Clean Energy Production Technologies Series Editors: Neha Srivastava · P. K. Mishra

Manish Srivastava Neha Srivastava · P. K. Mishra Vijai Kumar Gupta *Editors*

Nanomaterials in Biofuels Research



Clean Energy Production Technologies

Series Editors

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The consumption of fossil fuels has been continuously increasing around the globe and simultaneously becoming the primary cause of global warming as well as environmental pollution. Due to limited life span of fossil fuels and limited alternate energy options, energy crises is important concern faced by the world. Amidst these complex environmental and economic scenarios, renewable energy alternates such as biodiesel, hydrogen, wind, solar and bioenergy sources, which can produce energy with zero carbon residue are emerging as excellent clean energy source. For maximizing the efficiency and productivity of clean fuels via green & renewable methods, it's crucial to understand the configuration, sustainability and technoeconomic feasibility of these promising energy alternates. The book series presents a comprehensive coverage combining the domains of exploring clean sources of energy and ensuring its production in an economical as well as ecologically feasible fashion. Series involves renowned experts and academicians as volume-editors and authors, from all the regions of the world. Series brings forth latest research, approaches and perspectives on clean energy production from both developed and developing parts of world under one umbrella. It is curated and developed by authoritative institutions and experts to serves global readership on this theme.

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Nanomaterials in Biofuels Research



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Foreword



Renewable energy is a potential and bright replacement of fossil fuels in the form of bioenergy. Bioenergy options, like biohydrogen, biogas, biomethane, bioethanol, biomethanol, biobutenol, algal biofuels, and biodiesel, are the promising alternative source of renewable energy which can contribute towards green and sustainable environment by vomiting pollution. Numbers of tremendous efforts have been taken worldwide for the development of innovative technologies which can evenly distribute bioenergy options. Nevertheless, in spite of continuous practice towards the development of low-cost technology for easy commercialization of biofuels, practical viability is still far away and unequally distributed. The application of nanotechnology is emerging as a new area in the field of biofuels production via various means such as catalyst, enzyme, and microbial immobilizer. Nanomaterials have enough potential for commercial exploitation, and their industrial market is expected to become more broad in the combination of biofuels and nanotechnology area for viable and sustainable economic biofuels production.

Based on the current scenario, publication of the book *Nanomaterials in Biofuels Research* is much required effort in this series, and I am happy to write this positive message with great satisfaction. The book contains 11 essential chapters focusing on nanomaterial synthesis and characterization for biofuels application. Current global scenario of biofuels and role of nanotechnology in biofuels production, different immobilization patterns to improve biofuels production, and synthesis and mechanism of nanomaterial for economic biofuels production with the green approach are presented and discussed in details. The book suggests a new insight with advancement and sustainable solution to overcome existing loopholes in nanotechnology-based biofuels production process. The book predicated that nanotechnology will play a key role in the near future in the area of the biofuels production process, and hence, this book will serve as an asset for the researcher working in the relevant area including scientists, researchers, teachers, and students.

I am taking the opportunity to congratulate Dr. Manish Srivastava (DU, Delhi), Dr. Neha Srivastava (IIT-BHU, Varanasi), Prof. Dr. P.K. Mishra (IIT-BHU, Varanasi), and Dr. Vijai Kumar Gupta (TTU) for their effort to bring out the publication of *Nanomaterials in Biofuels Research* to cater the long-felt need of scientists, teachers, researchers, and students. My sincere thanks are for the editors for their untiring efforts, devotion, and dedication in this endeavor. All the authors and editors of this book deserve sincere appreciation for their commendable achievements.

Enhanced Composites and Structures Center, Dr. Vijay Kumar Thakur School of Aerospace, Transport and Manufacturing, Cranfield University, Bedfordshire, UK 21 March, 2019

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The editors are thankful to all the academicians and scientists whose contributions have enriched this volume. We also express our deep sense of gratitude to our parents whose blessings have always prompted us to pursue academic activities deeply. It is quite possible that in a work of this nature, some mistakes might have crept in text inadvertently, and for these, we owe undiluted responsibility. We are grateful to all the authors for their contribution to the present book. We are also thankful to Springer Nature for giving this opportunity to the editors and the Department of Chemical Engineering and Technology, IIT (BHU), Varanasi, UP, India, for all their technical support. We thank them from the core of our heart. Editor Manish Srivastava acknowledges the DST, Govt of India for awarding the DST-INSPIRE Faculty Award [IFA13-MS-02] 2014 and also Science and Engineering Research Board for SERB-Research Scientist Award-2019.

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1

Biofuel: Types and Process Overview

Ajay Kumar Chauhan

Abstract

Excessive use of conventional fossil fuels resulted in a hike in price, exhaustion, and change in climatic conditions. Therefore, a novel route to biofuel generation is another feasible option of sustainable process development. Continuous upgradation of technologies for biofuel generations from the first generation (1G) to the fourth generation (4G) gives new hopes to fulfill energy demands. Biofuel generation from multiple approaches such as physical, biological (includes microbial and enzymatic), chemical, and biochemical catalysis with nanotechnology from multiple feedstocks is the key to biofuel generation. Suitable conversion of cellulosic biorefinery and lignin biorefinery (via lignin valorization) is the key for complete utilization of lignocellulosic biomass. Utilization of the biological methods includes the use of microbial machinery from different domains which might open the door toward an environmentally benign process. Nanotechnology and its potential application with 1G to 4G have future promises for increasing yield and integration of technology. So, this book chapter covers a detailed process overview of biofuel generations and its challenges with the hope of overcoming it.

Keywords

Sustainable \cdot Microbial machinery \cdot Catalysis \cdot 1G \cdot 4G \cdot Lignin valorization \cdot Nanotechnology

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Abbreviations

1G	First generation
2G	Second generation
20 3G	Third generation
4G	Fourth generation
BGA	Blue-green algae
C	Carbon
CBDH	Cellobiose dehydrogenase
Centimeter	cm
CH4	Methane
CNG	Compressed natural gases
CO	Carbon monoxide
CO_2	Carbon dioxide
DME	Dimethyl ether
DNA	Deoxyribonucleic acid
ED	Entner-Doudorof
EMP	Embden-Meyerhof-Parnas
ETBE	Ethyl butyl ether
FAME	Fatty acid methyl esters
FT	Fischer-Tropsch
GHG	Greenhouse gases
H ₂	Hydrogen
HC	Hydrocarbons
HDO	Hydrodeoxygenation
Lignin peroxidase	LiP
LPG	Liquefied petroleum gas
LPMO	Lytic polysaccharide monoxygenase
Manganese peroxidase	MnP
Mn	Manganese
MPa	Mega Pascal
N ₂	Nitrogen
NO _x	Nitrogen oxide
NPs	Nanoparticles
PBR	Photobioreactor
PEG	Polyethylene glycol
PM	Particulate matter
SNG	Syngas

1.1 Introduction

Increase in population and economic growth has increased the energy demand, and fossil fuels fulfill this energy demand as per the data revealed from the International Energy Agency (IEA) in 2011. As the consumption of fossil fuels is nonrenewable and emits greenhouse gases (GHG) which severely deplete environmental conditions (Buragohain et al. 2010), therefore, the notable increase in renewable energy sources such as solar, wind, and biomass gives new hope of a sustainable process. Biofuels are the renewable energy sources defined as the energy derived from biological carbon fixation, which plays a pivotal role in new energy sources. Therefore, the production of biofuel requires extensive research and development of technologies. These biomass require suitable conversion of biomass into biobutanol, bioethanol, bio-dimethyl ether, biomethane, Fischer-Tropsch (FT) fuel, biohydrogen, algae, and halophilic lipid-based biofuels.

Plant biomass is an abundant source of renewable energy which comprises of the various components such as carbon, hydrogen, oxygen, and traces of minerals. Availability of plant biomass is more reliable compared to another energy sources such as solar energy, wind energy, and hydropower, and it can easily grow in variable environmental conditions. Moreover, the low density of biomass over fossil fuel makes it more feasible for storing and transportation purposes. In developing countries, utilization of plant biomass for household purposes (burning of wood) underestimates its value as a potential biofuel. The burning of fossil fuels and plant biomass is the main contributor to the increase in CO_2 level and has a direct impact on global warming. GHG emissions and global warming open up the door for the novel processes of biofuel generation.

Lignocellulosic biomass is made up of highly complex biopolymer of cellulose (40–60%), hemicellulose (10–40%), and lignin (15–30%) as major component which gives support to plant, and it is resistant to various microbial degradations as well as biochemical conversions (Himmel et al. 2007; DeMartini et al. 2013; Schutyser et al. 2018). Cellulose and hemicellulose mainly consist monomeric sugar unit, and after pre-treatment, lignocellulosic biomass sugar component via further saccharification is converted into suitable biofuel (Chen 2014). However, the lignin component is made up of the phenylpropane unit, which is more recalcitrant than the cellulose and hemicellulose. During kraft pulping process, lignin is utilized for the co-power generation, which can be separated, and suitably valorized into useful precursors for biofuel generation. However, the hydrogenation process is being used as a suitable method for converting lignin into a valuable product and converting raw material into biofuel (Chen 2014; Abdullah et al. 2017). Lignin valorization strategy utilizes the complete conversion of lignocellulosic biomass into biofuel generation.

Different methodologies are adapted from time to time for treating the plant biomass for the biorefinery route. This methodology has its advantages and limitations. Here, pulp and paper industries are the best examples of lignocellulosic biomass biorefinery, and they are emerging as fermentative production of bioethanol (Ragnar et al. 2000; Hahn-Hägerdal et al. 2006; Limayem and Ricke 2012). Various existing conventional techniques result in low yield of primary products obtained from the lignocellulosic biomass, but in case of lignin, it can be suitably valorized into valuable products (Davis et al. 2013). Further, the optimization process is done by understanding the process. Even biofuels productions have a direct impact on carbon sink; therefore, biofuels produced from oil-based fuels always have a better choice compared to others GHG emissions into the environment critics of biofuels, which ultimately depends on the route of production. Another selection of biofuel has the debate over food versus fuel; land used for cultivation for feedstock always competes with food demand and land uses for productions. Therefore, the governments of the countries take so many initiatives for the continuous growth of the biofuel industry, which includes providing fund for research and development along with mandating laws of Environmental Protection Agency (EPA) for blending of biofuel with conventional fuel. Renewable Fuel Standard (RFS) provides harness power of biofuels and infrastructure, and high productions of biofuels must be produced and widely available for customers with the competitive price of conventional biofuel (Richards 2013).

1.2 Classification of Biofuels

On the basis of origin of raw material biofuels are classified into first-generation (1G), second-generation (2G), third-generation (3G), and fourth-generation (4G) (Demirbas 2011). The first-generation biofuel, sugar, starch, vegetable oils, and fats are converted into bio-alcohol from (n = 1-4) fatty acid methyl esters (FAME). Second-generation biofuel depends on the carbon negative in terms of the carbon dioxide concentration in the environment, which majorly depends on the lignocellulosic biomass (plant material) (Gomez et al. 2008). The third and fourth generation of the biofuel utilize the algae and the blue-green algae (BGA) machinery for converting lipid into biofuel. In third generation, algae is directly used for the production of the biofuel, where in the fourth generation, metabolic engineered algae from the oxygenic photosynthetic microorganism create artificial carbon reservoir (Lü et al. 2011). Further, they are known as chemically synthesized biofuel and biologically synthesized biofuel. In chemical synthesis, biofuel is the generation with the help of a catalyst, and the action of various parameters such as pH, temperature, and pressure has played a significant role. Moreover, chemical methods, not benign environmental methods, in terms of the yield are preferable over biological methods. In the chemical process, the use of the catalyst is preferred over the various physical processes because of the more specific product formation and low cost of separation. Generally, physical processes yield more complex product formations, which are not easily separable and thus require a higher price for downstream processing. In biological methods, lignocellulosic biomass is microbially treated with bacteria, halophilic archaea, fungi, and algae. Moreover, other than these biological processes include various kinds of biocatalysts known as the

enzymes used for the production of the biofuels' generation. These enzymes are cellulases, xylanases, and lignolytic enzymes such as lignin peroxidase (LiP), manganese peroxidase (MnP), laccase, lytic polysaccharide monooxygenase (LPMO), multicopper oxidases, cellobiose dehydrogenase (CBDH), and lipases. Moreover, enzymatic use of the various processes of biofuel generations is the most environmentally friendly process which overcomes the numerous barriers of the chemical processes. Benefits of the enzymatic process yield specific product formation at optimum pH, temperature, and pressure. Therefore, enzyme from extremophile enzymes have added extra-advantages of various product formations at broad pH range and different temperature ranges from 4 to 80 °C, and they even act on higher pressure making them suitable for industrial process. To date, all the biological processes require a higher cost of production than the chemical process, and the cost-effectiveness of the biological process can only be reduced by the processes understanding of the microbial pathways, enzymatic reduction, and its substrate utilization strategy. However, new biological modified strains have shown various promises to overcome all barriers. In the last decade, there had been four industrial plants that started for the commercial production of the biofuel listed as "Project Liberty" by the joint venture of POET-DSM, 2G ethanol at Dupont, Abengoa Bioenergy Hybrid Kansas, and Crescentino by Beta Renewables in Europe (Valdivia et al. 2016).

Raw Materials

Availability of raw material is the key to success to achieve better economy and viability of the process. Biodiversity available on the earth is a good sink of energy as in the form of hydrocarbon and the production of hydrocarbon for biofuel generations are listed in the Table 1.1. As per requirement, feedstock can be classified on the basis of the source of origin. Feedstock can suitably be converted into biofuel and the abundance of this material in various forms are the source for different biofuel generations.

Biomass type	Examples		
Forest product	Wood, logging, residue, trees, shrubs, and wood residues, sawdust, bark		
Bio-renewable	Agricultural waste, crop residues, mill wood waste, urban wood wastes,		
waste	urban organic waste		
Energy crops	Short-rotation woody crops, sugar crops, forage crops, oilseed crops,		
	switchgrass, Miscanthus		
Aquatic plants	Algae, water weed, water hyacinth, reed, and rushes		
Food crops	Grains, oil crops		
Sugar crops	Sugar cane, sugar beets, molasses, Sorghum		
Landfill	Hazardous waste, nonhazardous waste, inert waste, liquid waste		
Organic wastes	Municipal solid waste (MSW), industrial organic		
Algae	Prokaryotic algae, eukaryotic algae, kelps		
Mosses	Bryophyta, Polytrichales		
Lichen Crustose lichens, foliose lichens, fruticose lichens			

 Table 1.1
 Classification of biomass feedstock (Speight 2011; Sikarwar et al. 2017)

1.3 First-Generation Biorefinery

Concept of the biorefinery comes in the limelight for the researcher's interest due to the limited resources of conventional energy sources and the rising price of the oil in the global market. 1G is produced from the food crops such as corn, sugar cane, rapeseed, cassava, etc. The first-generation biofuels are biodiesel, ethanol, and biogas, which are primarily producing so far. Biodiesel is the alternative biofuel to the diesel which is produced by the transesterification of the various vegetable oils and fats. However, small modification in the properties of biodiesel can make it a better substitute for the diesel. Whereas, bioethanol is the better substitute for the gasoline and is also known as the flexi fuel. Moreover, bioethanol is produced from various sources via fermentation of the sugar or starch material. Bioethanol is used as a feedstock for ethyl butyl ether (ETBE), which can easily blend with the gasoline. Another biofuel like biogas and biomethane is produced from the anaerobic digestion of the household and municipal waste. Biodiesel, bioethanol, and biogas can easily be produced from the food crop feedstock, but the increasing demand of the edible oils makes it difficult to use food crop for the biorefinery (Lee et al. 2014b). The big challenge of the production of the first-generation biofuel is the food to feedstock, high uses of the groundwater, fertilizer, and pesticide use on the crop.

1.3.1 Transesterification

The oil obtained from the various feedstock suitably transesterified into the fatty acid methyl esters (FAME) with the help of alcohol. The alcoholic group readily reacts with the triglyceride units in the presence of a suitable catalyst (homogenous or heterogeneous) with oil group and releases biodiesel and glycerol as high-value coproduct (Kulkarni et al. 2006; Meher et al. 2006). This mechanism can suitably be applied to all kinds of oil obtained from using algae, halophilic isolate, animal fat, and vegetable oil. The transesterification reaction is an equilibrium reaction in which continuous mixing is required for getting methyl esters (biodiesel). However, biodiesel obtained from transesterification reactions (given below) has similar characteristics as compared to diesel.

1.3.2 Ethanol Production

Ethanol is produced from sugar, starch, and cellulose-containing crops. Alcohol produced from the food crops and lignocellulosic biomass is known as the bioethanol, which can be easily produced by the biochemical process (Minteer 2016). Structure of starch consists of the polymeric unit of Dglucopyranose, which is first converted into glucose molecule with the action of enzymes "amylase." Amylase produces the maltodextrin by liquefaction process. The process of saccharification of dextrin and oligosaccharides is hydrolyzed by the enzymes pullulanase and glucoamylase. The action of these enzyme converts the substrate into the smaller unit

of glucose, maltose, and isomaltose. Then further, fermentation is carried by the yeast at 30 °C for ethanol production (Lee et al. 2014b).

1.3.3 Fermentation

The fermentation process converts organic compounds into simpler products with the action of the microbes and enzymes. Ethanol production from glucose utilizes fermentative consumption of pyruvate as an intermediate product (Ingram et al. 1987). The process can be classified as the aerobic and anaerobic among microbes depending up on oxygen requirement. Many microorganisms such as yeast (Saccharomyces cerevisiae), bacteria (Zymomonas mobilis), and halophilic bacteria (Halanaerobium saccharolyticum) convert butyrate and ethanol from glycerol and hydrogen from senegalensis (Kivistö et al. 2010). Most eukaryotes and prokaryotes follow Embden-Meyerhof-Parnas (EMP) also known as glycolysis pathway, and Zymomonas mobilis follows Entner-Doudorof (ED) pathway. Z. mobilis produces up to 97% of the theoretical yield of ethanol and also produces a 2.5-fold higher yield compared to S. cerevisiae (Weber et al. 2010). Moreover, Z. mobilis is suitably engineered for utilizing various types of sugar of lignocellulosic biomass such as glucose, mannose, and xylose for fermentation (Yanase et al. 2012). Another microorganism such as *Clostridia* can secrete hydrolytic complex to utilize both types of sugars (Lütke-Eversloh and Bahl 2011). However, these microorganisms are capable enough to ferment into C6 and C5 sugar or a mixture of both.

1.3.4 Anaerobic Fermentation

This process produces biogas under the absence of oxygen. However, anaerobic condition facilitates the growth of microorganism on organic biomass and converts it into methane (60–70%) and carbon dioxide (Naik et al. 2010). When we consider from the economic point, anaerobic digestion is an economically feasible process and also produces the biofertilizers for agricultural uses. The conventional process generally produces the gaseous fuel for the household purposes and electricity generation, but newer technology now focuses on the liquid fuel to overcome the separation process cost (Lee et al. 2014b). Utilizing simultaneous saccharification and fermentation (SSF) and anaerobic digestion in the case of birch wood by *Saccharomyces cerevisiae* efficiently converts sugar into ethanol, and stillage was subsequently utilized for the biomethane production (Kalyani et al. 2017).

1.3.5 Whole-Crop Utilization

In this concept, the whole crop utilization is applied for conversion of the whole plant into a useful product. Like *Jatropha*, oilseed contains 30–40% of oil. In the whole-crop utilization, different parts of the crop are treated separately. For

example, oil produces the biodiesel, which can further be used for the conversion into oleochemicals. Remaining de-oiled cake is used for the gasification purposes, which can further convert into the syngas. Syngas finally is utilized for the production of various fuels.

1.4 Second-Generation Biofuels

Second-generation biofuels are produced from the nonfood crops such as agricultural residues (wheat straw, rice straw, and sugar cane bagasse, etc.) and organic waste from municipal solid waste. 2G biofuels have a key advantage over the first generation in terms of no competition with the food resources, their product formation with less emission of GHG, and less land requirement for the raw material. However, the mixed feedstock concept involved heterogeneous feed. When compared with 1G, the technological risk of operation is higher and also requires higher input cost for the process development. Process understanding in terms of the route selection requires higher technical knowledge and process modification (Margesin and Schinner 2001). Plant biomass consists of the plant cell, which comprised of the 75% of polysaccharide as a potential source of carbon (Pauly and Keegstra 2008). Lignocellulosic biomass is converted into the advanced biofuels through hydrolysis and fermentation (bioethanol and biobutanol) or gasification (FT biodiesel, dio-DME, and SNG). Raw materials for the advanced biofuels are rotational crops (poplar and Eucalyptus), Miscanthus, switchgrasses, and agricultural waste residues (Naik et al. 2010). Advanced bioethanol can be a suitable alternative of gasoline, and the sugars obtained from the hydrolysis are converted into Fischer-Tropsch diesel or biomass to liquids for the full substitution of the conventional diesel. In this, the lignocellulosic biomass with the help of suitable gasification is converted into syngas. Further, these syngas is utilized as the source of liquid hydrocarbon (kerosene, diesel, or bio-SNG), or this syngas can have transformed into the methane or dimethyl ether (DME) (Naik et al. 2010). 2G biofuels involve two main route strategies. First one is the thermochemical conversion route in which biomass conversion utilizes the thermal decay and chemical reformation in the presence of different concentrations of the oxygen. In the second one, biochemical conversion utilizes the conversion with the help of microbes using catalysts/biocatalysts. The major advantage of the thermochemical conversion over the biochemical is the conversion of the whole organic component into biofuel, but in the case of the biochemical conversion, it is only the polysaccharide that is converted into biofuel (Gomez et al. 2008).

1.4.1 Physical Process

1.4.1.1 Mechanical Extraction

In the mechanical extraction processes, crude oil from the oilseeds is recovered from the action of an external mechanical pressure generated by screw press. However, a small amount of the de-oiled cake (around 15–20% of oil) from the

oilseed also comes out with the recovered oil. In some mechanical extraction processes, pre-pressing is followed by the solvent extraction for the high oil recovery around 30–40% (Stevens and Verhé 2004). On the other hand, in the full pressing system, high pressure of 9.5 MPa is applied to take out as possible oil content.

1.4.1.2 Briquetting

Various biomass resources obtained from agricultural, forest residues and other waste biomass come under second-generation fuel and are difficult to use as a direct biofuel due to uneven bulk. Therefore, this problem is overcome by the method of densification of the material to give near uniform compact shape. These densification methods are employed as pressing and maceration or combinations of both methods. In press, the density of the material increases by applying mechanical pressure in the early stage of compression. Further, an increase in the pressure has a negative impact on the density of material until it reaches the density of water, whereas in maceration, no such correlation of pressure exists, but chopping, grinding, and pulverization of the large material (like branches of the tree) are more effective methods. These methods are more effective compared to ultrafine grind material (Stevens and Verhé 2004; Osamu and Carl 1989).

1.4.1.3 Distillation

In this process, raw materials are crushed followed by steam distillation. Further, collective vapors are condensed separately which then turn into liquid form. Advanced distillation process known as molecular distillation is applied where conventional methods cannot be applied for the productions of the fragrances (Stevens and Verhé 2004).

1.4.2 Thermochemical Conversion

Thermochemical conversion involves heating of biomass followed by chemical conversion into biofuel. This process includes combustion, gasification, liquefaction, and pyrolysis under oxygen-deficient or oxygen-controlled conditions and results in the generation of syngas majorly comprising of H_2 and CO. Further, this syngas is processed into gaseous or liquid products (Lee et al. 2014b).

1.4.2.1 Combustion

In the combustion process, the chemical reactions usually take place in the air where fuel and air react with each other. Therefore, the products formed during this process are carbon dioxide and water, and it liberates energy in the form of heat (Lee et al. 2014b). Combustion is the simplest way to use biomass into energy with the low yield of products.

1.4.2.2 Gasification

Gasification is a type of pyrolysis carried out at a very high temperature and produces a mixture of gases known as producer gas (CO, H_2 , CH_4 , CO_2 , and N_2). In the

case of biomass, gasification producers are methanol, ammonia, and ethylene. Gasification utilizes conversion of the lignocellulosic biomass into the biofuel since the past 30 years. Gasification has many processes which simultaneously go on, such as drying, pyrolysis, and partial oxidation (Bridgwater and Boocock 2013). The syngas produced from the gasification process can be through catalytic and non-catalytic processes. In the non-catalytic process, higher temperature ranges 1000–1300 °C for the syngas production; in the catalytic process, it is operated at around 900 °C due to more advances in catalysis (Lee et al. 2014b). The key in gasification is controlling H₂/CO ratio for getting the different product in higher concentration (Melin et al. 2015).

1.4.2.2.1 Biomethanol

The reaction of the biomass requires oxygen to produce producer gas, which is the mixture of various gases such as CO, CO_2 , H_2 , CH_4 , and N_2 (known as syngas). These producer gases have the potential to produce stationary power generation. Gases are first cooled down to 300 °C. Further, gases are fed to shift reactor with the controlled condition of H_2/CO ratio provided to 2.1 for the methanol synthesis. Remaining acid gases are then removed by scrubber or dissolved in solvent (Vamvuka et al. 2004). Further, high pressure of 6.1 MPa is used to compress purified syngas to increase methanol synthesis:

$$CO + 2H_2 \leftrightarrow CH_3OH$$

 $CO_2 + 3H_2 \leftrightarrow CH_3OH + H_2O$

1.4.2.2.2 Methane

All conditions for the preparation of the syngas follow the same procedure. Here, the use of catalyst which catalyzes tar with methane decomposition requires higher H_2/CO ratio of 3.1, which forms methane at 31 bar of pressure and temperature between 300–600°C, whereas the chemical reactions are:

$$\begin{array}{c} \mathrm{CO} + 3\mathrm{H}_2 \leftrightarrow \mathrm{CH}_4 + \mathrm{H}_2\mathrm{O} \\ \mathrm{CO}_2 + 4\mathrm{H}_2 \leftrightarrow \mathrm{CH}_4 + 2\mathrm{H}_2\mathrm{O} \end{array}$$

1.4.2.2.3 Bioethanol Production

Biomass is majorly used for the power steam generation. Wherein, SO₂ feed with the steam explosion around 205 °C for a short time of 5 min. Then, steam is recovered by forming the product. This product flash and vapor formed during flash is condensed to recover furfural. After this steam recovery, the liquid is recovering with the help of additional water use for the simultaneous fermentation. Saccharification to form the simplest sugar is done at low temperature up to 40 °C. Therefore, saccharification forms the simplest sugar of hexoses and pentoses, which further are converted into bioethanol via the fermentation process:

$$C_6H_{10}O_5 + H_2O \rightarrow C_6H_2O_6$$

$$C_{5}H_{8}O_{4} + H_{2}O \rightarrow C_{5}H_{10}O$$

$$C_{6}H_{12}O_{6} \rightarrow 2CO_{2} + 2C_{2}H_{5}OH$$

$$2C_{5}H_{10}O_{5} \rightarrow 3CO_{2} + 3C_{2}H_{5}OH$$

Combine production of the methanol and ethanol is achieved by using the following strategy. First, hydrolyzed biomass of cellulose and hemicellulose is used for ethanol production. The remaining part such as lignin with the remaining residue of the hydrolyzed biomass is used for the methanol production (Melin et al. 2015). In other strategies, biomass is hydrolyzed and coupled with hydrogenation with the use of a catalyst such as lignin with the remaining residue of the hydrolyzed biomass used for the methanol production (Melin et al. 2015). In other strategies, biomass used for the methanol production (Melin et al. 2015). In other strategies, biomass is hydrolyzed and coupled with hydrogenation with the use of a catalyst such as Ru at 160 °C and 50 bar pressure. The major advantage of this method is the stability of sugar which produces alcohols:

$$C_6H_{12}O_6 + H_2 \rightarrow C_6H_{14}O_6$$

1.4.3 Liquefaction

Biomass is subjected to the presence of alkali, glycerine, propanol, butanol, or direct liquefaction (Demirbas 2004). This technique product is water insoluble, with high viscosity, which requires solvents, carbon monoxide (reducing gases) or hydrogen, or catalyst to be present in biomass.

In thermochemical conversion, lignocellulosic biomass is directly converted into the liquid as heavy liquid oils.

1.4.4 Pyrolysis of Biomass

The basic thermochemical process converts biomass into hydrocarbon-rich gas mixture in the absence/controlled oxygen conditions (Demirbaş 1998). In this process, biomass is converted into the vital product known as bio-oil, and other liquid products like acetic acid, acetone, and methanol. Solid products like charcoal, and gaseous products like non-condensable gases are also formed on heating 477 °C in the presence of air. However, around 70% of bio-oil are produced on a mass basis, whereas this process economically viable does not require pre-treatment of the biomass (Alonso et al. 2010). Therefore, the corrosive nature of the bio-oil towards equipment, and poor thermal stability has to be overcome by upgrading oxygen content, whereas, hydrogenation leads alkali removal and oil require catalytic cracking (Demirbaş 1998). Pyrolysis of lignocellulosic biomass such as wood is a zonal process, in which thermal degradations of hemicellulose, cellulose, and lignin occur (Chum 1991). Moreover, pyrolysis comprises of five stages (Demirbaş 2000):

First stage: Removal of moisture with some volatile loss Second stage: Degradation of hemicellulose and emissions of CO and CO_2 Third stage: Exothermic reaction and emission of CH_4 , H_2 , C_2H_6 Fourth stage: External energy supply Fifth stage: Completion of the process

1.4.4.1 Fast Pyrolysis

This process is performed in the higher temperature range of 577-977 °C with fast heating rate achieved in short residence time of 0.5–10 s. In fast pyrolysis, biomass is converted into vapors, aerosols, and char. With further cooling and condensation of vapor, the aerosol's dark color liquid is formed, whereas in terms of yield, 60–75% bio-oil, 15–25% char, and 10–15% non-condensed gases are formed (Shafizadeh 1982).

1.4.4.2 Flash Pyrolysis

Flash pyrolysis occurs in higher temperature than fast pyrolysis of 777–1027 °C with the fast heating rate of more than 723 °C/s (<0.5 s short retention time) and very fine particle size of <0.2 mm. Bio-oil is formed from flash pyrolysis, is mixed with char, and makes a mixture of bioslurry. Further, this bioslurry is fed to the gasifier at 26 bar pressure with 654–954 °C to convert into syngas with the conversion efficiency of 70%. This bio-oil formed from flash pyrolysis is used for the engines and turbines for the power generation (Demirbas 2004).

Biomass-derived syngas is used for different biofuel productions contaminated with various particulates, tars, nitrogen, sulfur contaminants, and halides with trace elements. These contaminants are removed before entering into the catalytic reactor system (Sikarwar et al. 2017).

Production of the 2G biofuel not an issue for the feed to food compared with 1G biofuel, but fiber crops used as raw material, compete with the food crops. Moreover, the pesticides and fertilizer use also the primary concern for the production of the second-generation biofuel.

1.5 Third-Generation Biofuels

In the early stage of development, algae are used for the generation of the 3G biofuel. Algae include the microalgae, macroalgae from seaweed, and cyanobacteria, also known as blue-green algae (BGA). More than 800,000 algal species are exist on the earth, and algae can rapidly grow and harvested in a month, and minimal nutrient is required for their growth; even economic production in wastewater is an added advantage (Bowyer et al. 2018). The potential of oil production from algae is 100 times greater than in area basis than the soya bean and canola oils (Hu et al. 2008). For the better growth of the algae, the optimal temperature range is between 20 and 30 °C and slow growth observed is lower than 16 °C, whereas above 35 °C is deleterious for the algae growth (Menetrez 2012). Microalgae produce lipid

Method	Adv Wang antages	Disadvantages
Oil expeller	Easy to operate No solvent required	Large biomass size required Slow operation Higher power utilization and scale up problem
Ultrasonication	Reduced extraction time Low solvent required Release of cell content higher in solvent	Filtration and centrifugation add extra cost for removal of solid residue Efficiency poor, target compound, volatile in nature
Microwave	Economical Environmental friendly Reduced solvent use, time Improved extraction yield	Extraction time long Large volume solvent required, which is toxic and highly flammable Solvent recovery is highly energy intensive High operational and infrastructure cost
Solvent extraction	Simple and economical viable High efficiency, better for small scale	
Supercritical CO ₂	Reduced time Low toxicity of solvents Flexibility to change pressure and temperature and improves selectivity Favorable for mass transfer Solvent-free extract	
Wet extraction	Energy saving for drying biomass Reduced solvent uses	Low-quality lipid extracted

Table 1.2 Various lipid extraction methods with their advantages and disadvantages Halim et al. (2011), Harun et al. (2010), Wang et al. (2015), Kanda et al. (2013) and Mubarak et al. (2015)

through photosynthesis, and lipid content reaches around 20–80% of dry weight depending on the species. However, changes in the nutritional requirement can change the product yield of the lipid, protein, and carbohydrate (Shen et al. 2009). After that, lipid extracted by different extraction processes is used for the production of the biodiesel, and the leftovers, carbohydrate and starches (residual), are further processed and can be used for the production of ethanol (Shen et al. 2009; Coppola et al. 2009). Lipid extraction process includes mechanical and chemical extractions (Halim et al. 2012). Table 1.2 summarizes the various method of extraction.

1.5.1 Open Pond

A natural water reservoirs like lake and lagoon and artificial reservoirs system like ponds, shallow ponds, and tanks are the open pond system. Advantage of the open system is that it is easy to construct and operate, but poor utilization of the light, evaporation loss, loss of the CO_2 to the environment, and chances of contamination

are the significant key challenges of the open pond system. This system can be suitably installed with the wastewater treatment plant (Beal et al. 2012). But the major drawbacks of this system lacks control of various parameters.

1.5.2 Photobioreactor (PBRs)

The system is closed, therefore it provides a better internally maintained control system for the growth of pure algal culture as they maintain various parameters of inadequate amount such as CO₂, water, temperature, light intensity, mixing density, and pH of the system (Lee 1999; Mussgnug et al. 2007). In PBRs, algae are cultured in glass or plastic tube, where they are grown in light of the particular wavelength which supports the optimal growth of the algae. Approximately, 7.6–10 cm light penetrates inside the PBRs due to the turbidity of the algal growth into PBRs (Lee 1999; Mussgnug et al. 2007; Müller et al. 1993; Rapala and Sivonen 1998). For the easy operations of the PBRs, various conditions optimized like volume size to the area for the optimum light penetration, spatial distribution of light, CO₂ transfer rate, and monitoring of the growth inside the reactor (Menetrez 2012). Their installation and operational costs are higher, but product yield in terms of oil is obtained around 40–55% (Hu et al. 2008; Chisti 2007). Due to the less expensive operation of the optimum, it can be suitably hybrid with the closed type of PBRs for optimizing cost and product yield (Menetrez 2012).

Algae are an excellent source to produce various biofuels such as biodiesel, ethanol, and petroleum. From various reports, productivity from the algae is 602 times higher than various crops like corn and switchgrasses (Menetrez 2012). The significant advantage of algae is higher productivity, and small land for the cultivation is required. Transesterification of oil recovered from algae produces biodiesel chemically and biologically. Moreover, biological production utilizes a lipase enzyme which converts lipids into biodiesel at room temperature. This operation at room temperature cuts the energy requirement for biodiesel production (Fig. 1.1).

1.6 Fourth-Generation Biofuel

The use of the new synthetic biological tools has emerged as the fourth-generation biofuel. In this generation, the breakthrough has made promises to photobiological biofuels and electro biofuels (Aro 2016). Synthetic biology of the algae and cyanobacteria involves various designs of the new promising strains by using recombinant



Fig. 1.1 Biofuel from algae

Generation	Source	Current conditions	Problems
First	Food crop sources Starch and sugar Oil crops	Commercially available. Technically developed	Competition with food supply and land uses Benefit of greenhouse gases is dependent on the feedstock
Second	Lignocellulosic biomass Agro-waste and energy crop	Advanced status of development and demonstration step	Higher cost, selection cheap, and sustainable raw material Additional cost of pre-treatment Enzyme cost is higher
Third	Algae	Fast-growing, cultivation land not required, Various potential algae strains are available, "under research and development"	High cost of downstream processing
Fourth (extension of third generation)	Algae and cyanobacteria	Primary stage of genetic engineering and new recombinant strains development, more promising than 3 G	Under research.

Table 1.3 Comparisons of different biofuel generations and its challenges

deoxyribonucleic acid (DNA) technologies. This technology uses the synthesis of the new strain by using new devices and the integration of the new biological parts or redesigning of the biological system for the biofuel generation.

The fourth-generation biofuels are inexhaustible, cheap, and readily available resources compared in (Table 1.3) with other generation of biofuel. During photosynthesis process, water molecule splits into hydrogen and oxygen by the action of solar energy with the interaction of photosystems I and II which is the key for the biofuel production at a large scale (Inganäs and Sundström 2016). Moreover, artificial photolysis is emerging as the new door for the biofuel generation (Inganäs and Sundström 2016). Even CO₂ water can also use as an alternative strategy for biofuel production, which utilizes carbon-based solar fuel.

1.6.1 Direct Process for Solar Fuel

This technology utilizes the combination of sensitizers and catalyst for performing individual reactions and generating approximately 30% of the solar fuel from water, but no complete device to date is made for direct fuel productions (Inganäs and Sundström 2016). In this process, oxidation of the water and reduction of the substrate are principle mechanisms. Further, proton from substrate utilized for the hydrogen fuel generation. The catalyst used is generally made up of abundant

metals like cobalt, iron, and manganese, etc. In another way, an artificial photosynthesis follows the molecular mechanism; it involves the enzymes which are catalytically active used as biocatalyst like hydrogenase enzyme, which is used for hydrogen production by dark fermentation (Tamagnini et al. 2007). In direct process, with the help of the nanotechnologies, hydrogen production follows the non-molecular mechanism light-driven catalysis on the surface. This surface area can be metal, semiconductors, or fabrication of nanostructured carbon-based material. Limitations of the non-molecular mechanism are expensive, and rare metals are used as the catalyst. Involvement of molecular and non-molecular mechanism involved makes the process complex thus less understood. Limitations of the non-molecular mechanism are the use of rare metals as the catalyst which makes it expensive.

1.7 Microbial Conversion

Production of the biofuel is still a challenge because the production cost is more than the raw material. In bioprocessing, microbial conversion step converts lignocellulosic biomass into biofuel by microbial action (no action of enzyme). This method has gained more interest from researchers over the first-generation biofuel production in terms of the cost (Lynd et al. 2005). The new trend of bioprocess states that the potential of ethanol production through microbial route or its combinations mainly utilizes the cellulose machinery with other components (hemicellulose and lignin) of the pre-treated biomass with the high yield of the ethanol. Various aerobic fungi can degrade pre-treated feedstocks directly into the CO₂, and only a minor amount of ethanol is produced. However, the electrons yielded are transferred to the respiratory chain for oxidative phosphorylation, and the metabolic pathway does not utilize the substrate level phosphorylation for ethanol production. The rule of thumb of alcohol production from sugar is that 2 moles of alcohol is produced from each mole of hexose sugar. Yeast like Saccharomyces cerevisiae uses the glycolytic pathway to form alcohol. All the industrial processes are operated at higher pH and temperature range, which suitably use consortia of potent fungal strain like Sarocladium strictum with thermophiles like Halomonas for the improved degradation of wheat straw (Cortes-Tolalpa et al. 2018) (Table 1.4).

1.8 Enzymatic Conversion to Biofuel

Biofuel production from lignocellulosic biomass has given the new option to fulfill global energy demand but has a greater challenge of global warming. Production of biological-derived energy known as "bioenergy" is a major concern with the biocatalysis (utilizes the various enzymes to catalyze biomass conversion) (Rubin 2008). The complex structure of the lignocellulosic biomass first gets hydrolyzed

		Biofuel yield	
Microorganism	Biofuel	(g/L)	References
Clostridium acetobutylicum	Butanol	3	Lütke-Eversloh and Bahl (2011)
Clostridium thermocellum	Isobutanol	5.4	Lin et al. (2015)
Escherichia coli	Butanol	30	Shen et al. (2011)
Escherichia coli	Ethanol	25	Romero-García et al. (2016)
Saccharomyces cerevisiae	Fatty acids	0.38	Yu et al. (2016)
Saccharomyces cerevisiae	Isoprenoid based biofuel	40	Westfall et al. (2012)
Pseudomonas putida	Butanol	0.05	Nielsen et al. (2009)
Cryptococcus	Lipids	7.8	Deeba et al. (2016)
Zymomonas mobilis	2,3 butandiol	10	Yang et al. (2016)
Zymomonas mobilis	Ethanol	-	Kremer et al. (2015)
Caldicellulosiruptor bescii	Ethanol	0.7	Chung et al. (2014)
Trichoderma reesei	Ethanol	10	Huang et al. (2014)
Yarrowia lipolytica	Fatty acids	55	Beopoulos et al. (2009)
Synechococcus sp.	Limonene	0.04	Davies et al. (2014)
Synechococcus elongatus	1,3 propanediol	0.28	Hirokawa et al. (2016)

Table 1.4 Microorganism for producing biofuels (Kumar and Kumar 2017)

into its simplest fractions with various hydrolyzing enzyme cocktails. However, the use of enzyme cocktail proved the maximum conversion into industrial product formation from lignocellulosic biomass conversion. Enzyme consortium is another way used to deconstruct recalcitrant substrate into a useful product (Lopes et al. 2018). This enzyme consortium conversion requires detailed studies on various enzymes and their action to work synergistically. To date, Sprizyme[®] from Novozyme, a commercial enzymatic cocktail is available in three versions (in the market) for the complete conversion of sugar. Moreover, Cellic®CTec3 (2017) from Novozyme is using cellulose conversion into ethanol. Enzyme cocktails enable the enzyme to hydrolyze cell wall structure due to their specific action. Moreover, the product of one enzyme acts as the substrate of another enzyme and also gives an option to the individual enzyme to act on an available substrate. During enzymesubstrate reaction, enzyme inhibitors are also produced, which can inhibit the hydrolysis process. Therefore, a complete enzyme mechanism can be studied to increase lignocellulosic biomass conversion. Wherein, each substrate in cellulosic biomass has its characteristic and feature which require the distinct condition to convert it into its simplest form.