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DOI: 10.2478/eces-2024-0002 ECOL CHEM ENG S. 2024;31(1):19-30

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MODELLING AND SIMULATION STUDIES ON THE CHARACTERISTIC BEHAVIOUR OF MACROALGAE DERIVED BIO-OIL

Abstract: With the expanding need for renewable and sustainable energy resources, the demand for biomass derived bio-oil particularly microalgae or macroalgae based bio-oil is growing. In this regard, understanding the characteristics of bio-oil as a function of several influencing operational parameters is essential. In this study modelling and simulation technique based on Aspen HYSYS was applied to investigate the characteristics of macroalgae *Ulva prolifera* derived bio-oil from Hydrothermal Liquefaction (HTL) process as a function of the influencing parameters like temperature, kinematic viscosity, and weight percent of the bio-oil assay, cut yield behaviour, and standard liquid density. Modelling and simulation help to optimise the process parameters and design the process layout for large-scale production. According to the simulation results the cut yield of off gases, light naphtha, heavy naphtha, light distillate, heavy distillate, gas oil and residues are shown at specific final boiling point (FBP) temperatures of 70 °C, 70 °C - 110 °C, 110 °C - 221.1 °C, 221.1 °C - 304.4 °C, 304.4 °C - 371.1 °C, 371.1 °C - 537.8 °C and 537.7 °C respectively. Whereas, above a temperature of 300 °C, the weight percentage of aromatic components increased steadily. The increase in percentage composition of the aromatic components is due to the reduction of the paraffinic components. The density of the liquid bio-oil was steadily increasing until a temperature of 200 °C.

Keywords: Aspen HYSYS, bio-oil assay, HTL simulation, modelling, macroalgae

Introduction

With the continued growth of the world's population, fulfilling the rapidly escalating demand for energy is a major global challenge. Fossil fuels being the major source of energy for most industrial and commercial activities, the issue of emission and depletion of fossil fuel resources is a crucial development and environmental concern. Excessive dependence on petroleum compromises national security, exacerbates environmental pollution and climate change [1] Major fossil fuel sources like coal are associated to environmental menaces, including release of higher sulphur, nitrogen oxide, carbon dioxide, and particulate matter [2].

On the other hand, renewable energy sources like wind and solar technologies that are meant to address the challenges linked to fossil fuels also suffer from disadvantages of high installation and maintenance cost and ecological imbalances [3, 4].

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Therefore, biomass based eco-friendly energy resources like biofuel can be suitable alternatives. Biomass based fuels are conventionally classified into three generations based on the complexity of the technologies and the type and nature of raw materials used. First generation biofuels are based on food energy sources. Second generation biofuels refer to agricultural waste biomass which particularly contain lignocellulosic materials those produced using more sophisticated manufacturing techniques compared to those in the first generation [5]. This type of biomass although they were thoroughly researched, their effectiveness is limited due to the complexity of the processes and the scarcity of the raw materials. The third generation biomass fuels are based on waste biomass including algal biomass that are abundant in nature [6]. First and second generation biofuel resources require vast space to produce. Thus, cannot meet the ever-growing global demand for transportation and industrial fuel on a long-term basis. Therefore, third generation biofuel from algae has a promising future to fulfill this demand. With regard to the yield and productivity, large scale algae production by aquaculture amounted to about $3.1 \cdot 10^{12}$ g of algae in the year 2006 compared to $2.2 \cdot 10^{10}$ g algae (dry mass basis) produced from wild algae stocks [7]. The best ten nations reaping wild stocks are topographically dispersed all throughout the world, however the production from aquaculture is centered around Asia with China representing 72 % of worldwide yearly generation [8].

A significant number of published works show the importance of algal use in biofuel production. According to Andrew Benson, Melvin Calvin and James Bassham, chlorella is a freshwater species of single-celled green algae that belongs to the phylum Chlorophyta. Chlorella's great photosynthetic efficiency makes it a potential source of both food and energy [9]. The first large-scale algae plant is expected to be operational by the coming decade, delaying the anticipated success for commercial scale algal plants in the year 2020. Algal biofuels are expected to contribute 50 % of total biofuel production by 2030 for the realisation of which facilities with $1.7 \cdot 10^{11}$ L capacity will need to be constructed [10].

Due to fluctuating fossil fuel prices and the constant depletion of resources, scientists are searching for a renewable substitute. The bigger problem is that if we are successful in using up every known fossil fuel, by the year 2300, atmospheric carbon dioxide levels will have increased by a staggering 5 times, from 408 parts per million (ppm) to 2000 ppm, resulting in a disastrous 8 °C increase in average world temperature. Algae biofuel has the potential to reduce global GHG emissions while improving the efficiency of transportation fuel. As a result, the application of biofuels as a renewable fuel source increase. Thus, macroalgae as a feedstock could provide a variety of advantages to meet global energy demand as an abundant renewable biofuel [11].

Micro- and macroalgae have gained significant attention due to their various applications in environmental remediation, biotechnology, and medicine. Microalgae, also known as phytoplankton, are unicellular organisms that can photosynthesise and thrive in diverse aquatic habitats. Macroalgae, also known as seaweeds, are multicellular organisms that grow in marine and freshwater environments. Both micro- and macroalgae possess unique biochemical and physiological properties that make them useful in various industries [12, 13].

Several studies have been conducted on micro and macroalgae to explore their potential applications. For instance, microalgae are known to produce high amounts of lipids and carbohydrates, which can be used to produce biofuels, bioplastics, and other high-value products. Macroalgae have been extensively used in the food industry due to their high nutritional value, and they also have the potential to be used to produce biofuels,

fertilisers, and other value-added products [14]. In addition to their commercial applications, micro- and macroalgae also have significant environmental benefits. Microalgae can be used to remediate wastewater by removing heavy metals and other pollutants. Macroalgae can help mitigate climate change by absorbing carbon dioxide from the atmosphere and reducing ocean acidity [15, 16].

In this regard, hydrothermal liquefaction represents one of the most effective thermochemical liquefaction technologies (HTL), which is a technique used for the treatment of both wet and dry biomass, including lignocellulosic and organic waste, without restriction on lipid content. Since the product of this process is an energy-dense intermediary that may be transformed into a variety of liquid fuels, it is referred to as bio-crude. HTL produces bio-crude from organic matter because of unique properties, such as the presence of water in the temperature range between 250 °C and 400 °C and pressures between $1 \cdot 10^7$ Pa and $3 \cdot 10^7$ Pa [17]. Temperature has a significant impact on bio-oil yield and quality during HTL. Bio-oil yield increases with temperature until it reaches an optimal range, then decreases at higher temperatures. The optimal temperature range for macroalgae HTL is typically between 250 °C and 350 °C. Higher temperatures (above 400 °C) can cause thermal degradation, resulting in lower bio-oil yield and quality [18].

Hydrothermal liquefaction of marine algal biomass is currently considered as the most efficient method for bio-oil production due to higher energy density, faster reaction time, usability with a larger variety of materials. To the contrary, algal biomass pyrolysis produces liquid products with high oxygen content which enhances instability and makes it challenging to use the product as a fuel. Instead, hydrothermal liquefaction has the potential to result in a liquid with far less oxygen. Dry biomass concentration was determined gravimetrically [6].

Several studies on the effects of temperature on viscosity, weight, and liquid density of bio-oil during HTL have been conducted. Fan et al. [19] investigated the HTL of sewage sludge and found that the bio-oil viscosity decreased from 11 $N \cdot s / m^2$ at 250 °C to 0.0013 $N \cdot s / m^2$ at 340 °C. According to the study, the weight of the bio-oil yield increased from 39.5 % at 250 °C to 55.2 % at 340 °C, while the liquid density decreased from 1.2 g/cm^3 at 250 °C to 0.95 g/cm^3 at 340 °C.

Guo et al. [20] examined biomass HTL and discovered that bio-oil viscosity decreased from 47.5 Pa at 240 °C to 0.18 Pa at 300 °C. The weight of the bio-oil yield increased from 37.3 % at 240°C to 54.5 % at 300 °C, while the liquid density decreased from 1.22 kg/m^3 at 240 °C to 0.89 kg/m^3 at 300 °C. The effects of these parameters on cut yield and bio-oil yield can be simulated using Aspen HYSYS. Using Aspen HYSYS to model the HTL of microalgae, Akhtar et al. [21] discovered that increasing the temperature from 280 °C to 340 °C increased the bio-oil yield from 42 % to 68 %, while increasing the residence time from 20 minutes to 60 minutes increased the bio-oil yield from 46 % to 62 %. The study also showed that increasing the pressure from $1 \cdot 10^6$ Pa to $2.5 \cdot 10^6$ Pa increased the cut yield of light gases and naphtha while decreasing the yield of bio-oil.

The properties of water change as a function of temperature, making it easier to separate products and byproducts and requiring less energy to purify them. By breaking down the macroalgae biomass, which is made up of a variety of complex macromolecules, into smaller chains and then allowing those small molecules to combine again to form a variety of new compounds, including short-chain carbon compounds like hydrocarbons, subcritical water can produce the liquid fuel known as algae bio-oil (bio-crude) [10].

Moreover, the relationship between temperature and other key operational and characteristic parameters was also studied using modelling tools. Aspen HYSYS was used to simulate the hydrothermal liquefaction process of *Ulva lactuca* and optimise the operating conditions. The authors found that the optimal conditions for bio-oil production were a reaction temperature of 320 °C, a residence time of 15 min, and a biomass-to-water ratio of 1 : 5 [18]. Aspen HYSYS was also used to model the hydrothermal liquefaction of *Ulva prolifera* and investigate the effects of process parameters on bio-oil yield. The authors found that the highest bio-oil yield was obtained at a temperature of 300 °C, a residence time of 30 min, and a biomass-to-water ratio of 1 : 5 [16].

Madina et al. [8] used Aspen HYSYS to simulate the hydrothermal liquefaction process of various biowaste including *Ulva* spp. and conduct a techno-economic analysis. The authors found that biowaste mixture had the highest bio-oil yield among the species studied and that the process was economically feasible at a large scale.

Mastalinezhad et al. [22] investigated the hydrothermal liquefaction of *Ulva pertusa* for bio-oil production and optimised the process using response surface methodology (RSM) and Aspen Plus simulation. The authors found that the optimal conditions for bio-oil production were at a reaction temperature of 300 °C, a reaction time of 20 min, and a biomass-to-water ratio of 1 : 10. The bio-oil yield under these conditions was 44.7 %, and the bio-oil contained high levels of oxygen, nitrogen, and sulphur compounds. The authors also conducted an economic analysis and found that the process was economically feasible, with a net present value of $5.5 million over a 10-year period [23].

The research trend in the part 8 to 10 years show that hydrothermal liquifaction process was found to be an effective hydrothermal degradation process for various species macroalgal based bio-oil or crude production. Yet the hydrothermal liquefaction process for bio-oil production from macroalgal biomass of *Ulva prolifera* group has not been well investigated and no significant research attempts were made to model experimental findings on the effect of various operational parameters like temperature and other influencing characteristic parameters like cut-yield, composition of aromatics and other organics, viscosity, standard liquid volume and PNA content of macroalgae (*Ulva prolifera*) derived bio-oil. In this study, Aspen HYSYS was used to model the relationship between bio-oil yield and the influencing parameters mentioned above.

Methodology

Sample collection, characterisation, and experimental study

The *Ulva prolifera* used in this study was collected from Kovalam beach area near Chennai. Characterisation of the algal biomass was carried out by measuring the weight loss of the heated biomass at different stages corresponding to the moisture (m), the volatile matter (vm), the fixed carbon (fc), and the ash (a) content of the sample. Algal cell growth pattern was monitored by measuring the absorbance of the culture at 685 nm by a UV-Vis spectrophotometer for 30 consecutive days. Then the macroalgae (Ulva), feedstock and water were infused into the HTL reactor. The blend ratio varied in accordance with each individual examination being carried out. Subsequently, heat was administered to reach an optimal temperature, which it sustained for a pre-planned duration before shutting down completely. As soon as room conditions were once again achieved within the reactor environment, a sample was collected for assessment purposes. The effects of the influencing parameters like temperature, composition of aromatics and

other organics, viscosity, standard liquid volume and PNA content were investigated against cut yield.

Modelling and simulation

In this study, the bio-oil assay characterisation was investigated using Aspen-HYSYS modelling tool. The hydrothermal liquefaction process was selected to produce whole bio-oil mixture from macroalgae *Ulva prolifera*. The temperature and pressure conditions assumed for the bio-oil production from the HTL were 280 °C and 2.5 x 10^7 Pa respectively. The whole bio-oil mixture is later fed into a three-phase horizontal separator after cooling and depressurising the bio-oil to a condition of 25 °C and 1 atm. Figure 1 displays the simulated process flow diagram for processing bio-oil in the Aspen Hysys platform. Then the gases and heavy components that are separated from the purified bio-oil are introduced to a heater 90 °C and later to vertical separator. The bottom product from the vertical separator is introduced to a cooler (25 °C) and the purified bio-oil is transferred to a receiving tank. The suitable fluid package used for this simulation is Peng-Robinson fluid package, it is ensured that all experimental findings are considered in accordance with the model's specifications.

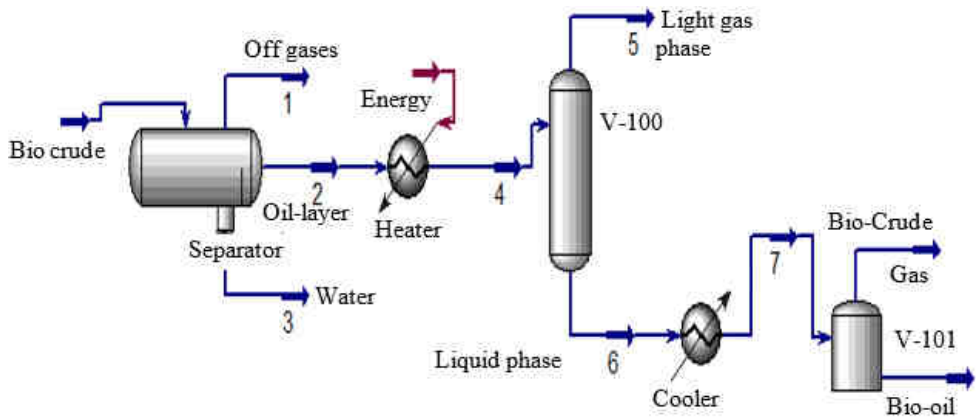


Fig. 1. Algae bio-oil characterisation using Aspen-HYSYS simulated diagram

Based on the simulation shown in Figure 1, the algae bio-oil compositions were investigated in terms of paraffinic aliphatic and aromatic components and the relationship of paraffinic, naphthenic and aromatic (PNA) components other physio-chemical characteristic and operating parameters. The results of the simulation study from Aspen HYSYS were replotted on excel and presented in the result and discussion section.

Results and discussion

Correlation between temperature and kinematic viscosity of bio-crude oil

The relationship between kinematic viscosity and temperature of the bio-crude was simulated based on the input data for the bio-oil in Aspen HYSYS. The results of the simulation are shown in Figure 2. Bio-oil yield showed a gentle linear increase with temperature in the range of 25 °C to 400 °C. However, with increasing temperature above

420 °C was observed to increase exponentially. The sudden change in temperature increasing trend may be attributed to the increase in cut yield of gas oil and residue composition of the bio-oil components and reduction of other components like light distillates and paraffins due to thermal degradation which resulted in the substantial increase in kinematic viscosity [19].

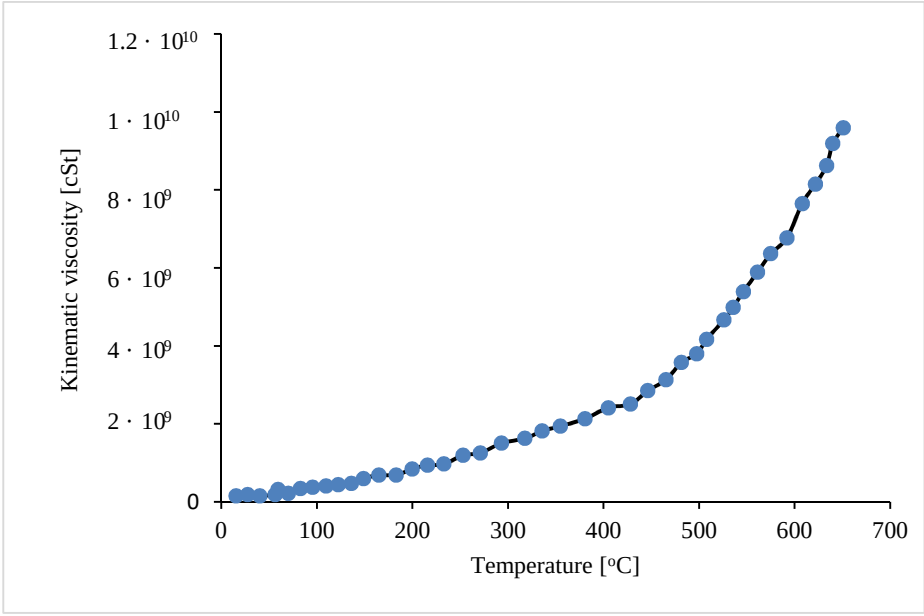


Fig. 2. Temperature effect with kinematic viscosity of bio-crude oil

Temperature and weight relationship for bio-crude oil

Figure 3 shows the relationships between temperatures and the weight percentages of the paraffinic, and the aromatic components of the bio-oil. The paraffinic content varies from 95 % at 0 °C to 50 % at 100 °C. The paraffinic component started to rapidly decline beyond 193.5 °C and completely disappeared at 400 °C. Interestingly, the simulation results showed the degradation trend of the paraffinic components where the percentage weight of light hydrocarbons showed sharp reduction until a temperature of 105 °C, further increase in temperature until 190 °C did not result in any substantial percentage weight reduction as the heavier paraffinic components resist the thermal degradation due to the long chain aliphatic groups where the bonds require higher energy to undergo breakdown. Nonetheless, beyond 193.5 °C the heavier paraffinic components showed steady reduction until they completely disappeared at 400 °C [24]. To the contrary, in the temperature range of 100 °C to 300 °C, the percentage weight of the aromatic components increased from 25 % to 50 %. Above a temperature of 300 °C, the weight percentage of aromatic components increased steadily. The increase in percentage composition of the aromatic components is due to the reduction of the paraffinic components. This simulation graph in general demonstrates the strong temperature dependence of the aromatic and paraffinic

composition of the bio-oil [2]. In addition to the paraffinic and aromatic components, the naphthene components were also considered in the simulation.

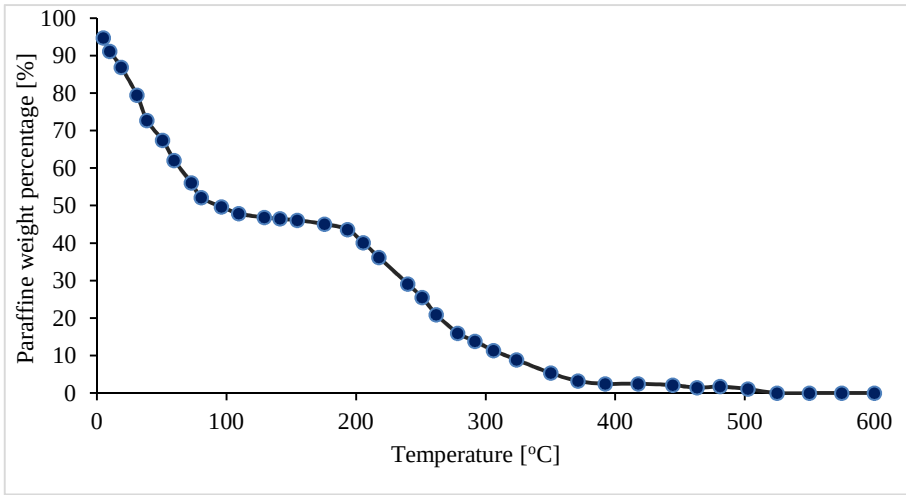


Fig. 3. Temperature effect of (bio-oil) paraffinic compounds IBP-FBP with weight percent

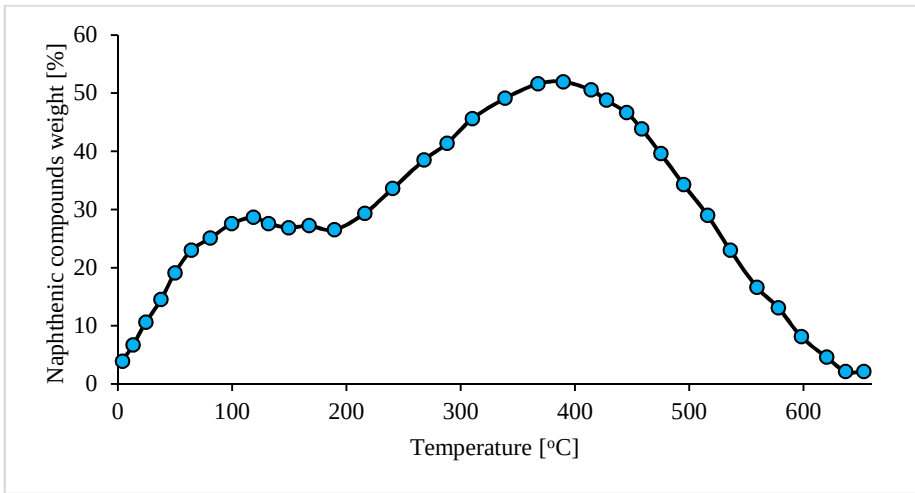


Fig. 4. Temperature effect of (bio-oil) naphthenic compounds IBP-FBP with weight percent

As shown in Figures 4 and 5, respectively, the profile of the simulated graph for naphthalene and aromatics components increases temperature of 100 °C and weight percentage. The naphthenic components like the aromatic components showed further increase up until a temperature of 370 °C and a corresponding weight percentage of 50%. Beyond the temperature of 370 °C the naphthenic component reduced steeply while the aromatic component increased steadily due to the resistance for thermal degradation [2]. The composition of paraffinic, naphthenic and aromatic (PNA) influences the yield and

quality of bio-oil produced from HTL. Aspen HYSYS can be a useful tool for predicting the effect of PNA composition on bio-oil production and for optimising operating parameters to produce bio-oil with desired properties and composition [6].

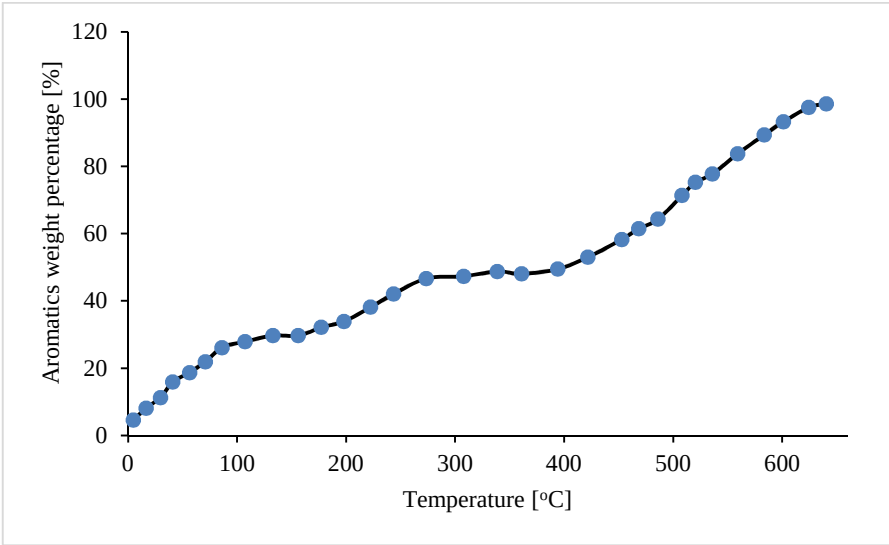


Fig. 5. Temperature effect of (bio-oil) aromatics compounds IBP-FBP with weight percent

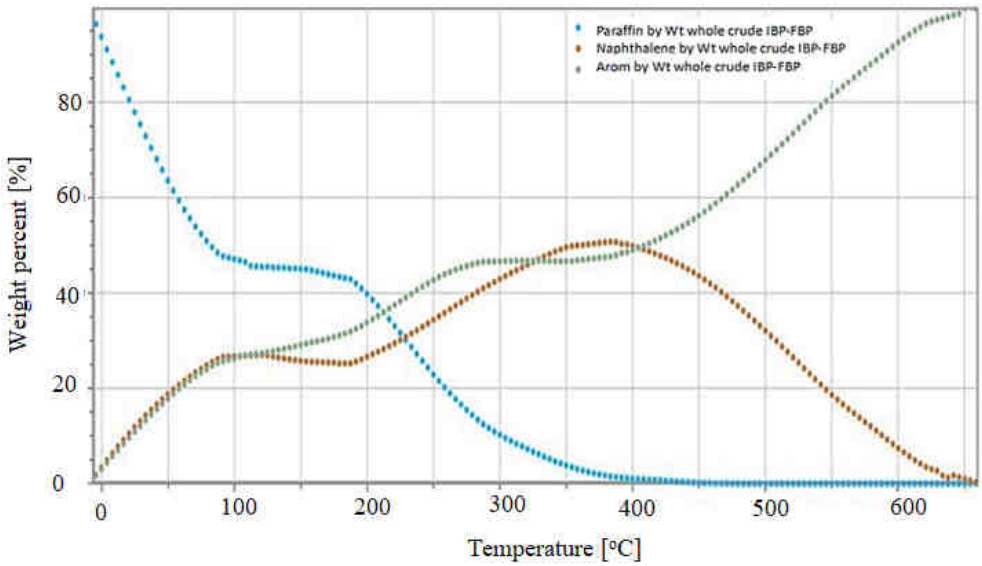


Fig. 6. Temperature effect on bio-crude oil with weight percent

Cut yield vs bio-oil yield

Figure 7 shows the macroalgae bio-oil assays with respect to the cut yield weight percent graph. Bio-oil consists of several components including off gaseous, light naphtha, heavy naphtha, light distillate, heavy distillate, gas oil and residues are shown in the above chart. The simulated results shown in Figure 7 portray the cut yield of a particular product at a given temperature. According to the simulation results the cut yield of off gases, light naphtha, heavy naphtha, light distillate, heavy distillate, gas oil and residues are shown at specific final boiling point (FBP) temperatures of 70 °C, 70 °C - 110 °C, 110 °C - 221.1 °C, 221.1 °C - 304.4 °C, 304.4 °C - 371.1 °C, 371.1 °C - 537.8 °C and 537.7 °C respectively. The cut yield percentages in Figure 7 show the influence of temperature on the separability of the components of the bio-oil assay. The findings form this simulation study about the composition of the bio-oil assay are comparable to previously published works [12]. In liquefaction, apart from producing a lower yield of amino acids, they undergo decarboxylation and deamination producing amines, carbonic acid and another substituent like organic acid and ammonia. On leaving the residue untouched over time, may get a chance of getting depolymerised to form lengthy chain hydrocarbons along with indole or pyrrole, which is an aromatic ring-type structure of phenols [25].

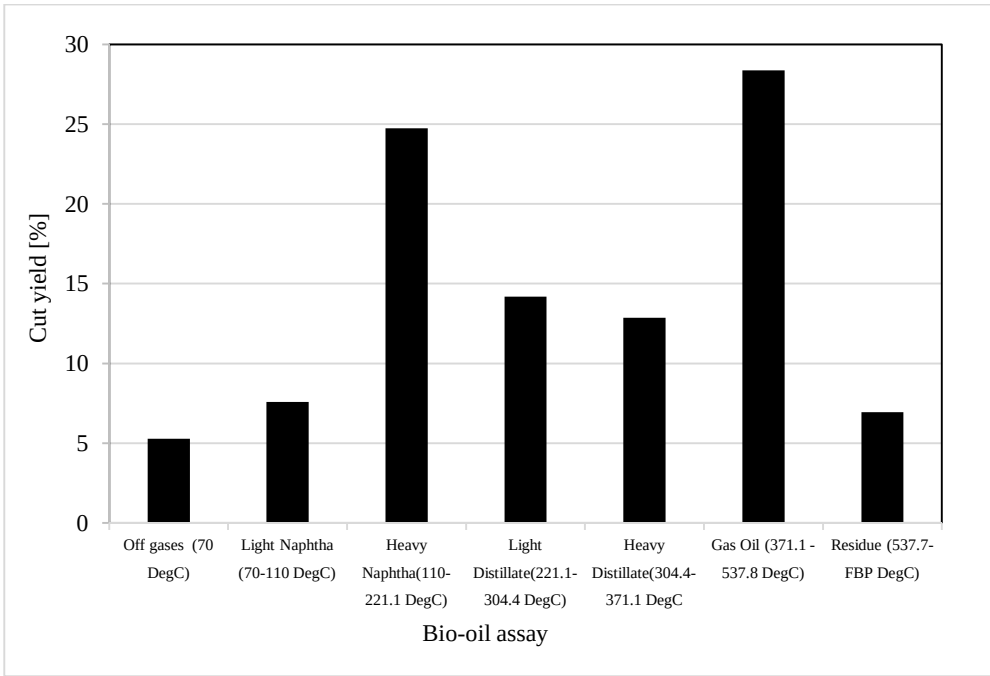


Fig. 7. Algae bio-oil assay vs cut yield

Effect of temperature on weight of algae bio-oil crude IBP-FBP

Figure 8 represents the relationship between temperature and weight percentage of macroalgae bio-oil as a whole crude. The results from the simulation study clearly show

that temperature has a linear relationship with the bio-oil weight percentage presented as whole crude which constitutes all the components discussed in the above section. As it was discussed earlier, the percentage weight of the paraffinic, naphthenic, and aromatic fractions shows a linearly increasing cumulative trend up until a temperature of 370 °C due to the increasing trend from naphthenic and aromatic components and further until a temperature of 495 °C due to the aromatic components only. When the temperature goes above 495 °C all components except aromatics disappear showing excessive increase in aromatic components. The finding from this simulations study accords well to published experimental research [8, 15].

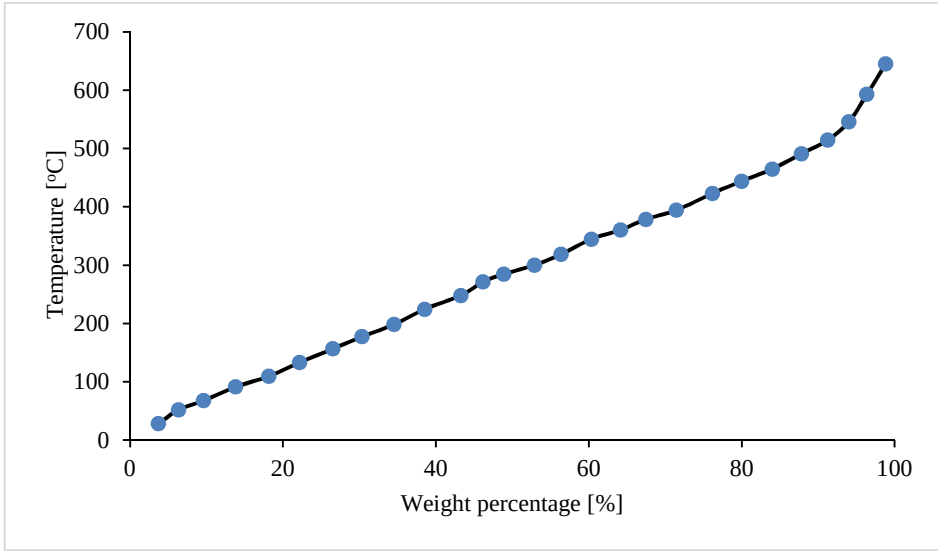


Fig. 8. Temperature vs weight of macroalgae bio-oil whole crude IBP-FBP

Effect of temperature on standard liquid density of algae bio-oil crude IBP-FBP

Figure 9 shows the influence of temperature on standard liquid density of the bio-oil composition. As shown in the simulated graph, the density of the liquid bio-oil was steadily increasing until a temperature of 200 °C. Further increase in temperature result in slight change of the density until a temperature of 420 °C attributed to the limited thermal degradation of the intermediate paraffinic, aromatic and naphtha components. Beyond the temperature of 420 °C the liquid density increased sharply mainly due to the heavy bio-oil aromatic and residual components. The above data of the standard liquid density has good agreement with previous work [12, 26]. The effect of temperature on kinematic viscosity of the bio-oil shown in Figure 2 perfectly matches with the trend shown in Figure 9 for the standard density. Thus, it can be inferred that increasing temperature results in the increase of both density and kinematic viscosity of the bio-oil as per the simulation output.

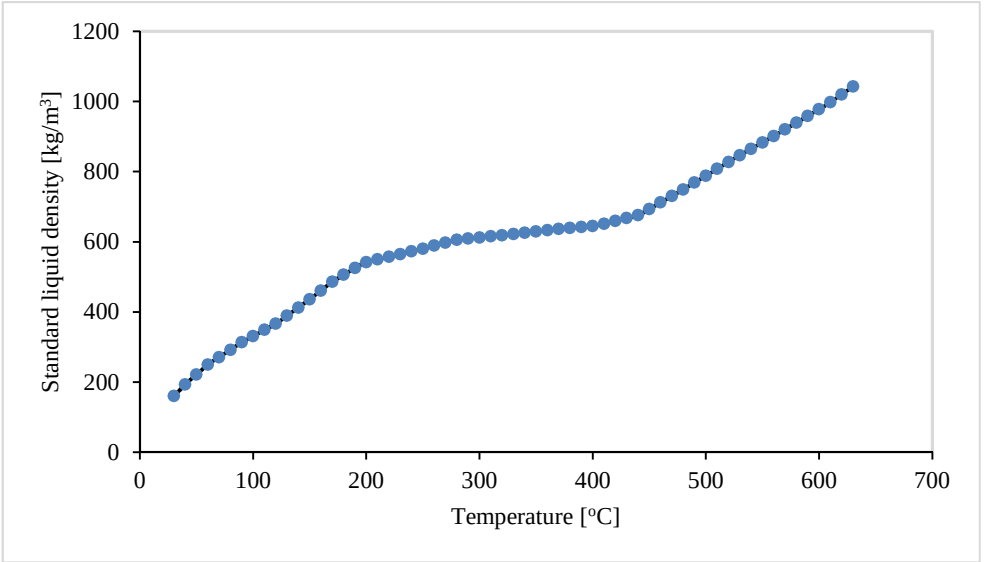


Fig. 9. Algal bio-oil standard liquid density vs temperature

Conclusion

In conclusion, this study discussed the characteristics of macroalgae *Ulva prolifera* derived bio-oil using Aspen HYSYS modelling tool. Macroalgae derived bio-oil has the potential to produce higher heating value renewable and sustainable fuel. Hence studying the characteristics of the bio-oil helps to determine the optimum treatment conditions to maximise the quality and effectiveness of the bio-oil to be produced ultimately. The current study is based on modelling and simulation techniques that provide valuable insight about macroalgae based bio-oil as a function of the influencing parameters like temperature, kinematic viscosity, and weight percent of the bio-oil assay, cut yield behaviour, and standard liquid density. The findings of the current study would support further optimisation efforts on process parameters and design of process layout. The sudden change in temperature may be attributed to the increase in cut yield of gas oil and residue composition of the bio-oil components and reduction of other components like light distillates and paraffins due to thermal degradation which resulted in the substantial increase in kinematic viscosity. Above a temperature of 300 °C, the weight percentage of aromatic components increased steadily. The increase in percentage composition of the aromatic components is due to the reduction of the paraffinic components. According to the simulation results the cut yield of off gases, light naphtha, heavy naphtha, light distillate, heavy distillate, gas oil and residues are shown at specific final boiling point (FBP) temperatures of 70 °C, 70 °C - 110 °C, 110 °C - 221.1 °C, 221.1 °C - 304.4 °C, 304.4 °C - 371.1 °C, 371.1 °C - 537.8 °C and 537.7 °C respectively. The density of the liquid bio-oil was steadily increasing until a temperature of 200 °C. In depth research is needed, however, to validate the simulation results and optimise the process conditions for large-scale production. Overall, bio-oil derived from macroalgae has the potential to play a significant role in the transition to a more sustainable and renewable energy future.

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