the process energy required during biomass and fuel production without adding indirect energy use such as for electricity and natural gas provisioning or material manufacturing. The high energy use during the winter growth stage arises from culture circulation when productivity is low, thus requiring long residence times in the pond. Davis et al. [6] examine economic performance when the assumed productivity and biomass compositions are achieved. The results will apply only to those locations at which the assumed parameters are achieved.

Table 5 displays the corresponding life cycle energy use and GHG emissions for the Davis et al. productivity of 30 g/m²/d and for a more conservative case with 18 g/m²/d annual average productivity. The latter value, 18 g/m²/d, is the average over spring, summer, and fall productivity in a recent LCA study of AHTL[2]. That study used a validated biophysical growth model for *Chlorella*, climate data, and a thermal pond model to compute productivity at over one thousand sites in the Gulf Coast region. We assumed that the 3-season 18 g/m²/d average was achieved as an annual average for HLSD.

The fractionation pathway had lower energy use and had fewer GHG emissions compared to low-sulfur petroleum diesel at both productivities. Petroleum energy use in this pathway arises from the use of hexane for oil extraction and from the transportation and distribution of material inputs and outputs. The reciprocal of the fossil energy use in Table 5 is the energy return on (fossil) energy invested on a life-cycle basis within the analysis system boundary.

Infrastructure cycle emissions in Table 5 are those related to facilities construction. The fractionation pathway facilities are dominated by the cultivation and harvesting equipment, which are identical to those described in the 2012 ALU study [8]. The fractionation pathway and the 2012 ALU study differ in several items of equipment, e.g., the addition of the acid pretreatment equipment and the absence of high-pressure homogenization (HPH), but Canter et al. [8] found that the emissions from process equipment and buildings are not significant compared to those from ponds and concrete use. Therefore, the infrastructure emissions from Canter et al. [8] for the 2012 ALU pathway were adopted as an approximation for the infrastructure cycle emissions in the fractionation pathway.

The fractionation pathway was explored by comparing it to the 2012 ALU study [1], which is adopted as a reference pathway. Table 6 shows that the fractionation pathway emissions and energy use were 40% lower than those in the reference pathway, but such a direct comparison is unfair because the fractionation pathway definition utilized algae with higher lipid content (Table 7). This difference confounds understanding the energy consequences of differences in carbohydrate handling, biomass pretreatment, and lipid extraction.

The reference pathway was modified to provide a clearer basis for comparison. Figure 5 compares the GHG emissions in the reference pathway (first bar) with the emissions when the lipid content of the algae feed is increased to 41% to match the fractionation pathway (second bar). This change dropped the reference pathway emissions from 58 gCO₂e/MJ to 51 gCO₂e/MJ.

Since the emissions for the reference pathway exceed those for the fractionation pathway when the two have equivalent feedstock compositions, the fractionation pathway outperforms the reference pathway with regard to emissions. Two things contribute to the improvement. First, the fractionation pathway disrupts the cell during the acid pretreatment stage, which eliminates the HPH step that was used in the reference pathway for cellular disruption prior to hexane extraction. HPH is energy intensive and is driven with electricity while weak-acid hydrolysis is a thermal process. Electrical operations often have higher emissions than thermal operations because of the thermodynamics of electricity generation. Second, converting carbohydrates to ethanol increases the total fuel yield. Even if the HPH energy demand in the reference pathway is replaced with the (lower) energy required for the acid pretreatment (third bar in Figure 5), the ALU emissions still exceed those in the fractionation pathway (43 vs. 33 g CO₂e/MJ fuel).

In summary, for the data and assumptions in [6], converting carbohydrates to ethanol increased the fuel yield by 35% (on a mass basis). At 30 g/m²/d productivity, the GHG emissions for the fractionation pathway were 32% of those for conventional low-sulfur diesel. At 18 g/m²/d productivity, the GHG emissions were 38% of conventional diesel. In this lower productivity scenario, the fractionation pathway used 41% of the fossil fuel and 7% of the petroleum as compared to producing conventional low-sulfur diesel. Thus, the fractionation pathway could provide a method to increase the total fuel yield per mass of algae feedstock while achieving good GHG and energy performance if it achieves the performance targets assumed in Davis et al. [6]. Future work should consider feeds with lower lipid levels, should be better tied to observed productivities and compositions, and should develop additional experimental work to further validate the empirical basis for key data such as yields, energy use, mass balances, recycle rates, and generation of waste streams requiring treatment.

Figure 4. Direct process energy consumption in the Fractionation pathway. All energy is electrical, except as noted in the legend. Electrical demand during conversion was negligible.

Table 5. Energy use and GHG emissions results for the fractionation pathway. The fuel cycle includes operations and materials used during fuel production while the infrastructure cycle considers energy and materials use associated with constructing the algae farm and biorefinery.

	Productivity (g/m ² /d)	Fossil fuel use (MJ/MJ RD_e)	Petroleum use (MJ/MJ RD_e)	WTW GHG emissions, g CO ₂ e / MJ RD_e
Fuel cycle				
Algal RD	30 ^a	0.41	0.062	29
	18 ^b	0.45	0.064	33
	12 ^c	0.50	0.065	37
Fuel cycle plus infrastructure cycle				
Algal RD	30 ^a	0.44	0.068	31
	18 ^b	0.50	0.072	36
	12 ^c	0.57	0.078	41
Low sulfur diesel ^d		1.22	1.08	96

^a Fuel use and emissions results are fuel weighted annual averages

^b Computed for a single productivity of 18 g/m²/d – not a fuel weighted average

^c Computed for a single productivity of 12 g/m²/d – not a fuel weighted average

^d Results from GREET [12]

Table 6. Life cycle GHG emissions comparison between the fractionation and reference pathways

Pathway	Productivity, g/m ² /d	Fuel cycle emissions, g CO ₂ e/MJ	Total emissions, g CO ₂ e/MJ	Pathway model source
Reference	18	58	65	[1]
Fractionation	18	33	36	This study[6]

Table 7. Biomass properties and fuel yields comparison between the fractionation and reference pathways

	Fractionation (This study)[6]	Reference ([1])
Lipids, (wt%)	41%	25%
Carbohydrates (wt%)	38%	28%
RD from lipids alone, g RD/ g afdw of algae	0.319	0.184
Total fuel yield*, g /g afdw of algae	0.430	0.184

*Includes ethanol for the fractionation pathway. Both pathways produce a small amount of naphtha, which is neglected here but was included in fuel yield reported in Table 4.

Figure 5. Comparison of the fractionation and reference pathways. The reference pathway (2nd bar) assumed a different lipid content (25 wt%) than did the fractionation pathway. The lipid content in the reference pathway was adjusted to match the fractionation pathway (41 wt%) in the 3rd bar to allow comparison between the two.

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288 public, and perform publicly and display publicly, by or on behalf of the Government.

289 **Abbreviations**

290 AD-Anaerobic Digestion

291 afdw- ash free dry weight

292 AHTL-Algae Hydrothermal Liquefaction

293 ALU- Algae Lipid Extraction and Upgrading

294 DAF- Dissolved Air Flotation

295 GGE- Gallon of Gasoline Equivalent

296 GHG-Greenhouse Gas

297 GREET-Greenhouse Gases Regulated Emissions and Energy Use in Transportation

298 HLSD- High Lipid Scenedesmus

299 HPH- High Pressure Homogenization

300 LCA-Life Cycle Analysis

301 LHV-Lower Heating Value

302 MJ-Megajoule

303 NG- Natural Gas

304 NREL- National Renewable Energy Laboratory

305 PTW-Pump To Wheel

306 RD- Renewable Diesel

307 RD_e- Renewable Diesel equivalent

308 TEA-Techno-economic Analysis

309 WTP-Well To Pump

310 WTW-Well To Wheel

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