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REVIEW



Conversion of biomass to biofuels and life cycle assessment: a review

Ahmed I. Osman¹ • Neha Mehta^{1,2} • Ahmed M. Elgarahy^{3,4} • Amer Al-Hinai⁵ • Ala'a H. Al-Muhtaseb⁶ • David W. Rooney¹

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Abstract

The global energy demand is projected to rise by almost 28% by 2040 compared to current levels. Biomass is a promising energy source for producing either solid or liquid fuels. Biofuels are alternatives to fossil fuels to reduce anthropogenic greenhouse gas emissions. Nonetheless, policy decisions for biofuels should be based on evidence that biofuels are produced in a sustainable manner. To this end, life cycle assessment (LCA) provides information on environmental impacts associated with biofuel production chains. Here, we review advances in biomass conversion to biofuels and their environmental impact by life cycle assessment. Processes are gasification, combustion, pyrolysis, enzymatic hydrolysis routes and fermentation. Thermochemical processes are classified into low temperature, below 300 °C, and high temperature, higher than 300 °C, i.e. gasification, combustion and pyrolysis. Pyrolysis is promising because it operates at a relatively lower temperature of up to 500 °C, compared to gasification, which operates at 800–1300 °C. We focus on 1) the drawbacks and advantages of the thermochemical and biochemical conversion routes of biomass into various fuels and the possibility of integrating these routes for better process efficiency; 2) methodological approaches and key findings from 40 LCA studies on biomass to biofuel conversion pathways published from 2019 to 2021; and 3) bibliometric trends and knowledge gaps in biomass conversion into biofuels using thermochemical and biochemical routes. The integration of hydrothermal and biochemical routes is promising for the circular economy.

Keywords Biomass · Biofuel · Thermochemical · Biochemical · Life cycle assessment

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Introduction

In recent decades, urbanisation, modernisation and industrialisation linked to energy production and utilisation have been a fundamental loop in various economic, scientific and social sectors (Ahmad Ansari et al. 2020; Shrivastava et al. 2019). The depletion of non-renewable fuel sources, accompanied with greenhouse gas emissions, has become a critical issue (Fawzy et al. 2020; Osman et al. 2021). Therefore, the necessary shift for exploring alternative options to overcome the world-scale looming energy crisis, considering the environmental concerns and its mitigation, while confronting the spiralling energy demand has become an urgent need of the hour.

Biomass, unlike other sustainable energy sources such as wind, solar, geothermal, marine and hydropower, can directly produce fuel along with chemicals (Quereshi et al. 2021; Farrell et al. 2019; Farrell et al. 2020). Thus, it is not feasible to substitute fossil-based fuels with the aforementioned sustainable energy sources; hence, biomass



utilisation to produce fuel and chemicals is required (Bharti et al. 2021). Biomass is classified as non-lignocellulosic or lignocellulosic in nature and exists in various forms such as woody, herbaceous, aquatic debris, farming manure and byproducts and other forms (Osman et al. 2019; Kaloudas et al. 2021). Various technologies are used to convert biomass into fuel or chemicals, such as gasification, combustion, pyrolysis, enzymatic hydrolysis routes and the fermentation processes (Abou Rjeily et al. 2021; Peng et al. 2020).

A recent review discussed integrating hydrothermal and biochemical routes in biomass utilisation from a circular bioeconomy approach (Song et al. 2021). The thermochemical methods usually involve a high energy intake along with solvent or catalyst addition. Meanwhile, the biochemical route has a lengthy cycle period and is less efficient in breaking down recalcitrant biomass materials. Thus, combining those two routes can be promising by incorporating the benefits of both methods in biofuel processing. They proposed a schematic route where hydrothermal routes are being used in the pretreatment stage to prepare the appropriate biomass feedstock for the following biological routes to improve the overall process efficiency and final product yields and vice versa,

as shown in Fig. 1. There are unprecedented challenges with the integration of thermochemical and biochemical routes. For instance, the catalysts or solvent utilisation of the thermal routes may result in poisoning or kill the microorganism or generate various inhibitors that can affect the biological progress routes. Furthermore, this integration may lead to additional costs.

Identifying sources of biofuels such as biodiesel and biochar can potentially reduce the environmental impacts of fossil fuels (Balajii and Niju 2019; Gunarathne et al. 2019). Biofuels can also counter the increasing use of fossil resources and prevent pressure on non-renewable sources (Peng et al. 2020; Hassan et al. 2020). However, it is important to use practical, scientific and robust tools to evaluate the real benefits of using biofuels over conventional energy sources (Chamkalani et al. 2020; Kargbo et al. 2021). Life cycle assessment (LCA) has been identified as a comprehensive evaluation approach (Astrup et al. 2015) to measure environmental impacts over the entire production chain of biofuels (Collotta et al. 2019).

Therefore, this review aims to critically evaluate existing biomass to biofuel pathways and associated studies

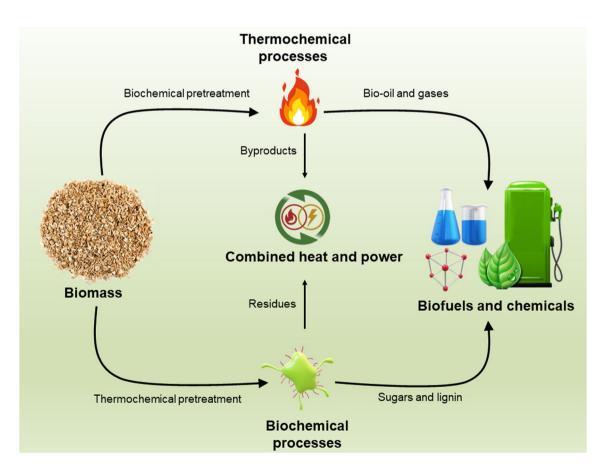


Fig. 1 Integration of hydrothermal and biochemical routes in biomass utilisation from a circular economy approach. Firstly, the biomass is pretreated using a biochemical process for the following thermochem-

ical route or vice versa by thermochemical pretreatment for the following biochemical route and eventually producing biofuel or chemicals



which evaluated environmental impacts for the entire life cycle. The main objectives were to: (1) critically review recent advances in biofuel production processes, (2) analyse existing LCA studies and highlight key methodological approaches and present critical findings.

Bibliometric analysis

Bibliometric research methodology: TOPIC: 'biomass pyrolysis' OR TOPIC: 'biomass gasification' OR TOPIC: 'biomass combustion' OR TOPIC: 'biomass hydrothermal liquefaction' OR TOPIC: 'biomass torrefaction' OR TOPIC: 'biomass fermentation' OR TOPIC: 'anaerobic digestion' AND TOPIC: 'biomass into fuels' AND TOPIC: ('thermochemical' OR 'thermo-chemical') AND TOPIC: 'biochemical'. The document type selected in the bibliographic search was articles, the timespan: All years.

It is a complex process to assess the sustainability of biofuels. This is because the use of energy crops can cause the transformation of natural and agricultural land for the cultivation of these crops. Moreover, various technical pathways range from biological to thermochemical conversion processes, all involving range of products and co-products. Therefore, it is imperative to conduct the LCA of the biofuel production chain. This study provides an overview of LCA approaches by recent studies. Keywords: 'biomass', 'biofuel', 'life cycle assessment', 'environmental impact assessment', were used for literature search in the Web of Science database. Forty most complete studies published from 2019–2021 were selected for analysis in the present study.

Biomass conversion technologies

Harnessing various renewable energy resources is considered affordable, reliable and sustainable solutions for their excessive availability, such as agriculture wastes, domestic wastes, forest residues, industrial wastes and human excreta. Among them, biomass is the most significant contributor with 9% (~51 EJ) of the global overall primary energy supply, out of which about 55% is traditionally used in daily living activities such as heating and cooking, especially in developing countries (Chan et al. 2019). Slade et al. (2014) revealed the possibility of biomass, wastes and energy crops for sharing up to ~100 EJ in the world energy supply (Slade et al. 2014).

Economically, biomass combustion is not the best strategy to utilise biomass because of causing severe environmental damage as well as not recovery of the total energy stored in the substrates (Ullah et al. 2015). In this context, biomass conversion into solid, liquid and gaseous forms is deemed an efficient and green energy supplier for various

sectors comprising heat, power and transport fuel (Kargbo et al. 2021; Wang et al. 2020a). For this purpose, two grouped distinct routes, namely thermochemical and biochemical, are currently used. Thermochemical methods use the whole biomass in the presence of a heat source and controllable oxygen atmosphere to modify it to different energy forms.

In contrast, biochemical methods employ enzymes, bacteria or other engineered organisms to transform it to liquid fuels such as drop-in-biofuels (Shen and Yoshikawa 2013; Singh et al. 2016). During the past decades, the biomassderived fuel synthesis process has upgraded from the firstgeneration biofuel to fourth-generation biofuel, passing by second and third generations. Food crops, inedible biomass, macro/microalgal biomass and genetically bioengineered algal and microbial systems-based biofuels are examples for the first, second, third and fourth generations, respectively (Martin 2010; Adelabu et al. 2019; Aro, 2016; Ben-Iwo et al. 2016). Innumerable biomass-based fuels, chemicals and organic compounds such as methane, ethane, propane, butane, ethylene, methanol, ethanol, butanol, dimethyl ether, ammonia, acetic acid, formaldehyde, gasoline, diesel, wax, paraffin, bio-jet fuels and others have been produced throughout different biomass to liquid routes and presently available in the markets throughout the world (Demirbas and Demirbas, 2010).

Thermochemical conversion methods

Thermochemically, diverse technologies including direct combustion, torrefaction, hydrothermal liquefaction, pyrolysis and gasification have been implemented to produce liquid fuels from biomass, as shown in Fig. 2. Practically, the biomass is decomposed in controllable operational conditions to produce solid, liquid and gas (syngas), which need a supplementary catalytic promotion process to produce liquid fuels called drop-in biofuel. One of its most important features is its capabilities to utilise any biomass type as a biomaterial feedstock, unlike biochemical conversion methods (Raheem et al. 2015).

The thermal-based processes are also classified into low-temperature, which typically operate at < 300 °C, such as torrefaction, hydrothermal carbonisation and high-temperature that operate at > 300 °C, such as gasification, combustion and pyrolysis biomass conversion methods (Quereshi et al. 2021). The direct combustion (flaming or smouldering) of biomass to produce energy usually operates in the temperature range of 1000-2000 °C in the presence of air. However, there are emissions associated with such processes as NOx and pollutants (Osman 2020). Hence, gasification which typically operates at 800-1300 °C is seen as a potential substitute to produce energy and chemicals as well. The synthesis gas produced from the gasification process can



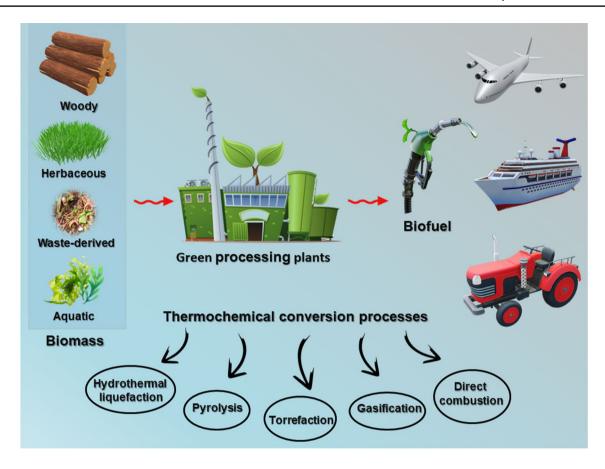


Fig. 2 Thermochemical conversion of biomass, including hydrothermal liquefaction, pyrolysis, torrefaction, gasification and direct combustion processes. Different types of biomass are treated in pro-

cessing plants into liquid biofuels for energy purposes or for the production of value-added chemicals

be used for electricity production, as well as the conversion into liquid fuel via the Fischer–Tropsch route. Interestingly, pyrolysis is considered a promising route requiring a lower temperature of up to 500 $^{\circ}\text{C}$ compared to the gasification process.

Direct combustion

Biomass utilisation as fuel was closely linked with the beginning of human civilisation. Moreover, it is the highest contributor source of clean energy globally (Mladenović et al. 2018). Biomass combustion is described as a group of chemical reactions involving carbon dioxide and water formation resulting from the transformation of carbon and hydrogen, respectively, by oxidation reactions. Improper oxygen quality can result in incomplete combustion associated with release of atmospheric polluters (i.e. CO, NO_X , SO_2 and particulate matter) (Yang et al. 2020). Nowadays, the usage of effective fabricated combustion systems such as combustion control systems that simultaneously use conventional and alternative biomass resources has become a predominant feature industrially.

In waste-to-energy facilities, biomass can be separately burned or combined in co-firing with coal to produce steam, used later in electricity generation. The net electricity efficacy generated from coal/biomass in co-firing power plant system varied from 36 up to 44% based on as-used strategy and biomass specifications (quantity and quality). Despite the present feasibility of 20% of co-firing as energy basis in addition to a theoretical achievability of 50%, only less than 5% and sometimes surpasses to 10% of biomass contribution continuously. It has been estimated that only 10% of biomass usage in co-firing systems in power plants can decline the release of CO₂ to the atmosphere from 45 to 450 million tonnes/year by 2035 (Sahu et al. 2014). Considering the physicochemical properties of fuel and its required volume to air for avoiding any troubles in the fuel-to-air ratio, excessive air can be forwarded to the reaction to control the temperature of the burning system and ensure complete combustion (Vicente and Alves, 2018). Majorly, combustion physicochemical features of biomass can be categorised into macroscopic and microscopic. Comprehensively, macroscopic features are provided by macroscopic analyses such as proximate analysis such as moisture tenor, sulphur,



particle magnitude, calorific value, bulk density, fixed carbon and ash fusion. This is coupled with ultimate analysis such as C, H, N, O and S %, whereas microscopic analyses include chemical, thermal and mineral data (Khodaei et al. 2015).

Main combustion reactions are as follows: (1) drying out of biomass, (2) pyrolysis, (3) pre-combustion reaction, (4) primary combustion, (5) secondary combustion and (6) effluent stack gas. Parameters that influence the combustion process include biomass magnitude, specific gravity, moisture tenor, ash percentage, elemental composition and anatomical structure as lignin, cellulose and hemicellulose. Different researchers highlighted that about 95–97% of the global bioenergy production is based on direct biomass combustion (Fouilland et al. 2010; Zhang et al. 2010). Given the massive quantity of ash produced from coal burning (~780 million tonnes), a less quantity of biomass ash (~ 480 million tonnes/year) was assumed to be generated coincided with the burning of 7 billion tonnes of biomass (Vassilev et al. 2013; Izquierdo and Querol, 2012). Retrofitting investment cost characterised to various power plants was USD 430–500/kW, USD 760–900/kW and USD 3000–4000/kW for co-feed plants and separate feed plants indirect co-firing, respectively. These investigated costs were totally more minuscule than the specified outlays of 100% biomass power generations facilities (Sahu et al. 2014). Table 1 presents the energy content in MJ/kg for several kinds of biomass, whereas Table 2 displays the differences in the physicochemical characteristics of biomass and coal-based fuels.

Table 2 Physicochemical characteristics of biomass and coal-based fuels (Demirbas, 2004)

Property	Biomass	Coal
Particle size (mm)	3	100
Fuel density	500	1300
Carbon content*	43-54	65-85
Oxygen content*	35–45	2-15
Sulphur content*	Max 0.5	0.5-7.5
AL ₂ O ₃ content*	2.4-9.5	15-25
SiO ₂ content*	23-49	40-60
K ₂ O content*	4–48	2–6
Fe ₂ O ₃ content*	1.5-8.5	8-18
Ignition temperature	418–426	490-595
Heating value	14–21	23–28

^{*(}wet % of dry fuel)

As a solid fuel, fossil-based fuels are still dominating this sector for power generation and heat. However, biomass utilisation in the co-combustion along with fossil-based fuel is seen as a cost-effective and interesting option (Variny et al. 2021). Co-combustion of those two feedstocks offers higher power generation than biomass alone, and biomass ash is acting as a sulphur capture and mitigates the sulphur oxide emissions. On the other hand, some challenges arise when mixing fossil-based coal with biomass, leading to higher corrosion, slagging and fouling due to the high alkali contents within the biomass. In fact, the projected gradual

Table 1 Energy contents of biomass sources, in MJ/kg

Biomass type	Energy content (MJ/kg)	Biomass type	Energy content (MJ/kg)	Biomass type	Energy content (MJ/kg)
Greenwood	8	Sour cherry stalk	17.59	Coconut shell	20.00
Manure	8.650	Tobacco leaf	17.97	Oven dry plant matter	20
Sewage	10.510	Corncob	17.99	Wood bark	20.3
Mustard stalk	10.73	Soybean cake	18.30	Hazelnut shell	20.47
Corn stover	10.730	Peanut shell	18.46	Spruce wood	20.5
Rice husk	13.524	Cereals	18.61	Wood bark	20.57
Peat	15.30	Ailanthus wood	18.93	Peach stones	20.657
Cotton gin	15.500	Tobacco stalk	19.02	Olive cake	21.57
Olive refuse	15.77	Hazelnut seed coat	19.2	Olive husk	21.8
Fuelwood	16.10	Eucalyptus (Grandis)	19.35	Oak bark	22
Peach bagasse	16.24	Poplar	19.38	Olive pits	22
Potatoes	17.00	Colza seed	19.38	Apricot stones	22.082
Potato peel	17.18	Spruce wood	19.45	Tuncbilek lignite	23.212
Corncob	17.3	Coir pith	19.50	Rape seed	27.80
Sugar beet	17.40	Black locust	19.71	Tyres	36.800
Cotton cake	17.50	Groundnut shell	19.80	Methane gas	55

Data sourced from: Sami et al. 2001; Demirbas 2001; Demirbas 2005; Atimtay 2010; Haykiri-Acma 2003; Borjesson 1996; Raveendran and Ganesh 1996; Erol et al. 2010; Friedl et al. 2005



shutdown of fossil-based power plants will limit this cocombustion approach. Technologies such as gasification and pyrolysis should be the main focus, to be competitive and fully available in the near future.

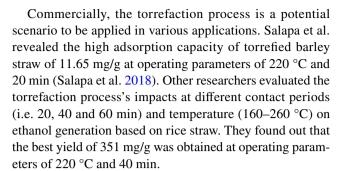
Torrefaction

Torrefaction is an endothermic pretreatment pathway that mainly proceeded at temperatures ranging from 200 to 300 °C and a non-accelerated heating rate of less than 50 °C/ min in an oxygen-free atmosphere. This process is used for upgrading the solid biomass to produce a torrefied product used later as a suitable alternate to coal (Cahyanti et al. 2020). Three transformational reactions, including: volatilisation, polymerisation and carbonisation, occur during the torrefaction process. The process efficacy is influenced by temperature, reaction time, particle magnitude, carrier gas type and flow, catalyst and performance index (Chen et al. 2021). This strategy significantly improves the physicochemical properties of utilised biomass such as hydrophobicity, grindability, mass/energy density, ignitability, moisture expelling and homogeneity (Chen and Kuo 2011).

Commonly, the torrefied product is termed bio-coal or green coal and biochar when used as fuel and soil amendment. Based on the mode of operation, the torrefaction process can be practically classified into dry, wet and steam modes (Barskov et al. 2019). In dry torrefaction, biomaterial feedstock can be torrefied in non-oxidative; for example, nitrogen and carbon dioxide are carrier gases or oxidative mediums such as air, flue gases and other streams with various oxygen concentrations at working temperatures ranging from 200 to 300 °C. Attributing to oxygen presence, the oxidative scenario has a better reaction rate than the other non-oxidative scenario and minimises the reaction time (Thanapal et al. 2014; Lynam et al. 2011).

Contrarily, in the wet torrefaction, the biomass is torrefied in a wet environment, typically water and dilute acids at 180–260 °C and 5–240 min for surrounding temperature and holding time, respectively, and the produced solid is termed as hydrochar. Under these subcritical conditions, physicochemical properties of water such as density, diffusivity, dielectric constant and viscosity alter and improve the biomass degradation process, which further upgrades the torrefaction process (Bach and Skreiberg 2016; Balat et al. 2008).

Besides the two mentioned routes, steam torrefaction by introducing steam with elevated temperature and pressure is conducted to torrefy the biomass at 200–260 °C and 5–10 min for environmental temperature and contact period, respectively. The subsequently accelerated venting of the pressure will allow steam to bulge the biomass and split it with minor loss in the feedstock.



Additionally, they confirmed that the torrefied biomass enhanced the yield of ethanol production at a value of 50.67% compared with the untreated one (Chiaramonti et al. 2011; Sheikh et al. 2013). An enhancement in the ethanol production yield is based on torrefied sugarcane bagasse and waste jute caddies by 19.34 and 20.28%, respectively, compared with the untreated biomass (Chaluvadi et al. 2019).

Igalavithana et al. demonstrated that torrefied product could be positively utilised in soil improvement because it increases air space, water retention efficiency, plant prosperity, microbial community and enzymes activity (Igalavithana et al. 2017). Ogura et al. observed an increase in the growth of the J. curcas when it was exposed to varied ratios of 250 °C torrefied biomass of 1, 3 and 5% (Ogura et al. 2016). The feasibility of using torrefaction condensate in plant safeguarding pathways (pest repellent, insecticide and herbicide) was confirmed considering its minimum amounts of polycyclic aromatic hydrocarbons and phenolic compounds. Comparing the as-generated condensate resulting from different biomass feedstocks such as pine bark, forest residue, wheat straw and willow biomass, the condensate based on willow had the best pesticide performance (Hagner et al. 2020). Table 3 presents more details about the physicochemical properties of numerous biomass after the torrefaction process.

Hydrothermal liquefaction

Hydrothermal liquefaction is defined as a thermochemical pathway at which the lignocellulosic feedstock, whether wet or dry, is effectively decomposed into renewable liquid fuel (Guo et al. 2015). Based on the mode of operation, it can be divided into two main subclasses: (1) indirect liquefaction and (2) solvent liquefaction (Mika et al. 2018). In the first scenario, biomass or its liquefied products are first converted into syngas followed by a subsequent fuel synthesis (i.e. alcohols and alkanes). In contrast, in the second, direct conversion of biomass into liquid fuel occurs by the action of proton solvents such as water, alcohols, phenols, sulpholane and ionic liquid. Solvent liquefaction has the priority to be implemented over the other scenario because of its remarkable merits, moderate operational conditions and higher yield of products (Gollakota et al. 2018).



Table 3 Physicochemical properties of biomass after torrefaction

Biomass	Fixed carbon (wt.%)	Volatile matter (wt.%)	Ash content	O/C ratio	H/C ratio	Higher heating value (MJ/kg)	References
Cotton stalk	16.48–34.01	53.62-75.44	4.79–9.88	0.25-0.83	0.06-0.14	18.68-25.43	Chen et al. (2014a),
Corn stalk	18.41–41.19	35.30–69.32	9.18–20.52	0.18-0.85	0.06-0.15	18.26–23.61	Chen et al. (2014b), Chen et al. (2014c)
Rice straw	17–48	44.5–79.6	6–8	0.24-0.81	0.09-0.16	19.0–28.6	Nam and Capareda (2015)
Oil palm fibre	5–25	3–48	6–7	0.22-0.62	0.02 – 0.10	11.13-23.98	Chen et al. (2013)
Cryptomeria japonica	12–25	5–54	0	0.23-0.63	0.02-0.09	21.94–28.81	
Coconut fibre	2–27	2–40	0.5-3	0.22 - 0.55	0.02 - 0.09	11.86-26.46	
Eucalyptus	15–28	10-61	0	0.28-0.64	0.02-0.09	20.59-26.28	
Bamboo	25.05-47.03	48.05-70.20	1.43-1.95	0.34-0.63	0.07 - 0.10	21.02-27.26	Chen et al. (2012a),
Rice husk sugar cane bagasse Madagas- car almond	22.27–41.17	33.33–61.88	13.18–23.11	0.32-0.70	0.06-0.10	17.68–21.48	Chen et al. (2012b)
Oil palm	32.58-43.67	39.60-53.76	9.97-13.52	0.29-0.54	0.06-0.08	20.61-23.54	
Reed canary grass	13.3-21.3	70.5-80.3	6.4-8.3	0.67-0.74	0.11-0.13	20.0-21.8	Bridgeman et al.
Wheat straw	15.6–38	51.8-77	7.4–10.2	0.48-0.72	0.10-0.12	19.8-22.6	(2008)
Olive tree pruning	20.4	74.5	2.4	0.89	0.16	N/A	Martin-Lara et al. (2017)
Olive trimmings	16.54-32.57	55.96-77.65	3.87-7.87	0.18-0.70	0.08-0.12	19.6-28.4	Martin-Lara et al.
Olive pulp	19.98-50.40	41.99-77.22	1.50-6.67	0.24-0.72	0.08-0.12	20-26.8	(2017), Volpe
Orange peel waste	38.9-62.2	37.8-61.1	3.3-5.6	0.24-0.65	0.06-0.10	21.45-28.74	et al. (2015)
Lemon peel waste	35.7-61.8	38.2-64.3	3.5-6.3	0.22 - 0.68	0.06-0.10	21.09-28.75	
Jatropha seed residue	13.5–35.5	55.5–79.8	6.25–7.60	0.29-0.49	1.14–1.50	21.8–23.8	Hsu et al. (2018)

The main process parameters that directly influence the hydrothermal liquefaction process include biomass composition, particle size, pressure, temperature, heating rate, residence time, feed/solvent ratio and presence of the catalyst and reducing gas. Generally, the hydrothermal liquefaction process operates at mild operational parameters of temperature in the temperature range of 250–500 °C and pressure of 5–35 MPa and contact periods of 5–60 min. These processes are conducted in the presence of solvents such as sub/supercritical water, organic solvents and mixed solvents such as the combination of water+organic solvent (Yang et al. 2019; Akalın et al. 2017).

The utilisation of water as a solvent has numerous advantages over conventional organic solvents due to its natural occurrence in biomass and eco-friendliness. On the contrary of water state at ambient conditions, the compressed water in a liquid state at the following critical conditions of temperature and pressure of 374 °C and 22.064 MPa, respectively, generates higher ionic products (i.e. $\rm H_3O^+$ and $\rm OH^-$) ions. Proximity to the mentioned critical conditions, the physicochemical properties of water such as density, viscosity, dielectric constant, polarity and permittivity change (Arun et al. 2021). Decrease in the

viscosity as well as dielectric constant and weakening of water hydrogen bonds consequently enhance the solubility of hydrophobic organic compounds associated with an improvement in the catalytic activity of acid-base reactions. This results in biomass conversion into four main fractions: bio-crude fuel (liquid), water-soluble products, solid residue and gases.

Moreover, hydrothermal liquefaction is regarded as lesser energy consumption and a higher efficiency strategy than pyrolysis because of the better physicochemical properties. The produced bio-crude from the hydrothermal liquefaction process has an oxygen content of 10-20 wt.% and a heating value of 30-35 MJ/kg, which is typically higher than those obtained from the conventional pyrolysis process (Guo et al. 2019). During the hydrothermal liquefaction process, the oxygen contained in the biomass is partially eliminated by dehydration, decarboxylation and decarbonylation reactions associated with producing CO, CO₂ and H₂O. Despite its higher quality, the higher oxygen content produces a highly sticky and acidic bio-crude product with a low heating value. Distinctly, its quality enormously varies depending on the operational parameters and biomaterial feedstock composition (Scarsella et al. 2020).



Hence, the importance of downstream upgrading treatment such as catalytic cracking and hydrotreating for the obtained bio-crude cannot be ignored for further utilisation as a transportation drop-in fuel. In situ upgrading using various acidic or alkaline homogenous catalysts such as HCl, H₃PO₄, K₂CO₃, Na₂CO₃, NaOH, KOH and Ca(OH)₂ and heterogeneous catalysts has been studied in detail (Perkins et al. 2019). Despite its cost and energy-saving nature, it has some disadvantages, such as operating at high pressure that results in the necessity of (1) solid/water slurries reliable pumping, (2) suitable unit metallurgy to avoid the potentially corrosive nature of slurries at the operational parameters of elevated pressures and mild temperatures and (3) usage of heat exchangers with high surface areas to overcome the problem of low heat transfer coefficients (Beims et al. 2020). Li et al. (2017) liquefied wheat straw biomass at a pre-adjusted temperature of 270 °C for 120 min, and the resulting oil was forwarded for manufacturing of bio-polyols and polyurethane foams (Li et al. 2017).

Crude oil originated from liquefaction of bark biomass was directed to produce bio-based phenol-formaldehyde formable resole as reported by Li et al. (Li et al. 2016). Spent

coffee grounds of approximately 15 and 17.4% for lipids and proteins, respectively, were liquefied to output crude bio-oil under N₂ atmosphere at (200–300 °C) and (5–25 min). The highest yield of acetone-recovered bio-crude oil (47.3 wt%) was obtained at 275 °C and 10 min with an estimated higher heating value of 31.0 MJ/kg⁻¹, better than that of spent coffee grounds of 20.2 MJ/kg (Yang et al. 2016). Xiu et al. performed a study on swine manure composed of < 1%, 17.1%, 22.3% and ~35% for lignin, crude protein, ash content and saccharide, respectively. They successfully liquefied into bio-crude oil under N₂ atmosphere at a temperature range of 260-340 °C and a contact period of 15 min with a yield of 24.2 wt.% and higher heating value of 36.05 MJ/kg (Xiu et al. 2010). Table 4 presents the proximate as well as ultimate investigations of crude bio-oil prepared from several biomaterials through the hydrothermal liquefaction process.

Pyrolysis

Pyrolysis is counted as one of the most as-used thermochemical scenarios to degrade the carbonaceous biomass, such as cellulose, hemicellulose and lignin (Aravind et al.

Table 4 Proximate and ultimate analyses of bio-crude oil prepared from several biomass through hydrothermal liquefaction process. Where, the higher heating value is abbreviated as HHV

Feedstock		ntal comp er Hydro				Physicoche ucts	emical propert	ies of th	e prod-	References
	C (%)	H (%)	N (%)	O (%)	S (%)	Ash %	Moisture %	HHV	Yield %	
Spirulina algae	68.9	8.9	6.5	14.9	0.86	_	_	33.2	32.6	Vardon et al. (2011
Anaerobic sludge	66.6	9.2	4.3	18.9	0.97	_	-	32.0	9.4	
Cladophora glomerata	26.8	3.53	2.14	20.48	0.22	36.5	9.1	10.29	_	Plis et al. (2015)
Nannochloropsis gaditana	40.3	5.97	6.30	14.49	0.37	28.3	4.1	18.53	_	
Almeriansis	73.2	9.3	5.1	0.8	11.7	35.8	20.0	_	42.6	López Barreiro et al. (2015)
Gaditana	74.2	9.3	4.0	0.6	11.8	36.2	12.4	_	50.8	
Swine manure	71	8.9	4.1	0.21	14.2	35	-	_	61	Toor et al. (2011)
Algae Dunaliella tertiolecta	63.55	7.66	3.71	_	25.8	30.7	_	_	25.8	
Porphyridium	66-83	5-11	0-12	0-1	8-27	22.8-36.9	_	_	5-25	
A.esculenta	73.8	8	3.8	14	0.8	_	_	33.8	17.8	Anastasakis and Ross (2015)
L.digitata	70.5	7.8	4.0	17	0.7	_	_	32	17.6	
L.Saccharina	31.3	3.7	2.4	26.3	0.7	24.2	9.2	12	_	
Fucus vesiculosus	32.88	4.77	2.53	35.63	2.44	11.8	_	15.0	_	Ross et al. (2008)
Laminaria hyperborea	34.97	5.31	1.12	35.09	2.06	11.2	_	16.54	_	
Miscanthus	46.32	5.58	0.56	41.79	0.2	2.1	_	19.08	_	
A.azuera	40.82	5.56	0.63	52.99	_	10.61	6.03	52.99	_	Aysu et al. (2016)
Nannochloropsis Oceana	77.6	4.9	3.4	_	0.3	_	_	37.70	54.2	Caporgno et al. (2016)
Aspen wood	75.2	8.2	0.5	15.8	0.3	0.48	3.8	34.3	_	Pedersen et al. (2016)
Arthrospira platensis	74.5	10.2	6.8	7.5	1.0	_	_	38.65	30	Lavanya et al. (2016)
Tetraselmis	71.4	9.5	5.7	12.3	1.1	_	_	35.58	29	
Nannochloropsis Salina	55.16	6.87	2.73	33.97	1.27	2.48	4.95	25.40	_	Toor et al. (2013)
Seaweed meal	43.99	5.95	5.21	36.13	1.02	7.7	7.92	18.35	_	Ferrera–Lorenzo et al. (2014)
Laurel algae	48.97	6.38	3.02	41.63	_	10.53	9.95	19.77	_	Ertas and Alma (2010)



2020). This results in the generation of solid, liquid and gas biofuels in an oxygen-free atmosphere via endothermic reaction (Perkins et al. 2018). The yield of the pyrolytic products is influenced by factors, including feedstock composition such as structure and complexity. This is also coupled with pyrolysis impacting factors such as particle size, temperature, heating rate, residence time, inert gas type, inert gas flow, catalyst type and others (Azizi et al. 2018).

Complex reactions such as dehydration, decarboxylation, decarbonylation, hydrogenation, isomerisation, aromatisation, depolymerisation and charring are involved in the thermal decomposition process of biomass. Typically, pyrolysis of biomass undergoes the following steps: (1) transfer of heat from its source to biomass to initiate the reaction, (2) elevated pyrolysis temperature of primary vapours contributes to volatiles and char formation, (3) because of the influx of hot vapours to the biomass, heat migration continues between unpyrolysed fuel and hot volatiles, (4) volatiles condensation associated with secondary reactions leads to tar formation and (5) autocatalytic secondary pyrolysis reactions take place in conjugation with primary pyrolytic reaction (Dabros et al. 2018).

The impact of different pyrolysis operational parameters occurs at different stages such as dehydration, decomposition and reforming. With elevated heating rate, minimum vapour contact periods and a surrounding temperature of 500 °C, liquid yields can be maximised (Chintala, 2018). These conditions directly prohibit (1) thermal or catalytic cracking of the primary decomposition products due to char presence to lesser non-condensable gas molecules as well as (2) their polymerisation to char (Kasmuri et al. 2017). Table 5 presents different working modes of the pyrolysis process. Other pyrolysis types such as catalytic and assisted microwave, carbon dioxide, additives, solar and hydro-pyrolysis can be performed to upgrade the product's yields.

Slow pyrolysis is a process in which organic materials are slowly heated at a low heating rate between 5 and 50 °C min-1 and the longest residence time above 10 s in the absence of oxygen, typically producing about 80% of carbon as the main product (Antoniou and Zabaniotou, 2013). Fast pyrolysis is a strategy by which organic materials are quickly heated at faster heating rates of > 103 °C s⁻¹ and shorter contact periods of up to 3 s without air existence.

shorter contact periods of Table 5 Different working

modes of the pyrolysis process (Bulushev and Ross, 2011)

Mode of action	Working tem-	Residence time (seconds)	Yields	(wt. %)	
	perature (°C)		Char	Liquid	Gas
Slow	450	Very long (>30)	35	30 (70% water)	35
Medium	500	Moderate (10-30)	25	50 (50% water)	25
Fast	500	Short (<2)	13	70 (30% water)	12
Flash	500	Very short (< 0.5)	< 13	75	< 12
Gasification	>800	Long	10	5	85

The primary product is liquid oil (Bridgwater, 2012a). Ultrafast or flash pyrolysis is a highly accelerated pyrolysis at a high heating rate with major gas and oil products. The operating conditions are as follows temperature (medium–high $(700-1000\,^{\circ}\text{C})$, shortest residence time and fastest heating rate. Yields of outputs are: (1) liquid condensate (10-20%), (2) gases (60-80%) and (3) char (10-15%) (Priharto et al. 2020).

Catalytic pyrolysis is a process in which catalysts such as natural zeolite, Cu/Al₂O₃, Co/Mo/Z, Zeolite-B, Fe₂O₃ and Ni-CaO-C are used to decrease the reaction operating temperature and increase the selectivity towards desired products. This process is used to optimise the biomass conversion into liquid fuels with improved physicochemical properties (Cai et al. 2019; Chai et al. 2020). Catalytic co-pyrolysis of biomass and plastic waste showed promising results in upgrading the oil quality by removing oxygen from the biomass and producing more aromatics and olefins (Wang et al. 2020b). This is due to the high hydrogen and carbon contents within the plastic waste and consequently acting as a hydrogen donor in the catalytic co-pyrolysis process and thus eliminates the oxygenated compounds. This approach is seen as a sustainable, efficient and economical approach to upgrading the bio-oil quality, along with extending the catalyst's lifetime.

Microwave-assisted pyrolysis is a new thermochemical process that transforms biomaterial feedstock into liquid oil using microwave input heat energy. In contrast to the conventional process, microwave-assisted pyrolysis is a more effective and controllable technique. CO_2 -assisted pyrolysis is a process by which CO_2 is delivered as a reactive medium instead of inert N_2 in utilising the pyrolysis process and enhancing the syngas yield and declines the produced oil but also decreases the greenhouse gas emissions (Kwon et al. 2019).

Additive—assisted pyrolysis is a type of pyrolysis at which metal salts such as sodium, potassium and calcium salts and inorganic additives (zeolites, biochar) are added and thus having some advantages over conventional pyrolysis. It has a great potential to decrease the required operating temperature, cracking time and solid residue yield, in addition, to increase the cracking efficacy of wastes and improves the quality of pyrolysis products (Wang et al. 2019a).



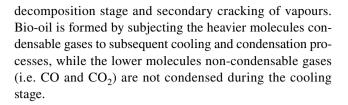
Solar pyrolysis is a process in which solar renewable energy can be introduced as thermal input sources rather than electrical energy that maximise biofuel production and reduce CO₂ emissions. Hydropyrolysis is a particular type of pyrolysis at which biomaterial feedstock is decomposed with the assistance of high pressurised hydrogen. Using the mentioned technique above, a higher yield of hydrocarbons with improved structures can be attained (Marcilla et al. 2013).

Pyrolysis products can be categorised as solid, liquid and gases that can be exploited to generate chemicals, energy, electricity and transportation fuels. Proximate and ultimate analyses are beneficial to characterise the obtained products. Char, pyrolytic char or biochar, is the produced solid, chemically not pure carbon, and contains carbon as the main constituent, hydrogen, nitrogen, ash and some volatiles. The highly porous char is used in several applications as adsorbent and soil amendment for wastewater treatment and enhancement of crop yields. Bio-oil (tar) is a dark brown, sticky liquid produced from the thermal degradation of biomass. Typically, it consists of more than 400 chemical compounds (i.e. aldehydes, alcohols, amines, acids, esters, ethers, ketones, phenol derivatives, ketones, guaiacols, furans, oligomers, syringols and sugars) (Henrich et al. 2016). Considering its low carbon, nitrogen and sulphur content, CO, SO_x and NOx emissions are low, hence preparing bio-oil and conventional fuels.

Table 6 displays a comparison between the properties of prepared bio-oil (after water removal) and conventional liquid fuels. Concurrently, Table 7 shows physiochemical properties of as-formed bio-oil resulting from the pyrolysis of several biomaterials feedstocks without applying any upgrading strategy as well as comparing it with other conventional fuels, respectively. Both condensable and non-condensable gases are generated throughout the first

Table 6 Comparison of bio-oil properties with conventional fuels (Bridgwater, 2012b)

Property	Bio-oil	Diesel	Heating oil	Gasoline
рН	2.0-3.0	_	_	_
Viscosity at 40 °C (cP)	40-100	2.4	1.8-3.4	0.37-0.44
Density at 15 °C (kg/m ³)	1200	820-950	865	737
Heating value (MJ/kg)	18-20	42	45.5	44
Pour point (°C)	-15	-29	-6	-60
Flash point (°C)	48-55	42	38	40
Solids (wt%)	0.2-1.0	_	_	_
Ash	< 0.02		< 0.01	
Carbon	42-47	87.4	86.4	84.9
Hydrogen	6–8	12.1	12.7	14.76
Oxygen	46-51	_	0.04	_
Nitrogen	< 0.1	392 ppm	0.006	0.08
Sulphur	< 0.02	1.39	0.2 – 0.7	_



Gasification

Gasification is a process by which carbonaceous materials are thermochemically converted into valuable gases, commonly referred to as synthesis gases in the presence of a gasifying agent such as air, oxygen, steam, CO₂ or a combination of them at a temperature above 700 °C. Primarily, the produced gas consists of CO, H2, CO2 and CH4 (Shahabuddin et al. 2020). Generally, the gasification process comprises four main steps: (1) heating or drying (100–200 °C) to decrease its moisture content, (2) pyrolysis, (3) oxidation or partial combustion and (4) gasification. Firstly, the moisture content (30-60%) of the biomass is vaporised at about 200 °C. Then, pyrolysis includes the decomposition of different biomass, including cellulose, hemicellulose and lignin, into solid residues and volatiles occurs (Thomson et al. 2020). Oxidation or partial combustion is the third stage in which the resultant volatiles and char residues are oxidised to CO, CO₂ and H₂O with gasifying agent assistance beyond 700 °C (exothermic reaction).

By the action of CO_2 or steam as gasifying agents, carbon and volatile compounds react with them in terms of reduction reaction to produce syngas at a temperature over $800\,^{\circ}\mathrm{C}$ in an endothermic reaction (Situmorang et al. 2020; Hanchate et al. 2021). Briefly, simultaneous exothermic and endothermic reactions are included in the gasification process, and the first previously mentioned reactions are considered as heat suppliers for the endothermic one. Main reactions involved in the gasification include carbon reaction such as primary or secondary steam reforming, hydrogasification, oxidation, shift reaction, mechanisation and steam reaction.

The efficiency of the gasification process is impacted by different operational parameters such as feedstock composition, moisture content, ash content, granulometry, pressure, temperature, gasifier's type, gasifying agents, equivalence ratio and steam to biomass ratio (Díaz González and Pacheco Sandoval 2020). Basically, the gasifier's selection: fixed or moving bed (dry ash/slagging), fluidised bed (circulating, bubbling) and entrained flow (upflow, downflow), is controlled by several factors, for instance, feedstock composition, gasifying agent and product requirements (Mehrpooya et al. 2018). Syngas chemical composition and its heating values vary based on the as-used gasification method, as presented in Table 8.



Table 7 Physiochemical properties of bio-oil produced from different biomasses without using any upgrading technique. Where, the higher heating value is abbreviated as HHV

Saccharina Flui Japonica		temperature	(wt.%)			(wt.%)	•						Note the second
		(50)	% 2%	% Z	%0	%Н	Hd	Density (g/ml)	Viscosity	Water content (wt.%)	HHV (MJ/kg)	Acid value (mg of KOH/ g)	
	Fluidised bed	300	60.15	5.77	16.48	7.74	5.9	1	I	1.76	28.63	. 1	Choi et al. (2017)
Corn stover Fixe	Fixed bed	400	ı	ı	I	ı	2.67	1.25	2.6 ^b	I	15.3	ı	Sundaram et al. (2016)
	Fixed bed	400	1	I	I	1	2.77	1.25	2.1 ^b	ı	14.9	I	
Prairie cordgrass Fixe	Fixed bed	400	ı	ı	I	ı	2.59	1.25	2.5 ^b	ı	15.2	I	
Pinewood Aug	Auger reactor	450	ı	I	I	1	3.08	1.18	55.2°	16.9	22.49	69.5	Hassan et al. (2009)
Sweetgum Aug	Auger reactor	450	ı	I	I	1	2.65	1.16	8.26°	38.3	2.65	119.2	Wang et al. (2012)
Corn stover Aug	Auger reactor	450	ı	ı	I	1	5.66	1.08	1.60°	54.7	2.66	85.8	
Saccharina Fixe Japonica	Fixed bed batch	470	69.2	3.7	15.4	8.3	1	1.13	1	34.7	35.0	43	Choi et al. (2014)
p	Pilot-scale reactor	500	42.64	0.22	49.59	7.55	1	1.21	178.2^{c}	23.5	18.9	ı	Zhang and Wu, (2014)
Oak Flui	Fluidised bed	500	54.9	0.07	38.7	6.28	I	1.24	57 ^d	20.3	ı	110	Elliott et al. (2014)
Loblolly pine Flui Chips	Fluidised bed	500	50.1	0.53	42.7	6.65	I	I	16.4 ^b	19.8	I	I	Meng et al. (2014)
Hardwood Flui	Fluidised bed	500	61.35	0.24	32.07	6.34	ı	I	715 ^b	8.93	23.5		Martin et al. (2014)
Eucalyptus Free Bark pyro	Free-fall pyrolysis unit	550	I	I	I	1	2.78	1.13	I	26.07	12.45	I	Pidtasang et al. (2013)
Pine nuts Confined	Continuous fixed bed	550	58	0.3	33.5	8.2	4.84	1.09	1244 ^d	9.36	19.31	I	Xu et al. (2018)
Walnut shell Spor	Spouted bed	550	37.91	1.19	25.02	8.78	4.28	0.95	3.29 ^d	23.29	ı	I	Zhu et al. (2017)
Napier grass Fixe	Fixed bed	009	45.32	0.81	46.60	7.17	2.95	1.05	2.71 ^b	26.01	20.97	ı	Mohammed et al. (2017)
Rice husk Flui	Fluidised bed	009	ı	I	I	ı	3.59	1.15	58.16^{d}	15.82	22.99	I	Zhang et al. (2014)
Oakwood –		009	59.99	0.92	31.91	7.18	1	I	173.35^{a}	15.15	24.87	ı	Farooq et al. (2019)
Softwood		009	39.96	0.11	52.19	7.74	ı	1.20	67.39^{a}	28.05	15.27	79.23	Jiang and Ellis (2010)



Table 8 Composition of syngas from several biomass types and operating parameters, where N.A is abbreviated for not available

Biomass type	Operating parameters	neters				Syngas Composition (% v/v)	tion (% v/v)			Lower heating
	Temperature (°C)	Steam: biomass Gasification reactor	Gasification reactor	Gasifying agent Equivalence ratio	Equivalence ratio	00	CO ₂	CH_4	H ₂	value (MJ/Nm³)
Bamboo	400–600	I	Fluidised bed	Air	0.4	23.5–30.6% m/m	59–63% m/m 4–5% m/m	4–5% m/m	6.6–8.1% m/m 1.6–1.9	1.6–1.9
Ватьоо	400–600	0:1, 1:1	Fluidised bed	Air & air-steam 0.4	0.4	23.5–30.6% m/m (air); 36.1–40.3% m/m (air–steam)	N.A	N.A	6.6–8.16% m/m N.A (air);10.9– 16.5% m/m (air–steam)	N.A
Empty Fruit Bunch	700–1000	I	Fluidised bed	Air	0.15-0.35	21–36	10–65	5–14	10–38	7.5–15.5
Empty Fruit Bunch	850	I	Fluidised bed	Air	0.15-0.35	32–45	16.6–36	12–15	18.3–27.4	12.3–15.3
α-cellulose	800	0-1.5	Fluidised bed	Air-steam	0.27	6.5–11.2	26.3–27.7	2.2-3.7	13.5–18.5	6.5-7.6
Palm oil wastes	800	0.67–2.67	Fixed bed	Steam	ı	14–33	14–26	3-6	47–58	8.7-12
Pine sawdust	700–900	2.7	Fluidised bed	Air-steam	0.22	35-43	18-20	6-10	21–39	7.4–8.6
Palm oil wastes	750–900	1.3	Fixed bed	Steam	I	15–25	20–25	4-5	48–60	9.1–11.2
Olive kernel	950	I	Fixed bed	Air	0.14-0.42	15-20% w/w	40-55% w/w	40-55% w/w 10-12% w/w 20-30% w/w	20–30% w/w	8.8–10.4

Data sourced from Ahmad et al. (2016), Lv et al. 2004), Luo et al. (2009), Mohammed et al. 2011), Skoulou et al. (2008)



Other gasification types such as plasma, supercritical and microwave have been operated to improve gasification yields. In plasma gasification, an intense plasma thermal process is used to catalyse and ionise the organic compounds in biomass and gas, respectively, into syngas with slag using a plasma torch powered by an electric arc (over 2500 °C). Supercritical water gasification is a type of gasification typically performed in the presence of a vast amount of water for the generation of $\rm H_2$ and $\rm CH_4$ (Rodriguez Correa and Kruse, 2018). The process yield is very high, mainly affected by different parameters such as temperature, catalyst and biomass/water ratio.

Microwave gasification is a compelling scenario for transforming biomass. This technique's benefits over the traditional methods include uniform distribution of temperature, efficacy for large particle handling and higher heating values (Chen et al. 2015). Different technologies such as scrubbers (i.e. spray, dynamic wet, cyclonic, impactor, venture and electrostatic) and filters (fabric bag, fibrous ceramic, metallic foam and granular bed) have been used to clean up the syngas from different contaminants and hence improve its quality for numerous applications. A list of global biofuel-based facilities comprising its manufacturer, country, starting-up year, feedstock composition, downstream products and asused technology is shown in Table 9.

Biochemical conversion methods

Biochemical conversion pathways such as anaerobic digestion and fermentation can be employed to generate various biofuels from waste biomass, as shown in Fig. 3. Biochemical conversion methods have numerous merits, including high product selectivity, high product yield and flexibility to be operated at ambient temperature and pressure conditions (Singh et al. 2016). Ethanol and bio-hydrogen can be produced from the fermentable biomass via alcoholic fermentation and dark fermentation/photo-biological routes, respectively, whereas biogas can be produced anaerobic digestion as follows (Osman et al. 2020).

Fermentation

Fermentation is a process by which biological activities are utilised conjugated with air existence known as aerobic fermentation or without air called anaerobic fermentation (Karimi et al. 2021). Bioconversion of biomass to biofuel comprises of sequential stages: pretreatment, hydrolysis (acid/enzymatic) and fermentation (Alvira et al. 2010).

The pretreatment step aims to damage the cell wall as well as exhibit cellulose and hemicellulose for subsequent hydrolysis. It can be classified into four main categories, including: (1) physical, e.g. grinding, (2) physicochemical, e.g. wet oxidation, (3) chemical, e.g. oxidising agents

and organic solvents, and (4) biological or combination of them (Haghighi Mood et al. 2013). Secondly, pretreated biomass is decomposed into monomers by the action of acid/enzymatic hydrolysis. Lastly, the intermediate monomers are converted into alcohols using yeast/bacteria (Liu et al. 2015).

Based on the consolidation degree of the mentioned stages, ethanol production can be configured into four routes: (1) separate hydrolysis and fermentation, (2) simultaneous saccharification and fermentation and (3) simultaneous saccharification and co-fermentation and (4) consolidated bioprocessing. In separate hydrolysis and fermentation scenarios, enzyme generation, hydrolysis, hexose and pentose fermentation are employed in separate OR individual reactors. Despite execution of hydrolysis in addition to fermentation at their optimised conditions, accumulation of cellobiose and glucose enzymes during hydrolysis process negatively prohibits the efficiencies of cellulases (Margeot et al. 2009).

In the simultaneous saccharification and fermentation scenario, cellulose hydrolysis and hexose fermentation simultaneously run at the same reactor that overcomes cellulase inhibition because of instant consumption of sugars by fermenting microorganisms (Hahn-Hägerdal et al. 2007). In the simultaneous saccharification and cofermentation scenario, two genetically modified strains of *Saccharomyces cerevisiae* and *Zymomonas mobilis* are used to co-ferment glucose and xylose in the same reactor. In the consolidated bioprocessing scenario, only one microorganism is simultaneously utilised for hydrolysis and fermentation, which decreases the operation cost and enhances the process efficacy (Lin and Tanaka, 2006).

Different modes of fermentation can be briefly viewed as follows: photo-fermentation is a fermentative transformation of organic substrate to produce bio-hydrogen driven by miscellaneous groups of photosynthetic bacteria. This is occurring throughout a set of biochemical reactions in three steps like anaerobic conversion. The difference between photo-fermentation and dark fermentation is its proceeding in the presence and absence of light, respectively.

Alcoholic fermentation is another type of fermentation driven by yeast by which sugars are transformed into cellular energy associated with the generation of ethanol and carbon dioxide. Considering its occurrence in the absence of oxygen, it can be categorised as an anaerobic integration process. Heterotrophic algae or yeast can transform sugars into lipids inwards their cells associated with using suitable solvents to break down the cells (Łukajtis et al. 2018). The resultant lipids can be further purified and improved to liquid forms of transport fuels by hydro-treated vegetable oil diesel scenario, whereas genetically modified bacteria consume sugars and consequently produce short-chain gaseous



Table 9 Second-generation biofuel plants, including manufacturer, country, starting up a year, feedstock composition, downstream products and as-used technology

Company/Institute/University Startiname Lahti Energia Oy 1998 CHP Agnion Biomasse 2001 Heizkraftwerk Pfaffenhofen	ing-Up Year					
	J- S-	Starting-Up Year Feedstock composition	Downstream products	As-used Technology	TRL-scale	Country
		Wood waste	Renewable diesel (HVO) (70 MWth)	Circulating Fluidised Bed gasifier	TRL 9 commercial	Finland
		Wood waste (80,000 t/year)	Synthetic natural gas (32.5 MWth)	Agnion Heatpipe-Reformer	TRL 4–5 pilot	Germany
CHOREN Industries GmbH 2003		Dry wood chips from recycled wood and residual forestry wood	Fischer–Tropsch liquids (53 t/year)	Not available	TRL 4–5 pilot	Germany
Vienna University of 2005 Technology/BIOENERGY 2020+		Syngas from FICFB gasifier (5 m³/h)	Fischer-Tropsch liquids (5 kg/day)	Not available	TRL 4–5 pilot	Austria
West Biofuels 2007		Clean wood, waste wood (5 t/day)	Fischer–Tropsch liquids	Dual fluidised bed thermal reforming	TRL 6–7 demonstration	United States
Bio SNG Guessin 2008		Syngas from gasifier (350 m^3/year)	Synthetic natural gas (576 t/ year)	Not available	TRL 6-7 demonstration	Austria
TUBITAK MRC—ENERGY 2009 INSTITUTE—TURKEY		biomass	Synthetic natural gas (0.2 MW)	Downdraft fixed bed gasifier	TRL 4–5 pilot	Turkey
Greasoline GmbH 2011		Bio-based oils and fats, residues of plant oil processing, free fatty acids used bio-based oils and fats (3 t/year)	Diesel type hydrocarbons (2 t/year)	Catalytic cracking of biobased oils + fats primarily produces diesel fuel-range hydrocarbons	TRL 4–5 pilot	Germany
BioTfueL-consortium 2012		Forest waste, straw, green waste, dedicated crops	Fischer–Tropsch liquids (60 t/year), jet fuel component	Not available	TRL 4–5 pilot	France
TUBITAK 2013		Combination of hazelnut shell, olive cake, wood chip and lignite blends (0.2 t/h)	Fischer–Tropsch liquids (250 t/year)	Pressurised fluidised bed gasifier	TRL 4–5 pilot	Turkey
Goteborg Energi AB 2014		Forest residues, wood pellets, branches and treetops	Synthetic natural gas (11,200 t/year)	Repotec indirect gasification technology and Haldor Topsoe fixed bed methanation	Demonstration	Sweden
BioMCN 2017	_	Wood chips	Methanol (413,000 t/year)	Not available	TRL 8 first-of-a-kind	Netherlands
Go Green Fuels Ltd 2018		Refuse derived fuel and waste wood (7500 t/year)	Synthetic natural gas (1500 t/year)	Not available	TRL 8 first-of-a-kind commercial demo	United Kingdom
ECN 2019		Not available	Synthetic natural gas (300 MW)	Not available	TRL 6–7 demonstration	Netherlands
Red Rock Biofuels 2019		Not available	Diesel type hydrocarbons (1 t/y ear)	Not available	TRL 8 first-of-a-kind commercial demo	United States
Fulcrum BioEnergy Sierra 2019 Biofuels Plant		Waste (20,000 t/year)	Fischer–Tropsch liquids (314,913 t/year)	Not available	TRL 9 commercial	United States



Canada demonstration **FRL**-scale TRL 6-As-used Technology Not available Downstream products Ethanol (30,000 t/year) Starting-Up Year Feedstock composition Y. 2019 Company/Institute/University Table 9 (continued) Greenfield Ethanol)

Data were taken from (Molino et al. 2018)

alkenes that can be transformed by oligomerisation and hydro-treatment into jet/gasoline.

Electro-fermentation is a novel fermentation pathway by which microorganisms can be induced by using an electric field that can positively: (1) stabilise and optimise metabolisms of fermentation integration process by regulating redox and pH imbalances, (2) stimulate whether breakdown or elongation of carbon chain via different oxidative/reduction conditions, (3) enhance the synthesis of adenosine triphosphate and upgrade the yield of microbial biomass, (4) extract purposed products via selective membranes and (5) the possibility of directing the fermentation reaction towards the manufacture of a single and specific product (Schievano et al. 2016).

Anaerobic digestion

Anaerobic digestion is a biochemical, cost-effective and environmentally sustainable approach for upcycling biomass (Al-Wahaibi et al. 2020). It is a recovery process by which biodegradable organic substrates' bioconversion into renewable biogas occurs by several anaerobic organisms in an oxygen-free environment (Kainthola et al. 2019). Typically, the produced biogas comprises 50–75% of CH₄, 30–50% of CO₂, (0–3% of N₂, ~6% of H₂O and 0–1% of O₂. The biogas could also contain other minor impurities such as ammonia, hydrogen, hydrogen sulphide, nitrogen and water vapours (Wainaina et al. 2020).

The growth rate of biogas production was 11.2%, with approximately 58.7 billion Nm³ in 2017. It has been investigated that the production outlay of biogas resulting from anaerobic digestion plants will be declined by about 38% in 2050 compared with 2015. Collectively, more than 17,240 operating anaerobic digestion facilities in Europe generated 63.3 TWh of electricity based on biogas, which represented about 14.6 million European households of the global consumption rate per year in 2014. The American Biogas Council announced that about 2,000 anaerobic digestion plants were operated to handle the residues from municipal wastewater treatment facilities, food waste and animal manure digestion in 2015 (Shrestha et al. 2017).

In addition to biogas energy generation, the anaerobic digestion process contributes to nutrient recovery, mitigation of greenhouse gas emissions as well as depletion of dissolved oxygen (Bharti et al. 2021). Several parameters, including alkalinity, organic loading rate, temperature, pH, feedstock composition, hydraulic retention time and concentration of volatile fatty acids, directly affect the anaerobic digestion process and the physiochemical properties of biogas (i.e. composition and heating value). Considering the organic substrates, the anaerobic digestion process can be categorised into wet, semi-dry and dry due to varying % of total solids (Feng and Lin 2017).



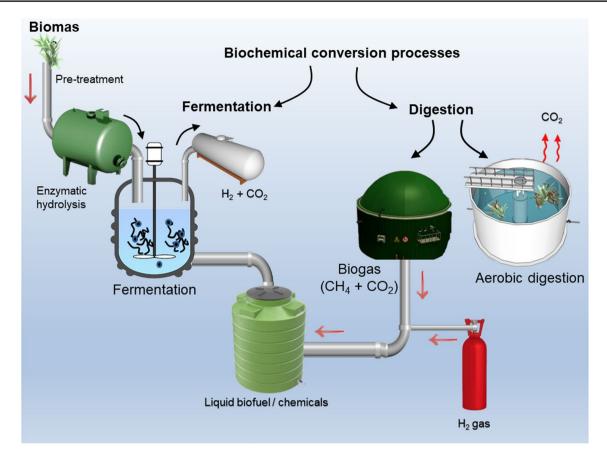


Fig. 3 Biochemical conversion route of biomass utilisation into biofuel, including fermentation and anaerobic digestion processes. Two types of digestion: aerobic digestion, which produces carbon dioxide and fertiliser, while anaerobic digestion produces biogas which in

turn, along with hydrogen, could be converted into liquid fuel. The other route is the fermentation process, where the biomass is firstly pretreated and then followed by enzymatic hydrolysis and fermentation, and finally, the production of liquid biofuel

Commonly, an anaerobic digestion scenario comprises four consecutive steps, including hydrolysis, acidogenesis, acetogenesis and methanogenesis, catalysed by different microorganisms, whereas hydrolysis is deemed as the ratedetermining step.

Hydrolysis is the first stage of anaerobic digestion at which complex biopolymers (i.e. carbohydrates, lipids, proteins, polysaccharide and nucleic acid) are converted into simple soluble compounds (i.e. amino acid, fatty acids, monomers, sugar, purines and pyrimidine) by the action of enzymes (i.e. amylases, lipases and proteases) produced from hydrolytic bacteria (Sawatdeenarunat et al. 2015).

Secondly, acidogenesis is the second stage of anaerobic digestion at which the simplified amino acids, sugar, fatty acids and monomers are converted into intermediate biomolecules (i.e. alcohols, volatile fatty acids, propionic and butyric acids) by fermentative bacteria.

Acetogenesis is the third step at which the mentioned acidogenesis products serve as a substrate to produce acetate by homoacetogens bacteria. Methanogenesis is the last step at which both acetate and carbon dioxide are directed to produce methane (biogas) by two sets of methanogens: acetoclastic and hydrogen utilising organisms (Matheri et al. 2018; Ganesh Saratale et al. 2018). The first group (acetoclastic methanogens) convert acetate into methane and carbon dioxide, while the other ones (hydrogen-utilising methanogens) generate methane by applying hydrogen and carbon dioxide as electrons donor and acceptor, respectively.

Several enhancement techniques as pretreatment steps such as: (1) physical, e.g. milling, (2) chemical, e.g. ionic liquid and surfactant, thermophysical, e.g. microwave irradiation, (3) thermochemical, e.g. supercritical CO₂, ammonia fibre explosion and ammonia fibre percolation, and (4) biological (microbial and enzymatic) can precede to enhance anaerobic digestion process (Gautam et al. 2020). To upgrade the quality of biogas (impurities removing), an additional cleaning step can be added to capture CO₂, H₂S and water vapours and avoid mechanical and chemical appliances throughout its utilisation. Different materials (i.e. silica gel) can be used to tackle H₂S and water vapours.

In contrast, other techniques (i.e. water scrubbing, organic scrubbing, membrane separation, cryogenic technology and



pressure swing adsorption) can be delivered to sequester CO₂ from the product and subsequently elevate its calorific value (Nag et al. 2019). The raw biogas can be used for producing electrical energy, whereas the improved biogas can be directly inserted into the natural gas grid or utilised as fuels for vehicles. From the economic point of view, two substrates can be simultaneously mixed (anaerobic codigestion) to overcome the disadvantages of mono-digestion and enhances its feasibility. Numerous types of reactors, including submerged packed beds, fluidised beds and other types, have been employed for the anaerobic digestion treatment process of wastewater with high biochemical oxygen demand (Paudel et al. 2017).

Production of liquid biofuel such as methanol and bio-oil from biomass

Historically, more than a hundred years ago, Giacomo Ciamician mentioned in his manuscript entitled 'Photochemistry of the Future' about the urgent need for the sustainable transfer from non-renewable to renewable sources (Sharma et al. 2020; Qasim et al. 2020). In 2017, the global energy consumption was rated at 13.5 billion tons of oil (~656 exajoules) by a yearly growth rate of 1.7%. Relatedly, an increase in the uncontrolled population has directly deepened the negative effects of the ascending pressure on non-renewable resources globally (Pradhan et al. 2018). Considering the new United Nations reports, it has been stated that with an introduction of approximately 83 million people to our globe per year, the current global population of 7.6 billion is anticipated to increase to 8.6, 9.8, and 11.2 billion by 2030, 2050 and 2100, respectively. Based on the United States Energy Administration (EIA) estimations, the global energy requirement is increasing annually and projected to rise by almost 28% in 2040 (~739 quadrillions Btus) (Sharma et al. 2020). Majorly, high pressure on energy consumption originates from countries with robust economic growth. A total enhancement in energy consumption has been investigated by non-OECD (Organisation for Economic Co-operation and Development) countries (~473 quadrillions Btus) by 2040, compared with its counterpart of Organisation of the Petroleum Exporting Countries (~266 quadrillions Btus) (Kumar et al. 2020).

The main route of biomass into liquid fuel 'drop-in' is through the gasification process. On a small scale, woody biomass gasification outperforms combustion and pyrolysis in terms of technological and economic impacts, while pyrolysis has been identified as the best large-scale method for upgrading woody biomass (Solarte-Toro et al. 2021). The biomass into liquid fuel such as bio-methanol starts with biomass gasification under low pressure using downdraft gasifier owing to its low tar formation along with long

residence time (Li et al. 2021). Where steam and oxygen (95% vol.) are commonly the gasification agents, the heat required for the gasification is supplied by biochar combustion. The gasification gas usually has a low content of light hydrocarbon and high-water content; thus, an in situ reformer with steam is used to convert them into carbon monoxide and hydrogen, followed by cooling of the hightemperature reformer effluent gas before subjecting it to gas composition adjustment (compression and sulphur removal steps). This is then followed by the water gas shift reaction process, where steam is introduced into the unit to increase the hydrogen to carbon monoxide ratio to 2, and then, carbon dioxide is removed before the methanol synthesis stage. Finally, the compressed synthesis gas is pre-heated before entering the methanol reactor, where carbon monoxide hydrogenation produces bio-methanol.

Interestingly, bio-methanol derived from biomass feedstocks can be used to produce light olefins of 230 million tonnes demand worldwide (Li et al. 2021). Ethylene, propylene and butylene as light olefins are commercially produced from petroleum-based hydrocarbon via steam cracking, where currently, biomass into olefins route gains interest through bio-methanol, dimethyl ether or Fischer–Tropsch process.

Bio-methanol can produce biodiesel via the transesterification process, where triglycerides/lipids are transformed into fatty acid methyl ester using a catalyst and alcohol, mainly methanol (Al-Mawali et al. 2021; Al-Muhtaseb et al. 2021; Hazrat et al. 2021). There is also a non-catalytic route for microalgal biodiesel production via subcritical and supercritical methanol (Karpagam et al. 2021).

As a type of biomass, algae recently showed some merits in producing biofuels, such as high lipid productivity, carbon dioxide capture, high growth rate, limited land requirement and high production yields (Sekar et al. 2021; Peter et al. 2021). Then again, there are still challenges, such as the post-processing of algae and the cultivation process. Besides the biofuels mentioned earlier, microalgae can produce biooil via different processes, most commonly pyrolysis and others such as gasification and liquefaction as thermochemical routes. Pyrolysis is preferred herein due to its simplicity, speed, better yields, along with operating conditions.

Bio-oil is the main pyrolytic product in fast and flash pyrolysis, along with biochar and gaseous products (Xiao et al. 2021). However, when applying bio-oil directly in petroleum (gasoline and diesel), engines will not produce sufficient heat due to its low calorific value, a high number of oxygenated compounds (> 300) and high water content (20–40 wt.%), which is negligible in hydrocarbon fuels (gasoline and diesel) (Gupta et al. 2021). Furthermore, due to its high viscosity and the presence of acidic compounds, it will provide a flow barrier when it passes into injectors and engines, resulting in engine corrosion. Besides, crude



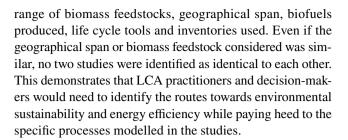
bio-oil will generate coking complications in the combustion stage due to the presence of a high number of solid particles. Therefore, upgrading the bio-oil via the integrated refinery is crucial for its commercialisation and producing value-added chemicals, char utilisation and gasoline grade fuel. The bio-oil upgrading process starts with moisture separation either by distillation (fractional or azeotropic), using catalysts, additional pyrolysis or biomass pretreatment techniques (demineralisation and torrefaction). This is followed by value-added chemicals extraction from the aqueous phase (acids, ketones, alcohols, ethers and esters) to improve the overall economics of the process. Some chemicals could also be extracted from the organic phase. The final organic residue of the bio-oil is then upgraded into a transportation fuel via various techniques such as deoxygenation, emulsification, hydrocracking, esterification, catalytic cracking and heavy fuel blending.

However, there are challenges associated with the upgrading routes for bio-oil, mentioned above, as they are still not commercialised due to the high cost of the catalyst, short catalyst life and complex operating conditions (high-pressure, special reactor requirements). Furthermore, extracting the chemicals in their low concentration is expensive and will require more investigation on the low-cost solvent, catalyst and process optimisation, primarily as the physicochemical characteristics of the bio-oil rely on the catalyst used. The catalyst minimises the heteroatom content of the biooil and increases the hydrogen-to-carbon ratio (H/C). This consequently lowers the harmful emissions of NO_x, SO_x and increases the calorific value of the bio-oil (Nagappan et al. 2021). Selling the biochar produced during pyrolysis can also increase the overall economics, which can be used in the carbon sequestration, adsorption of the contaminants, soil amendments and catalytic supports in bio-oil upgrading that enhances the circular bioeconomy of the process (Fawzy et al. 2021).

Life cycle assessment of biomass to biofuel conversion processes

Life cycle assessment is recognised as an effective framework for assessing impacts on natural environment, humans and natural resources for processes, products and systems. It provides evidence-based data to policymakers to make long-term strategic decisions and improve environmental sustainability. The four main stages defined by ISO 14,040 and ISO 14,044 for conducting LCA are: (1) goal and scope definition, (2) life cycle inventory analysis, (3) environmental impacts assessment and (4) life cycle interpretation (Lewandowski et al. 2000).

Herein, we analysed 40 LCA studies published from 2019 to 2021 on biofuels (Table 10). These studies covered a wide



Goal and scope definition

Goal and scope definition includes defining specific purpose, aim and objectives for conducting LCA. This stage is imperative to understand overall results and LCA findings. It incorporates defining functional unit and respective system boundaries. Functional units are quantified description of the performance requirements that the product system fulfils and are linked with functions of the product rather than with physical products. It was observed that about 32% of the reviewed studies used 'units of bioenergy in J or kWh' as the functional unit, while about 22% recorded LCA results for 'amount of biofuels produced' such as in kg (Fig. 4).

The system boundaries included in the LCA studies control what processes will be considered for computing environmental impacts. Figure 5 shows the generalised three crucial phases for biofuel production: (1) Phase 1 includes biomass cultivation, fertiliser application, impacts of fertiliser on soil, carbon emissions from land use, use of marginal and/or forest land, transportation of produced biomass to the production system, (2) Phase 2 incorporates chemical, thermal, biochemical, thermochemical processes for conversion of biomass to biofuels and related environmental impacts due to chemicals, electricity and energy procurement, upgradation of biofuels for final purpose, and (3) Phase 3 involves environmental impacts due to co-products management and emissions due to biofuel use.

The system boundaries included in the LCA studies varied for processes and systems considered. It was observed that about 90% of the reviewed studies considered Phase 1, Phase 2, while only 25% of the studies included Phase 3 (Fig. 6). This highlights a paucity of research in the biofuel LCA field containing a holistic approach and includes all the phases of the biofuel production chain. Even where the studies focused on Phase 3, they primarily dealt with anaerobic digestion. Most of the studies focused on specific processes of the biofuel production chain, focusing on critical areas of concern rather than evaluating the overall impacts of the entire production chain. Interestingly, use of biofuels has increasingly been recognised as a measure to reduce greenhouse gas emissions; however, very few studies addressed the use phase of biofuels and compared them to conventional sources.



Table 10 Characteristics of life cycle assessment research analysed in the present study

Reference	Region	Functional unit	Feedstock	Process	Product	LCA tools	Database
Aberilla et al. (2019)	Southeast Asia	1 kWh of electricity produced	Rice and coconut residues	Comparison of: Combustion Anaerobic digestion Gasification	Electricity	GaBi 7.3	Ecoinvent 3.1
Amezcua-Allieri et al. (2019)	Southern Mexico	1 MJ of energy produced	Sugarcane bagasse	Boiler	Bioenergy	n/i	Ecoinvent
Ardolino and Arena (2019)	EU-27	1 MW of biomethane	Biowaste	Comparison of: Anaerobic digestion with Gasification	Biomethane	SimaPro 8.2	n/i
Bacenetti (2019)	North Italy	1 ha of vineyard	Pruning residues from grapevine cultivation	Combustion for use in Heat and Cold boiler followed by Chiller		n/i	Ecoinvent 3.5
Banerjee et al. (2019)	India	Mass of CO ₂ equivalent released per MJ of fuel produced	Microalgal biomass	Comparison of Transesterification (Fe ₂ O ₃ catalyst with HCl catalyst)	Biodiesel	GaBi 7	n/i
Chung et al. (2019)	n/i	1000 kg of biodiesel produced per day	Waste cooking oil	Transesterification (waste chicken egg- shell derived CaO catalyst)	Biodiesel	openLCA 1.8	Agribalyse and NEEDS
Dasan et al. (2019)	n/i	100,000 kg of dry algae biomass for 340 days of yearly operation	Microalgal biomass from Chlorella vulgaris	Lipid extraction followed by lipid recovery and transesterification	Biodiesel	n/i	n/i
Derose et al. (2019)	n/i	Mass of CO ₂ equivalent released per MJ of fuel produced	algae harvested from an algal turf scrub- ber	Comparison of: Biochemical process- ing with Thermochemical processing	Biodiesel	n/i	GREET
Di Fulvio et al. (2019)	EU-27 + United Kingdom	Biodiversity loss per ha of land use for spatially explicit LCA	Biomass from land	n/i	Biofuel	GIS^a	GLOBIOM model
Dupuis et al. (2019)	East England, United Kingdom	Mass of CO ₂ equivalent released per unit of delivered feedstock	Lignocellulosic Biomass from Mis- canthus and Wheat	n/i	High octane biodiesel	n/i	DNDC and STAMINA models
Zhu et al. (2019)	Hubei province, China	1 MJ of electricity output	Cotton straw	Combustion	Electricity	n/i	Water footprint database ^b
Wang et al. (2019b)	China	1 kg hydrogen	Biomass and water	Gasification	Hydrogen	n/i	n/i



Reference	Region	Functional unit	Feedstock	Process	Product	LCA tools	Database
Wagner et al. (2019)	Germany	1 GJ of electricity	Miscanthus	Anaerobic digestion followed by combined heat and power generation	Electricity	n/i	Ecoinvent 3.4 and KTBL
Timonen et al. (2019)	Southern Finland	g CO ₂ eq per MJ of energy produced and kg CO ₂ eq per kg of nitrogen (N) in digestate	Pig slurry	Anaerobic digestion	Heat and electricity	SimaPro	LIPASTO
Tanzer et al. (2019)	Brazil and Sweden	1 GJ of fuel	Agroforestry residues	Comparison of: Gasi- fication Fast pyrolysis hydro- treatment Hydrothermal lique- faction	Marine fuels	CMLCA 5.2	Ecoinvent 2.2
Quispe et al. (2019)	Peruvian north coast	1 MJ of energy from rice husk	Rice husk	Combustion	Electricity	n/i	Ecoinvent 3.1
Krzyżaniak et al. (2019)	Poland	1 Mg of dry biomass	Poplar	n/i	Biofuel	SimaPro	Ecoinvent
Im-Orb and Arporn- wichanop, (2020)	n/i	kg CO ₂ eq per kg of biofuel produced	Rice straw	Integrated biomass pyrolysis, gasification, and methanol synthesis	Methanol	LCSoft version 6.1 Aspen Engineering 8.4	ASPEN Plus
Jeswani et al. (2020)	United Kingdom	1 MWh of heat	Woodchips from forestry and sawmill waste	Boiler	Heat	GaBi 4.4	Ecoinvent
Lan et al. (2020)	Southeast United States	1 MJ of biofuel produced	Blended feedstock: switchgrass and pine residues	Fast pyrolysis followed by combustion	Gasoline and diesel	n/i	GREET 1.8 Ecoinvent
Lask et al. (2020)	Southwest Germany	1 kWh of electricity produced	Perennial wild plant mixtures and maize silage	Anaerobic digestion	Electricity	openLCA 1.8.0	Ecoinvent
Mendecka et al. (2020)	Italy	Treatment and upgrade of 1 kg of biomass	Olive pomace	Hydrothermal carbonisation	Bioenergy	n/i	European Reference Life Cycle Database and Ecoinvent 3.2
Reaño, (2020)	Southeast Asia	1 kg of biofuel produced	Rice husk	Comparison of: Alkali water electrolysis Gasification Dark fermentation	Bio-hydrogen	openLCA 1.10	Ecoinvent 3.5



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Reference	Region	Functional unit	Feedstock	Process	Product	LCA tools	Database
Spatari et al. (2020)	United States	1 kg of bio-oil	Forest residues	Comparison of: Fast pyrolysis Catalytic fast pyrolysis	Biodiesel	ASPEN Plus GaBi 9	GaBi 9 LCA
Thengane et al. (2020) California, United States	California, United States	1 MJ of torrefied rice husk pellets	Rice husk	Torrefaction	Torrefied rice husk pellets	openLCA 1.8.0	Ecoinvent 3.4
Ubando et al. (2020)	Taiwan	l kg of torrefied microalgal biomass	Microalgal biomass from Chlorella vulgaris	Torrefaction	Torrefied microalgal biomass	Simapro 8.5.2	Simapro 8.5.2 LCA
Nilsson et al. (2020)	Sweden	1 ha of land under cultivation	Perennial grass	n/i	Bioenergy	$\mathrm{GIS}^{\mathrm{a}}$	DNDC agricultural model
Aristizábal-Marulanda Colombia et al. (2021)	Colombia	I MJ of energy	Coffee cut stems	Comparison of: Enzymatic hydrolysis followed by gasifi- cation system Gasification only system	Ethanol and electricity + steam	SimaPro 8.3	Ecoinvent 3
Brassard et al. (2021)	Metropolitan France	1000 kg of dry bio- mass	Primary forestry residues	Pyrolysis	Bio-oil, biochar) and gaseous (non-condensable gases)	Simapro 9.0	Ecoinvent 3
Cusenza et al. (2021)	Sicily, Italy	I kWh of electricity feed into the grid	Residual biomasses: olive pomace, chicken and bovine manure, whey, citrus processing waste, and Hedysarum coronarium silage	Anaerobic digestion	Electricity and heat	i/u	Ecoinvent 3
Da Silva et al. (2021)	Region state of Rio Grande do Sul, Brazil	1 kg of methanol	Rice straw	Gasification	Methanol	Aspen Plus	Aspen Plus
Fu et al. (2021)	Dongying, Shandong province, China	1 kg of ethanol	Sweet sorghum	Combustion	Ethanol	GIS^{a}	Harmonised world soil
Martillo Aseffe et al. (2021)	Ecuador (Los Rios and Guayas)	1 ton of seed-corn produced	Corncob	Comparison of: Combustion Gasification	Electricity	SimaPro 8	SimaPro 8 LCA
Schonhoff et al. (2021)	Germany	100 GJ thermal energy	Sida hermaphrodita	Comparison of: Chopping Pelletisation Splitting	chips, pellets and briquettes as carriers of Thermal energy	GaBi 9.2	Ecoinvent 3.5



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Reference	Region	Functional unit	Feedstock	Process	Product	LCA tools	Database
Schmidt Rivera et al. (2020)	United Kingdom	I tonne of spent coffee grounds treated	Spent coffee grounds	Comparison of: Incineration Landfilling, Anaerobic digestion, Composting fol- lowed by direct application to land	Biodiesel, compost	GaBi 8.7	Ecoinvent 3.3
Al-Mawali et al. (2021)	Gulf cooperation council countries	1000 kg of biodiesel	Waste date seeds	Esterification using a magnetic catalyst	Biodiesel	SimaPro	SimaPro LCA
Yang et al. (2021)	China	1 ton of crop residues	Crop residues	Slow pyrolysis	Biofuel and biochar	GaBi 8.7	PRC 2019 ^c
Saranya and Ramachandra, (2020)	Karnataka, India	Biomass achievable in Microalgal biomass I ha area	Microalgal biomass	Comparison of: Acid catalysis Biocatalysis	Biodiesel	OpenLCA 1.10.3	Ecoinvent 3.6
Bora et al. (2020)	United States	1000 kg of wet poultry litter	poul- Poultry litter	Comparison of: Direct land use Pyrolysis Gasification Hydrothermal processing	Biofuel and biochar	n/i	Ecoinvent 3
Al-Muhtaseb et al. (2021)	Pakistan	1000 kg of biodiesel	Waste loquat seeds	Transesterification using a CaO/CeO ₂ catalyst	Biodiesel	SimaPro	SimaPro LCA

n/i: Not included, EU-27: The European Union; GREET: Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation

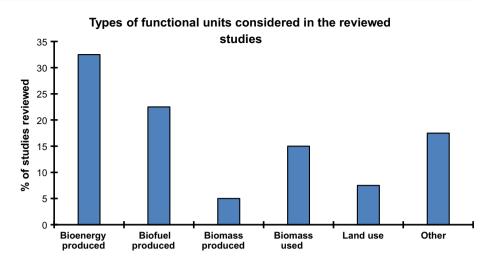
^a GIS: Some life cycle assessment (LCA) studies analysed environmental impacts of land use for biofuel production and thus used geographic information systems software

^b Water footprint database: Data sourced from the water footprint assessment manual

^c PRC 2019; Data sourced from the national development and reform commission of the People's Republic of China (PRC)



Fig. 4 Types of functional units used in the life cycle assessment studies reviewed in the present work (N=40)



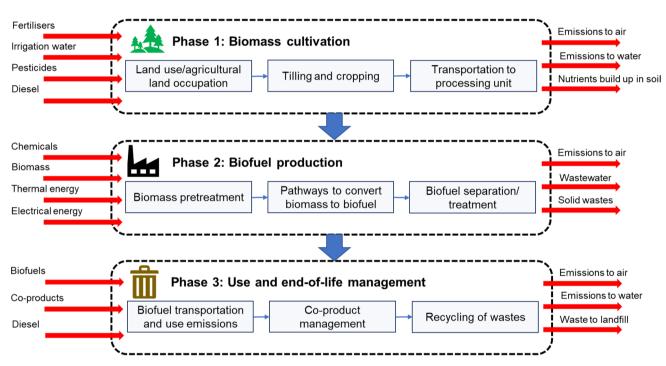
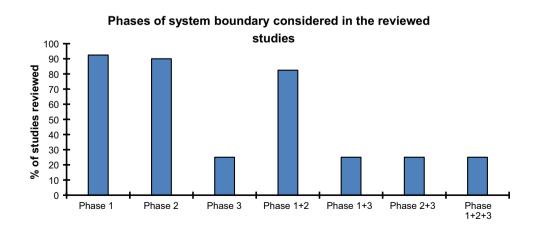


Fig. 5 General system boundary for conducting life cycle assessment of conversion of biomass feedstocks to biofuels

Fig. 6 Phases of system boundary considered for conducting life cycle assessment of converting biomass feedstocks into biofuels in the reviewed literature (*N*=40)





Life cycle inventory analysis

Inventory analysis involves quantifying all the inputs and outputs for the processes considered in the system boundary of the LCA. This includes raw material requirements, energy input, emissions to air, wastewater production, solid waste generation, emissions to land and others. It should be noted that more the systems involved in the system boundary, greater would be the need for data for inventory analysis, which also is explained in Fig. 6 with only some studies considering all the phases of biofuel production.

Table 10 shows databases for conducting inventory analysis, used in the reviewed studies such as SimaPro LCA databases, Ecoinvent, GREET (Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation). Some LCA studies also used agricultural models for considering the impacts of land use on overall environmental sustainability, such as GLOBIOM, DNDC and STAMINA (Di Fulvio et al. 2019; Dupuis et al. 2019; Nilsson et al. 2020).

Environmental impacts assessment

Mid-point indicators

In this stage of LCA, key environmental impacts are quantified and distributed in various environmental categories depending upon the functional unit, system boundary, modelled systems and need of the decision-makers. Some studies computed net energy ratio to evaluate the usability of biofuels as energy sources (Al-Mawali et al. 2021; Al-Muhtaseb et al. 2021; Dasan et al. 2019; Im-Orb and Arpornwichanop 2020; Reaño 2020; Saranya and Ramachandra 2020), which is defined as the ratio of output energy to input energy for the overall process (Pleanjai and Gheewala 2009).

Mid-point categories used for expressing life cycle environmental impacts were: global warming potential (100 years), which includes greenhouse emissions is generally expressed as kg CO₂ equivalent for a time horizon of 100 years. Some studies also considered greenhouse gas emissions for a temporal scale of 20 years, in accordance with the life span of infrastructure (Aberilla et al. 2019; Cusenza et al. 2021). Abiotic depletion reported as kg Sb equivalent corresponds to the depletion of fossil fuels, minerals, clay and peat. Abiotic depletion (fossil fuels, recorded as MJ) is linked to the depletion of fossil deposits. Ozone layer depletion (kg trichlorofluoromethane equivalent) is typically accounted for a time scale of 40 years.

Ecotoxicity potential evaluated in kg 1,4 dichlorobenzene equivalent or cumulative toxic units is calculated in three separate categories, which examine damage to terrestrial, freshwater and marine sources for the entire production process. Photochemical oxidation recorded in kg non-methane volatile organic compounds equivalent refers to emissions

of reactive substances injurious to human health and ecosystems. Acidification measured in kg SO_2 equivalent is caused by the emission of acidifying substances. Land use calculated in m^2 is categorised as the transformation of urban land, agricultural land and natural land such as forests. Water depletion (m^3) is the use of water for the entire production chain of biofuels. Particulate matter formation expressed as PM2.5 equivalent and/or PM10 equivalent relates to the emission of PM 2.5 (particulate matter with \leq 2.5 μm in diameter) and/or PM10 (particulate matter with \leq 10 μm in diameter). Eutrophication consists of the effect of releasing an excessive amount of nutrients reported as kg PO_4 equivalent. Ionising radiation (kg U235 equivalent) transfers energy into the body tissue and may thereby interfere with the structure of molecules (Table 11).

Finally, human toxicity is recorded as kg 1,4 dichlorobenzene equivalent or cumulative toxic units. Human toxicity (carcinogens) is an index that corresponds to potential harm of a unit of cancer-causing chemical released into the environment and is based on both the inherent toxicity of a compound and its potential dose. Human toxicity (non-carcinogens) index is associated with non-carcinogenic chemicals release, doses and exposure.

Endpoint indicators

The mid-point categories are aggregated to present results as endpoint categories. It is argued that the environmental impacts should be presented as mid-point categories to prevent oversimplification or misinterpretation of environmental impacts (Kalbar et al. 2017). Nevertheless, some studies did not present detailed mid-point indicator impacts but only endpoint indicators (Amezcua-Allieri et al. 2019; Martillo Aseffe et al. 2021; Bora et al. 2020).

The endpoint categories used in reviewed studies were: (1) human health (disability-adjusted life year) is related to the impacts of environmental degradation that results in an increase in and duration of loss of life years due to ill health, disability or early death, and (2) ecosystem quality (species × year) is linked to the impact of global warming potential, ozone layer depletion, acidification, ecotoxicity, eutrophication and indicates biodiversity loss. It is recorded as local species loss integrated over time, and (3) resources are related to the depletion of raw materials and energy sources expressed generally in US dollars (\$), representing the extra costs involved for future mineral and fossil resource extraction (Al-Muhtaseb et al. 2021).

Uncertainty, scenario and sensitivity analysis

Life cycle assessment (LCA) studies are models which are simplified versions of the real-world system and thus are inherently uncertain (Wang and Shen 2013). These



Table 11 Environmental impacts and main findings recorded in the life cycle assessment case studies analysed

Reference	Environmental impacts considered	Findings
Aberilla et al. (2019)	Global warming (20 years), ozone layer depletion, particulate matter formation, photochemical oxidant formation, freshwater eutrophication, marine eutrophication, terrestrial acidification, freshwater ecotoxicity, marine ecotoxicity, terrestrial ecotoxicity, fossil depletion, mineral depletion, water depletion, agricultural land occupation, natural land transformation, urban land occupation, human toxicity, ionising radiation potentials	The study compared anaerobic digestion, combustion and gasification for rice and coconut residues in South East Asia to produce 1 kWh of electricity as the end product. It was observed that anaerobic digestion was the best option for 14 out of 18 impacts (the remaining four impacts categories were: global warming, photochemical oxidant formation, particulate matter formation and terrestrial ecotoxicity potentials) Global warming potential for anaerobic digestion = 100 kg CO_2 eq Global warming potential for gasification and combustion =- $30 \text{ to } 30 \text{ kg CO}_2$ eq Ozone layer depletion for anaerobic digestion = -36 \mug CFC -11 eq Human toxicity (carcinogens) for anaerobic digestion = $-74 \text{ g } 1.4 \text{ DB}$ eq Human toxicity (carcinogens) for gasification and combustion = $-74 \text{ g } 1.4 \text{ DB}$ eq Human toxicity (carcinogens) for gasification and combustion = $-72 \text{ to } 182 \text{ g}$ 1,4 DB eq
Amezcua-Allieri et al. (2019)	Abiotic resource depletion, acidification, eutrophication, ozone layer depletion, freshwater aquatic ecotoxicity, marine ecotoxicity and terrestrial ecotoxicity potentials	Although all these environmental categories were analysed, the results were only presented as normalised or % change in the value of environmental impacts compared to fossil fuel. Potential environmental impact index (PEI) recorded using a summation of all environmental categories was 2528 (for the use of sugarcane bagasse) vs 20,200 PEI/GI for using fossil fuel in a sugar mill
Ardolino and Arena (2019)	Global warming, non-renewable energy and respiratory inorganics potentials	Both the biomethane scenarios consisting of using biomethane produced from anaerobic digestion and gasification were better than the diesel scenario, improving 56% and 65% for global warming potential, and 31% and 52% for non-renewable energy potential, respectively
Bacenetti (2019)	Global warming (100 years), ozone layer depletion, human toxicity (carcinogens), human toxicity (non-carcinogens), particulate matter formation, photochemical oxidant formation, terrestrial acidification, freshwater eutrophication, terrestrial eutrophication, freshwater ecotoxicity, mineral fossil and renewable resource depletion potentials	When compared with the business-as-usual scenario where the pruning residues are wasted, pruning residues for bioenergy production showed better environmental impacts in most categories. Below are some of the results recorded for business-as-usual and reuse of residues for 1 ha of vineyard land as a functional unit (in respective order) Global warming potential = 89 and -807 kg CO_2 eq Ozone layer depletion = 5 and -43 mg CFC -11 eq Human toxicity (carcinogens) = 9.6×10^{-7} and -1.1×10^{-6} comparative toxic units Human toxicity (non-carcinogens) = 3.7×10^{-5} and -4.3×10^{-5} comparative toxic units
Banerjee et al. (2019)	Global warming, eutrophication, human toxicity, ozone layer depletion potentials	Biodiesel production from wet biomass and using Fe ₂ O ₃ nanocatalyst in the transesterification process reported less environmental impacts as compared to the conventional biodiesel production process in all four environmental categories

Table 11 (continued)		
Reference	Environmental impacts considered	Findings
Chung et al. (2019)	Global warming, ozone layer depletion, eutrophication, terrestrial acidification, ecotoxicity, fossil depletion, minerals depletion, land use, human toxicity(carcinogens), ionising radiation, respiratory inorganics and respiratory organics potentials	The highest environmental impacts were observed during transesterification of the entire biodiesel production process. The transesterification process contributed a 1010 MJ surplus on fuel consumption due to the high demand of electrical energy for 1000 kg of biodiesel produced
Dasan et al. (2019)	Net energy ratio and global warming potential (10,000 years)	This study assessed the impact of different microalgae cultivation systems on biomass productivity. The net energy ratio was below 1 for all microalgae cultivation systems (open pond, tubular, bubble column photo-bioreactors)
Derose et al. (2019)	Global warming potential	Biochemical and thermochemical pathways were compared for bioenergy production (1 MJ of fuel produced) Global warming potential for biochemical pathway = 111.2 g $\rm CO_2$ eq
Di Fulvio et al. (2019)	Biodiversity loss	Global warming potential for thermochemical pathway = -2 g CO ₂ eq Increased cultivation of perennial crops for bioenergy production could be an option for climate change mitigation. However, it could negatively impact biodiversity through loss of species habitats in EU-27 and the UK
Dupuis et al. (2019)	Global warming potential per unit of delivered feedstock (100 years)	Perennial energy crops rather than wheat husk should be considered biofuel in East England based on the life cycle assessment of the use of wheat husk and energy crops
Zhu et al. (2019)	Water footprint in L per MJ of bioenergy	The total life cycle water use intensity for using cotton straw as a bioenergy source via combustion was 11.708 L/MJ, lower than bio-oil power generation, however, much greater than other renewable energy sources (such as geothermal, solar photovoltaic and wind power). The study also noted that biomass agricultural production accounted for 84.61% of the total water use
Wang et al. (2019b)	Global warming potential	An increase in gasification temperature and decrease in biomass slurry concentration and pressure could decrease global warming potential
Wagner et al. (2019)	Global warming, agricultural land occupation, freshwater ecotoxicity, human toxicity, marine ecotoxicity potential, freshwater eutrophication and marine eutrophication potentials	This study noted that marginal German electricity mix, when substituted by miscanthus-based biogas, can lead to net benefits in the impact categories of global warming potential, human toxicity, marine ecotoxicity, freshwater ecotoxicity and freshwater eutrophication. This was observed even when the biomass is transported over longer distances, biomass yields are lower, or a lower heat utilisation rate is applied
Timonen et al. (2019)	Global warming potential	The study compared the use of pig slurry with grass and pig slurry with food waste as feedstocks for anaerobic digestion. The highest global warming potential was observed when pig slurry and grass from uncultivated fields were used for anaerobic digestion. This could be due to due to prominent N ₂ O emissions from grasslands, while the lowest level of global warming potential was in the scenario, which involved the digestion of pig slurry and side streams from the food industry due to avoided emissions from co-feedstock procurement (covered storage and zero emissions to produce food industry sludge as waste)



Table 11 (continued)		
Reference	Environmental impacts considered	Findings
Tanzer et al. (2019)	Global warming potential (100 years), NO_x and SO_2 emissions	All biofuel systems of agroforestry residues have substantially lower life cycle environmental impacts compared to heavy fuel oil (HFO, marine fuel reference)
		Global warming potential for biofuels = -30 to 40 kg CO_2 eq/GJ
		Global warming potential for HFO reference = 90 kg CO_2 eq/GJ
		SO_2 emissions all biofuels < 0.3 kg SO_2/GJ
		SO_2 emissions for HFO reference = 2.5 kg SO_2/GJ
		NO_X emissions from biofuels $\approx HFO$ reference = 1.6 kg NO_X/GJ
Quispe et al. (2019)	Global warming (100 years), acidification potential, eutrophication and water depletion potentials	For generating 457 MJ of bioenergy from fuels considered, the results obtained are summarised below
		For rice husk, with yield $= 9.5$ t/ha and dryer efficiency as 0.7:
		Global warming potential = $1.96 \text{ kg CO}_2 \text{ eq}$
		Water depletion potential = 2.3 m^3
		Eutrophication potential = 12.5 g PO_4 eq
		Acidification potential = $50.3 \text{ g SO}_2 \text{ eq}$
		For coal system:
		Global warming potential = $61 \text{ kg CO}_2 \text{ eq}$,
		Water depletion = 23 m^3
		Eutrophication potential = 63 g PO_4 eq
		Acidification potential = $462 \text{ g SO}_2 \text{ eq}$
		The study concluded that if all rice husk available in Peru were used for dry-
		ing processes instead of coal, it could mitigate around 708,540 metric tons
		or CO ₂ cy per year



Table 11 (continued)		
Reference	Environmental impacts considered	Findings
Krzyżaniak et al. (2019)	Global warming, particulate matter formation, terrestrial acidification, freshwater eutrophication, human toxicity, freshwater ecotoxicity, fossil depletion potentials	This study compared poplar production (1 tonne of dry matter) as lignocellulosic feedstock for use in bioenergy systems considering four fertilisation systems: unfertilised, mineral, lignin and lignin+mineral, and observed the following results
		Global warming potential for lignin + mineral fertilisation system = -20 kg CO ₂ eq
		Global warming potential for unfertilised system = 20 kg CO_2 eq
		Particulate matter formation potential for lignin + mineral fertilisation system = $0.3 \text{ kg PM}10 \text{ eq}$
		Particulate matter formation potential for unfertilised system = $0.3~\mathrm{kg}$ PM10 eq
		Terrestrial acidification potential for lignin + mineral fertilisation system = $0.7~{\rm kg~SO_2}$ eq
		Terrestrial acidification potential for unfertilised system = 0.3 kg SO_2 eq
		Freshwater eutrophication potential for lignin + mineral fertilisation system = $0.01~\mathrm{kg}$ P eq
		Freshwater eutrophication potential for unfertilised system $= 0.3~\mathrm{kg}$ P eq
		Human toxicity potential for lignin + mineral fertilisation system = 9 kg 1,4 DB eq
		Human toxicity potential for unfertilised system = $19 \text{ kg } 1,4 \text{ DB}$ eq
		The study concluded that fertilised systems could cause negative environmental impacts for poplar production in Poland
Im-Orb and Arpornwichanop (2020) Net energy ratio and potential environmen	Net energy ratio and potential environmental impact	The study analysed the life cycle impacts of methanol production. The impact categories considered global warming, ozone layer depletion, acidification, eutrophication, photochemical oxidant formation, ecotoxicity, respiration effects, human toxicity (non-carcinogens) and human toxicity (carcinogens) were all merged as potential environmental impact. The highest net energy ratio achieved was 0.612. The lowest PEI/kg methanol was observed as 0.18



	Findings
	Environmental impacts considered
lable 11 (continued)	Reference

Reference	Environmental impacts considered	Findings
Jeswani et al. (2020)	Global warming, abiotic depletion, abiotic depletion for fossil fuels, acidification, eutrophication, freshwater ecotoxicity, human	The study compared the use of biomass for boiler compared to fossil resources (i.e. natural gas, oil and electricity). The findings demonstrated that heat from biomass boilers had lower impacts than from the coal and oil equivalents in all categories (except terrestrial ecotoxicity potential). Some of the environmental impacts observed for the production of 1MWh of heat are summarised below
	toxicity, marine ecotoxicity, ozone layer depletion, photochemical oxidant formation and terrestrial ecotoxicity potentials	Global warming potential during biomass use = 14.7 kg CO_2 eq
		Global warming potential during use of fossil resources = $233.9-468.4 \text{ kg}$ CO ₂ eq
		Terrestrial ecotoxicity potential during biomass use = 0.6 kg DB eq
		Terrestrial ecotoxicity potential during use of fossil resources = $0.03-0.11 \ \mathrm{kg}$ DB eq
		Human toxicity potential during biomass use = $29 \text{ kg } 1,4 \text{ DB eq}$
		Human toxicity potential during use of fossil resources = 4-192 kg 1,4 DB eq
Lan et al. (2020)	Primary energy consumption and global warming potential (100 years)	The life cycle energy consumption and global warming potential of 1 MJ of gasoline and diesel production utilising switchgrass and pine residues were observed as 0.7 to 1.1 MJ and 43.2 to 76.6 g $\rm CO_2$ eq, respectively
Lask et al. (2020)	Global warming potential	When considering land-use scenarios due to maize silage and wild energy crop production, blended feedstock consisting of both showed the lowest global warming potential as 198 kg CO_2 eq/kWh of electricity produced
Mendecka et al. (2020)	Global warming, acidification, eutrophication and	For treatment and upgrade of 1 kg of olive pomace, following impacts were observed
	freshwater ecotoxicity potentials	Global warming potential = 0.468 to 1.024 kg CO_2 eq
		Acidification potential = 0.0007 to 0.0015 mol H+eq Eutrophication potential = 0.036 to 36.290 \circ P eq
		Freshwater ecotoxicity potential = 165.8 to 254.5 comparative toxic units
Reaño (2020)	Net energy ratio and	The study compared alkali water electrolysis, gasification and dark fermentation for hydrogen production. Dark fermentation pathway for the production of 1 kg of H ₂ was recorded as the most efficient process based on:
	Global warming (100 years), acidification, eutrophication, and terrestrial ecotoxicity potentials	Net energy ratio = 1.25 ,
		Global warming potential = $46 \text{ kg CO}_2 \text{ eq}$
		Acidification potential = 75 g SO_2 eq
		Eutrophication potential = 350 g PO_4 eq
		Terrestrial ecotoxicity potential = 2.5 mg 1,4 DB eq

Table 11 (continued)		
Reference	Environmental impacts considered	Findings
Spatari et al. (2020)	Global warming potential (100 years)	For 1 MJ of biodiesel produced:
		Global warming potential for catalytic pyrolysis = -80 g CO_2 eq
		Global warming potential for fast pyrolysis = 20 g CO_2 eq
Thengane et al. (2020)	Global warming potential	Global warming potential = 6.04 ton CO ₂ eq/ton torrefied rice husk for severe torrefaction
Ubando et al. (2020)	Global warming, water depletion, fossil resource scarcity, mineral resource scarcity, land use, human toxicity(carcinogens), human toxicity (non-carcinogens), marine ecotoxicity, freshwater toxicity, terrestrial ecotoxicity, marine eutrophication, freshwater eutrophication, terrestrial acidification, ozone layer depletion, particulate matter formation, ionising radiation potentials	Global warming potential was observed as 12.72 CO $_2$ eq for the entire process from cultivation to torrefaction. For all other categories, only normalised results as $\%$ were recorded
Nilsson et al. (2020)	Global warming (100 years) and eutrophication potentials	The mean global warming potential was 1170 ± 460 and 1200 ± 460 kg CO ₂ eq/ha year for a 140 and 200 kg N/ha fertilisation rate. The eutrophication potential was observed as 11 ± 6.1 kg N-eq/ha to use 1 ha of land to produce perennial grass as a biofuel source in five different sites in Sweden. The variation between sites highlights the use of temporal and spatial data for assessing whole life cycle impacts
Aristizábal-Marulanda et al. (2021)	Global warming, ozone layer depletion, terrestrial acidification, freshwater eutrophication, human toxicity, photochemical oxidant formation, particulate matter formation, freshwater ecotoxicity, water depletion, fossil depletion and agricultural land occupation potentials	The study compared the use of coffee-cut stems for ethanol production concerning the production of electricity and steam. Lesser environmental impacts were observed for the latter. Some of the findings for both 1 MJ of ethanol and 1 MJ of electricity and steam are: Global warming potential for 1 MJ of ethanol = 1.37 kg CO ₂ eq
		Global warming potential for 1 MJ of electricity and steam = 0.27 kg CO $_2$ eq Ozone layer depletion potential for 1 MJ of ethanol = 2.5 \times 10 $^{-8}$ kg CFC-11 eq
		Ozone layer depletion potential for 1 MJ of electricity and steam = 0.7×10^{-8} kg CFC-11 eq
		Fossil depletion potential for 1 MJ of ethanol = 0.05 kg oil eq Fossil depletion potential for 1 MJ of electricity and steam = 0.01 kg oil eq



Table 11 (continued)

Reference	Environmental impacts considered	Findings
Brassard et al. (2021)	Global warming (100 years), ozone layer depletion, human toxicity (carcinogens), freshwater eutrophication, mineral resource use, terrestrial, freshwater acidification, water scarcity, photochemical ozone formation, human toxicity (non-carcinogens), respiratory inorganics, marine eutrophication, terrestrial eutrophication, freshwater ecotoxicity, ionising radiation, resource use (energy carriers), land use potentials	This study compared the use of primary forestry residues for pyrolysis to the business-as-usual of no use and concluded that pyrolysis could result in better environmental impacts in nine out of a total of 16 categories. Some results recorded for the use of 1000 kg of feedstock are given below Global warming potential = 906 kg CO_2 eq during pyrolysis Global warming potential = 1750 kg CO_2 eq during no use Ozone layer depletion potential = 3×10^{-5} kg CFC-11 eq during pyrolysis Ozone layer depletion potential = 1.5×10^{-5} kg Sb eq during pyrolysis Mineral resource use potential = 1.5×10^{-3} kg Sb eq during no use Human toxicity(non-carcinogens) = 5×10^{-5} cumulative toxicity units, during
		pyrolysis Human toxicity(non-carcinogens) = 2.25×10^{-4} cumulative toxicity units, during no use
Cusenza et al. (2021)	Cumulative energy demand, and	The study analysed environmental impacts for residual biomass feedstocks to produce 1 kWh of electricity to feed in the grid: Cumulative energy demand = 3.8 MJ
	Global warming (20 years), ozone layer depletion, human toxicity (non-carcinogens), human toxicity (carcinogens), particulate matter formation, ionising radiation, photochemical oxidant formation, acidification, terrestrial eutrophication, freshwater eutrophication, marine eutrophication, ecotoxicity and abiotic deletion potentials	Global warming potential = 1.1 kg CO_2 eq
		Ozone layer depletion potential = 3.3×10^{-8} kg CFC-11 eq. Abiotic depletion potential = 2.9×10^{-8} kg Sb eq. Human toxicity(non-carcinogens) = 3.6×10^{-8} cumulative toxicity units. Human toxicity(carcinogens) = 9.6×10^{-9} cumulative toxicity units. Particulate matter formation potential = 1.1×10^{-4} kg PM2.5 eq. The study conducted an environmental performance of bio-waste valorisation practices considering the double perspective of producing renewable energy and treating bio-wastes.
Da Silva et al. (2021)	Global warming potential (100 years)	Global warming potential $= 0.8 \text{ kg CO}_2$ eq for 1 kg of methanol produced using rice straw through gasification
Fu et al. (2021)	Net energy gain and global warming potential	For 1 kg of ethanol produced using rice straw through combustion in the boiler
		Net energy gain potential = 4.33 MJ Global warming potential = $0.005 \text{ kg CO}_2 \text{ eq}$



Table 11 (continued)		
Reference	Environmental impacts considered	Findings
Martillo Aseffe et al. (2021)	Global warming potential	The study analysed the use of corncob for electricity production through gasification and combustion and its impacts on the seed-corn supply chain. It concluded that if waste-to-energy scenarios are included with the agribusiness sector, it could reduce environmental impacts
	and	Global warming potential = 913 kg CO2 eq/t seed-corn through combustion
Schonhoff et al. (2021)	Endpoint indicators: human health, ecosystem quality, resource depletion Global warming (including biogenic carbon), global warming (excluding biogenic carbon), acidification, freshwater eutrophication, terrestrial eutrophication, marine eutrophication, human toxicity (carcinogens), human toxicity (non-carcinogens), land use, particulate matter formation, water depletion, freshwater ecotoxicity, ionising radiation, photochemical zone formation, abiotic depletion potentials	Global warming potential = 797 kg CO2 eq/t seed-corn through gasification. The study concluded that different production and processing pathways for Sida hermaphrodita biomass did not significantly differ in the overall environmental impacts. Moreover, when comparing Sida hermaphrodita biomass was performed with alternative biomasses like wood or Miscanthus; in most cases (except Miscanthus and mixed logs), beneficial properties of Sida hermaphrodita biomass were observed
		For 100 GJ of thermal energy from Sida hermaphrodita biomass, some of the environmental impacts recorded were:
		Global warming potential (including biogenic carbon) $\approx 12,\!000~\mathrm{kg}~\mathrm{CO_2}$ eq
		Global warming potential (excluding biogenic carbon) $\approx 1500 \text{ kg CO}_2$ eq Ozone laver denletion notential $\approx 10 \times 10^{-5} \text{ kg CFC-}11 \text{ eq}$
		Abiotic depletion potential $\approx 0.15 \text{ kg Sb eq during pyrolysis}$
		Human toxicity(carcinogens) = 14 to 19×10^{-5} cumulative toxicity units
		Human toxicity(non-carcinogens) $\approx 10.5 \times 10^{-4}$ cumulative toxicity units
Schmidt Rivera et al. (2020)	Global warming, primary	The study compared waste treatment cases for spent coffee grounds. It concluded that overall, incineration (with electricity and heat recovery) showed the least environmental impacts compared to landfilling, biodiesel production and composting, followed by the spreading of compost on land. It showed that use for waste as an energy source should follow not only waste hierarchy but also life cycle assessment. Some of the impacts recorded for incineration (with electricity and heat recovery—best case) and biodiesel production using 1 tonne of spent coffee grounds are below
	energy demand, fossil depletion, metal depletion, particulate matter formation, stratospheric ozone depletion, photochemical oxidant	Global warming potential for biodiesel production = 31 kg CO_2 eq
	(ecosystems), photochemical oxidant (humans) formation, freshwater eutrophication, marine eutrophication, terrestrial acidification, human toxicity (carcinogens), human toxicity (non-carcinogens), freshwater ecotoxicity, marine ecotoxicity and terrestrial ecotoxicity potentials	Global warming potential for incineration=525 kg CO_2 eq
		Ozone layer depletion for biodiesel production = $0.25~\mathrm{g}$ CFC-11 eq
		Ozone layer depletion for incineration = 0.18 g CFC-11 eq
		Human toxicity (non-carcinogens) for blodiesel production = $33/ \text{ kg } 1,4 \text{ DB}$ eq
		Human toxicity (non-carcinogens) for incineration = 5 kg 1,4 DB eq



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Reference	Environmental impacts considered	Findings
Al-Mawali et al. (2021)	Net energy ratio and	The study conducted LCA to analyse the transformation of waste date seed oil to biodiesel via esterification. It concluded that it is environmentally feasible to produce biodiesel from waste-derived feedstocks
	Global warming (100 years), ozone layer depletion, abiotic depletion (fossil Net energy ratio = 2.17 fuels)	Net energy ratio = 2.17
	abiotic depletion, human toxicity, freshwater ecotoxicity, marine ecotoxicity	Global warming potential = 1.11 kg CO_2 eq/kg of biodiesel produced
	terrestrial ecotoxicity, photochemical oxidant formation, acidification, and eutrophication potentials	Abiotic depletion potential = 2×10^{-5} kg Sb eq/kg of biodiesel produced
		Abiotic depletion (fossil fuels) potential = 19 MJ/kg of biodiesel produced
		Human toxicity potential = 0.6 kg 1,4 DB eq/kg of biodiesel produced
Yang et al. (2021)	Global warming (100 years), ozone layer depletion, abiotic depletion, human toxicity, freshwater ecotoxicity, marine ecotoxicity	The study used LCA to evaluate the potential of biochar for carbon sequestration produced using crop residue in China. The carbon sequestration potential of a country-level biochar system was estimated at 500×10^{12} kg CO ₂ eq for a year. This accounts for 4.50% of the total greenhouse gas emissions from China, which could be mitigated by use of biochar
	terrestrial ecotoxicity, photochemical oxidant formation, acidification, and eutrophication potentials	Global warming potential = -5×10^{14} kg CO_2 eq/kg of biochar
		Abiotic depletion potential = 4.5×10^7 kg Sb eq/kg of biochar
Saranya and Ramachandra, (2020)	Net energy ratio and	The study compared acid catalysis and biocatalysis for the use of microalgal biomass feedstocks (produced in 1 ha of land—FU) for biodiesel production. The study concluded that biocatalysis was the best route to use with the following impacts
	Global warming (100 years), abiotic depletion, abiotic depletion (fossil fuels)	Net energy ratio = 18.8
	photochemical oxidant formation, acidification, and eutrophication potentials	Global warming potential = $2000 - 3000 \text{ kg CO}_2 \text{ eq/FU}$
		Abiotic depletion potential = $0.004 \text{ kg Sb eq/FU}$
Bora et al. (2020)	Endpoint indicators: global warming potential, human health, ecosystem quality, resource depletion	This study computed 15 mid-point indicators for thermochemical conversion of poultry litter to fuel and biochar in the USA. However, the results were aggregated to present endpoint indicators for 1000 kg of poultry litter (FU)
		Global warming potential of thermochemical conversion = 659 to 1192 kg ${\rm CO}_2$ eq/FU
		Global warming potential for spreading on fields = $1410 \text{ kg CO}_2 \text{ eq/ FU}$



Table 11 (continued)		
Reference	Environmental impacts considered	Findings
Al-Muhtaseb et al. (2021)	Net energy ratio and	The study conducted LCA to analyse the transformation of waste date seed oil to biodiesel via esterification. It concluded that it is environmentally feasible to produce biodiesel from waste-derived feedstocks
	Global warming (100 years), ozone layer depletion, abiotic depletion (fossil Net energy ratio = 2.23 fuels), abiotic depletion, human toxicity, freshwater ecotoxicity, marine ecotoxicity	Net energy ratio $= 2.23$
	terrestrial ecotoxicity, photochemical oxidant formation, acidification, and Global warming potential = 1.13 kg CO ₂ eq/kg of biodiesel produced eutrophication potentials	Global warming potential = 1.13 kg CO_2 eq/kg of biodiesel produced
		Abiotic depletion potential = 3×10^{-6} kg Sb eq/kg of biodiesel produced
		Abiotic depletion (fossil fuels) potential = 26 MJ/kg of biodiesel produced

CFC-11: Trichlorofluoromethane; DB: dichlorobenzene; EU-27: The European Union; eq: Equivalent; FU: Functional unit; HFO: Heavy fuel oil; LCA: Life cycle assessment; NER: Net energy ratio; PEI: Potential environmental impact; PM2.5: Particulate matter with < 2.5 µm in diameter; PM10: Particulate matter with < 10 µm in diameter uncertainties arise due to statistical variation, subjective judgement, linguistic imprecision, variability in space and time, inherent randomness, expert disagreement and model approximations. These uncertainties can be propagated through the model using Monte Carlo simulations for parameter uncertainties or by considering different scenarios for biofuel production. Though it was noted that about 60% of the reviewed studies considered scenario analysis, only 10% of the studies did uncertainty analysis by accounting for parameter uncertainties (Fig. 7).

Sensitivity analysis identifies which process of the biodiesel production life cycle contributes directly to the burdensome environmental footprints. Relatedly, if impacts in environmental categories are to be minimised, these will be the processes where future research and development should focus on (Al-Muhtaseb et al. 2021). 50% of the studies reviewed here conducted sensitivity analysis.

Therefore, for reliable and robust decision-making, it is necessary to analyse sensitivity and uncertainty for various scenarios. There were about ten studies identified with no analysis on sensitivity or uncertainty (due to parameters and scenarios). Moreover, the presence of only four studies with scenario, sensitivity and uncertainty analyses poses questions on the applicability of findings presented in the LCA studies. There was no study identified that included all three phases of the life cycle of biofuel production system boundary (described in Fig. 5) and uncertainty, scenario and sensitivity analyses in the 40 reviewed studies. This presents a considerable knowledge gap when it comes to the use of LCA studies for strategic decision-making.

Interpretation of results

This stage of the assessment includes making interpretations, drawing conclusions, identifying the phases or processes that can be improved in the life cycle of a biofuel production chain to improve the environmental feasibility of the environmental system. This stage could also involve presenting and communicating results to stakeholders.

Key points observed in life cycle assessment studies

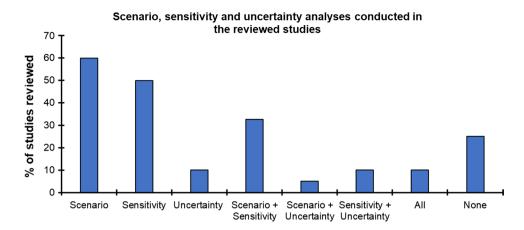
We conducted an intensive critical review of 40 LCA studies, including methods and findings. In this section, we present key points observed in these studies.

There was no study identified that included all the three phases of biofuel production along with uncertainty, sensitivity and scenario analyses.

Moreover, most of the studies that computed net energy ratio (Al-Mawali et al. 2021; Al-Muhtaseb et al. 2021; Dasan et al. 2019; Im-Orb and Arpornwichanop 2020; Reaño 2020; Saranya and Ramachandra 2020), which is defined as the ratio of output energy/input energy for the overall process



Fig. 7 Details of the scenario, sensitivity and uncertainty analyses conducted in the reviewed studies (N=40)



(Pleanjai and Gheewala 2009), recorded net energy ratio > 1, showing the importance of biofuels as energy sources.

Generally recognised contentious issue to produce biofuels was land use which could occur due to natural land, agricultural and urban land transformation. This was highlighted by the fact that 7% of the studies used land use as a functional unit (Fig. 5); furthermore, about 15% of the reviewed studies analysed impacts on land use (Aberilla et al. 2019; Chung et al. 2019; Ubando et al. 2020; Aristizábal-Marulanda et al. 2021; Brassard et al. 2021; Schonhoff et al. 2021). It was also noted that the use of perennial energy crops is an interesting approach towards mitigation of greenhouse gas emissions; however, it could result in loss of biodiversity for the European Union and UK (EU-27 + UK) (Di Fulvio et al. 2019) (Table 11).

Other studies focused on water depletion and concluded that while producing biofuels can mitigate greenhouse gas emissions, it is also necessary to compute water depletion during crop production and biomass processing (Aberilla et al. 2019; Quispe et al. 2019; Ubando et al. 2020; Aristizábal-Marulanda et al. 2021; Schonhoff et al. 2021). In fact, Zhu et al. (2019) concluded that water depletion for biofuel production from cotton straws was lower than biooil power generation, however, much greater than observed for other renewable sources of energy (such as geothermal, solar photovoltaic and wind power). Most of the water use occurred due to biomass agricultural production, accounting for 84.6% of the total water use.

The impacts on land use and water depletion due to energy crop production show that waste-derived feedstocks could provide more sustainable energy sources. Waste-to-energy applications for biomass could mitigate the use of land, fertilisers and water for agriculture of energy crops. This is also in accordance with zero-waste hierarchy for management of waste biomass (Refuse/ redesign > Reduce > Reuse > Recycle > Material and chemicals recovery > Residuals management > Unacceptable, e.g. landfilling of non-stabilised waste/energy recovery) (Simon, 2019). In fact, there was

only one study identified that highlighted that for spent coffee grounds, incineration is a better route compared to biofuel production (Schmidt Rivera et al. 2020).

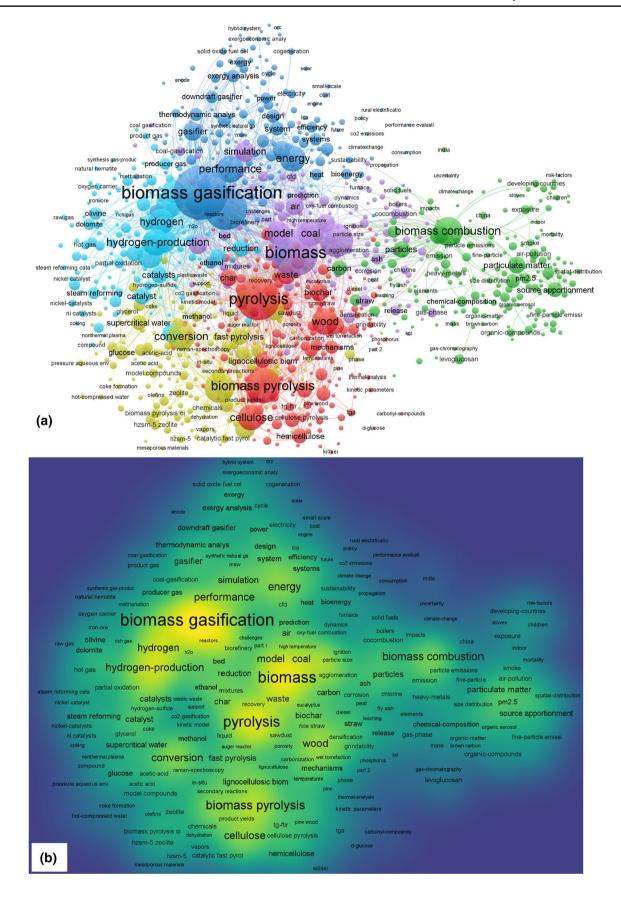
Finally, focusing on the comparison of biological and thermochemical pathways, in general, it was observed in the comparative studies that thermochemical processes showed lesser environmental impacts compared to biological processes for the same biomass and geographical and temporal span (Ardolino and Arena 2019; Derose et al. 2019). Even in Aberilla et al. (2019), which showed higher environmental impacts in the thermochemical process, the greenhouse gas emissions for gasification were lower than that of anaerobic digestion for rice and coconut residues.

Bibliometric analysis

Figure 8a, b depicts the bibliometric analysis mapping originated from the Web of Science core collection for the network visualisation and density visualisation, respectively. Firstly, the data were exported 500 entries at a time of 9947 results and then were fed into the VOSviewer software that plotted the data. The type of analysis used herein was cooccurrence, and all keywords were included, as well as the fractional counting method. We observed direct clusters connecting identifiable keywords to broad topics such as thermochemical, biochemical and processes associated with those two routes (gasification, pyrolysis, hydrothermal liquefaction, combustion, torrefaction, anaerobic digestion and fermentation). This enabled the visualisation of most of the significant keywords in publications in the period of 1970–2021 that were associated with the thermochemical and biochemical conversion routes of biomass.

It is evident from Fig. 8a, b that keywords that have seen a significant increase in popularity and, as a result, progress in keyword research such as biomass gasification, pyrolysis and combustion as they part of the thermochemical route in biomass conversion. In addition, other correlated keywords







∢Fig. 8 Bibliometric mapping of biomass conversion processes into biofuels **a** network visualisation and **b** density visualisation maps. The bibliometric mapping was performed between 1970 and 2021

have shown in the bibliometric mapping, such as hydrogen production, catalysts and fuel performance. This signifies that the process of thermochemical conversion is at a very mature stage in terms of research and development, as demonstrated by the prominence of publication keywords over the last 51 years. Because of its higher productivity, economic viability and existing infrastructure compatibility resources, it is ultimately readily available and easily scaled up for the industrial sector.

Although gasification appears to be most researched in the thermochemical route, this could be down to the fact that gasification technology has existed longer than counterpart technologies, apart from combustion. This also does not indicate that it is the most efficient thermochemical technology in process efficiency and product quality. For example, when using gasification, there is a need to remove the hydrogen sulphide and clean the synthesis gas produced and other requirements.

On the other hand, the biochemical conversion route is less favourable since it suffers from certain limitations, such as its time-consuming process and low product yield and product inhabitation. Biochemical conversion keywords are shown in Fig. 8a, b, such as ethanol production, bio-hydrogen and others.

Conclusion

Biomass as an affordable, reliable and sustainable energy source contributes 9% (~51 EJ) of the global overall primary energy supply. Thermochemical and biochemical technologies are the two main routes employed to convert biomass into biofuels. The former route includes hydrothermal liquefaction, pyrolysis, torrefaction, gasification and combustion processes, while the latter route consists of fermentation and anaerobic digestion processes.

Herein, we critically reviewed each individual route along with the integration between hydrothermal and biochemical routes of biomass utilisation from a bioeconomy perspective. Both routes have drawbacks: the former method usually involves a high energy intake along with solvent or catalyst addition. In contrast, the latter route has a lengthy cycle period and is less efficient in breaking down recalcitrant biomass materials. Thus, combining those two routes can be promising by incorporating the benefits of both methods in biofuel processing. However, there are outstanding challenges associated with integration between those two routes. For instance, the catalysts or solvent utilisation of the thermochemical route can result in poisoning or killing the

microorganism or generate various inhibitors that can affect the biological progress routes. Furthermore, this integration may lead to additional costs.

Moreover, to understand the recent advances in evaluating environmental impacts due to biofuel production, we conducted an intensive critical review of 40 life cycle assessment (LCA) studies published from the years 2019–2021, including methods and findings. The important methods and key findings observed were:

- Only eight studies included all three phases of biofuel production (which includes biomass cultivation, biofuel production process and biofuel use and end-of-life management phase).
- Waste-derived feedstocks could provide more sustainable energy sources by mitigating impacts on land use and water depletion incurred during the production of energy crops.
- Focusing on thermochemical and biological processes for the same biomass feedstock and geographical and temporal span, thermochemical processes caused lesser greenhouse gas emissions compared to biological pathways.

This review has suggested interesting new avenues for evaluation of environmental impacts of the biofuel production chain and key outcomes from a range of biofuel production processes. Based on the bibliometric mapping (network and density visualisation maps) from the Web of Science core collection, we have identified that the thermochemical conversion route of biomass is more researched and far outweighed and understood than the biochemical counterpart route of research outputs. This indicates that the biochemical route suffers from specific gaps in the research, as shown from the lack of impact in the bibliometric mapping analysis, thus opening doors for a scope for further research in this area.

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Declarations

Conflict of interest The authors declare no conflict of interest.



Ethical approval The views and opinions expressed in this review do not necessarily reflect those of the European Commission or the Special EU Programmes Body (SEUPB).

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