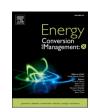
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Development of a mobile, pilot scale hydrothermal liquefaction reactor: Food waste conversion product analysis and techno-economic assessment

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ABSTRACT

The purpose of the present work is to investigate the feasibility of hydrothermal liquefaction (HTL) on food waste using a mobile pilot scale reactor and assess its techno-economic potential as a renewable energy technology that can be commercialized in the future. A 35 L pilot scale reactor (0.15 Gal·min⁻¹, 300 °C, and 60 min retention time) resulted in a higher biocrude oil yield than lab scale reactors (29.5 wt% vs 21.9 wt%). Biocrude oil qualities from pilot scale and lab scale HTL showed similar characteristics when comparing the elemental distribution, oil composition, and heating values. Further, techno-economic assessment (TEA) showed that the minimum selling price of the biocrude oil from a base case scenario was \$3.48 per gallon gasoline equivalent (GGE). The transportation cost of the feedstock and oil product was compared between onsite and mobile scenarios of HTL reactor operation. The results demonstrated that the mobile HTL reactor was more profitable when the sources of food waste were widely distributed (more than 106 miles). Combined pilot reactor results and assessments in different scenarios could be used to assess the sustainability of the HTL process for future large-scale implementation.

1. Introduction

It has been reported that nearly one third of the food produced for human consumption is wasted globally per year [1]. Of greater concern is the fact that the amount of food produced has exhibited an increasing trend in recent years. According to the 2018 U.S. Food Waste Investment Report, the United States alone spent approximately \$218 billion on the treatment of wasted food, amounting to as much as 52 million tons being sent to landfills and 10 million tons constituting on-farm losses [2]. Currently, there are three main methods to treat food waste: landfilling, incineration, and composting. However, all these methods generate greenhouse gas emissions (e.g. CO₂, CH₄) and have the potential to lead to surface and groundwater pollution [3].

Hydrothermal liquefaction (HTL) is an emerging thermochemical conversion technology that can convert wet biowaste into four different product fractions: biocrude oil, solid residue, aqueous phase, and gaseous product [4]. HTL normally operates at elevated temperatures (200–380 $^{\circ}$ C) and pressures (5–28 MPa) in an oxygen-free environment in order to transform the macromolecules of the feedstock into

bioenergy [5]. A myriad of feedstocks has been successfully converted into biocrude oil, such as algae, agricultural residues, animal manure, municipal solid waste, sewage sludge, waste plastics, wood, and other waste products [6-10]. In recent years, different studies have utilized HTL as a batch thermochemical technique for the treatment of food waste. Maag et al. performed HTL for the conversion of food waste using a CeZrOx catalyst and found that catalyst incorporation led to an energy recovery of 39%, a 159% increased oil yield, and a 12% reduction in the higher heating value (HHV) when compared with non-catalytic runs [11]. Gollakota et al. utilized ternary mixtures of food waste model compounds via fast HTL and reported that a yield of 23%, an energy recovery of 46%, and an HHV of 37.0 MJ/kg could be attained at a temperature of 600 °C using a retention time of 60 s [12]. Cheng et al. used mixture of microalgae and food waste in a two-stage process to improve both transesterification and HTL process by the regulating the C:N ratio in mixed feedstock [13]. Chen et al. investigated a two-step process, which is an alkaline pretreatment process followed by an HTL process. Highest energy yield of 56.5% was achieved with 10% of dry feedstock [14]. Aierzhati et al. compared the HTL of individual food

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waste compounds, as well as a mixture of food waste components [15]. This study found that high-lipid salad dressing resulted in a higher biocrude oil yield (78%), carbon content (88%), and energy recovery (96%) than the mixture of food waste compounds collectively (47%). Biocrude oil from food processing waste HTL was previously used to generate renewable diesel blendstocks [16], thereby signifying the potential of food waste-derived biocrude oil to contribute to the transportation fuel market. Despite the presence of batch HTL studies for the processing of food waste, continuous and pilot scale HTL experiments need to be conducted in order to assess the performance reliability of this process to provide renewable alternatives to conventional food waste treatment techniques.

Recently, researchers have focused on the continuous and up-scale processing of HTL technology in order to ascertain its commercialization potential. Because of the complexity and difficulty of constructing a continuous HTL system, only a few studies have reported the comparison of biocrude oil derived from bench and pilot scale reactors. Recently a critical review discussed the historic development of continuous HTL, specifically describing the impact of feedstock selection and dry matter content on the biocrude oil yield [17]. Wagner et al. demonstrated that a laboratory-scale continuous HTL reactor could be constructed at a low cost; this type of reactor only led to a maximum biocrude oil yield of 22%, which could have been attributed to the high ash content of the microalgae [18]. Pacific Northwest National Laboratory developed a 1 L tubular catalytic reactor which yielded 38-62.6%, 58.8% and 50-56% under 350 °C and 20 MPa with microalgae, macroalgae, and grape pomace, respectively [19-21]. Despite the fact that continuous stirred tank reactors (CSTR) [22], fixed beds [23], and packed bed reactors [24] have been previously used as continuous HTL reactors, plug flow reactors (PFR) still are the most popular design selection for continuous HTL. Feng et al. explored the feasibility of a pilot-scale continuous flow reactor with algal biomass cultivated in urban wastewater. Their CFR system ran 5% dry weight slurry at 350 $^{\circ}\text{C}$ and 17 MPa for 4 h and 28.1% biocrude oil yield was obtained [25]. Although most of the previous continuous HTL reactors were sized from 0.68 to 1.5 L [26,27], there have been some larger reactors reported. University of Sydney developed a 5 L PFR which was utilized to convert macroalgae in the presence of organic co-solvents, amounting to a yield up to 25% [28]. Aalborg University conducted co-liquefaction of aspen wood and glycerol experiments with two 5 L heat-traced serial reactors, which led to a biocrude oil yield of 20-33% [29]. Aarhus University successfully converted energy grass, microalgae, and sewage sludge with their 20 L HTL PFR with a yield ranging from 25 to 33% [30,31]. This study proposed a bigger pilot scale reactor which was one step closer to the commercialization of HTL technology. With an increasing amount of food waste generation in the world, a larger pilot scale reactor developed to investigate continuous food waste valorization is essential to broaden the application of continuous HTL technology. Although technical issues remain a barrier to the further upscaling of HTL technology and its eventual commercialization, the techno-economic feasibility and potential pecuniary gains from upscale HTL technology also pose roadblocks for commercialization of biocrude oil production technology.

Current TEA studies have emphasized the critical parameters influencing the economic uncertainty of thermochemical conversion technology for biocrude oil production for algae and woody biomass [32]. Previous studies have shown that the most significant factors influencing the TEA results for the upscale production of biocrude oil include: biocrude oil yield, capital expenditure for HTL reactors, and feedstock moisture and lipid content [33]. However, previous studies have also noted that transportation of low energy density feedstock from distributed sources to the reactor site could also represent a sizeable monetary cost. Nie et al. reported that biomass transportation costs constituted a large fraction (73%) of the total feedstock costs [34]. Nie et al. demonstrated that converting biomass to liquefied products first before transportation to a conversion facility could reduce the costs associated

with the feedstock by up to 48%. Jong et al. investigated centralized and distributed supply chains for biocrude oil production [35]. This study found that although downstream transport costs were marginal and roughly similar among all examined cases for woody biomass, upstream transportation costs amounted up to 10% of the total production costs. Overall, this study concluded that distributed supply chain configurations could decrease upstream transportation costs for large plant sizes (>75 PJ_{out}/year) [36]. Brinsemead et al. also reported that the transportation of the feedstock accounted for 23.3% and 23.9% of the total cost of biodiesel and biojet fuel for a nominal feedstock (crop residue) concentration of 50 tons/km²-yr [37]. Because of the wide distribution of food waste, it is even more important to investigate the feedstock transportation cost. Thus, establishing a methodology to minimize the transportation distance of the feedstock to the site of hydrothermal conversion with a mobile HTL unit could be a promising method to decrease the expenditure associated with biocrude oil production via HTL technology.

In order to address the knowledge gaps within the literature, a novel, mobile pilot-scale HTL reactor was developed and operated utilizing a previously unutilized feedstock for large-scale biocrude oil production: real food waste. Different from previous studies, this study aims to assess the viability of developing a mobile HTL reactor to offset costs associated with feedstock transportation, by conducting a TEA comparison of on-site fixed and mobile HTL based on pilot scale reactor data. Herein, this study aims to achieve the following goals: 1) Convert food waste into biocrude oil via a novel, mobile, 35 L pilot-scale HTL reactor; 2) Compare batch- and pilot-scale reaction results to determine the quality of the oil product; 3) Develop TEA process parameters for food waste HTL process; 4) Conduct TEA on mobile and on-site HTL to determine the commercial potential of up-scaling HTL by reducing feedstock transportation costs using a mobile pilot-scale reactor.

2. Material and methods

2.1. Reactor system and HTL conversion

The feedstock used for HTL reactions in this study was a food waste collected from a university dining hall (Champaign, IL). The volatile solid content in the feedstock was around 94.6%, and the higher heating value of the feedstock was 24.6 MJ·kg⁻¹. A 35L pilot scale plug flow continuous HTL reactor was used in this study as shown in Fig. 1. The system operated at 300 °C as highest temperature and at 10.7 MPa as pressure maintained by the back-pressure regulator (BPR). The use of back pressure regulator made sure the whole system was operating under the same pressure. The flow rate was 0.15 Gal·min⁻¹ on average controlled by a frequency-controlled diaphragm pump at feedstock supply of the reactor system. The retention time was 60 min. The food waste feedstock was ground with a meat grinder (Weston/Pro#22). After adding water and sieving (No. 4 Mesh 4750 µm), the solid content of the feedstock slurry was 13.3%. The reactor and rest of the system was stainless steel tubing with a half inch diameter. The operational data figure was in the supplementary material Fig. A.1. Seven heater hands were implemented to heat the reactor. A lab scale tubular reactor with the same diameter was used for comparison on the biocrude oil quality with pilot scale products under identical reaction condition. A 30 mL stainless steel tubular reactor with a high-pressure valve was built for lab scale experiments under the same reaction conditions as the pilot scale experiments. An electric resistance furnace (Thermo Scientific Lindberg Blue M) was used for heating the tubular HTL reactor. After the HTL process, dichloromethane (DCM, CH2Cl2) was dispensed into the HTL reactor to recover the products (biocrude oil, post HTL wastewater and solids). The mixture of liquid and solid phase was separated using a vacuum filter separation system. The biocrude oil was obtained by phase separation. Additional laboratory scale experimental details were the same as reported previously by the author [15].

The biocrude oil yield was defined as the total mass of the biocrude

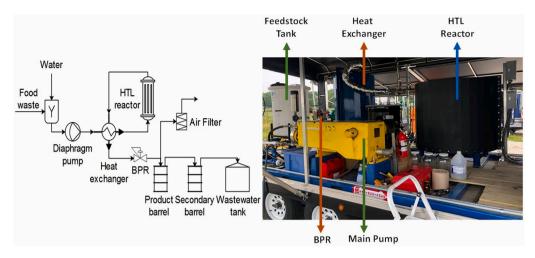


Fig. 1. Pilot-scale HTL process flow diagram and mobile reactor. (BPR: back pressure regulator).

oil divided by the total dry volatile organic compounds of the feedstock. The modified Dulong's formula was used to estimate the HHV of biocrude oils [38], as given in Eq. (1).

$$HHV = 0.0338 \times C + 1.428(H - \frac{O}{8})$$
 (1)

where HHV is higher heating value (MJ·kg⁻¹); and C, H, and O are the mass percentage of carbon, hydrogen, and oxygen (wt.%), respectively. The flow Reynolds numbers was calculated by Eq. (2), as follows.

$$Re = QD_H/vA \tag{2}$$

where Re is the Reynolds number; Q is the volumetric flow rate (m^3/s); D_H is the hydraulic diameter of the pipe (m); A is the pipe's cross-sectional area (m^2); and v (nu) is the kinematic viscosity (m^2/s).

The energy consumption ratio (ECR) was calculated to compare the thermochemical conversion energy of the process and energy recovered by biocrude oil, while accounting for energy loss and heat recovery efficiency. ECR values were calculated according to Eq. (3) [39]:

$$ECR_{HTL} = \left[C_{pw}W_i + (1 - W_i)C_{pf} \right] \Delta T[1 - R_h] / (Y(HHV)(1 - W_i)R_c)$$
 (3)

where C_{pw} is the specific heats of water (4.18 kJ·kg $^{-1}$ ·K $^{-1}$); R_h and R_c are the efficiencies of heat recovery and combustion assumed to be 0.5 and 0.7, respectively; W_i is the feedstock water content; ΔT is the temperature increase (assuming 25 °C as the initial temperature); Y is the biocrude oil yield; HHV (kJ·kg $^{-1}$) is the higher heating value of the biocrude oil; and C_{pf} is the specific heat of a dewatered feedstock (kJ·kg $^{-1}$), which is calculated with Eq. (4) as follows.

$$C_{pf} = 4.18(0.2X_{salt} + 0.34X_{carbohydate} + 0.37X_{protein} + 0.4X_{lipid})$$
(4)

where X represents the mass fraction % of each of the component groups [40]

2.2. Oil product characterization

A CE440 elemental analyzer (Exeter Analytical, North Chelmsford, MA) was used to measure the C, H, and N contents of the biocrude oils. The oxygen content was calculated with the difference subtracted from 100%.

A GC–MS system consisting of an Agilent 6890 gas chromatograph, an Agilent 5973 mass selective detector, and an Agilent 7683B autosampler was used. Gas chromatography was performed with a 60 m ZB-5MS column with 0.32 mm nominal diameter and 0.25 μm film thickness, using an injection temperature of 250 $^{\circ} C$ and Mass Selective Detector transfer line at 250 $^{\circ} C$. The oven temperature was initially set to

70 °C with a hold time of 2 min, then increased at 5 C·min⁻¹ until reaching 300 °C, and held constant for 5 min. The source temperature was 230 °C, electron ionization was set at 70 eV, and spectra were scanned from 30 to 800 *m/z*. Individual peaks were identified by matching fragmentation patterns against a NIST (NIST08) database. Analysis was performed based on the compounds with at least 0.5% relative peak area from the GC–MS tests of both pilot scale and lab scale biocrude oil.

Thermogravimetric Analysis (TGA) was performed with a Q50 TGA system, using a quartz crucible for the biocrude oils. While under a 100 mL·min $^{-1}$ nitrogen flow, the oven temperature was raised from 25 to 900 $^{\circ}\text{C}$ at a heating rate of 10 $^{\circ}\text{C·min}^{-1}$. Approximately 10 to 20 mg of biocrude oil samples were used.

In order to understand the molecular weight distribution of the bench and pilot-scale oil samples, MALDI-TOF-MS was performed. MALDI-TOF mass spectra measurements were conducted using a Bruker Autoflex Speed LRF instrument (Bruker Scientific Instruments, Germany) with dual microchannel plate detectors for both linear and reflection modes. In accordance with the manufacturer's instructions, MALDI-TOF was used with Flexcontrol software 3.0 (Bruker Daltonics) for the automatic acquisition of mass spectra in the linear positive mode within the range 2 to 20 kDa. The acceleration voltage was +25 kV, and ions were measured in reflection mode. Acetone was used as the matrix reagent. Samples used for MALDI-TOF analysis involved the addition of 10 L of liquid product and 30 μ l of the matrix solution, then 1 μ l of this mixture was placed on the MALDI sample holder for subsequent testing.

The functional groups in the biocrude oils were detected by using Fourier transform infrared spectroscopy (FTIR). All spectra were collected with a Thermo Nicolet Nexus 670 Fourier transform infrared spectrophotometer in transmission mode and with atmospheric compensation, between wavelengths of 800 and 4000 $\rm cm^{-1}$.

2.3. Simulation process overview

According to Natural Resources Defense Council (NRDC, 2017) [41], 1.6 kg of food waste was generated per person per week. This data assumes 75% moisture content (the moisture content of food waste varies seasonally, ranging from 76% in the summer to 72% in the winter). If the food waste generation is broken down to dry weight per hour, big cities like New York and Chicago generate 19,013 and 8662 dry kg/h, respectively. Midsized cities like Denver and Nashville generate as much as 1554 and 1524 dry kg/h. Given the assumed challenges of collecting all the food waste, 1000 dry kg/h scale was assumed for the base case scenario in this study. A wide range of feedstock solid contents were investigated by previous pilot scale HTL studies [42,43]. The solid content depends on both the pumping capability of the high-pressure

pump and the nature of the feedstock. Various pumps were tested to find the best fit [44], which was not the focus of this study. Thus, the feedstock solid content was assumed to be 20.0% in this economic analysis. The base case scenario process simulation parameters were based on author's previous work [15] and the pilot case scenario parameters were based on the pilot scale HTL process in this study as shown in Table 1.

The process simulation boundary includes three major parts, which are hydrothermal liquefaction (HTL), wastewater treatment, and biocrude oil upgrading. Since this is the first food waste HTL process simulation, HTL process was simulated based on a well-established large scale HTL process analysis [45] with the data collected from pilot scale reactor in this study as shown in Fig. 1. A deterministic steady-state chemical process modeling software, advanced system for process engineering (Aspen) plus V11, was utilized to build a process simulation for HTL and wastewater treatment approaches of the whole system. In the simulation, a thermodynamic model of the non-random two-liquid model (NRTL) was used. A shell and tube countercurrent heat exchanger was integrated to recover heat from the product stream. The total capital investment was calculated with the Aspen Process Economic Estimator V11. The energy returns of these scenarios were determined by the ratio of energy input (heating, pumping, mixing etc.) to energy output (biocrude oil, biohythane). According to the author's previous research [46], two stage fermentation was used to treat aqueous product in this study instead of catalytic hydrothermal gasification. The proposed biocrude oil upgrading process were simulated based on the protocol in the technical reports [45,47].

2.4. Techno-economic analysis

The TEA conducted in this study was based on previous HTL TEA research, and the method had been validated by several different studies [45–47]. A 30-year discounted cash flow rate of return (DCFROR) was used to evaluate the investment returns. The minimum selling price (MSP) of biocrude oil was also calculated for each process. MSP was presented as USD per gallon of gasoline equivalent (\$-GGE⁻¹) using energy densities of gasoline, and the calculation was based on the previous study [45]. Capital cost for HTL and wastewater treatment was estimated using the Aspen Process Economic Analyzer V11, whereas upgrading units (including hydrotreating, hydrocracking, and hydrogen generation units) were estimated by scaling quotes from the literature using the economies of scale power law with appropriate scaling factors [48]. The yearly operating cost of the facility was performed with a service factor of 0.9 [46].

To determine the MSP of the product, biofuel revenue (BR) from the sales of these biofuel components should be determined at the breakeven point where total revenues and total costs are equal [46]. For the

 Table 1

 Process simulation parameters of base case and pilot case.

Processes	Base case [15]	Pilot case
HTL		
Temperature (°C)	360	300
Pressure (MPa)	10	10
Product yields		
Biocrude oil	46.9%	29.5%
Aqueous	33.5%	39.5%
Gas	6.8%	3.5%
Solid	12.8%	27.5%
Gas compositions, wt.%		
CO_2	92.2%	89.0%
CO	5.6%	8.4%
CH ₄	1.1%	1.2%
H_2	1.1%	1.4%
Aqueous compositions, wt.%		
H ₂ O	93.2%	93.2%
Dissolved organics	6.8%	6.8%

basic design, total revenues include the revenues generated from biofuel and wastewater treatment benefit. The amount of biogas produced by wastewater treatment process should be expressed in terms of gasoline equivalent according to energy density of the products. The details of the economic analysis are shown in the supplementary material based on a well-established large-scale techno-economic analysis of HTL systems [45].

Sensitivity analysis shows how the uncertainty in the output of a mathematical or system (numerical or otherwise) can be apportioned to different sources of uncertainty in its inputs. In this study, the factors which significantly contributed to the economic viability of the scenarios were considered, i.e. reaction scale, feedstock cost, project lifetime, biocrude oil yield, subsidies, discount rate, tax rate, wastewater treatment operating conditions (dilution water and chemicals), price of utilities (electricity and natural gas) [49–51] as shown in Table 2.

3. Results and discussion

3.1. Biocrude oil characterization

The CHN test result and energy balance comparison are listed in Table 3. The CHN results show that the hydrothermal liquefaction process accumulated carbon and hydrogen in the biocrude oil phase. For the pilot scale biocrude oil, carbon content increased from 43.7% to 75.0%, hydrogen content increased from 6.9% to 9.1%, and the oxygen content decreased from 44.4% to 10.8%. There was no significant change for the nitrogen content. The biocrude oil from the pilot scale reactor had higher HHV in terms of higher carbon and lower nitrogen and oxygen content. These HHV results are consistent with other continuous reactor results reported by Pedersen et al. [29] and Barreiro et al. [22]. On the other hand, some previous results are lower [26,52], and some groups reported higher value [19-21]. The feedstocks were different in these studies, which resulted in a difference in HHV for the biocrude oil. All previously reported HTL biocrude oil HHV results are within the range of 33.2-39.3 MJ/kg. Lab scale biocrude oil had a higher hydrogen content. The pilot scale reactor had a higher biocrude oil vield (29.5 wt%) than the lab scale reactor (21.9 wt%), indicating that the continuous reactor led to a higher conversion of the feedstocks. This was attributed to the turbulent flow in the pilot scale reactor. The Reynolds number of the flow in the pilot reactor during the run was calculated as 7590 (>2900). This flow type helped in the mixing and sufficient contact of feedstock during the run, on the contrary, the lab scale tubular reactor didn't have mixing during the reaction. Because of better biocrude oil yield, the pilot scale reactor had a better performance in terms of energy balance. It presented a lower ECR, and its energy recovery was higher than the lab scale HTL reactor, as shown in Table 2. Another reason for this result was the pilot scale reactor had a heat exchanger that can recycle the heat from the reactor effluent. As such, the feedstock slurry was already preheated to 180 °C before it went into the reactor coils, and then, the products were cooled down to 80 $^{\circ}\text{C}$ after the heat

Table 2Parameters used for comparison of best and worst cases [46].

	Base case	Best	Worst
Project lifetime (year)	30	40	20
Feedstock cost (per dry ton)	0	-50	50
Discount rate (%)	0.067	0.067	0.15
Capital cost change (%)	0	-40%	40%
Tax rate (%)	0.17	0.05	0.35
Electricity price (USD·kWh ⁻¹)	0.0556	0.0556	0.2
Natural gas price (USD·kWh ⁻¹)	0.01	0.01	0.1
Dilution water reduction (%)	100	100	0
Chemicals reduction (%)	0	50	0
Project contingency (%)	0	10	20
Reaction scale (dry ton per hour)	20	1	0.5
Bio crude oil yield (wt. %)	80	47	10
Subsides for green fuel (\$/GGE)	2.65	0.757	0

Table 3 Elemental Analysis and energy balance comparison.

Samples	CHN test results			HHV (MJ/	Energy balance		
	C (%)	H (%)	N (%)	O (%)	kg)	ECR	ER (%)
Food waste	43.71 ± 0.13	6.90 ± 0.03	5.03 ± 0.36	44.37 ± 0.20	24.5	-	-
Pilot oil	$75.03 \\ \pm 0.08$	$\begin{array}{c} 9.13 \\ \pm \ 0.00 \end{array}$	$\begin{array}{c} 5.07 \\ \pm \ 0.02 \end{array}$	$10.78 \\ \pm 0.10$	36.5	0.53	57.8
Lab oil	$70.53 \\ \pm 0.55$	$\begin{array}{c} 9.59 \\ \pm \ 0.09 \end{array}$	$\begin{array}{c} 5.2 \pm \\ 0.03 \end{array}$	$14.77 \\ \pm 0.61$	34.9	0.74	29.6

exchanger, thus no additional cooling was required.

Fig. 2 presents biocrude oil categorized as a mixture of long chain organic acids (saturated fatty acids, monounsaturated fatty acids (MUFAs), polyunsaturated fatty acids (PUFA)), fatty acid amides and esters, N-heterocycle compounds, phenols, and others. The highest concentration compound group was fatty acid amides and esters, which resulted from recombination reactions of fatty acids from lipid and amines from protein degradation [53], Hexadecanamide, 9-Octadecenamide, (Z)- were present as the first and second highest concentration in both biocrude oils. The long chain organic acids, which include saturated fatty acid (n-Hexadecanoic acid, Octadecanoic acid, Tetradecanoic acid), MUFAs (Octadec-9-enoic acid), PUFAs (9-Octadecenoic acid), had the highest concentration. These long chain organic acids were hydrolyzed by sub-critical water from triacylglycerides (TAGs) [54]. There was also a high concentration of N-heterocyclic compounds. Cyclo (L-Phe-L-Pro), formed from cyclodehydration of amino acids, was the most present N-heterocyclic compound. In addition, biocrude oil from the lab scale reactor had more C₇ to C₁₅ N-heterocyclic compounds.

Hydrothermal liquefaction biocrude contains a wide range of compounds with different molecular weights, which results in a wide range of boiling points. Fig. 3 shows the results for the thermogravimetric analysis (TGA) and differential thermogravimetry (DTG). The majority of the weight loss for both of the oils were in the range of $149-232~^{\circ}C$ and $232-343~^{\circ}C$, according to the petroleum products classification [55], identified as kerosene and gas oil ranges, respectively. There was a notable weight loss in the light vacuum gas range ($343-371~^{\circ}C$) for lab

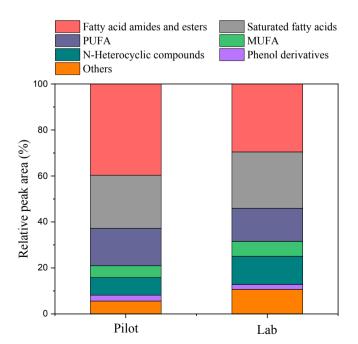


Fig. 2. Relative peak area percentage based on qualitative analysis of biocrude oils by GC–MS.

scale biocrude oil. The oil from the pilot scale reactor presented weight loss in the early part of heavy vacuum gas oil range (371–566 °C), which indicated a higher percentage of heavier compounds. After 500 °C, there were no significant weight changes for both of the samples. The unevaporated portion of the pilot scale biocrude oil was about three times greater than the lab scale biocrude oil. The MALDI results indicated the same trend with TGA results. The lab scale biocrude oil had the most molecular distributed from 101 to 200 kDa. However, the most accumulated molecular weight of the pilot scale biocrude oil was range from 301 to 400 kDa. These results showed the pilot scale biocrude oil was heavier than the lab scale biocrude oil, which is believed because of the polymerization reaction was accelerated by the turbulent flow in the pilot scale reactor.

3.2. Techno economic analysis of fuel production from food waste via hydrothermal liquefaction

As shown in Table 4, the cost of HTL units was about 32% of the total capital investment (TCI). Equipment cost and total installed cost of each HTL components are listed in supplementary material Table A.1. The wastewater treatment system was more expensive than the biocrude oil production unit, which cost about 45% of the TCI. Although the wastewater treatment process was chosen to be fermentation for this study, the wastewater treatment cost was in line with DOE reports [59]. The hydrotreating and hydrocracking, and hydrogen generation process was scaled down from previous reports [45] as experiments of HTL oil pilot scale upgrading was not within the scope of this research, which was 23% of the TIC. For the base case, the feedstock cost was considered as 0 because the food waste was assumed to be free in this case, and transportation cost of feedstock was not considered in this section either. Two different scenarios for feedstock transportation cost will be discussed in Section 3.3. Natural gas was chosen for heating the reactor to the desired temperature, which was 16% of the total operating cost. 19% of the operating cost was chemicals used during the wastewater treatment process. The electricity cost resulted from the pumps in the system. The minimum selling point of the base case biocrude oil was \$3.48/GGE, which is higher than the DOE goal of \$3/GGE by 2022 [59]. Thus, further investigation on means to reduce the product selling price of food waste HTL is still needed.

The scale of the process can affect the price of the product dramatically. Fig. 4 shows a price comparison of food waste from this study and different feedstocks from other reports. Based on previous publications on commercialization potential of promising HTL feedstocks, wastewater sludge [62], lipid extracted algae [60], and woody biomass [61] were compared. Among these feedstocks, food waste showed better economic feasibility by having lower biocrude minimum selling price at the same scales. The main reason was food waste had higher percentage of volatile compounds which led to higher yield of biocrude oil. At a certain scale, being around 1000 dry kg per hour for food waste, the fuel product from these biomasses can be cheaper than recent gasoline prices. All the biomass listed have the potential to be economically feasible at certain scales. At less than 20 dry ton per hour, food waste and municipal wastewater treatment plant sludge have better economic performance than woody biomass and algae. However, a woody biomass and algae conversion hydrothermal liquefaction facility can expectedly be scaled up to a larger size more easily than the other two feedstocks because of greater availability of the biomass feedstock. On the other hand, when the scale is greater, the transportation cost of the feedstock will also increase. The balance of feedstock transportation cost varies with the actual application.

Based on the range of the significant financial and manufacturing parameters listed in Table 2, the potential range of effects that could actually happen in the variation of a typical process was evaluated in Fig. 5. The minimum selling point of biocrude oil of the base case was \$3.48 per gallon gasoline equivalent. The most significant factor to the price was the biocrude oil yield. The biocrude oil yield was evaluated

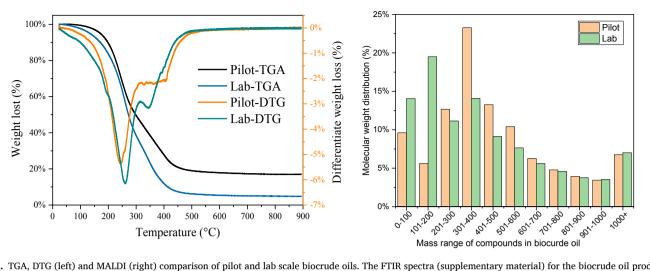


Fig. 3. TGA, DTG (left) and MALDI (right) comparison of pilot and lab scale biocrude oils. The FTIR spectra (supplementary material) for the biocrude oil products from the different reactors can be seen in Fig. 4. The C—H stretching from alkyl groups was noted in both the biocrude oils, presented as peaks between 2850 and 2960 cm⁻¹. The peak at 1708 cm⁻¹ was identified as ketones and aldehydes because of C—O stretching vibration from carboxylic groups [56]. Alkyl groups were identified by the C—H bending vibration at 1370 cm⁻¹ and alkylated compounds by the CH₂ bending vibration at 1460 cm⁻¹ [57]. Furthermore, peaks at 1150–1730 cm⁻¹ indicated the presence of N and O heteroatoms, according to Huang et al. [58]. The FTIR results corroborated similar results from the elemental analysis and GC–MS test.

Table 4Capital and operating cost for hydrothermal liquefaction of food waste.

Capacity: 1000 dry kg/h	Details	Amount (\$)
Total capital investment (TCI)		
Hydrothermal liquefaction	Reactor, heater, pump, cooling, product separation	3,744,530
Wastewater treatment	Reactor, heater, pump, product separation	4,120,340
Hydrotreating		1,613,871
Hydrocracking		372,432
Hydrogen generator		1,868,692
Total Installed Cost (TIC)		11,719,864
Warehouse	1% of TIC	117,199
Site development	9% of TIC	1,054,788
Additional piping	4.5% of TIC	527,394
Total Direct Cost (TDC)		13,419,245
Prorated expenses	10% of TDC	1,341,924
Field expenses	10% of TDC	1,341,924
Home office	20% of TDC	2,683,849
Project contingency	10% of TDC	1,341,924
Other costs	5% of TDC	670,962
Fixed Capital Investment (FCI)		20,799,829
Working capital	5% of FCI	1,039,991
Total Capital Investment (TCI)		21,839,821
Operating cost (per year)		
Feedstock		0
Natural gas		299,686
Catalysis/chemicals		339,811
Upgrading operating cost		65,565
Electricity		53,324
Water		83,376
Labor		338,256
Plant overhead	90% of Labor	304,430
Maintenance	3% of FCI	623,995
Insurance and tax	0.7% of FCI	145,599
Total operating cost		2,254,043
Capital depreciation	3% of Capital	655,195

from 10 wt% to 80 wt%, based on previous lab results of various food waste feedstocks under different reaction conditions [15]. The large yield ranges from different sources, attributed to the variation of the food waste biochemical components, could lead to a significant

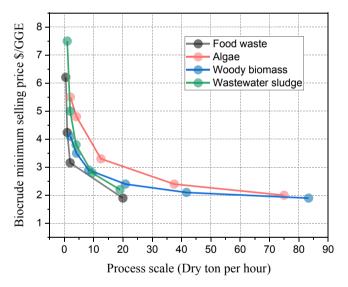


Fig. 4. Effect of process scale on minimum selling price of biofuel production via HTL (Algae: Zhu et al. [60]; Wood: Snowden-Swan et al. [61]; Wastewater sludge: Zhu et al. [62]).

difference in biocrude oil and the economic feasibility of the process. When the biocrude oil yield was higher than 46.9 wt%, up to 80 wt%, the MSP decreased from \$3.48/GGE to \$1.78/GGE. However, when the biocrude oil yield was only 10 wt%, the MSP became \$16.22/GGE. Notably, this indicated that a decent biocrude oil yield is the basis for the food waste HTL process to be economically feasible. Process scale was another important factor for the economic feasibility, which also proven by Fig. 4. Larger scales result in greater process feasibility. However, there is a limit to the scale, according to the amount and type of food waste that can actually be collected. Large scale food waste HTL process is more suitable for big cities with large amounts of food waste generated every day and efficient waste management supply chain networks. The next big factor was the natural gas price, which affected the operating cost for heating the system to the desired reaction temperature. This indicated that the location of the reaction site favors a place with lower natural gas prices. Feedstock price was also shown to have a big

Base Case 3.48/Gallon Gasoline Equivalent (1 dry ton per hour)

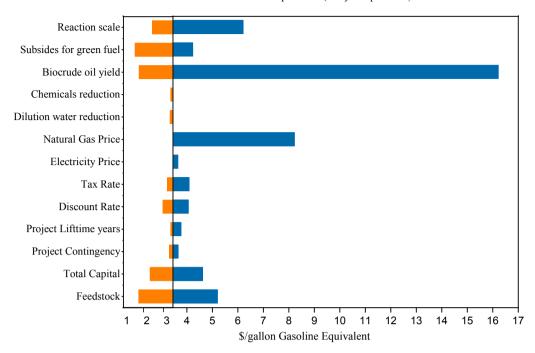


Fig. 5. Price of renewable diesel from food waste HTL process under different cases.

influence on the MSP. The base case assumed food waste feedstock as a free resource. However, the municipal solid waste disposal cost is \$50.60 per ton [63]. The HTL process can charge similar tipping fee to help decrease the MSP of the process further to \$1.71/GGE. The capital cost was not the biggest impact factor for making the process economical. Government support, tax rates, and discount rates also can help make the biofuel more competitive in the market. In addition, factors like chemicals usage, dilution water, electricity price, and project lifetime in the industrial practical range did not have huge impact on the selling price.

Fig. 5 showed the complete range of the MSP for the food waste HTL process by considering the wide range of parameters that could vary in an industrial process. In Fig. 6, the parameters were changed by a fixed ratio of $\pm 30\%$ to study which parameter had the highest influence on the process. The biocrude oil was the parameter that influenced price the most. This result aligned with previous algal HTL technoeconomic

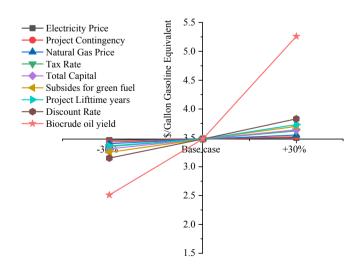


Fig. 6. Sensitivity analysis of the food waste HTL process (-30%/base case/+30%).

analysis results [64,65]. According to Aierzhati et al. [15], picking the right feedstock source for the process became significant for potential future commercial applications of HTL technology. In addition, more investigation on improving the process performance, which leads to a higher biocrude oil, is needed. Three of the remaining five high impact factors are related to the government policy, discount rate, subsides for green fuel, and tax rate. A favorable policy environment that supports renewable biofuel technology is essential for the development of the technology. Project lifetime and capital investment were not the most significant factors for the process economics.

3.3. Comparison with HTL of food waste via mobile trailer scenario and fixed location scenario

Transportation cost of the feedstock and biocrude oil is often neglected in TEA, however, it can be significant impact factor in food waste HTL process. Unlike algae, municipal wastewater sludge and woody biomass, food waste is usually generated in individual distributed sources, like restaurants and residentials. Under these circumstances, the discussions of feedstock and product transportation cost are valuable. Fig. 7 shows comparison of two cases, onsite reactor, and mobile reactor. In part a, the food waste feedstock was collected from a centralized source. The HTL reactor, upgrading and wastewater treatment units were located together. In this study, part a was called onsite HY (high yield) scenario which was the same as the base case scenario discussed in Section 3.2, using a flow rate of 1000 dry kg per hour, resulted in an MSP of oil product of \$3.48. As a comparison, part b included five 200 dry kg per hour mobile HTL HY reactors scenario. The total food waste conversion capacity of these two scenarios were the same (1000 dry kg per hour). However, the mobile HTL reactor scenario did not include wastewater treatment facilities, and there was a centralized refinery to upgrade the biocrude oil collected from those five 200 dry kg per hour mobile HTL reactors. Because of the lack of the scaling effect, the MSP of the 200-dry kg per hour mobile HTL reactor was calculated as \$4.81. Although it is higher than the MSP of the onsite HY scenario, if the transportation cost is part of the cost estimation, the advantage of mobile HTL scenario can be demonstrated. With the HTL



Fig. 7. Different HTL scenarios for centralized and distributed food waste sources; a) Onsite reactor; b) Mobile reactor,

reactor on a mobile trailer, the HTL reactor can be located next to the food waste generation site. There is no feedstock transportation cost involved in this scenario. It only involved transaction of biocrude oil, a more energy condensed material, to the centralized refinery location. On the contrary, the onsite HTL process was built with the refinery together. The transportation cost only included bringing feedstock to the site. When the results observed from pilot scale experimental demonstration as in Table 1 were used in the technoeconomic analysis, the MSP increased. The MSP for the onsite HTL scenario with pilot scale case operational and oil yield data was referred to as onsite LY (low yield) scenario, which was \$6.80. The mobile trailer scenario with pilot scale data resulted in a higher MSP, being up to \$9.09 in mobile LY scenario. These results again corroborated the significant impact of biocrude oil yield on the process economic feasibility.

Food waste delivery cost from their facility to a processor facility was estimated at \$0.50 per ton of food waste for every mile travelled [66]. The cost of transporting biocrude oil was calculated according to data reported by Marufuzzaman et al. [67]. As shown in Fig. 8, two scenarios were compared in the range from 0 to 175 miles. For high yield scenarios, although the MSP of the mobile HY Scenario was higher than the MSP of the onsite HY scenario, the increase in the transportation cost of

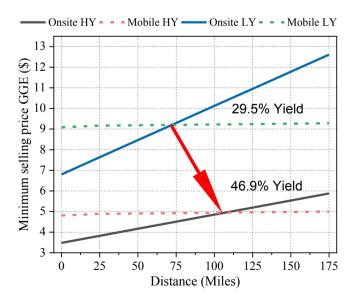


Fig. 8. Comparison of MSP considering transportation cost in different scenarios: onsite HY (46.9 wt% biocrude oil yield, 360 $^{\circ}$ C); mobile HY (46.9 wt% biocrude oil yield, 360 $^{\circ}$ C); onsite LY (29.5 wt% biocrude oil yield, 300 $^{\circ}$ C); mobile LY (29.5 wt% biocrude oil yield, 300 $^{\circ}$ C)).

the mobile HY Scenario increased slower than the onsite HY scenario (with a smaller slope). The MSP of two scenarios were equal at a transportation distance of 106 miles. When the distance of transportation is longer than 106 miles, the mobile HY scenario has an advantage with respect to the MSP of the oil product.

This analysis indicated that the mobile HY scenario can be an alternative solution for the future application of HTL technology. Because of the nature of the food waste feedstock, the HTL reactor can be situated at different food waste sites. The breakeven point for the onsite LY scenario and the mobile LY scenario for pilot scale reaction results was observed at 72 miles, which was lower than the high yield scenarios. When the biocrude oil yield is low, there is less amount of product needs to be transferred to the refinery. On the other hand, at a low biocrude oil yield, more feedstock is needed to produce equal amount of product. Although the scenarios with pilot scale reaction results are not as promising as the base case, higher solid content in the slurry and higher yield could be achieved in the future, thus improving process economics. Along with the development of the pilot case reactor technology, the breakeven point for the onsite HTL reactor and mobile HTL reactor should follow the red arrow in Fig. 8, which moves from the beginning of the arrow to the end. The mobile HTL reactor scenario in this study suggested an option between collecting feedstock or delivering biocrude oil products. In this study, the HTL reactor was located on a trailer to give mobility to the reactor. The capital cost of a warehouse, site development, additional piping, and home office were not included. The upgrading system and the wastewater system were not located on the mobile trailer with the reactor. The cost of HTL wastewater treatment via CHG accounted 45% of the total cost (except the feedstock cost) [59]. Without the onsite wastewater treatment plant, the capital cost and operating cost can be significantly reduced. With a mobile HTL reactor, which only have one-fifth of the scale of onsite HTL plant, the amount of wastewater produced was less and it could be discharged to an onsite sewer system. It was already reported that only 21% of organics remained in the wastewater [15]. Hence, the wastewater will not cause a shock for the wastewater treatment plant. Schideman et al. verified that the wastewater could be treated in a full-scale experiment in a wastewater treatment plant [68]. Given the nature of the scenario, the mobile HTL system will be moved to a new site after the accumulated food waste at one site is all converted. Thus, the wastewater from the HTL process is no longer a large-scale generation that can affect the COD, nitrogen, and phosphorus load of the local wastewater plant. The COD of the wastewater was 76500 mg/l. The wastewater treatment cost was estimated according to this study [69]. By avoiding the longdistance transportation of the feedstock with high moisture content, the mobile HTL reactor scenario is able to improve the economic feasibility. The produced biocrude oil products could be directly delivered to

the biocrude oil refinery for transportation fuels and value-added chemicals production.

Based on the base case parameters, on site HTL reactor operation is more suitable in big metropolitan areas and given large amounts of food waste availability within 106 miles. For the rural areas, which have widely distributed (distance more than 106 miles) food waste generation points, the HTL mobile unit is more economically feasible. It is worth noting that these scenarios are based on transporting the biocrude oil by truck. However, if the biocrude oil is transported by rail or even pipeline when the facilities are available, the scenario of putting the HTL reactor system on a mobile trailer will be even more economically feasible [70,71]. This indicated that the location of the HTL system is very important to its economic performance, and each individual case should be analyzed in detail to take into consideration the site-specific limitations and local policy incentives.

4. Conclusions

Investigating pilot-scale implementation and techno-economic assessment, this study demonstrated that HTL conversion of food waste is a promising process. The results indicated that the biocrude oil yield was higher in a continuous reactor than in a batch reactor; however, the pilot-scale biocrude oil had more heavy oil compounds and less N-heterocycle compounds. A techno-economic assessment of food waste HTL was developed to show the economic feasibility of this process. The mobile HTL unit can be more profitable when the biomass resources are widely distributed (more than 106 miles). This study provided a significant basis for food waste HTL pilot and commercial scale application by providing in-depth analysis to improve economic performance for future development.

CRediT authorship contribution statement

Aersi Aierzhati: Conceptualization, Writing - original draft, Methodology, Resources, Writing - review & editing, Investigation, Formal analysis. Jamison Watson: Writing - review & editing, Investigation, Formal analysis. Buchun Si: Writing - review & editing, Investigation, Formal analysis. Michael Stablein: Resources, Investigation. Tengfei Wang: Visualization. Yuanhui Zhang: Supervision, Funding acquisition, Project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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