

Introduction to Biofuels and Potentials of Nanotechnology

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Abstract In recent decades, the development trends of biofuel technology worldwide have been staggering. In the US, the transition from conventional biofuels (derived from food crops) to advanced biofuels (cellulosic and algae-based fuels) occurred in less than a decade. Advanced biofuel technologies involve breaking down cellulose in plant biomass or extracting lipids from algae biomass, both of which are expensive processes driving up production costs. As of today, advanced biofuels are still cost-prohibitive, especially as they compete with low crude oil prices or even conventional biofuel prices. Despite unfavorable economics, both cellulosic ethanol and algae-based fuels help reduce GHG emissions, while also counteract the tradeoff between food and fuel production, as given with conventional biofuels/feedstocks. Nanotechnology has been implemented in the biofuel production process as a potential solution to economic infeasibility of advanced biofuels either by altering the feedstock or by increasing the biomass content. Although nanotechnology bears potential opportunities for biofuels production, full implementation of this technology is still challenging, while some studies report potential risks as well. This chapter presents an overview of different conventional and advanced biofuels and feedstocks, their developments and production trends at the global and US level. It also points out current challenges for advanced biofuels and discusses potentials and risks related to nanotechnology application in biofuels production.

1 Introduction—Biofuel Types and Feedstocks

Establishing biofuels as a part of the energy portfolio worldwide has taken more than five decades. The beginnings of biofuels date back to 1900 when Rudolf Diesel (German inventor and mechanical engineer) presented an engine run on peanut oil at the World Exhibition in Paris. This invention was followed by a

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Table 1 Biofuels classifications, products, and feedstocks

Classification	Product	Feedstocks
First generation (food feedstocks)	Ethanol	Corn, cereals, sugar beet/sugarcane
	Biodiesel	Soybean, rapeseed, palm oil, animal fats, waste oils
Second generation (biomass)	Cellulosic ethanol	(a) Energy crops (switchgrass, miscanthus, wheat straw, poplar, willow, jatropha)
		(b) Green waste (corn stover and other field residues, e.g., stalks and stubble (stems), leaves, seed pods, as well as forest/park residues)
Third generation	Biodiesel/ethanol	Algae
Fourth generation	Biodiesel/ethanol	(a) “Drop in” biofuels, GM crops for biofuels
		(b) Renewable solar fuel (e.g., Joule)

Model T engine designed to run on what is today called corn ethanol, among other potential fuels, and was introduced by Henry Ford (American industrialist) in 1908. The revolution of vegetable oils as a feedstock for diesel fuels began in the 1930s and 1940s. The development of the biofuels technology (both ethanol and biodiesel) took different paths in different countries and was regulated with different policies.

In the United States (US), the 1970 Clean Air Act introduced by the Environmental Protection Agency (EPA) set the first standards for fuel additives. Since 1992, a strong governmental support for alternative fuels was enacted with the Energy Policy Act to ensure independence from foreign oil. In 2005, the Energy Policy Act implemented amendments to the MTBE (methyl tert-butyl ether) (additive in gasoline) due to its scientifically proven carcinogenic characteristics. This development opened a wide market niche for biofuels and facilitated a fast uptake of this technology. At the same time, a variety of different feedstocks have been explored to expand biofuels portfolio. The 2007 Energy Independence and Security Act substantiated those developments with Renewable Fuel Standards (RFS) that set minimum production and blending requirements for each biofuel type.

Biofuel types are determined and characterized by feedstocks used for their production (Table 1). The most common classification categorizes biofuels as “conventional” and “advanced” biofuels.¹ Conventional (i.e., first generation) biofuels encompass ethanol and biodiesel produced from eatable crops, which has raised questions about their competition for resources (e.g., water, energy, land) with food production (Rathmann et al. 2010; Harvey and Pilgrim 2011, Ajanovic 2011). Advanced biofuels (second- to fourth-generation biofuels) were defined by the 2007 Energy Independence and Security Act as liquid fuels from non-food/non-feed

¹In addition to this classification, other terms are known in the biofuels methodology, e.g., “bioalcohol” that describes a broader group of biofuels, including ethanol, methanol, propanol, and butanol. As this contribution is focused on cellulosic ethanol and algae-based fuels, other alcohols will not be discussed in this chapter.

sustainably grown feedstocks and agricultural (municipal) wastes. Accordingly, advanced biofuels need to meet sustainability requirements, i.e., reduce greenhouse gas (GHG) emissions by a higher percentage than conventional biofuels and not create any competition with food crop production.

In the discussion of the most economically feasible feedstocks, scientists are still divided with their research findings of energy efficiency of corn ethanol compared to gasoline (Shapouri et al. 2002; Pieragostini et al. 2014; Sheehan et al. 2003). On the other hand, research studies have been more unanimous about positive energy balance of advanced biofuels (i.e., cellulosic ethanol) and their lower environmental footprint compared to gasoline (Bansal et al. 2016; Schmer et al. 2008). They have also been commended as a viable option to counteract the competition for resources with food production. This chapter will address two biofuel types: cellulosic ethanol and algae-based fuels and ways how nanotechnology can impact their production and processing.

Cellulosic ethanol can be produced from crop residues or from energy crops planted specifically for biofuels production. Even though cellulosic ethanol unveiled prospective opportunities for the biofuels market, many challenges need to be overcome to make it a feasible solution in the long term. The key challenge is economic feasibility related to the complex process of breaking down cellulose, hemicellulose, and lignocellulose in plant materials, which requires high energy inputs and expensive enzymatic reactions. Nanotechnology has been found to provide a possible solution to this problem, and it will be discussed in more detail in Sect. 3.

In addition, algae-based (third generation) biofuels have expanded the biofuels market and have been embraced by private investors as the most energy-efficient biofuel with relatively low environmental impacts (Jones and Mayfield 2012; Singh et al. 2011). However, although algae biomass can produce between 10 and 100 times more oil per acre compared to traditional oil crops (e.g., oil palm) and can grow 20–30 times faster than food crops (Ziolkowska and Simon 2014), economic feasibility of algae-based fuels has also been a challenge (Doshi et al. 2016; Vassilev and Vassileva 2016). Nanotechnology has been applied in algae fuel production to increase efficiency of algae biomass and decrease production costs, thus making it a cost-competitive addition to the biofuel market.

In the context of biofuels discussion, the question of sustainability has played an important role in evaluating both economic, and environmental and social feasibility of each conventional and advanced feedstock. Accordingly, life cycle assessment, sustainability and environmental indicators, the energy concept, and uncertainty analyses have been among the most applied methods to determine short- and long-term sustainability of different biofuels and feedstocks (Chang et al. 2017; Saladini et al. 2016; Lazarevic and Martin 2016; Ziolkowska 2013, 2014a).

2 Biofuels Production Trends, Economic Feasibility, and Environmental Impacts

Global ethanol production has surged reaching more than 75 billion liters in 2009, doubling production volumes from 2004. The largest contributors to the ethanol market are the US (with its corn ethanol production of ~ 40 billion liters in 2009) and Brazil (with its sugarcane ethanol production of almost 30 billion liters in the same year) (Timilsina and Shrestha 2011). Other countries and country associations (like the European Union–EU), China, India, and Canada contribute to the global ethanol production to a smaller extent (Fig. 1).

A similar trend was recorded for biodiesel production at the global scale, with 2.3 billion liters in 2004 and 17 billion liters in 2009 (an increase by 672%) (Timilsina and Shrestha 2011). Production of biodiesel in the US has increased as a total, with temporary production variations over time. Brazil, France, and the rest of the EU have expanded their biodiesel production over time, while Germany's production was rather stable between 2004 and 2009. Also, Argentina and Italy contributed to the global biodiesel production at lower rates (Fig. 2).

Future projections anticipate continuous increase in biofuels production worldwide (OECD 2010). Accordingly, ethanol production is expected to go up to 160 billion liters by 2019, while biodiesel production is projected to increase to 41 billion liters in 2019, and increase by $\sim 113\%$ and $\sim 173\%$, respectively (Fig. 3). Moreover, the mix of different feedstocks in the global biofuels production has changed considerably over time. According to OECD (2010), the year 2016 will set the production peak for ethanol from coarse grains (including corn), while the

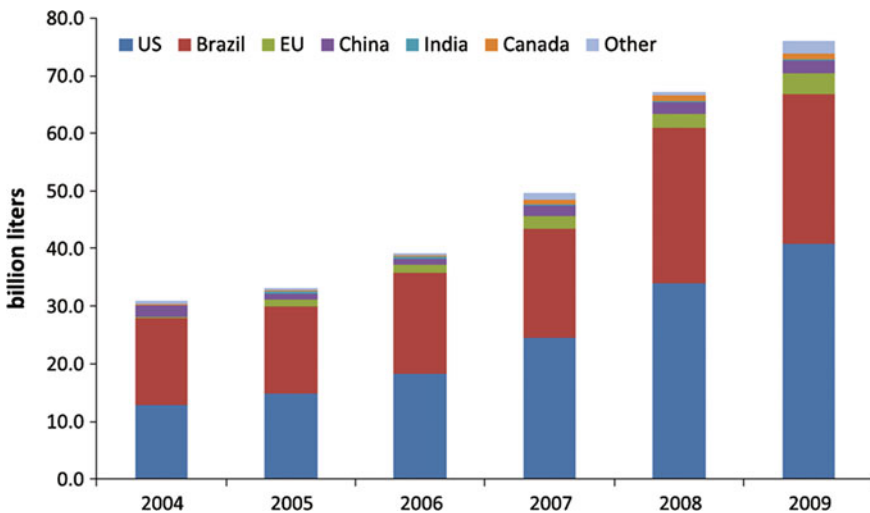


Fig. 1 Global ethanol production by country (2004–2009). Source Timilsina and Shrestha (2011)

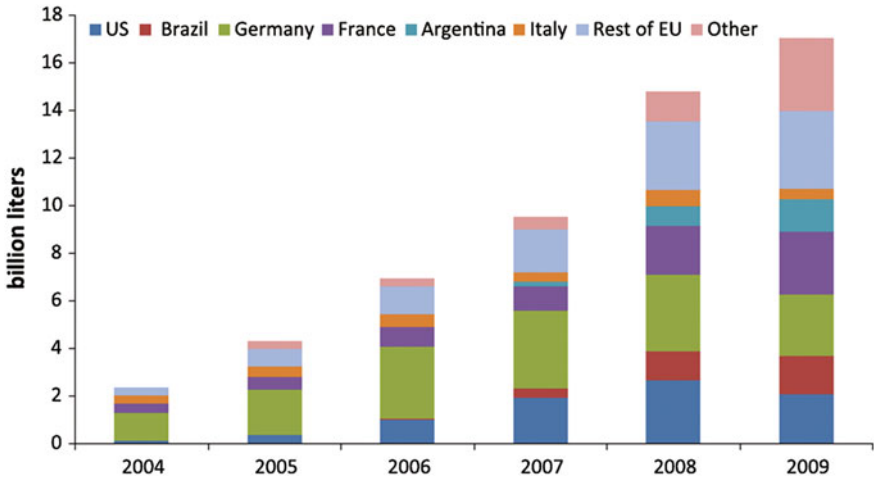


Fig. 2 Global biodiesel production by country (2004–2009). Source Timilsina and Shrestha (2011)

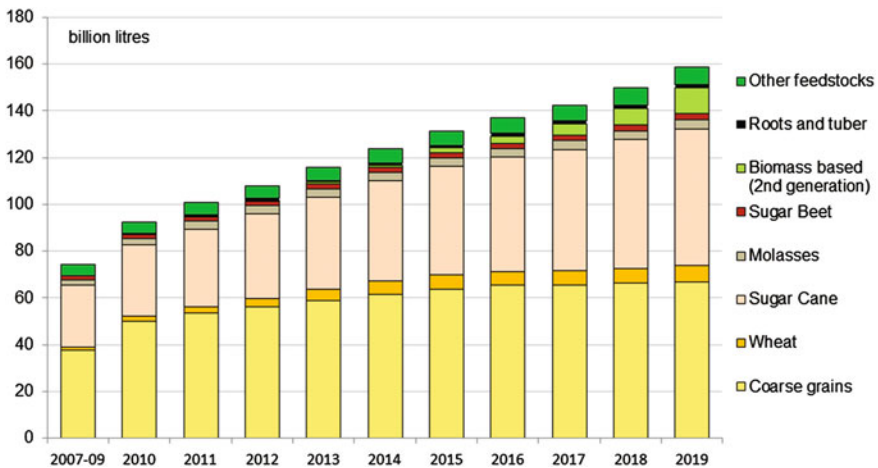


Fig. 3 Global ethanol production by feedstock—projections (2007–2019). Source OECD (2010)

production of sugarcane ethanol will increase throughout 2019. Furthermore, production of biomass ethanol (e.g., cellulosic ethanol) is anticipated to increase, reaching 11 billion liters in 2019.

Also, feedstock composition in global biodiesel production has varied over time. Use of vegetable oils has increased and is anticipated to grow up to 30.7 billion liters through 2019 (OECD 2010). Jatropha and other biomass feedstock for biodiesel production make a considerably smaller share; however, their use has increased over time and is anticipated to remain at this level in the future as well.

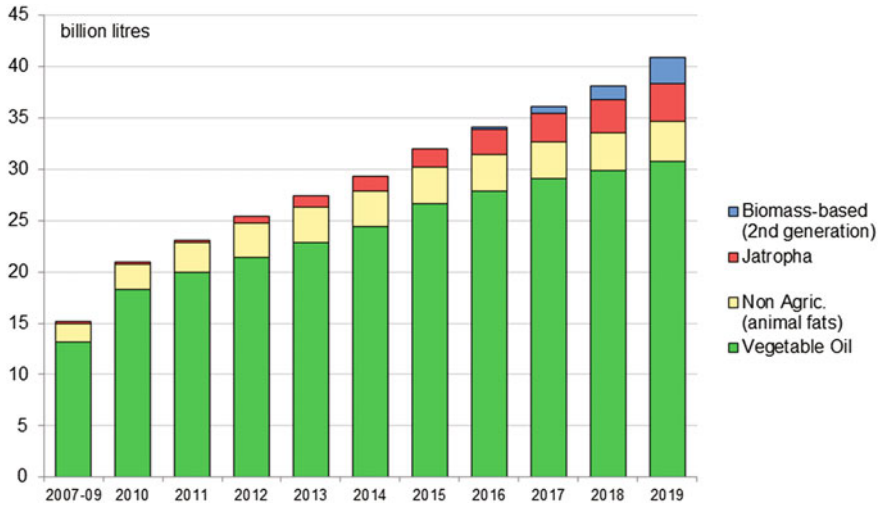


Fig. 4 Global biodiesel production by feedstock—projections (2007–2019). *Source* OECD (2010)

Production of biodiesel from animal fats has remained rather stable over time and is anticipated to reach the level of 3.9 billion liters by 2019 (Fig. 4).

On the US biofuels market, a significant increase in cellulosic ethanol production was sparked by the 2007 Energy Independence and Security Act that enacted Renewable Fuel Standards (RFS) as a mandate to expand the total quantity of renewable fuels blended into transport fuel from 9 billion gallons (34.07 billion liters) in 2008 up to 36 billion gallons (136.27 billion liters) in 2022. These totals were also divided into specific categories, with a requirement that each category of renewable fuel emits less GHG than petroleum fuel it replaces. Accordingly, starting in 2015, out of the total 36 billion gallons only 15 billion gallons (56.78 billion liters) can be provided on the market from conventional ethanol. The remaining volume needs to be produced from advanced feedstocks. In April 2010, the EPA announced the RFS2 that specified minimum quantities from specific feedstocks or biofuel types that need to be met toward the total mandate (FAPRI 2010a; Ziolkowska et al. 2010) (Fig. 5). Thus, the production of cellulosic ethanol is mandated to increase gradually and reach 16 billion gallons (60.5 billion liters) in 2022 (US EPA 2010) (Fig. 5).

Past and current developments indicate that cellulosic ethanol (and other biomass fuels, e.g., from algae feedstock) will play an increasingly important role in the future. However, at the same time, the economic feasibility of both cellulosic ethanol and algae-based fuels is still unfavorable for a large-scale market commercialization, especially in times of low prices of conventional fuels (i.e., gasoline). According to Colye (2010), production costs of cellulosic ethanol equal to \$2.65/gal, which is \$1 more than costs of corn ethanol (Fig. 6). High costs of

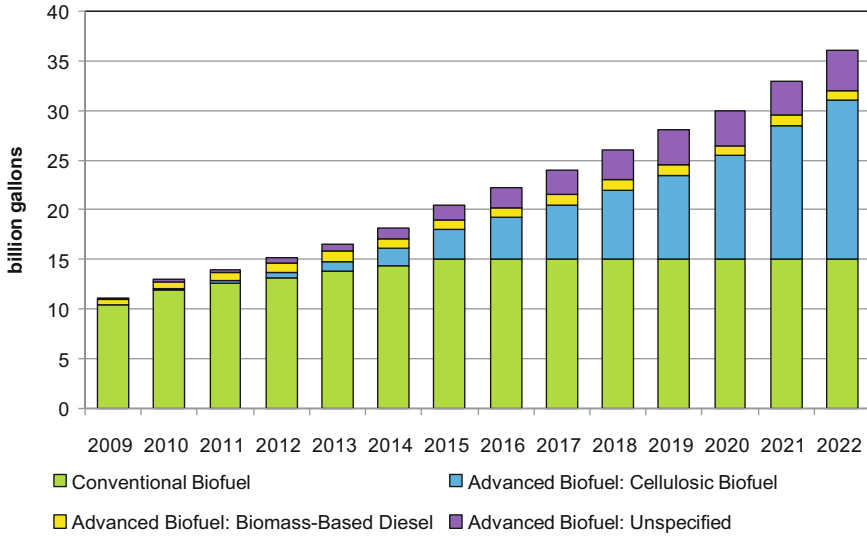


Fig. 5 Renewable Fuel Standard in the US (2009–2022). *Source* US EPA (2010)

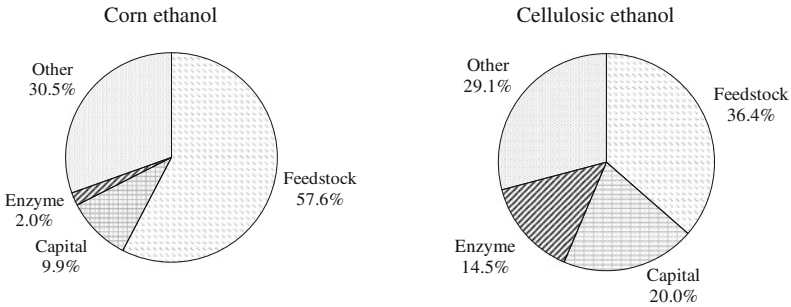


Fig. 6 Production costs of corn-based and cellulosic ethanol. *Source* Coyle (2010); Ziolkowska and Simon (2011)

enzymes needed to break down cellulose are among the main driving cost components in production of the cellulosic ethanol.

For algae-based fuels, economic feasibility presents even a bigger challenge. According to US Department of Energy (2008), the price for algae-based fuels produced on a large scale amounts to more than \$8/gal (compared to \$4/gal for soybean-based fuel). However, it needs to be mentioned that long-term cost trends over the past 30 years (and normalized to 2009 price values) indicate a decrease in production costs from \$6.09 in 1982 (Benemann et al. 1982) to \$2.41 in 1996 (Benemann and Oswald 1996; Gallagher 2011; Ziolkowska and Simon 2014).

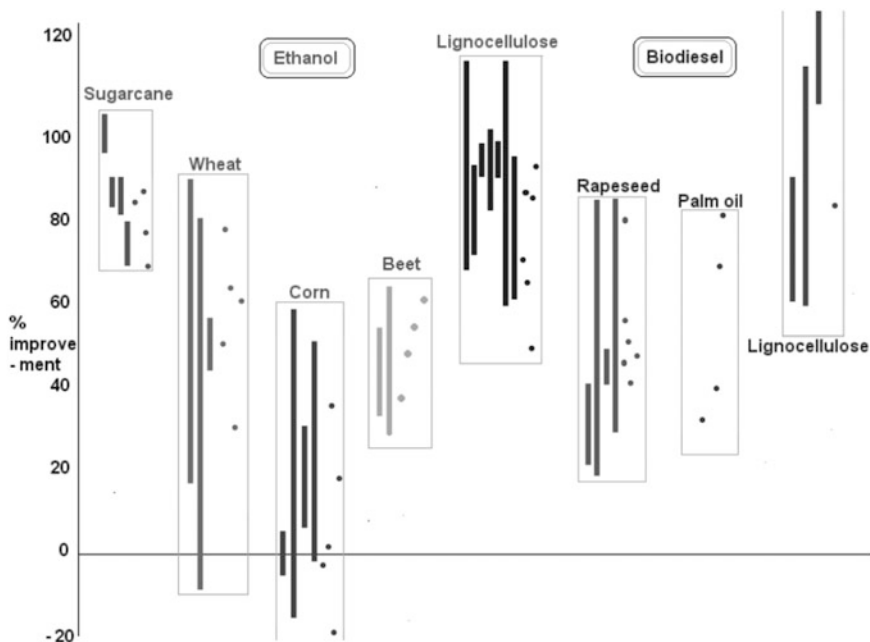


Fig. 7 Net life relative cycle GHG emission improvement of selected biofuels pathways as compared to gasoline and diesel fuels (without land use change). Legend: Bars and dots shown in the graph indicate the range and point estimates of improvements in net GHG emissions as elaborated from the data found in the reviewed studies. *Source* OECD (2008); Ziolkowska and Simon (2010)

Considering environmental concerns, the ecological footprint of cellulosic ethanol is significantly lower than of traditional gasoline or even conventional biofuels. According to OECD (2008), lignocellulosic ethanol can reduce GHG emissions in the range between 50 and 110% compared to gasoline and diesel fuels (w/o LUC—land use change), while lignocellulosic biodiesel can generate an environmental effect of 60% and more GHG reduction (Fig. 7). Also, algae-based fuels offer a number of environmental benefits that make them a desirable product on the biofuels market. Environmental footprint of algae has been described as negative (i.e., carbon neutral) as algae biomass requires 2 g of CO₂ for every g biomass generated (Pienkos and Darzins 2009), while one ton of CO₂ can be converted into 60–70 gallons of algae-based ethanol (Hon-Nami 2006; Hirayama et al. 1998). In addition, algae do not compete for freshwater as they can be grown by using waste/saline water, while they do not require productive land either (Ziolkowska and Simon 2014).

Given the economic challenges and environmental potentials of advanced (cellulosic and other biomass-based) biofuels, nanotechnology has been studied as a possible solution to reduce production costs and thus lower environmental footprint as described above. However, most recent studies indicate that nanotechnology can

also bear other health and environmental risks that have not been considered comprehensively in the discussion of biofuels yet. Section 3 will provide more insights into both new advances nanotechnology offers for biofuels production and potential short and long-term risks associated with its application.

3 Potentials and Risks of Nanotechnology for Biofuels Production

Scientific metric classification describes the “nano” unit as one billionth part of the factor of 1. Thus, for instance, 1 nanometer (nm) equals 10^{-9} m (0.000000001 m).

Nanotechnology (from the Greek word “nano”, i.e., dwarf) aims at manipulating materials at the atomic and molecular levels to create new molecular structures known as “nanomaterials”. Nanomaterials have unique and new characteristics that differ from characteristics of the original materials they are derived from, while their structured components are of at least one dimension less than 100 nm. As a comparison, the size of atoms is in the range of 0.1–1 nm, viruses are between 10 and 100 nm small, and bacteria between 1 and 10 micrometers (μ m) (=1,000–10,000 nm) (Warad and Dutta 2007). Thus, nanotechnology is able to “see” and control individual atoms and molecules (NNI 2016). It is important to emphasize that at the nanoparticle level, changes in electrical, chemical, magnetic, mechanical or biological properties of materials can occur that differentiate them from the bulk material, albeit with no change in chemical composition. Consequently, nanotechnology generates new (in many instances enhanced) characteristics related to, e.g., material flexibility, strength, conductivity, surface tension or color (Molins 2008).

Although nanotechnology has been successfully applied in many disciplines to solve complex environmental problems (e.g., oil spill cleanups) (Avila et al. 2014), this section will focus on nanotechnology in advanced biofuels production only.

The major question raised frequently is about ways and approaches how nanotechnology could be utilized to improve efficiency of biofuels production. According to Wegner and Jones (2009), nanotechnology can be applied in the following ways:

- (1) To manipulate nanoscale cell walls structures (also referred to as a nanofibril) within trees and plant materials to facilitate an easier disassemblment into constitutive materials for biofuels production (either through fermentation, gasification, or catalysis).
- (2) Through application of nanocatalysis to break down cellulose that makes 15–25% of the carbohydrate part of wooden materials.
- (3) Through application of engineered nanoscale enzymes or systems of enzymes (e.g., glycol hydrolases, expansins, and lignin-degrading enzymes) to improve conversion efficiency of cellulose into sugars. Also, in addition to the first approach, tree biology could be engineered for enzymes and enzyme systems to

be created and stored/sequestered in the living tree until harvest and then be activated for engineered woody biomass self-disassembly.

- (4) To create new symbiotic nanoscale biological systems which would work together to create ethanol or other biofuels.

In recent years, nanotechnology has been described as a technology of the future, research in this field has boomed to determine cons and pros of each of the abovementioned approaches, while public research funding and private investments in laboratory experiments increased considerably. This strong support for nanotechnology results from the many advantages this technology promises as well as the consecutive (currently yet unknown) advances as a spillover effect.

In regard to cellulosic ethanol, the mainstream of traditional material science with treating cellulose, hemicellulose, and lignin has been through breaking materials down to particles to regenerate them or create new materials. Enzymes have been applied to convert cellulose into simpler sugars that can further be fermented by bacteria to ethanol. Nanotechnology has been introduced as advancement to this traditional practice as it provides a potential to build materials through a designed arrangement of atoms into nanostructures of various types. This is possible as lignocellulosic biomass is made up of nanometer-size building block units that provide valuable properties to wood and other types of renewable lignocellulosic and cellulosic biomaterials. Accordingly, nanoparticles can immobilize beds of expensive enzymes that can be used over and over again to break down the long chain of cellulose polymers into simpler fermentable sugars for ethanol production (LTU 2009). Savings estimates range between \$32 million for each cellulosic ethanol plant and \$7.5 billion given that the RFS goal of 16 billion gallons of cellulosic ethanol is achieved (LTU 2009). For biodiesel production, nanocatalysts can be used for transesterification of fatty esters from vegetable oils or animal fats into biodiesel and glycerol (Lin et al. 2007).

Laboratory experiments triggered consecutive research and continuous development of nanotechnology. An increasing number of research studies unveiled new approaches for cellulose conversion (Munasinghe and Khanal 2010; Jiang et al. 2009), while economics, sustainability and renewable energies have been among driving research issues (Raman et al. 2015; Cacciatore et al. 2012; Serrano et al. 2009). While the number of research studies on nanotechnology for biofuels has increased over years (Li et al. 2016; Kizling et al. 2016; Babadi et al. 2016; Guo et al. 2012), many questions still remain open (Bhatia 2014; Guerin 2009).

In regard to algae-based fuels, research in nanotechnology has also raised scientific interest (Gavrilescu and Chisti 2005) to solve current challenges related to algal biomass. Those challenges have been identified by Pattarkine and Pattarkine (2012) as follows: (a) lack of consistent industrial-scale algae production, (b) high costs of algae harvesting and production, and (c) energy intensive lipid extraction. Nanotechnology could help with: (a) mitigating the existing limitations related to gas transfer, mixing, illumination, and biomass yield, (b) improving efficiency, lipid extraction and yield of algal biofuels (also through genetic engineering), and (c) improving harvesting technologies. As emphasized by Pattarkine and Pattarkine

(2012), application of silver nanoparticles for improved photoconversion, calcium oxide nanocrystals in transesterification, and mesoporous nanoparticles in biofuel separation would help with achieving those goals.

In addition, the new “nanofarming” technology can facilitate oil extraction from algae more efficiently as it supports a continuous process of “milking algae” for up to 70 days instead of destroying their cell and biomass structure as suggested by traditional material science (Vinayak et al. 2015; Chaudry et al. 2016; Ziolkowska and Simon 2014). Commercialization of this new technology has been discussed between the US Ames Laboratory and Catilin (a nanotechnology-based company specializing in biofuel production) with the aim to reduce costs and energy consumption of non-food source biofuels feedstocks. The pilot project of “nanofarming” has been funded by the US DOE (Office of Energy Efficiency and Renewable Energy Industrial Technology Program), Catilin company, and Iowa State University (Lin et al. 2009).

In summary, the application of nanotechnology in biofuels production has been evaluated as a promising approach to (1) reduce transportation costs of feedstocks, (2) break down the feedstock more efficiently, and (3) improve biofuels production efficiency, which would help lower prices of advanced biofuels. Despite those promising advantages, on the one hand, nanotechnology has raised concerns about potential short and long-term economic and environmental issues, on the other hand.

One of the major concerns is missing knowledge about potential impacts and side effects of nanotechnological modifications (Renn 2006), as well as effects and implications for living organisms (including ecosystems and humans), as many nanomaterials entering the environment might remain in it indefinitely (Colvin 2004). Once deposited on soils, nanoparticles can traverse several soil strata and leak into aquifers, while drinking water filtering systems might not be capable of filtering them out (Alargova and Tsujii 2001). Unlike nanoparticles originating as byproducts of combustion engines, manufactured nanoparticles do not agglomerate as much and thus could remain more reactive for longer periods of time. There is a risk that due to their very small size, they could enter the human bloodstream via the lungs after inhalation, the digestive tract, and the skin if applied or deposited on it. As consequences and impacts of nanoparticles in human body are unknown, more research is needed to be able to assess advantages and disadvantages of nanotechnology in general and for any kind of application, including biofuels production (Justo-Hanani and Dayan 2015; Hull and Bowman 2014). Also, policy regulations are needed that would establish a well-grounded oversight and monitoring procedure (Colvin 2003; Molins 2008).

In addition to traditional examples and applications of nanotechnology in biofuels production as described above, also experiments have been undertaken to explore utilization of bacteria and plant enzymes to break down cellulose and lignin (Ziolkowska 2014b). For instance, the US Department of Energy (DOE), the BioEnergy Science Center, and the University of California researchers developed the *Clostridium celluloyticum* bacteria capable of breaking down cellulose and enabling the production of isobutanol in one inexpensive step (Casey 2012a, b).

In addition, DOE also found engineered strains of the *Escherichia coli* bacteria to be able to break down cellulose and hemicellulose contained in plant cell walls, e.g., switchgrass (Ziolkowska 2014b). Also, a method has been developed at the University of Central Florida to break down cellulose and refine ethanol from orange peels by means of a tobacco enzyme. The tobacco enzyme is derived by cloning genes from fungi and bacteria. This process was found to be considerably less expensive than using synthetic enzymes (Casey 2012a, b). Combining nanotechnology with those processes might yield even higher economic benefits by limiting carbon footprint of biofuels production. However, environmental and health concerns remain and will require basic research and potentially policy regulations in the years to come.

4 Conclusions and Outlook

Biofuels technology has experienced a considerable progress in the past decades, expanding its scope from conventional to advanced feedstocks with lower environmental footprint and higher energy efficiency. This process revealed several key challenges, especially for advanced biofuels: cellulosic ethanol and algae-based fuels that face economic challenges related to breaking down cellulose and lignin, and extracting lipids, respectively.

Nanotechnology can potentially provide solutions to some of those challenges that have hindered and delayed commercialization of advanced fuels at a large scale. It can help with reducing transportation costs of feedstocks, breaking down the feedstock, extracting oils more efficiently, and improving biofuels yields and production efficiency, which could ultimately help with reducing biofuels prices. Even though nanotechnology provides promising potentials, uncertainties exist among scientists and regulatory agencies about the safety of nanotechnology both for humans and the environment, as some nanomaterials can be toxic. As nanotechnology is not regulated yet, and no rules exist for its application, there is a valid question of unknown potential long-term environmental and social impacts that could theoretically be irreversible.

Furthermore, as nanotechnology is a new approach, more research is necessary to discover and understand its potentials, also for biofuels applications, while minimizing any potential risks to the environment and humans. Governmental and environmental regulations are needed for companies producing nanomaterials as well as a clear monitoring and enforcement system. Also, the potential combination of new technologies (nanotechnology and genetic engineering) to reduce production costs of advanced biofuels might bring new developments and changes to the biofuel market in the long term.

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