Chapter 12
Progress and Perspectives of Nanomaterials for Bioenergy Production

Alka Pareek and S. Venkata Mohan

Abstract Bioenergy that comprises biodiesel, biogasoline, bioethanol, biobutanol, hydrogen, etc. is one of the emerging renewable energies capable of tackling climate change and promising long-term durability. There is an upsurge in the interest in scientific field to enhance the output of the biofuel industry that seeks intervention of nanotechnology to overcome the limitations. Nanotechnology is a tremendously growing field merging and effecting a wide range of technological, biological and pharmacological applications but still its usage for bioenergy production from biomass is at a budding stage. Employing nanomaterials in the production of bioenergy increases efficiency and reduces process cost. Nanosized materials enhance the reaction kinetics of catalysis process by providing more catalytic sites and considerably large surface area for interaction. Wide range of nanomaterials are synthesized with distinct properties and surface features to accommodate the demand of cost-effective and process-efficient biofuel industry. The promising role of nanotechnology in the biofuel industry can be realized from studies like increase in biodiesel production rate by nano-catalyst-based microbial enzymes, use of nanomaterial additives to enhance the biogas yield and improvement of anaerobic digestion process using magnetic nanoparticles. This chapter focuses on the role of bionanomaterials in biofuel production and highlights the impact of nanotechnology-based bioenergy generation through comprehensive literature study.

Keywords Bioenergy production · Nanomaterials · Biodiesel · Biogas · Microbial fuel cells · Carbon-based materials · Nanostructured materials

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12.1 Introduction

Ever-growing and huge energy demands are becoming the main challenge with respect to future energy requirements. It is well known that 90% of global energy is obtained from depleting carbon-based fossil fuels, which has been contributing to global warming. Moreover, surge in prices of crude oil and its refined products indicates continued depletion of fossil fuels (Waqas et al. 2018). Apart from incessant consumption of fossil fuels, there is also threat to environment causing serious global devastation. To accommodate these issues, bioenergy can be a lucrative alternative as bioresources are self-prevalent, ubiquitous, inexhaustible and extremely eco-friendly (Hoel and Kverndokk 1996; Lin and Huber 2009). Biofuels produced from biomass include biogas, bioethanol, biodiesel, biohydrogen and biomethane. Bioethanol production includes the following processes: pretreatment, hydrolysis, saccharification and fermentation using sugarcane, corn, wheat, potatoes, lignocellulose biomass, etc. (Kim Keon Hee 2018). On the other hand, biodiesel is synthesized by transesterification process in which reaction of triglycerides and an alcohol produces acid alkyl esters. Biodiesel exhibits less CO_2 emission, high combustion efficiency, flash point, lubricant efficiency and cetane number (Abbaszaadeh et al. 2012). Regardless of these advantages, bioenergy is still underutilized due to lack of reliable techniques that can harvest biomass in an efficient manner (Zebda et al. 2018). Therefore, presently highly specific and focused modification methods are required to untap the unused potential of biomass. To enhance the production of biodiesel and biogas numerous approaches are explored recently. One such emerging technique that can contribute progressively towards the biofuel industry is nanotechnology that provides promising economical and productive modification tools to enhance biofuel generation. Nanotechnology is a branch of science that deals with materials of dimension or surface features in the size range of approximately 1–100 nm (Rahman et al. 2016). Nanomaterials are advantageous in biofuel systems due to unique properties like high surface areas, degree of crystallinity, adsorption power, catalytic activity, stability, longevity and storage capacity that can cumulatively optimize and make the entire process efficient (Donaldson and Poland 2013) (shown in Fig. 12.2). Moreover, it also provides a system having higher probability for recyclability, reusability and recovery. Nanoparticles thus used as co-catalysts in a system could bring a biotransformation of microbial species that maximizes the bioproduct production and hence promotes bioenergy generation (Ingle et al. 2019). The tool of nanotechnology in the biofuel industry participates in numerous applications like hydrogenation, transesterification, pyrolysis, anaerobic digestion and gasification (Zhang et al. 2013b). Some of the applications of nanotechnology in the bioindustry are shown in Fig. 12.1 (Srivastava et al. 2017). The choice of nanoparticle depends on the type of bioprocess and required yield of biofuels. This chapter summarizes the impact of nanotechnology on bioenergy production and parametrial dependency of each other through a comprehensive literature review.

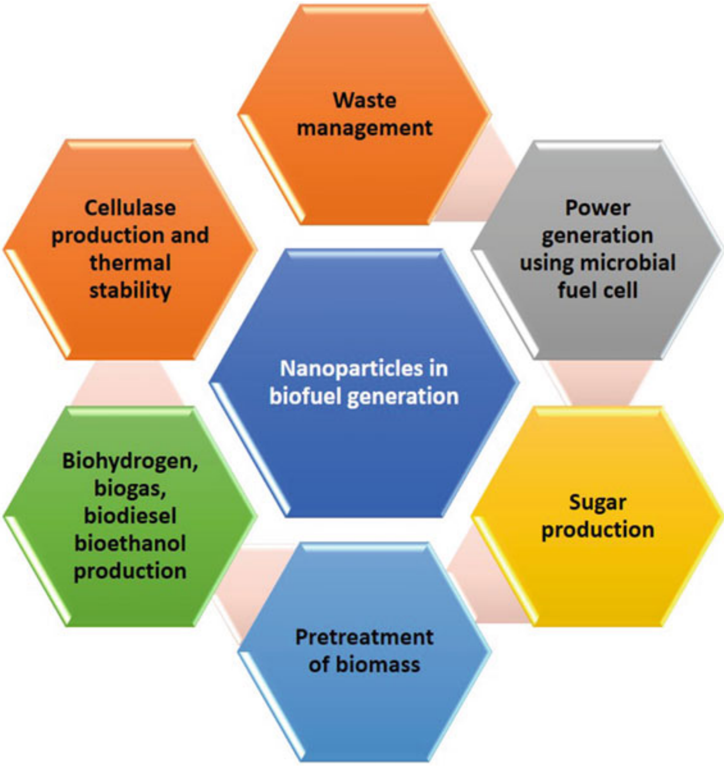


Fig. 12.1 Application of nanoparticles in the biofuel industry

12.2 Characteristics and Properties of Nanoparticles

Tremendous efforts are devoted to designing nanoparticles that can act as a functionalized catalyst for modifying biomass and making biosystems more efficient. These specially designed nanoparticles possess the potential for creating an economic, efficient, stable and durable biosystems capable of achieving higher bioproduct quality and yields. Therefore, it is imperative to study important characteristics and properties of nanoparticles relevant to bioenergy generation.

12.2.1 Characteristics of Nanoparticles

Recently, huge interest has risen in the organization of nanoscale structures into predefined superstructures due to their excellent physicochemical, optical, electrical and photoelectrochemical properties (Chandrasekharan et al. 2000). The small features in nanomaterials provide more functionality and accessible area in a defined

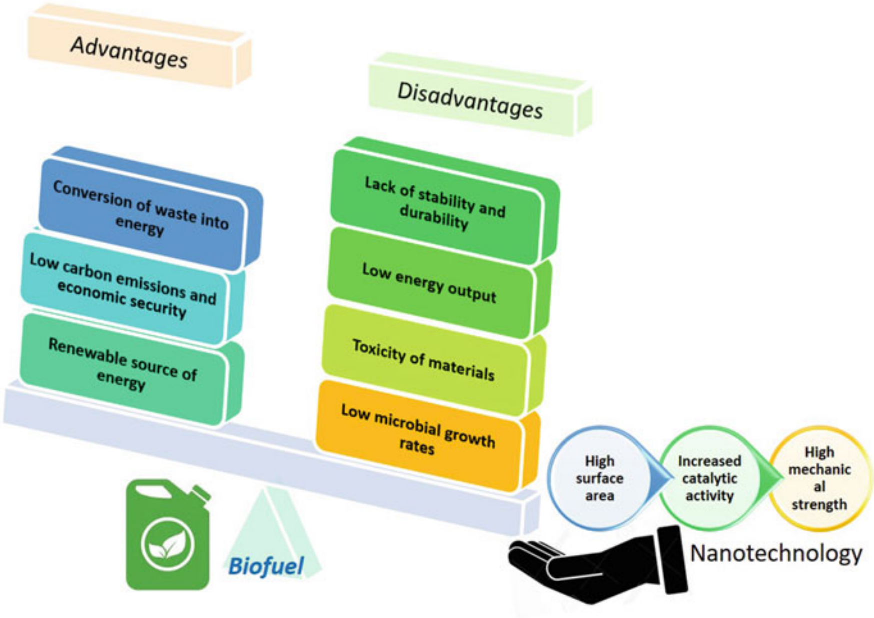


Fig. 12.2 Role of nanotechnology in the biofuel industry

space. Nanotechnology is not merely miniaturization from micro range to nanoscale but physical characteristics of nanomaterials distinctively vary from their bulk counterpart. Nanomaterials have low melting point, reduced lattice constant and enhanced catalytic properties as compared to bulk counterpart. For example, bulk aluminium is stable but highly combustible in nanoform; similarly macroscale metals like gold, silver and platinum are inert but their nanoparticles are highly reactive and possess catalytic properties. Even crystal structures of bulk materials that are stable only at high temperatures can be stabilized at a considerably low temperature in nanoform. These changes in properties are observed at nanoscale due to the fact that the population of surface atoms or ions significantly increases as compared to the total number of atoms and hence surface energy increases that controls the physical properties of nanomaterials. Nanoparticles are beneficial for various processes on account of their distinctive properties like enhanced Rayleigh scattering, surface plasmon resonance (SPR), Raman scattering in metal nanoparticles, confinement or quantization effects in semiconductor nanoparticles and superparamagnetic properties in magnetic materials (Schmid 2004). Nanotechnology plays an increasingly crucial role in many key technologies of the era like optoelectronics, catalysis, solar cells, water treatment, biomedical, electromagnetic, energy and nano-remediation (Guo et al. 2013). Owing to its versatility and important role in numerous applications, engineering of nanoparticles with different morphologies and surface properties is explored at an enormous scale. Researchers are working relentlessly towards either improving existing techniques or exploring new techniques to produce various shapes of nanoparticles like rectangular, triangular, flower, tubes, rods, wires and thin films. With nanotechnology being a vast interdisciplinary subject, various methods have been adopted to synthesize nanoparticles using physical, vapor, chemical, biological and hybrid techniques.

12.2.2 Nanoparticles for Bioprocesses

Nanoparticles along with liquid biomass exhibit an important part in water purification due to its antibacterial properties (Stoimenov et al. 2002). Utilization of nanomaterials has been proved to be the most beneficial technique for water treatment due to its high surface area and enhanced catalytic properties (Qu and Alvarez 2013). Recently, nanotechnology has also shown potential for applications like adsorption, photocatalysis, membrane processes, microbial control, disinfection, sensing and monitoring. Moreover, they are also employed in processes involving detection and subsequently removing chemical and biological substances like nutrients (phosphate, ammonia, nitrate), metals (Cd, Cu, Zn), cyanides, organic substances, algae species (cyanobacterial toxins), parasites, viruses and bacteria. Though some properties are useful for bioenergy production some may show adverse effects like toxicity and concentration of nanoparticles (Lazar 2011). Microorganisms have the tendency to respond to various nanoparticles that can significantly affect the efficacy of biological process. Antimicrobial properties of nanoparticles may impose potential adverse effects on microorganisms and harm cell membranes that leads to change in structures and causes more permeability in bacteria (Lazar 2011). This effect is dependent on various factors like size, shape and concentration of nanoparticles. Antibacterial property of nanoparticles is dependent on two parameters: firstly, physicochemical properties of nanoparticle and secondly, bacteria type.

In one of the studies, it is claimed that the Ag nanoparticle-treated coliform bacteria irradiated with ultrasonic waves enhanced the antibacterial activity. Even sometimes nanoparticles in close vicinity with a microbial community reduce the efficiency of anaerobic digestion process causing unsuccessful treatment process and release of contaminated effluent (Hoffmann and Christofi 2001). For similar reasons, silver nanoparticles are utilized in various medical processes like dental treatment, tubes including catheters and curing burn wounds (Klasen 2000). Adverse effects of silver nanoparticles were studied using *E. coli* and observed to be reliant on the concentration of nanoparticles and exposure time. Harmful effects of nanoparticles on bacteria are associated with the leakage of reducing sugars and proteins, cell disruption, enzyme inhibition and scattered vesicles that inhibits cellular respiration and hence cell growth. It is observed that the toxicity of nanoparticles can be largely controlled when nanoparticles are exposed to sludge. The effect of toxicity on bioenergy yield is dependent on the concentration, nanoparticle size, exposure time and microorganism type. Moreover, behaviour of nanoparticles and its interaction with biomass are also dependent on environmental factors like pH, light, ionic strength and natural organic matter (Klaine et al. 2008). Both size and shape of nanoparticles play a crucial role in bioprocesses; for example it is reported that nanoparticles of size less than 30 nm are cytotoxic towards *E. coli* and *S. aureus* (Martinez-Gutierrez et al. 2010) as compared to nanoparticles of size 80–90 nm (Martínez-Castañón et al. 2008). Similarly shape of nanoparticles is also important; for instance, the triangular shaped AgO nanoparticles exhibit the highest bactericidal effect on *E. coli* in comparison to spherical or rod-shaped AgO in both agar plate and broth cultures.

12.3 Role of Nanoparticles in Bioenergy Generation

12.3.1 Biodiesel Production

Biodiesel is an eco-friendly fuel that behaves like fossil diesel which is synthesized domestically using vegetable, animal and waste cooking oil (shown in Fig. 12.3) (Marchetti et al. 2007). The process of conversion of these oils to biodiesel is known as transesterification (Vasudevan and Briggs 2008). The transesterification process is the mechanism in which fat/oil (triglyceride) reacts with an alcohol to form esters and glycerol. This fuel possesses properties like density, air/fuel ratio and heat of vaporization that are comparable to mineral diesel. Moreover, it is beneficial for the environment as it reduces carbon monoxide, sulphur oxide and smoke emissions. Nanomaterials have been widely studied for the optimization of yield and quality of biodiesel. Magnetic nanoparticles are used as catalysts for industrial scale biodiesel production owing to their ease of separation from the final product, reusability and economic nature (Gardy et al. 2018, 2019). Similarly, metal oxides like TiO_2 (Gardy et al. 2017), CaO (Liu et al. 2008), MgO (Verziu et al. 2008) and SrO (Liu et al. 2007) nanoparticles show great catalytic activity for efficient biodiesel production. Enhanced biodiesel generation has been reported using carbonaceous materials like

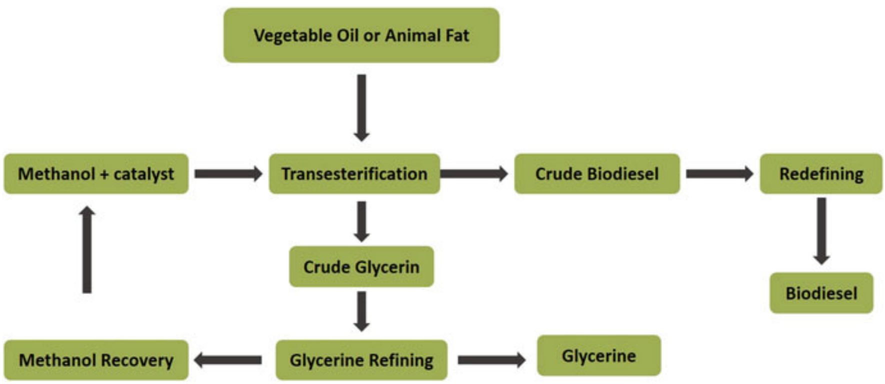


Fig. 12.3 Schematic showing the process of biodiesel production

graphene oxide (Mahto et al. 2016), carbon nanotubes (Guan et al. 2017), carbon nanofibres (Stellwagen et al. 2013) and biochar (Dehkhoda et al. 2010). Few mesoporous nanomaterials are also studied with excellent structural properties that exhibit improved catalytic activities for biodiesel production. Tangy et al. used microwave irradiation and studied SrO nanoparticle-decorated SiO_2 beads for generation of biodiesel from waste cooking oil (Peralta-Yahya and Keasling 2010). High conversion values as large as 99.4 wt % (in 10-s irradiation time) were obtained using composite nano-catalyst, leading to the development of economical biodiesel in a very short time. Jayanthi et al. studied the efficiency and emission characteristics of DI diesel engine filled with biodiesel using copper nanoparticle additives. Reports suggested that brake thermal efficiency was enhanced to B20+ 80 PPM CuO and also specific fuel consumption was reduced at full load conditions (Jayanthi and Rao 2016). There are some reports that studied improvement in biodiesel generation by employing alumina (Al_2O_3) and cerium oxide (CeO_2) nanoparticles. In one such study, Al_2O_3 and CeO_2 nanoparticles of each 30 ppm were used in DI diesel engine that improves the brake thermal efficiency by 12%, followed by reduction of 30%, 60%, 44% and 38% in NO, CO, hydrocarbon and smoke emission, respectively (Prabu 2017). Similarly, Ramesh et al. studied performance, combustion and emission characteristics of diesel engine by employing alumina nanoparticles as additive with poultry litter (Ramesh et al. 2018). Chaichan et al. reported the effect of alumina nano-fluid (aqueous) on diesel engine's performance and emission characteristics (Tariq et al. 2017). The group studied the impact of nano-Al_2O_3 (51 nm diameter) with varying weight fractions of 1%, 3%, 5%, 7% and 10% that resulted into improved brake thermal efficiency by 5.5% and decreased the relative fuel consumption by 3.94%. Kim et al. reported the design of nanoparticles (magnetic and non-magnetic), carbon nanotubes and carbon nanofibres as nano-immobilized biocatalysts for biodiesel generation (Kim Keon Hee 2018). Recently, Ajala et al. synthesized nano-catalysts using waste iron filling for biodiesel production (Ajala et al. 2020). Numerous groups have reported nanotechnological advancements towards efficient biodiesel production using nano-catalysts (Dantas et al. 2020; Xie and Wang 2020; Mofijur et al. 2020).

12.3.2 Biogas Production

Biogas is a kind of biofuel that is generated naturally from the decomposition or breakdown of organic waste such as food scraps and animal waste in an environment absent of oxygen (anaerobic environment). Decomposition of waste in anaerobic conditions releases a blend of gases, primarily methane and carbon dioxide, and the above process is also known as anaerobic digestion. Common sources of biogas generation constitute sewage treatment plants, landfills, organic industrial waste and mesophilic and thermophilic digestion of organic wastes (Ganzoury and Allam 2015). The anaerobic digestion process contains mainly four steps, namely, hydrolysis, acidogenesis, acetogenesis and methanogenesis. To improve the efficacy of

Table 12.1 Biogas production using different nanoparticles

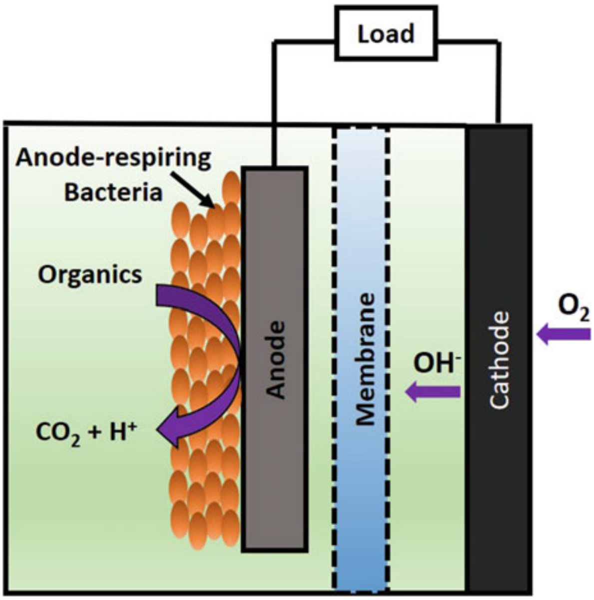
Type of material	Nanoform	Effect of nanomaterial on biogas
Transition metal oxides/zero-valent metals	Titania, ceria, nano-zero-valence iron (NZVI)	Outcome is dependent on the nanomaterial concentration and process digestion time
Metal oxides	ZnO, CuO, MnO_2, Al_2O_3	Reduction in the rate of biogas production
Zero-valent metals	Zero-valent iron nanoparticles	Leads to enhanced methane generation
Transition metal oxides	Metal oxide nanoparticles encapsulated in porous SiO_2	Considerable enhancement in methane
Nanoform ash and carbon nanostructures	Ag/Au nanoparticle	Biogas production depends on the concentration
	Micro/nano fly ash or bottom ash	Enhanced biogas
	C60 (fullerene) and SiO_2 nanoparticles, SWCNTs	Unaltered biogas

this process, numerous nanomaterials were utilized as additives to improve the biogas quality and yield. During anaerobic digestion process, employing of various nanoparticles like iron oxide, fly ash, zero-valence iron, bottom ash and metal oxides has promisingly increased methane production (Table 12.1). Though Mohamed et al. have extensively discussed a variety of nanoparticles employed till now for biogas production in the form of review paper, few studies are presented here (Ganzoury and Allam 2015). Abdelsalam et al. carried out biogas production using laser irradiation and Ni nanoparticles from anaerobic digestion. The combination of laser irradiation (irradiation time ~2 h) and 2 mg L^{-1} Ni nanoparticles obtained maximum specific biogas and methane generation of 679.5 mL and 453.3 mL, respectively (Abdelsalam et al. 2018). Unsar et al. studied anaerobic digestion using CuO, Ag and CeO_2 nanoparticles and elaborated their long- and short-term impacts using municipal waste-activated sludge (Ünşar et al. 2016). Casals et al. studied programming related to iron oxide nanoparticles disintegrated in anaerobic digesters that boosted biogas generation (Casals et al. 2014). Ambuchi et al. have shown enhancement in biogas production using Fe_2O_3 nanoparticles and multiwall carbon nanotubes (MWCNT) (Ambuchi et al. 2016). The study demonstrated that 0.75 g/L and 1.5 g/L concentration of Fe_2O_3 nanoparticles and MWCNT, respectively, caused faster substrate consumption and higher biogas generation. Abdelsalam et al. studied the effect of cobalt and nickel nanoparticles on methane and biogas production (Abdelsalam et al. 2017). The study revealed that the maximum biogas and methane generation was observed with 2 mg/L nickel nanoparticles showing 0.61 l biogas and 0.36 l methane, respectively. Duc et al. synthesized CeO_2 nanoparticles of size 192 nm and 10 mg/l concentration that increased the biogas production from UASB sludge by 11% (Nguyen et al. 2015). Similarly, 7.5 nm size TiO_2 nanoparticles of concentration 1120 mg/l by Garcia et al. enhanced the wastewater treatment sludge by 10% (García et al. 2012). In another report, Fe_3O_4 nanoparticles of size 7 nm were synthesized that increased the biogas yield by 180% and methane by 234% from wastewater sludge (Casals et al. 2014). Lo et al. reported the use of micro/nano fly and micro/nano bottom ash in anaerobic digestion that increased the biogas production by 2.9 times and 3.5 times, respectively (Lo et al. 2012). Al-Ahmad et al. reported the synthesis of nickel (Ni)-, cobalt (Co)-, iron (Fe)- and platinum (Pt)-encapsulated porous SiO_2 structures and their effect on anaerobic digestion process. It was found that methane production was increased in the range of 70%, 48%, 7% and 6% with Ni, Co, Fe and Pt nanoparticles, respectively (Al-Ahmad et al. 2014).

12.3.3 Bioelectrochemical Systems

Bioelectrochemical system (BES) is a promising technology for converting chemical energy of waste into electrical energy and other valuable products employing the technologies like microbial fuel cells (MFC) and microbial electrolysis cells (MEC). MFC is the most studied technology in which bacteria carry out a chain of redox reactions to transform organic mass into electric current (Butti et al. 2016). Generally, MFCs generally consist of two important parts: anode compartment and a cathode compartment divided by a proton-exchange membrane (PEM), as depicted in Fig. 12.4. The chemical mixture contained in anode gets oxidized using microbial metabolism following anaerobic conditions, which generates electrons and protons. The electrons migrate to anode and travel via external circuit producing electrical

Fig. 12.4 Schematic showing MFC cell set-up



current; on the other hand, protons are transferred to cathode via PEM. MFCs are categorized into two groups: mediator-assisted MFC (where mediators are introduced to the system) and mediator-less MFC. In mediator-less MFCs, bacteria consist of conductive pili or electrochemically active membrane-associated cytochromes secrete redox-mediating molecules that facilitates the flow of electron. MFC has matured as an emerging technology for biological treatment of municipal or industrial wastewater (He et al. 2005; Logan et al. 2006). Still, this technology is not apt for commercialization owing to its poor power outcome. To improve the output power of MFC, research is carried out in two broad aspects: (1) the bacteria that work efficiently in mediator-less MFC (2) and the material used to make electrode. Electrode material is a vital aspect as physical and chemical characteristics of various electrode materials influence microbial attachment, electron transfer, electrode resistance and rate of electrode surface reaction. Nanostructuring of electrodes is an easy and cost-effective way of improving MFC performance (Choudhury et al. 2017). Carbon-based materials are most dominantly followed by anode and cathode materials in history and present era of MFC technology owing to outstanding chemical, electrochemical and biological stability. Carbon-based materials are studied in different shapes and sizes like graphite rod, graphite fibre (Chaudhuri and Lovley 2003), brush, carbon cloth, carbon paper, carbon felt (Chen et al. 2012) and reticulated vitreous carbon (RVC) (Wei et al. 2011). But there are some shortcomings associated with this class of materials like limited electrical conductivity. In the case of graphite, electrical conductivity is 2–3 times lesser than metals. Metal electrodes were also explored as anode; though they offer good electrical conductivity they offer poor microbial adhesion or biofilm formation, electrochemical corrosion and metal ion-related water pollution. Few of them like Cu and Ag are even known to show antimicrobial properties, which restricts their candidature as anode material in MFC (Yamashita and Yokoyama 2018). To overcome these problems, nanostructured materials like CNT and graphene are studied that offer higher surface area for bacterial colonization, biocompatibility, great conductivity and extraordinary mechanical strength due to their distinct features and morphologies (Pareek and Mohan 2018, 2019). Moreover, they also offer excellent charge-storing capacity that further enhances output power density of MFC and provides possibility of constructing hybrid MFC. But synthesis of CNT (Mohanakrishna et al. 2012; Zhang et al. 2013a) and graphene (Singh et al. 2011; Gautam et al. 2016; Pareek et al. 2019a, b, c) includes complicated methods that increase the cost associated with MFC operation. Moreover, stacking of graphene sheets owing to strong van der Waals forces reduces their surface area as a result of negatively charged bacteria that experience electrostatic repulsions from graphene. Metal oxide nanomaterial (Mehdinia et al. 2014; Winfield et al. 2016) is another class of material, which can replace carbonaceous electrodes owing to its greater mechanical strength, morphological and electrochemical stability, biocompatibility, redox activity, electrochemical stability, low cost and eco-friendly nature. Still their performance in MFC is delimited due to relatively low conductivity, porosity and surface area. In view of this, reticulated vitreous or 3D sponge-like carbon nanomaterials were fabricated that provide hierarchically micro-, meso- or macrospores, establishing outstanding power density (Yang et al. 2016; ElMekawy et al. 2017). These hierarchical porous structures enhance the bacterium attachment due to the micro- and mesoporous pores, provide superior bacteria and electrode interaction enhancing the charge transfer process and form good-quality stable biofilm for long-term application. However, there is clogging of mesopores with time due to rapid bacterial growth, which prevents further electrolyte diffusion and limits further bacterial colonization of anode surface.

12.4 Conclusions

Nanomaterials exhibit a crucial role in enhancing the efficiency of bioenergy production by interconnecting biological processes with nanomaterials. It has impacted various bioprocesses like fermentation, enzyme hydrolysis, biomass pretreatment, product separation and microbial fuel cells. This chapter briefly discusses reports highlighting different nanomaterials that have been utilized to improve the efficiency of biofuels like biodiesel, biogas and bioelectrochemical systems. Wide range of nanomaterials are extensively explored as catalysts in biodiesel production and as additives in biogas production. A high surface area-to-volume ratio of nanosized particles provides greater reactivity and hence catalytic activity to them. On the other hand, the antibacterial activity of nanoparticles poses adverse effect or inhibits the bioprocesses, which is dependent on nanoparticle size, concentration of nanoparticles and time of exposure. There is enormous possibility to explore the effect of nanomaterials on biofuel generation using other biomasses, like agricultural waste and municipal solid waste (MSW). It is required to study optimum concentration and exposure time to reduce the toxicity of nanoparticles and also bioactive nanometal oxides.

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