Chapter 14
Nanotechnology Systems for Biofuels Production

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1 Introduction

Petroleum-based fossil fuels account for more than 90% of global primary energy consumption and are a major energy resource (Li et al. 2019). World energy consumption tends to increase over the years, energy demand is expected to be approximately 30 TW by 2050 and 46 TW by 2100 (Sadaf et al. 2018). One of the main reasons for environmental pollution is the generation of energy from fossil fuels, and the combustion of fossil fuels causes the emission of greenhouse gases, such as CO_2. In the last 150 years, the concentration of CO_2 in the environment has increased to 370 ppm (Li et al. 2019).

The development of renewable energy emerges as a solution for reducing greenhouse gas emissions (Pradhan et al. 2018). In this context, the main sources of renewable energy today are those from natural phenomena such as solar, wind, and biomass-derived bioenergy (Li et al. 2019). For the more, developments in technologies to convert biomass into sustainable fuel are being widely studied and implemented to reduce the damage caused by fossil fuels (Li et al. 2019).

In this regard, biofuels are classified into two groups, namely primary and secondary biofuels (Sekoai et al. 2019). Thus, first-generation biofuels are produced from sucrose, animal fats, plant oils, and crops such as corn, wheat, among others

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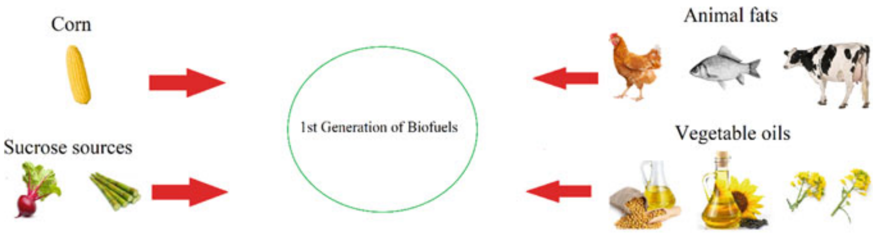

Fig. 1 Sources of the first generation of biofuels

(Fig. 1). Meanwhile, second-generation biofuels use non-food raw material as wood waste, agricultural waste, lignocellulosic biomass, among others (Leong et al. 2018; Zhang et al. 2018). In addition to these two types of biofuel generation, there is a third-generation employing processing microalgae to produce biofuel (Leong et al. 2018), and a fourth generation focused on modifying the metabolism of these organisms, lowering production costs and increasing production (Moravvej et al. 2019). Some examples of biofuels generated from biomass are biodiesel, bioethanol, biohydrogen, and biogas (Sekoai et al. 2019).

Biodiesel is a clean, biodegradable, non-toxic, and low pollutant biofuel. In addition, it can be obtained from various renewable sources (Nisar et al. 2017; Tian et al. 2017). Biodiesel has environmental technical and strategic advantages: It can reduce most exhaust emissions, it is biodegradable and has inherent lubricity (Rai et al. 2016). In recent years, the biodiesel industry has grown considerably, and globally the biodiesel industry will expand its production with an annual expansion of 7.3% to $54.8 billion in 2025 (Sekoai et al. 2019).

Other types of biofuels, such as bioethanol and biogas, are alternative candidates that can be used in the renewable energy market (Cesaro and Belgiorno 2015). Bioethanol is also considered a clean, renewable, and non-toxic fuel and can be obtained from various biomass raw materials such as sugar cane, cornstarch, and algae (Sirajunnisa and Surendhiran 2016). In 2017, global ethanol production was estimated at approximately 100 billion liters and is expected to double in the next decade (Aditiya et al. 2016). The technique for production of second-generation bioethanol is not yet stable compared to first-generation bioethanol. However, bioethanol obtained from non-food raw materials does not present socioeconomic questions because they are considered waste (Sekoai and Daramola 2017). On the other hand, biogas is a biofuel produced from anaerobic digestions of microorganisms (Bundhoo and Mohee 2016). It is mostly composed of 50–75% methane (CH_4), 25–45% carbon dioxide (CO_2), and other components in small quantities (Andritz Group 2013).

Biohydrogen is another biofuel, which has a high energy content that is considered three times higher than fossil fuels, uses various raw materials such as organic effluents and can be generated under operating conditions of pressure and ambient temperature, enabling the large-scale production (Sekoai et al. 2019). Biohydrogen production is increased by nanomaterials since these materials can potentiate the activity of microorganisms that possess physicochemical properties (Pugazhendhi et al. 2019).

Fortunately, the application of nanotechnology in recent years has intensified in various segments, such as the food, agricultural, cosmetic, pharmaceutical, and electronic industries, due to the ability to use various nanoscale materials, such as nanoparticles, in a range from 1 to 100 nm (Sekoai et al. 2018; Tyagi et al. 2018). This use of nanomaterials in various sectors is attributed to the properties of nanoparticles, which include high reactivity and structure/morphology (Sekoai et al. 2018; Tyagi et al. 2018). Nanoparticles have other characteristics such as high degree of crystallinity, catalytic activity, stability, durability, efficient storage, high recovery potential, reuse, and recycling make these materials exceptional candidates for biofuel systems (Nizami and Rehan 2018).

Due to these characteristics, many nanomaterials can be applied in biofuel production. For example, nanoparticles can be used mainly as catalysts because they perform an important job in electron transfer, improve anaerobic agent activities, and reduce inhibitory substances (de Vasconcellos et al. 2018). However, some nanosystems suffer from some technical issues and economic viability, but continuous researches are being developed and some of the future trends will be pointed at the end of this chapter, with recommendations for further studies.

2 Biofuels Production with Nanotechnology Systems

Application of nanotechnology in biofuel production, especially in biocatalysts, is being reported in several studies to increase process efficiency (Biswas 2019). The small size of nanoparticles and different forms in nature (metallic, semiconductor, or polymeric) imply in a big versatility and are an advantage in biocatalysis, due to higher surface area, which increases catalytic activity, better stability, and reusability (Ahmadi et al. 2019). They also present a high degree of crystallinity and could be synthesized using a top-down or bottom-up approach (Sekoai et al. 2019). This chapter will broach these and many other interesting characteristics and applications of nanotechnology in biofuels production, showing which barriers are being studied to be overcome in the future.

2.1 Biodiesel

Biodiesel is a very promising fuel as it is produced especially from renewable energy sources or biomass (Fig. 2), is biodegradable and contributes to the reduction of combustion emissions (Rosha et al. 2019; Sekoai et al. 2019). Biodiesel accounts for 82% of total biofuel production, making it the main alternative to diesel (Bozbas

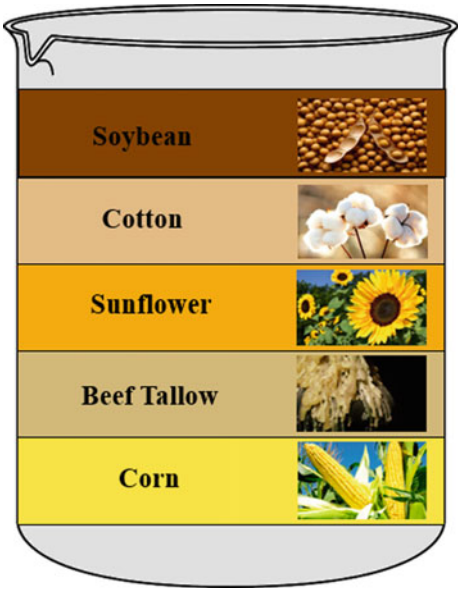

Fig. 2 Biomass for biodiesel production

2008; Sekoai et al. 2019), and vegetable oils are the main source of biodiesel production. However, due to the great competition with their use in cooking and food production, their use is becoming increasingly expensive (Muniru et al. 2018; Zhang et al. 2013). Animal fats, soaps, greases, inedible oils, used frying oils are examples of low-cost biodiesel feedstock, however, their quantities available are not sufficient to meet the current requirements of this biofuel (Fingerman et al. 2018). Thus, microalgae, an inedible raw material that does not interfere with the global food economy, is gaining great importance today (Banerjee et al. 2019). Given the need to reduce environmental impacts and ensure the same level of performance of existing fuels, microalgae can contribute to a reduction in the need for land due to their energy yield, high growth rate in a short time, high production of biomass, and the lack of agricultural land (Lee et al. 2010). Microalgae also stand out for having higher photosynthetic efficiency when compared to plants (Banerjee et al. 2019).

Many studies have also been performed on lipase immobilization by nanomaterials (Bezerra et al. 2017; Costa et al. 2016; Galvão et al. 2018; Monteiro et al. 2019; Rios et al. 2019; Souza et al. 2017). Lipase from *Candida rugosa* was covalently linked to a magnetic microsphere on Fe_3O_4/Poly (styrene-methacrylic acid) for biodiesel production from soybean oil (Xie and Wang 2014). In this nanosystem, the developed immobilized enzyme converted methanol and soybean oil to 86% FAME and the biocatalyst remained active for 4 cycles without significant loss of activity (Xie and Wang 2014). In another investigation with a nanosystem (Kalantari et al. 2013), lipase from *Pseudomonas cepacian* was bound to the nanocomposite particles of non-porous silica-coated magnetite agglomerates covalently amino functionalized by glutaraldehyde as a coupling agent. The authors tested the FAME conversion with the free enzyme, where the conversion was only 34% and with the immobilized enzyme, obtaining 54% conversion, conserved after 5 times reused (Kalantari et al. 2013). In nanosystem presented by Tran et al. (2012) (Tran et al. 2012), the lipase of *Burkholderia* sp. was immobilized on a ferric silica nanocomposite for biodiesel production. They coated the Fe_3O_4 core with silica shell to synthesize core-shell nanoparticles, where they achieved 90% FAME yield in 30 h in a batch operation, further validating the new support and good selection of the immobilization method and immobilized lipase was used for 10 cycles without significant loss of activity.

In another study on nanosystem, Wang et al. (2011) designed a four packed bed reactors (PBR) system for repeated and highly efficient lipase use. Both the conversion rate and stability obtained using the four accumulated bed reactor system were much higher than that obtained using the single PBR, and the biodiesel conversion was maintained at a high rate of over 88% for 8 days (Wang et al., 2011). According to the authors due to the longer residence time of the reaction mixture in the reactor and the reduction in inhibition of lipase-nanoparticle biocomposite by-products, it was possible to obtain a high conversion rate and considerable stability in the four-bed reactor, which further contributes to reducing the cost of biodiesel production. Thus, the advantages of using an immobilized nanobiocatalytic system could outweigh the costs of the immobilization step (Wang et al. 2011). These reactor scale studies highlight the possibility of designing and operating even industrial scale enzyme systems for biodiesel production (Verma et al. 2013).

Therefore, considering the above, it is important to emphasize that the interdisciplinary combination of biotechnology and nanotechnology represents a very promising opportunity, increasing the amount and efficiency of biodiesel production. The possibility of reusing stable and efficient nanobiocatalytic systems could greatly improve the economic viability of biodiesel.

2.2 Bio-Jet Fuels

Air transport is necessary for world tourism and trade. The International Air Transport Association (IATA) reported that about 4.0 billion passengers and 64 million tons of cargo were transported by air in 2018, generating 65 billion jobs and underpinning $2.7 trillion (3.6%) of the world's gross domestic product (IATA 2019). Indeed, the global aviation industry is projected to expand, creating 90 million jobs and nearly $6 trillion in annual economic activities by 2034; besides, the number of air passengers will double in the next 20 years (Yang et al. 2019).

The growth in air traffic requires a big consumption of jet fuels; in fact, according to US Energy Information Administration (EIA), excluding electricity, jet fuel consumption is projected to grow more than any other transportation fuel, rising 35% from 2018 to 2050 (EIA 2019), as well as the average price of jet fuel is expected to grow at a 2.7% annual rate from 2016 to 2050 (EIA 2019). Jet fuel costs account for about 24% of all the airline's operating costs in 2018, and it is mainly related to oil prices (IATA 2019). Furthermore, the large consumption of jet fuel increases greenhouse gas emissions. As it was reported by Air Transport Action Group, worldwide, flights produced 895 million tons of CO_2 in 2018; thus, the global aviation industry produces more than 2% of all human-induced CO_2 emissions and 12% of the transport sector (ATAG 2019). As a result, the aviation industry aims to achieve a 50% reduction in CO_2 emission by 2050 as compared to 2005s level (Wei et al. 2019). Therefore, to ensure sustainable development of the aviation industry, to rely only on improving fuel efficiency is not enough, it is crucial to develop an alternative renewable and environmental innocuous fuel to meet the growing demand and reduce the dependency of fossil fuel (Wei et al. 2019).

Bio-based jet fuels or, for short, bio-jet fuel (also known as synthetic paraffinic kerosene) consists of renewable hydrocarbons which properties are almost identical or, in some cases, superior to those of fossil jet fuel (Gutiérrez-Antonio et al. 2017). The properties of bio-jet fuels are related to their chemical composition, which mainly defines the performance characteristics (low-temperature fluidity, thermal oxidation stability, combustion property, fuel compatibility with current aviation system, fuel volatility, and fuel metering and aircraft range) of bio-jet fuels in turbine aero-engines (Braun-Unkhoff and Riedel 2015).

For bio-jet fuel production, as for other biofuels and bioenergy field in general, nanotechnology has different applications, such as the modifications of feedstocks, development of more efficient catalysts, among others (Andrade et al. 2011).

In this sense, depending on the characteristic of the triglyceride's feedstocks, it is necessary to remove oxygenated compounds and obtain n-alkanes, which ensures the energy density of the fuel, by hydrodeoxygenation (Itthibenchapong et al. 2017). For upgrading bio-oils to produce bio-jet fuels by removal of oxygen using hydrogen, noble metal catalysts, which are environmentally friendly nanoparticles doped on porous or acid/base solid supports, such as Pt, Pd, Rh, and Ru have been used (Yang et al. 2019).

As an example, Gutierrez et al. (2009) studied the upgrading of a bio-oil by hydrodeoxygenation using zirconia-supported mono- and bimetallic noble metal (Rh, Pd, Pt) catalysts, which were active and selective in the hydrodeoxygenation guaiacol at 300 °C (Gutierrez et al. 2009). In addition, silica nanoparticles, an environmentally friendly catalyst support, have been widely used in the hydrodeoxygenation reactions. For instance, Duan et al. (2017) reported an efficient hydrodeoxygenation of water-insoluble bio-oil to alkanes by using a catalyst that combines highly dispersed palladium and ultrafine molybdenum phosphate nanoparticles on silica (Duan et al. 2017).

Besides bio-oils, bio-jet fuels can be produced from different kind of biomass feedstocks, like hemicellulose, cellulose, and lignin. Lignin is the only large-scale biomass source with an aromatic functionality; nevertheless, its unique aromatic structure is quite stable. Indeed, the transformation of lignin into C8–C15 cycloparaffins and aromatics provides a pathway for the development of aromatic components in bio-jet fuels using lignin (Yang et al. 2018). As an example, Bi et al. (2015) reported the transformation of lignin into jet and diesel fuel, involving directional production of C8–C15 aromatics with HZSM-5 nanocomposite and C8–C15 cycloparaffins by the hydrogenation of aromatics over palladium on carbon nanocatalyst (Bi et al. 2015).

By the above, bio-jet fuel has been emerging as an alternative and complementary to jet fuels, once it mitigates CO_2 emissions and helps to meet the increasing demand of fuels of aviation industry. In this regard, nanosystems have been mainly used to modify bio-jet fuels feedstock in order to upgrade the properties of the biofuel.

2.3 Biohydrogen

Considered as an efficient and promising "energy-carrier," hydrogen has gained global attention focus because it is a fuel with high energy content and carbon-free, therefore the only final combustion by-product is water, which makes it favorable for reduction in GHG emissions (Chezeau et al. 2019). It can be applied either in combustion engines or to generate electricity by using fuel cell technologies, since its similarity to electricity is greater than fossil fuels in terms of energy systems (Chandrasekhar et al. 2015; Ghimire et al. 2015). The major hindrance of its use is related to the production for commercial applications, which is performed mainly by using fossil fuels in a thermo-chemical process, and the storage (Al-Mohammedawi et al. 2019; Ghimire et al. 2015; Ren et al. 2019). Hydrogen biological generation is an alternative pathway that allows an eco-friendly and attractive renewable production (Fakhimi et al. 2019; García-Depraect et al. 2019).

Biohydrogen is a second-generation biofuel that can be obtained by biological pathways which requires a lower energy supply and is less harmful in terms of CO_2 emission. Its production can be performed by some process using the biomass of several categories (Dinesh Kumar et al. 2019; Ghimire et al. 2015; Ren et al. 2019). The use of lignocellulosic substrates presents several advantages for they are abundant and renewable materials cultivated in the no arable soils and thus it does not cause any competition for food production (Kumar et al. 2015; Sambusiti et al. 2015). The bioprocess could be even more attractive when the cost is reduced with the utilization of low-value material, as agro-industrial residues and organic municipal waste and wastewater (Ghimire et al. 2015).

Biohydrogen can be generally obtained by some process, as dark fermentation, photo-fermentation, water biophotolysis, and indirect biophotolysis which have their own positive and negative points (Seelert et al. 2015). Due to the possibility to use cheap organic substances as substrate, as food and industrial waste, photo and dark fermentation are the most explored and reported techniques in literature (Cai et al. 2019; Wimonsong et al. 2014). Figure 3 shows mechanisms for biohydrogen production.

Photo-fermentation is a widely studied method for biohydrogen production for it allows a relatively high substrate conversion and can make use of several substrates as carbon source. Besides that, it can be performed at ambient conditions, which allows a lower energy demand (Al-Mohammedawi et al. 2019; Mirza et al. 2019). In

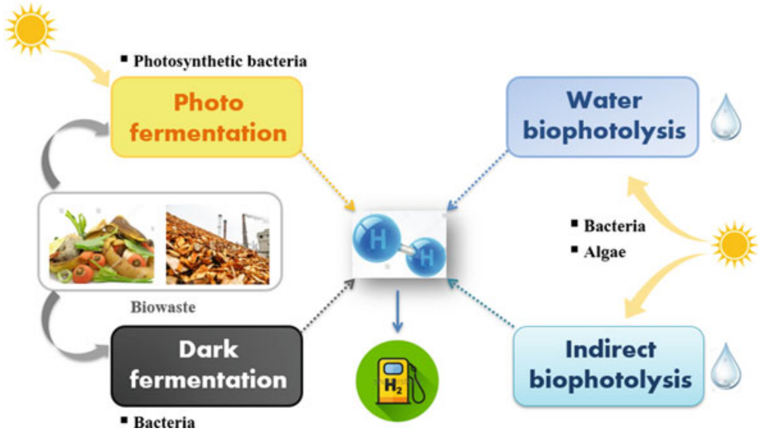

Fig. 3 Mechanisms and sources for biohydrogen production

this process, the production is regulated by two key enzymes, the membrane-bound hydrogenases and the nitrogenases (Dolly et al. 2015; Show et al. 2019)

In an anaerobic dark fermentation, the biohydrogen production is achieved by a redox reaction catalyzed by hydrogenase (Chezeau et al. 2019). The dark fermentation presents some good environmental and economic benefits for the biohydrogen production that makes it a viable way to that fuel obtaining (Malik et al. 2014; Palomo-Briones et al. 2019). The process can be operated under mild conditions of temperature and pressure, it presents a better resistance against contamination, it accepts different substrate species and generally presents a greater efficiency when compared to the other biological process for that production (García-Depraect et al. 2019; Malik et al. 2014).

However, several barriers remain a challenge to be overcome for successful biohydrogen production and commercialization, as the low substrate conversion and production rates, which decrease the processes efficiency (Chandrasekhar et al. 2015; Fakhimi et al. 2019; Show et al. 2019). In this context, nanotechnology systems have attracted attention because it may enhance several process of hydrogen production, both by dark and by photo fermentation (Malik et al. 2014; Wimonsong et al. 2014). Because of the high specific surface of nanoparticles, its simple addition influences directly on the reaction rate. Besides that, these particles might induce the interactions inside the microorganism improving the electrons transference which benefits the production kinetics (Jafari and Zilouei 2016).

In this point, organic and nonorganic nanoparticles have been studied as a carrier for cell immobilization aiming the improvement in biohydrogen production (Kumar et al. 2016). Several studies involving metallic nanoparticles have been realized because of their ability to react quickly with the electron donors, which enhance the reaction kinetic (Dolly et al. 2015). The addition of metal nanoparticles, like hematite, gold, nickel, and silver, to dark fermentation process has showed to improve the process efficiency, by increasing the hydrogen yield and microbes bioactivity, for example, and also by reducing the lag phase for the biohydrogen production (Hsieh et al. 2016; Pugazhendhi et al. 2018; Zaidi et al. 2019). Some of those studies attribute these results to stimulation of hydrogenase enzyme activity increase caused by the metal addition (Hsieh et al. 2016).

2.3.1 Iron and Nickel Nanoparticles as Nanosystems for Biohydrogen Production

Bulk and nanoparticles form of iron sulfate were studied as a supplement on photo-biohydrogen production and both forms achieved the highest cumulative hydrogen production at the same concentration in mg/L (Dolly et al. 2015). However, this maximum result obtained for the nano form was 1.2 fold higher than the bulk form, which testifies the enhancement caused by the utilization of nanosized particles (Dolly et al. 2015).

The addition of Fe and Ni to the fermentation has shown a direct influence on the activity of *hydrogenase*, the key enzyme in the hydrogen production by dark fermentation bioprocess, because these metals are present in its active site and many studies have demonstrated an improvement of results by adding iron and nickel salts (Elreedy et al. 2017; Gadhe et al. 2015). Besides that, the Fe and Ni nanoparticles utilization may enhance also the activity of the *ferredoxin oxidoreductase*—which can indirectly generate hydrogen in dark fermentation—for they can ameliorate the electron transfer rate due to surface increase, which makes this utilization attractive (Gadhe et al. 2015). In process of anaerobic digestion of an industrial wastewater containing mono-ethylene glycol (used in petrochemical productions), nanoparticles composed of Ni and graphene were added as a supplement to the process and there was an increase of more than 100% in the biohydrogen production (Elreedy et al. 2017).

Iron (II) oxide nanoparticles were added to an anaerobic process, conducted at a thermophilic condition, aiming the biohydrogen improvement using glucose as the substrate. The results showed a rate augmentation with no relevant modifications in the metabolic pathway and the authors considered this addition as a vital factor to the required enhancement achievement. Another nanosystem using molasses-based distillery wastewater to the biological production of hydrogen, iron oxide nanoparticles were added in the process and showed to make the hydrogen production increase (Malik et al. 2014). Ferrihydrite nanoparticles present an enhancement effect in a dark fermentation for hydrogen production with an increase of more than 60% in the production (Zhang et al. 2019a, b). The Fe supplementation has attracted attention also in photo-fermentation because it is part of the structure of the key enzymes—hydrogenase and nitrogenase—and it can work in the electron transfer chain (Dolly et al. 2015).

2.3.2 Hematite Nanoparticles for Biohydrogen Production

Hematite (Fe_2O_3) and nickel oxide (NiO) nanoparticles added to a process of biohydrogen generation from dairy wastewater presented an enhancement in the production by shortening the lag phase and increasing the microbial activity (Gadhe et al. 2015). Both individual and conjoint addition presented a positive effect, however best results were obtained when the conjoint addition was performed. A significant positive effect was obtained by the addition of TiO_2 and magnetic hematite (Fe) nanoparticle in a dark fermentation by *Clostridium pasteurianum* for the biohydrogen production (Hsieh et al. 2016). By performing gene expression measurements, the authors noted that the nanoparticles addition effect was not at the hydrogenase gene level, which was speculated in the previous literature, however, it was attributed to the enhanced electron transference.

2.3.3 Others Nanosystems for Biohydrogen Production

Literature reports that gold can be applied as the catalyst in a great variety of reactions. Usually, the activity of the gold particles is related to the size and other factors, as the support nature and the preparation method (Sekoai et al. 2019). Different metallic nanocatalysts (Au/Fe-Zn-Mg–Al-O) supported onto hydrotalcite were applied in dark fermentation with sucrose as the substrate for biohydrogen production (Wimonsong et al. 2014). The greatest achieved increase in the hydrogen yield (2 times higher than the control) was attributed to the Au, that can act as electron sinks, supported on a surface having Zn, which can work as the active site for the key enzyme hydrogenase.

Nano-spray dried particles of ferric citrate, ferrous sulfate, nickel chloride, and nickel acetate were added to biohydrogen process production by dark fermentation of crude glycerol generated by biodiesel industry (Sarma et al. 2014). It was observed that only ferric citrate particles showed an enhancement of hydrogen production, with a yield value more than 2 times higher than the standard process. Nano-titanium dioxide was applied in the pre-treatment of sugarcane bagasse for using in a dark fermentation process and the results showed an improvement of fermentation efficiency (Jafari and Zilouei 2016).

In this way, it can be noticed that nanosystems from different nature have shown an enhancement effect in different process for biohydrogen generation, by increasing different production parameters.

2.4 Algae-Derived Systems for Biofuels Production

Algae-derived biofuels present great potential. However, they suffer from high production costs and other technical barriers. For example, wastewater-algal biomass could have microbial contaminants compromising the production yield (Limayem et al. 2016). Nanotechnology and nanomaterials could be used to improve production efficiency and reduce processing cost.

2.4.1 Nanotechnology Applied to Algae Biofuel Production

Algae Cultivation

To cheap production costs and increase biomass growth, nanomaterials could be applied in light emitting diodes (LEDs) for artificial illumination of algae, manipulating illumination properties (Pompa et al. 2006). Metal nanoparticles can also be coupled to localized surface plasmon resonance and this technique could avoid photo-inhibition and optimize light frequencies preferred by algal species (Torkamani et al. 2010). For insurance of an adequate provision of CO_2 and nutrients for the growing cells, the use of nanosystems is also possible: using a mix of micro- and nano-bubbles in airlift loop bioreactors can increase mass transfer efficiency, the residence time of the bubbles and reduce energy consumption of the provision process (Zimmerman et al. 2010, 2011).

As it was presented, there is a problem with microbial contaminants of wastewater-algal biomass, but nanotechnology can overcome it. A combination of natural chitosan and zinc oxide nanoparticles was applied, with positive results. The nanoparticles were proven effective inhibitory agents against microbial lytic groups, like *Pseudomonas* spp., *Micrococcus luteus*, and *Bacilus pumilus.* Another advantage of the process was that did not compromise algae cells, showing great potential for in situ interventions (Limayem et al. 2016).

Algae Harvesting

Nanosystems are also being applied to extract oil from algae without breaking their cell. The spongiosum characteristic of some mesoporous nanoparticles enables an entrapping of lipids molecules and catalysts such as oxides of strontium and calcium can also be introduced into the pore structure and transesterify their molecules in situ (Akubude et al. 2019; Zhang et al. 2013).

In this way, the application of magnetophoretic separation technology using magnetite nanoparticles can provide rapid growth and high efficiency for algal harvesting. The process needs further studies to become economically practical, but a good strategy is coating the nanoparticles with a cationic polymer, reducing the costs and enabling reuse. The use of UV radiation increases algal harvesting efficiency and can also bring potential for large-scale production of algae-derived biofuels (Ge et al. 2014). A successful similar nanosystem process was also applied, obtaining 99% of harvesting efficiency of *Chlorella* sp. KR-1 using chitosan-Fe_3O_4 nanoparticles, without any adverse effects on microalgal growth, enabling medium recycling and therefore, lowering the cost of the employment of the biofuel from this source (Abo Markeb et al. 2019; Lee et al. 2013).

Biomass Transformation and Biofuel Employment

The use of enzymes to catalyze biomass transformations suffers from high cost and low stability of the catalysts. Using nanoscale structures can eliminate these problems, improving product quality, with high specific surface area, high catalytic activity, high resistance to saponification, and good rigidity (Akubude et al. 2019). Nanostructured supports are used to immobilize enzymes, like lipases, and this was already proved to increase transesterification yields for biodiesel production from different sources of oil (algae included), when comparing with traditional supports (Tahvildari et al. 2015; Teo et al. 2016). Besides that, nanomaterials can also be used as additives for the algae biodiesel, improving brake thermal efficiency and reducing exhaust emissions (Karthikeyan and Prabhakaran 2018; Karthikeyan and Prathima 2017).

2.5 Nanosystems for Biogas Production

The development of researchers to investigate the effects of nanomaterials (1–100 nm) on the production of biogas is currently intensified by the increasing demand for green energy (Baniamerian et al. 2019). As described by Zhang et al. (2016), biogas is a gaseous mixture containing mainly methane (CH_4) and carbon dioxide (CO_2) (Zhang et al. 2016). Sekoai et al. (2019) report CH_4 percentages of 50–75%, followed by 25–45% of CO_2 (Sekoai et al. 2019). Some impurities, like hydrogen sulfide (H_2S) and ammonia (NH_3), are also presented (Kadam and Panwar 2017). Moreover, typical biogas has some nitrogen (N_2), oxygen (O_2), carbon monoxide (CO), water vapor, siloxanes, dust particles, halogenated and aromatic compounds (Abdeen et al. 2016).

In this way, the anaerobic digestion of organic matter by microorganisms and enzymes, which results in biogas production, takes place in four complex steps: hydrolysis, acidogenesis, acetogenesis, and methanogenesis (Sahota et al. 2018). As stated by Kadam and Panwar (2017), the total biochemical process depends on different experimental factors, e.g., type of organic substrates, temperature changes, pH and inoculum concentration (Kadam and Panwar 2017). Several liquids and solids substrates can be treated by anaerobic digestion, including microalgae, agricultural residues, animal manure, wastewater, waste activated sludge, food waste, yard waste and solid organic waste (Mushtaq et al. 2016; Vasco-Correa et al. 2018).

Remarkably, the decomposition of municipal solid waste (MSW) in landfills is also interesting (Lima et al. 2018; Seman et al. 2019). According to Kormi et al. (2017), landfills are among the three largest anthropogenic sources of methane emissions in the world (Kormi et al. 2017). Thus, in addition to energy benefits, proper recovery of landfill biogas reduces local greenhouse gas emissions (Villanueva-Estrada et al. 2019). Xu et al. (2019) report that the amount of landfill produced biogas can be improved by controlling moisture conditions for MSW microorganisms (Xu et al. 2019). Besides, biogas production rates are affected by the type and age of landfill waste, properties of landfill leachate and environmental conditions (Plocoste et al. 2016). Recent works indicate that nanomaterials often found in landfills, due to the disposal of diverse commercial products, can also influence the biogas generation(Xu et al. 2019; Plocoste et al. 2016).

Temizel et al. (2017) investigated the effect of nano-ZnO on biogas generation from sanitary landfills (Temizel et al. 2017). Two conventional and two bioreactor landfills, operated at mesophilic temperature (35 °C) for about 1 year, were used in the experiments. The results suggest that the presence of nano-ZnO in MSW may decrease the potential of landfill biogas production. In the reported work, conventional and bioreactor landfills produced 15% less biogas in comparison with the control reactors. The authors suggested that the release of Zn^{2+} from nano-ZnO affects adversely methanogenesis phase by reducing the activity of key enzymes (Temizel et al. 2017). The same hypothesis was raised by Mu and Chen (2011) when they studied the impact of nano-ZnO on anaerobic digestion of waste activated sludge (Mu and Chen 2011). In another work, Mu et al. (2011) concluded that nano-ZnO has an inhibition effect on anaerobic digestion of waste activated sludge much lower than nano-TiO_2, nano-Al_2O_3, and nano-SiO_2 (Mu et al. 2011). Zhang et al. (2017) revealed that the Zn^{2+} released from nano-ZnO reduces the biogas production and it can be one of the main reasons for the positive impact on volatile fatty acids (VFAs) accumulation in sludge anaerobic digestion (Zhang et al. 2017).

In another approach, Luna-del Risco et al. (2011) studied the influence of copper and zinc oxide particles size on biogas production from mesophilic anaerobic digestion of cattle manure (Luna-del Risco et al. 2011). The tests were performed in gas-tight closed serum bottles during a period of 14 days. The results showed that both nano-Cu and nano-ZnO have inhibitory effects on biogas production. Relative to total biogas obtained in the control sample, a nano-CuO concentration of 15 mg/L reduced in 30% the biogas production. In the presence of 120 and 240 mg/L of nano-ZnO, the biogas production decreased by 43 and 74% (Luna-del Risco et al. 2011). Due to their toxicity on the methanogens, other nanomaterial additives, such as Mn_2O_3 (Gonzalez-Estrella et al. 2013) and Al_2O_3 (Alvarez and Cervantes 2012), are also associated with negative influence on biogas production.

On the other hand, application of nanosystems of supplementation of solid organic wastes (SOWs) with trace of metal nanoparticles is a promising way to enhance biogas production in view of the fact of many of the enzymes associated with methanogenesis step require a co-factor for its activation (Choong et al. 2016). As an example of nanoparticles commonly added into SOWs, Zhang et al. (2019a, b) quote zero-valent metals, metal oxides, nano-ash, and carbon-based materials (Zhang et al. (2019a, b). The work of Liu et al. (2015) demonstrated that zero-valent iron nanoparticles have great potential to promote significant enhancement in biogas yields obtained from waste activated sludge (Liu et al. 2015). In relation to such substrate, at mesophilic conditions, Su et al. (2013) found that the addition of 0.1 wt% zero-valent iron (ZVI) nanoparticles improves in 30.4% the biogas production rates (Su et al. 2013). About biogas production from anaerobic digestion of cattle dung slurry, Abdelsalam et al. (2017) reported that trace of metal nanoparticles such as 1 mg/L Co, 2 mg/L Ni, 20 mg/L Fe, and 20 mg/L Fe_3O_4 has demonstrated positive effects (Abdelsalam et al. 2017).

Abdallah et al. (2019) studied the impact of Ni-Ferrite and Ni-Co-Ferrite nanoparticles on biogas production from anaerobic digestion of cow manure (Abdallah et al. 2019). The experiments were carried out in 35 days, at mesophilic conditions. The results indicated that the use of Ni-Ferrite nanoparticles, at concentrations of 20, 70, and 130 mg/L, resulted in biogas enhancements of 30.8%, 28.5%, and 17.9%, respectively. Besides, Ni-Co-Ferrite nanoparticles, also at concentrations of 20, 70, and 130 mg/L, were able to increase the biogas production by 6.6%, 5.9%, and 32.9%, respectively (Abdallah et al. 2019). According to Sekoai et al. (2019), nanoparticles provide a large surface area for microorganisms to bind in active sites of molecules, which results in the increase of the substrate conversion by hydrolysis. In addition, the positive effects of nanomaterials on anaerobic digestion are attributed to their high reactivity and specificity (Sekoai et al. 2019).

Nanomaterials can also affect the biogas treatment for contaminant removal purposes. Ma and Zou (2018) investigated the effect of Cu and CuO nanoparticles on the removal of hydrogen sulfide in biogas using methyl diethanolamine (MDEA) as solvent (Ma and Zou 2018). The results showed that both nanoparticles tested, Cu and CuO can promote the gas-liquid mass transfer in the desulfurization process of biogas by MDEA. However, MDEA-based CuO nanofluids presented better absorption efficiency than MDEA-based Cu nanofluids (Ma and Zou 2018). All these works can conclude that nanoparticles could affect positively or negatively in biogas production, depending on the biogas source or in which step nanoparticles are employed.

2.6 Bioethanol

Modern use of nanosystems: enzymes-(magnetic nanoparticles) can increase ethanol production by optimizing the production of second-generation ethanol (E2G) from agricultural waste. The nanosystem for biofuels from lignocellulose biomass is still in its initial stage. The E2G ethanol has the advantages of not competing with food; advance technology still under development to reduce the cost of conversion and environmentally friendly (Rodríguez-Couto 2019). The biofuels industry has been looking for new biocatalysts to meet the growing demand for ethanol.

In this context, the prospect of using the cellulase enzyme complex to hydrolyze lignocellulosic biomass trapped in magnetic nanoparticles for E2G production is presented from the following characteristics: higher pH and temperature tolerance, higher storage stability, reusability in reaction cycles, and higher substrate affinity (Zhang et al. 2015).

Lupoi and Smith (2011) studied the hydrolysis of microcrystalline cellulose for cellulase-silica nanoparticles and obtained ethanol yields of 2.1 (P 1/4 0.06) to 2.3 (P 1/4 0.01) times higher in simultaneous saccharification and fermentation (SSF) compared to the enzyme in solution. These results indicate more technical and economic reliability for the use of nanosystems in the ethanol (E2G) production process (Lupoi and Smith 2011).

Different biomass was also studied to produce E2G ethanol using nanosystems. The jackfruit biomass and sugarcane leaves using the nanosystem cellulase supported on MnO_2 nanoparticles for E2G production (yield of 21.96 g/L) (Cherian et al. 2015). The biomass from sesbania aculeate using the nanosystem cellulase bound magnetic nanoparticles with E2G yields (5.31 g/L) (Baskar et al. 2016).

Carli et al. (2019) studied the use of nanosystems: enzymes β-glucosidase and endoglucanase supported on to functionalize magnetic nanoparticles for sugarcane bagasse hydrolysis. The hydrolysis of sugarcane bagasse alcohol insoluble residue treatment (AIR) and bagasse pretreated with dimethyl sulfoxide (DMSO) for incubation of nanosystems presented the total reducing sugar released with DMSO treated sugarcane were about 1.9-, 3.0-, and 2.1-fold higher than that released from AIR. The study also noted the improvement of the catalytic activity, high optimum temperature, with easy recovery using the magnetic field.

The hydrolysis feedstocks are available in bulk and in industrial facilities, resulting in an industrial facility, reducing costs compared to others biomass presented. Thus, the development of new technologies, nanosystems, should be designed in such a way that also encompasses solutions for the regional problems. Agribusiness brings different possibilities, especially in the use of energy residues, which even used, still does not have its full potential.

2.7 *Other Biofuels Production Using Nanotechnology Systems*

Several other biofuels have the potential to be obtained using nanosystems, among which we can mention biomethanol, biobutanol, green diesel, among others, and these are briefly discussed in the present topic (Fig. 4)

In addition to the biofuels already discussed, we can also mention another very important one, biomethanol. In general, the term biomethanol is used to describe methanol produced from two different methods, being Fischer–Tropsch reaction of syngas or biomethane (Minteer 2011). Methanol is the simplest alcohol with the chemical formula (CH_3OH) and compared to ethanol, is more volatile and more toxic. The raw materials that can be used for methanol production can be concentrated carbonaceous materials of any kind, such as biomass, coal, or even carbon dioxide and solid waste (Melikoglu et al. 2016). Among the main benefits of methanol, we can mention that it is a distributed energy source for power generation (Suntana et al. 2009) and after its combustion can easily be decomposed into carbon dioxide and water vapor (Shamsul et al. 2014).

In water resource recovery facilities (WRRFs), methanol is a largely used exogenous carbon source for denitrification, in order to achieve low total nitrogen levels

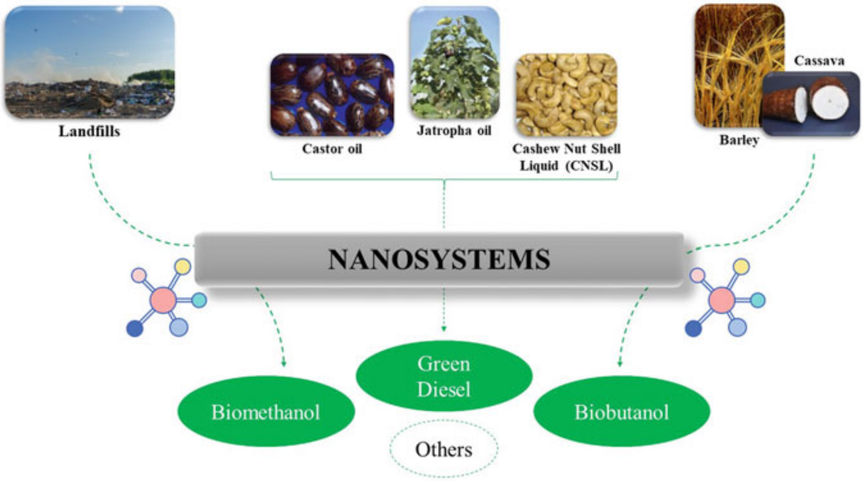

Fig. 4 Other examples of biofuels

(Cherchi et al. 2009). However, methanol used for denitrification is cost dependent on competing for demand from the chemical industries. Besides, there are certain safety concerns related to methanol storage and handling, making it difficult to purchase and transport commercial methanol to the facilities (Su et al. 2019). Biogenic methanol production can be an attractive solution to reduce dependence on external carbon sources and benefit the sustainability of these facilities. Biogenic conversion of methane to methanol has been previously demonstrated on a laboratory scale (Taher and Chandran 2013). However, the need for the use of high temperatures and the low conversion efficiency obtained made it more difficult to apply this technology to full scale (Shamsul et al. 2014).

Another biofuel that stands out is biobutanol. It is a straight-chain primary fuel of molecular formula C_4H_9OH with an alcohol (–OH) functional group included (Bharathiraja et al. 2017; Tigunova et al. 2013). Butanol which is produced from the alcoholic fermentation of natural, biodegradable, or renewable biomass is considered biobutanol (Pugazhendhi et al. 2019). It has great potential as a biofuel, as it has similar properties to gasoline and can be used directly in the car engine without modification (Malik et al. 2014; Xue et al. 2016). Besides, it can be mixed with gas, diesel, or gasoline in any percentage and normally used as a transportation fuel (Pugazhendhi et al. 2019). Biobutanol is considered superior to biomethanol, as it has some advantages such as a greater mixture with gasoline, higher energy efficiency, less need for engine modification and high performance, among others (Dürre 2008).

The use of biomass feedstock for biobutanol production via fermentation began in the early 1900s (Pugazhendhi et al. 2019). Since then, several cellulosic raw materials have been used for its production, such as cassava bagasse, changed grass and miscanthus, barley straw, maize, and wheat, as well as glucose and cornstarch (Huang et al. 2015). However, lignocellulosic biomass is the most promising raw material for biobutanol production (Jiang et al. 2019). Microorganisms of the genus clostridia are the main fermentative organisms used for biobutanol production, is known for their potential to ferment different types of renewable biomass in butanol through the fermentation pathway known as acetone-butanol-ethanol (ABE).

Although that certain microbial strains have intolerance to butanol accumulation in the fermentative medium, alternatives to solve this problem have been studied, such as increasing the resistance of these strains to butanol accumulation through the use of genetically modified or mutant strains with potential for production of biobutanol, searching for low-cost substrates for anaerobic fermentation, choosing the most suitable fermentation system, and also adapting the production system already used to nanosystems, thus seeking to improve this bioprocess (Pugazhendhi et al. 2019).

Biodiesel is a biofuel that has been previously discussed and despite having several advantages, it also has problems related to chemical stability, low calorific value, cold flow and filterability due to oxygen present in its structure (Knothe 2010; Santillan-Jimenez et al. 2013). As an alternative, green diesel consists of a drop-in biofuel, which has no oxygen in its composition and is different from those from petrochemical sources (Scaldaferri and Pasa 2019). Their present some features, like full compatibility with petro-diesel, has a high calorific value (44 MJ/kg), and low specific gravity (0.78), is very storage stable and promotes lower combustion emissions (Kalnes et al. 2009). Drop-in biofuels also have higher combustion heat compared to oxygenated biofuels (bioethanol and biodiesel) and are not hygroscopic (Scaldaferri and Pasa 2019).

For green diesel production, many raw materials can be applied, and the most used are cashew nut shell liquid (CNSL), jatropha oil, and castor oil (Orozco et al. 2017; Scaldaferri and Pasa 2019; Yenumala et al. 2019). Starting from these materials, the mentioned biofuel can be obtained from pyrolysis, catalytic cracking, and hydrodeoxygenation (HDO), commonly prepared under high pressure and in the presence of hydrogen, being widely accepted due to the high yield of product obtained (Yenumala et al. 2019). However, it is a less economic and ecological route, because it consumes a large amount of conventional hydrogen for the reaction to occur (Huber et al. 2007). Some alternative routes have been studied, such as the production under a free hydrogen atmosphere, better known as the deoxygenation process (DO). It is a route whose reaction conditions are lighter compared to the HDO process (Lestari et al. 2010).

3 Future Trends

Nanosystems have a wide range of applications for improving biofuel's production. However, some issues need to be solved to increase commercial availability for some of these systems. For this purpose, many studies are being developed, and this topic shows some of the prospective in this technology.

The use of immobilized enzymes is proven to be a powerful mechanism in biofuels processing, but it is impossible to predict and control enzyme stabilization with different supports and immobilization media (Kočar et al. 2015; Pavlidis et al. 2014). Although nanosystems have a potential stabilizing effect in the application of nanostructured materials, this mentioned effect is enzyme-specific, being difficult to understand the conformational changes as they interact with nanomaterials (Zeng et al. 2019). To expand studies of this type, it is necessary to lower the cost of the whole nanosystem, from the nanoparticle's synthesis to the immobilization protocol.

Another possible strategy is to combine enzyme immobilization with genetic modification. The last one is primarily used to produce soluble enzymes and these are not reusable and have low stability. The combination could bring novel biocatalysts with both qualities from the individuals' research fields and high efficiency for industrial applications (Bernal et al. 2018)

Biofuel cells were not mentioned in this chapter since they are on an early stage of research and more issues to be solved. However, nanomaterials are proven to increase electron transport between electrode surfaces and biocatalysts. Their exploration will increase due to their excellent electrocatalytic activity and that will enhance the current and power densities of biofuel cells (Zhao et al. 2017).

For nanosystems applying nanoengineered materials for bioelectrode development (metal nanoparticles carbon nanotubes and graphene), it is necessary to understand the interactions with the biomolecule/enzyme aggregated, making the appropriated modifications to their structure and immobilization and functionalization protocols to obtain practical nanobiocatalytic systems (Adeel et al. 2018). Those characteristics are linked and therefore need a lot of studies for optimization of the mentioned systems, providing better performance in the field of a biological fuel cell. The use of redox mediators to improve the communication between the enzyme's redox center and the electrode surface could be a way to increase the performance of the cells (Adeel et al. 2018).

The use of another type of nanosystems using simple synthetic methods to nanostructure biomimetic materials is also being studied, without the use of enzymes. These materials mimic the native environments of the biocatalysts, extending their longevity and therefore extending the cell's lifespan (Zhao et al. 2017). For biohydrogen cells, it is also necessary to improve the storage and distribution infrastructures and overcome other scale-up problems (Aricò et al. 2013).

For biodiesel production, the integration of nanosystems with biofuel production could be the key to improve their development to its full potential, and one strategy to increase the yield of the process is using co-immobilization technique, facilitating the contact of various enzymes in each step of the reaction or just improving the hydrolysis of complex substrates for biofuel synthesis (Lee et al. 2013).

On another step from biofuels production chain, the separation is primally made with inorganic materials, but nanosystems using nanoporous carbons can also be used as effective, selective, and economic absorbents for the separation of biomolecules (Peluso et al. 2019). More recently, biocompatible mesoporous nanoparticles with property to absorb hydrophobic molecules have been developed and investigated as a mechanism to harvest fatty acids from algal cultures (without killing them) and are subsequently used to produce biodiesel by esterification. The fact that the lipids are stored between the cell membrane and the cell wall makes this harvesting strategy possible without interfering with the cell membrane, putting mesoporous nanoparticles that withdraw oils from the cell and into the pores. To prevent transesterification in situ, strong oxides can be introduced into the pore structure. Nanofarming could provide another sustainable component in biodiesel production, produced directly without biocatalyst disruption (Biswas 2019)

Nanosystems could also bring revolutionizing changes to biogas production, improving the stability of cellulose enzymes, enhancing the catalytic capacity in biohydrogen production, and improving biological and chemical digestion with biocompatible nanoparticles. These topics need deeper comprehension and a cheaper production to increase the use of this technology in the future (Faisal et al. 2018). Anaerobic photo-fermentation reactors using visible-light photoactive metal oxides could be a way to increase methane production and turn this type of system more economically attractive (Ganzoury and Allam 2015).

4 Conclusion

Many researches had been made to increase biofuels production efficiency with the employment of nanosystems. For biocatalytic nanosystems, it is necessary to improve stability, biocompatibility, and economical availability. Strategies like co-immobilization of enzymes, nanofarming, synthesis of nanostructured biomimetic materials among others previously discussed could bring the full potential to biofuels production field, but many further researches are necessary to reach this point.

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