A Technological Overview of Biogas Production from Biowaste
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Abstract

The current irrational use of fossil fuels and the impact of greenhouse gases on the environment are driving research into renewable energy production from organic resources and waste. The global energy demand is high, and most of this energy is produced from fossil resources. Recent studies report that anaerobic digestion (AD) is an efficient alternative technology that combines biofuel production with sustainable waste management, and various technological trends exist in the biogas industry that enhance the production and quality of biogas. Further investments in AD are expected to meet with increasing success due to the low cost of available feedstocks and the wide range of uses for biogas (i.e., for heating, electricity, and fuel). Biogas production is growing in the European energy market and offers an economical alternative for bioenergy production. The objective of this work is to provide an overview of biogas production from lignocellulosic waste, thus providing information toward crucial issues in the biogas economy.

Keywords:
Anaerobic digestion
Biogas
Sustainable energy
Lignocellulosic waste
Microbial ecology

1. Introduction

The continuing use of fossil fuels and the effect of greenhouse gases (GHGs) on the environment have initiated research efforts into the production of alternative fuels from bioresources. The amount of GHG emissions in the atmosphere is rising, with carbon dioxide (CO2) being the main contributor. In addition, the global energy demand is increasing rapidly, with approximately 88% of the energy produced at the present time being based on fossil fuels [1,2].

Moreover, the security of the energy supply is a crucial challenge because most natural energy resources (i.e., oil and gas reserves) are found in politically unstable regions. In this context, biogas from waste and residues can play a critical role in the energy future. Biogas is a multilateral renewable energy source that can replace conventional fuels to produce heat and power; it can also be used as gaseous fuel in automotive applications. Biomethane (upgraded biogas) can also substitute for natural gas in chemicals production. Recent evaluations indicate that biogas produced via anaerobic digestion (AD) provides significant advantages over other forms of bioenergy because AD is an energy-efficient and environmentally friendly technology [3,4].

In comparison with fossil fuels, AD technology can reduce GHG emissions by utilizing locally available sources. In addition, the by-product of this technology, called digestate, is a high-value fertilizer for crop cultivation and can replace common mineral fertilizers. In Europe, the production of biogas reached $1.35 \times 10^7$ t in 2014 [5]. Germany is the pioneer country in global biogas production, with approximately 25% installed capacity due to the strong development of agricultural biogas plants on farms. At the end of 2014, more than 8000 agricultural biogas production units were in operation in Germany [6]. Several countries have already become involved in the development of new pathways for biogas production from biomass and biowaste. Many European countries have established favorable conditions for electricity production from biogas. It is remarkable to note that the agro-biomass available for AD is as high as $1.5 \times 10^9$ t in Europe [7].

The United States, China, and India are also investing in alternative technologies for biogas production from cellulosic resources, and are likely future producers [8,9]. Although biogas (and/or biomethane) based on waste is a promising substitution for, or contribution...
to, the natural gas network, the amount produced is limited in comparison with the annual global consumption. There is no clear answer regarding which feedstock is most appropriate for the biogas economy. In general, carbohydrates, fats, and proteins can be used in many applications. The necessity for global sustainable waste management has led to research interest in alternative fuels based on agro-waste and biowaste [10,11]. This study discusses recent trends in biogas production and provides a summary of the current problems and barriers affecting different biogas production pathways. It also analyzes potential issues and trends in biotechnological conversion performance.

2. Current status of biogas production in Europe

Although the advantages of biogas as an alternative fuel have been reported since the 19th century, the current rekindling of interest in biogas production—and hence in methane capture via upgrading—is due to the depletion of natural gas reserves and the increase in GHG emissions [12]. At the beginning of the 20th century, the high value of fertilizer (i.e., compost) produced from waste enhanced AD technology and favored the biogas economy [13]. Moreover, Europe was prompt in applying sustainable waste management and simultaneously becoming independent from foreign oil-providing countries to a high degree. Toward this end, European bodies implemented new research programs to support an alternative-fuels future based on renewable resources. Biogas technology has been widely used in Europe for several decades, and biogas production has grown from approximately 7934 T onne of oil equivalent in 2009 to 14 120 toe (1.6548 × 10^10 L) in 2016, as can be seen in Table 1 [14].

There has been a significant effort in Europe to encourage industrial activities to manufacture fuels from biomass and biowaste by adjusting tax exemptions and encouraging biogas research and development programs. According to the European Biogas Association (EBA), Germany is the leading biogas producer in Europe, with more than 8000 biogas plants currently in operation, and its biogas amount corresponds to an approximate total electricity capacity of 4 TW·h (in Table 2 [13,15,16], the top five European biogas producers are given) [6]. In 2010, it was reported that despite the global economic burden, biogas production continued to expand rapidly and converge significantly to the economic development of rural communities in Germany [7,17].

However, the performance of waste conversion into gaseous fuels still remains a crucial issue, which has caused research initiatives to focus on easily accessible resources such as agro-industrial waste.

3. Waste for biogas production

3.1. Feedstock types and characteristics

A wide range of waste types can be used as substrates for biogas production using AD technology. Large quantities of lignocellulosic waste are collected from agriculture, industrial, municipal, and other activities. The most typical forms of waste used in the European energy industry are: (i) animal manure and slurry, (ii) sewage sludge, (iii) municipal solid waste, and (iv) food waste. Table 3 [18,19] compares the production amount and energy potential for the different feedstocks that can be utilized for biogas production.

Biomass contains carbohydrates, proteins, fats, cellulose, and hemicellulose, which can be used as feedstocks for biogas production. In current practice, co-substrates are usually added to increase the organic content and thus achieve a higher gas yield. Typical co-substrates include organic wastes from agriculture-related industries, food waste, and/or collected municipal biowaste from households. The composition and yield of biogas depend on the feedstock and co-substrate type. The typical feedstocks in the biogas plants in Germany are given in Fig. 1 [20]. Even though carbohydrates and proteins show faster conversion rates than fats, it is reported that the latter provide a higher biogas yield [21–24]. To avoid process failures, feedstock pretreatment is necessary. The application of pretreatment methods enhances the degradation of substrates and therefore the process efficiency. Chemical, thermal, mechanical, or enzymatic processes can be applied to speed up the decomposition process, although this does not necessarily result in a higher biogas yield [25,26].

3.2. Lignocellulosic molecular constituents

Cellulosic waste such as energy crops, agricultural residues, and sewage sludge have great potential for biofuel production. As Fig. 2 [27] shows, lignocellulose consists of three main organic components: cellulose, hemicel lulose, and lignin [28,29].

Cellulose is a primary structural component that is related to the mechanical strength of plant cell walls, while hemicellulose macromolecules are synthesized by repeating the polymers of pentoses
and hexoses. Lignin contains three aromatic alcohols (coniferyl alcohol, sinapyl alcohol, and p-coumaryl alcohol) that are produced through a biosynthetic process [30–32]. The composition of lignocellulose varies highly among different sources as it depends on diverse conditions such as material, origin, and season [33–35].

Cellulose is a linear polymer that is linked by several β-1,4-glycosidic bonds (Fig. 2) [36]. Its structure contains parts with a crystalline structure and parts with an amorphous arrangement [37].

According to Deguchi et al. [38], crystalline cellulose can be converted into cellulose with a non-organized structure by applying a temperature of 320 °C and a pressure of 25 MPa. Cellulose is the most plentiful organic compound on earth and makes up over 25% of plant biomass [39]. Lignocellulose is a complex and changeable structure that consists of different polymers such as pentoses (xylose, arabinose), hexoses (mannose, glucose, and galactose), and sugar/uronic acids (glucuronic, galacturonic, and methylgalacturonic acid). Its molecular structure is given in Fig. 4 [36]. The dominant compound in the hemicellulose arrangement is xylan (up to 90%), although the composition varies depending on the origin of the feedstock. Recent studies indicate that hemicellulose requires a wide variety of enzymes to be fully hydrolyzed into free monomers [40–44].

Hemicellulose has a low molecular weight and short lateral chains, and its structure consists of numerous sugars in polymers that are easily hydrolyzed [37]. Hemicellulose forms a linkage between lignin and cellulose molecules, thus increasing the compactness of the entire cellulose-hemicellulose-lignin network [40]. The solubility of the different hemicellulose compounds is directly related to temperature. The solubility of higher molecular polymers cannot be predicted because of unknown melting points [41,45]. Bobleter [46] reports that hemicellulosic compounds start to dissolve in water at 180 °C in a neutral environment. Garrote et al. [47] report that parts of hemicellulose are dissolved at 150 °C. It is important to note that this solubilization is connected to various parameters such as temperature (thermal-chemical sensitivity), pH (i.e., acid or alkaline environment), and moisture content [48–50].

Lignin is a naturally occurring heteropolymer of the cell wall. Its structure is complex as it consists of three phenylpropane-based units (p-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol) that are held together by linkages [51]. The structure of lignin provides resistance to microbial attack and oxidative stress. The difficult solubility of lignin in water results in low degradability [28]. Bobleter [46] reports that lignin dissolves in water at 180 °C in a neutral environment, just like hemicellulose. The solubility of lignin in acidic, neutral, or alkaline environments is related to which phenylpropane-based unit is present in the lignin [52]. Lignin is a significant component of wood (making up 30%–60%); its structure is shown in Fig. 5 [36]. Agricultural residues and grasses contain 5%–30% lignin, whereas crop residues are mainly composed of hemicellulose [53].

Recent studies report that lignin characteristics such as composition and structure can positively affect the hydrolysis process and thus increase biogas production efficiency [54]. Grabber [52] reports that a higher lignin content in biomass leads to a lower degradation efficiency.

4.1. Overcoming the recalcitrance barrier

Lignocellulose degradation requires the use of enzymes for...
hydrolyzation. Although lignocellulosic waste is a promising feedstock for biogas production, the complex structure of lignocellulose creates an economic and technical barrier for the operation of bio-refineries. The constituents of lignocellulose (cellulose, hemicellulose, and lignin) enhance the linkages between molecules, resulting in a compact and strong structure [55]. Recent studies report that the bioprocess efficiency of lignocellulose is related to pretreatment performance. Pretreatment technologies mainly aim to make AD faster, increase the biogas yield (Fig. 6), and provide a wide range of new and/or locally available substrates for use. Table 4 presents the advantages and disadvantages of different pretreatment technologies; a more extensive analysis of a variety of pretreatment processes is given in the literature [56–72]. The choice of treatment process is very important because each material has different characteristics and requires specific treatment.

Developments in pretreatments aim to enhance the product yields from lignocellulosic feedstocks and lower the methane emissions to the atmosphere, thus positively contributing toward environmental protection. The effect of pretreatment techniques on AD has only recently been investigated, and it is still necessary to optimize these techniques in terms of efficiency, cost, and application range. Further research will likely focus on whole-process engineering, in which pretreatment is integrated into the digester, rather than viewing pretreatment as a separate process.

Pretreatment must overcome the structural barriers of lignocellulose and its polymers (cellulose and hemicellulose) by subjecting them to microbial breakdown activities, resulting in enhanced biomass degradation and increased biogas yield [73]. Pecorini et al. [74] report that autoclaving and microwaving result in the hydrolysis of a significant fraction of non-biodegradable substances in municipal waste that is recalcitrant to AD. Micolucci et al. [75] applied a pilot-scale pressing system to pretreat biowaste, which resulted in higher biogas yields. Ideal biomass pretreatment aims to make the substrate more accessible to microorganisms by completely or partially decomposing the feedstock into fermentable sugars, thus eliminating the lignin resistance and decreasing the crystalline structure of the cellulose. The problem of recalcitrance has not yet been solved; further research is required to investigate new genetic engineering approaches to solve problems related to degradation and thereby succeed in achieving higher efficiency [76].

4.2. Multiple-stage and high-pressure AD

A considerable number of research projects have been developed to evaluate different configurations (e.g., single- or multiple-stage reactors) in order to enhance the efficiency of AD. Recent studies report that the separation of the AD process into two stages, such that hydrolysis/acidogenesis and acetogenesis/methanation are carried out in separate reactors (Fig. 7) [77], can increase the conversion rate of organic material to methane; however, the cost of such a complex system is a significant drawback [78].

The application of multiple bioreactor systems generally has a specific goal, such as improved process stability and higher efficiency. A multiple-stage bioreactor system permits different conditions (e.g., organic loading rate and temperature) to be applied. At present, few multiple-stage AD units operate to produce biogas fuel on a commercial scale. The complexity and high cost of this multiple-stage technology are barriers to commercial use [79,80]. Colussi et al. [81] investigated the two-stage AD of maize, which resulted in higher chemical oxygen demand removal efficiency and a higher yield of biogas. Marín Pérez and Weber [82] report that the physical separation of AD into two stages permits the adoption of different process conditions for specific species of bacteria; as a result, hydrolysis (which is the main rate-limiting step) can be accelerated, leading to faster degradation of organic matter. Yabu et al. [83] investigated the two-stage AD of garbage combined with ammonia stripping to prevent ammonia inhibition. Park et al. [84] compared the single- and two-phase AD of kitchen garbage and found that the two-phase AD resulted in a higher yield of methane.

![Fig. 6. Pretreatment can increase the rate of AD (Case b) or increase the methane yield (Case c).](image)

### Table 4

<table>
<thead>
<tr>
<th>Technology</th>
<th>Advantages</th>
<th>Disadvantages</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Milling</td>
<td>• No production of inhibitors (e.g., furfural and HMF)</td>
<td>• High energy requirements</td>
<td>[56–64]</td>
</tr>
<tr>
<td></td>
<td>• Increased methane (5%–25%)</td>
<td>• High maintenance cost</td>
<td></td>
</tr>
<tr>
<td>Extrusion</td>
<td>• Increased surface area</td>
<td>• Increased energy demand</td>
<td>[56–63]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>• High maintenance cost</td>
<td></td>
</tr>
<tr>
<td>Steam pretreatment</td>
<td>• Increased cellulose fiber reactivity</td>
<td>• Risk of producing inhibitors (e.g., furfural and HMF)</td>
<td>[64–66]</td>
</tr>
<tr>
<td>Steam explosion</td>
<td></td>
<td>• Less digestible biomass because of lignin condensation</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>• Precipitation phenomena</td>
<td></td>
</tr>
<tr>
<td>Liquid hot water</td>
<td>• Solubilized hemicellulose and lignin products are present in lower</td>
<td>• High heat demand</td>
<td>[67,68]</td>
</tr>
<tr>
<td></td>
<td>concentrations</td>
<td>• Only effective up to a certain temperature</td>
<td></td>
</tr>
<tr>
<td>Microwave</td>
<td>• 4%–7% more biogas produced than untreated</td>
<td></td>
<td>[69]</td>
</tr>
<tr>
<td>Diluted or strong</td>
<td>• Solubilizes hemicellulose</td>
<td>• High cost of acids</td>
<td>[70,71]</td>
</tr>
<tr>
<td>acid pretreatment</td>
<td></td>
<td>• Risk of forming inhibiting compounds</td>
<td></td>
</tr>
<tr>
<td>Alkaline pretreatment</td>
<td>• Hemicellulose and parts of lignin are solubilized</td>
<td>• Corrosion problems</td>
<td></td>
</tr>
<tr>
<td></td>
<td>• Increased methane production</td>
<td>• Risk of producing inhibitors</td>
<td>[72]</td>
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HMF: hydroxymethylfurfural.
A two-stage AD process can be applied to a variety of waste that reaches high microbiological activity. Blonskaja et al. [85] used a two-stage system to process distillery waste and observed a higher growth rate of methanogenic populations, resulting in increased gas generation. Kim et al. [86] studied a four-stage AD system using activated sludge, which showed significantly higher digestion efficiency than a single-stage system. In addition, recent studies have indicated that multistage AD processes have a higher hydrogen yield. Nasr et al. [87] evaluated bio-hydrogen production from thin stillage and concluded that a two-stage system enhances the performance of the AD process.

An alternative technique has been developed that is based on a high working pressure (up to 100 bar, 1 bar = 100 kPa); with this technique, the production of biogas with more than 95% methane content is feasible. The aim of the technique is to integrate biogas production and in situ increased-pressure purification into a single process in order to produce clean biogas (99% methane) that can be fed directly into the natural gas networks.

Lindeboom et al. [88] report that pressure of up to 20 bar can increase methane yield and that in situ upgrading is successful with a high-pressure autogenerative method. They report that the biogas produced by this method can contain less than 5% CO2, because more CO2 dissolves in the water under high pressure. Previous work has shown that working pressures of up to 90 bar can provide methane-enriched biogas because pressure can influence the microbial processes [89,90]. Merkle et al. [91] studied AD at up to 100 bar using grass and maize silage hydrolysate as the substrate. Their results showed a significantly high methane yield; however, more research is required to determine the pressure dependence of the microbial processes. Nevertheless, the use of multistage and high-pressure approaches can promote and accelerate the future use of lignocellulosic feedstocks for biogas production.

4.3. Microbial ecology: Microbiological dynamics

The conversion to methane of most waste products, such as pentoses, hexoses, volatile products, and soluble lignin, is feasible by using a mixture of microorganisms, which is a way of improving AD [92]. During hydrolysis, the first step of the process, extracellular enzymes produced by hydrolytic microbes decompose complex organic matter into simple soluble molecules. Carbohydrates, fats, and proteins are hydrolyzed into sugars, fatty acids, and amino acids, respectively [93]. These compounds, which have smaller carbon chains, are then converted into a mixture of volatile fatty acids (VFAs) and other minor products, such as alcohols, by acidogenic bacteria (acidogens). Acetogenic bacteria (acetogens) further convert the VFAs into acetic acid (acetate), CO2, and hydrogen, which are important substrates for biogas production. The last step is methanogenesis, in which methanogens produce biogas.

The dynamics of the different microbial groups are complex and interactive. The quantities of the microbial groups are disproportional and influence the overall process reaction rate [94]. It is reported that among the four microbial groups involved in AD, methanogens have the slowest growth rate and are the most sensitive to changes in process conditions such as temperature, pH, redox, and inhibitors. Hence, methanogenesis is the key pathway for biogas production and is commonly considered to be the rate-limiting step of the whole process [95]. Recent studies report that one strategy to improve the process economics is the optimization of the metabolic pathways in order to genetically modify the metabolic efficiency of the microbes. The investigation of different metabolic pathways has led to energy-rich biofuels [96]. In addition, an alternative strategy that is commonly used in bioethanol production is metabolic redirection, which blocks undesirable metabolic pathways and redirects the metabolism targets of bacteria [97].

Little is known about the different types of microbes that are responsible for the metabolic activities in the AD process. Low percentages of bacteria and archaea have been isolated so far, but little information is available about the dynamics and interactions between these microorganisms. This lack of knowledge results in “sour” digesters due to malfunctions and unexplainable failures. Research initiatives currently focus on investigating the structures of microbe communities in AD using molecular techniques [98–102].

5. Recent issues in biogas production

5.1. The gap between biotech research and commercialization

Large-scale lignocellulose-to-biogas production has significant potential, and research efforts toward its further development have already been carried out. These processes typically have technical problems that stem from a poor understanding of optimal reactor operation. The complexity of AD and the risk that is involved in investment in new technologies are two of the major constraints affecting AD improvement.

The goal of R&D departments in this field is to mature AD technology in order to facilitate the implementation of biomethane in the transportation fuel markets. The key to identifying the biotechnology and research gap (Fig. 8) lies in understanding the science and technology and evaluating the impacts of important technical, economical, and ecological barriers. Benefits and costs must be analyzed. For example, for cost reduction, it is necessary to identify the critical technological steps (e.g., the cost of multiple-stage AD application or the use of enzymes) that have the greatest effect on the overall economics. The analysis of such steps will provide essential information for evaluating research priorities for development [103].

The type and amount of microorganisms and/or biocatalysts that are selected for the degradation of organic waste affect the conversion rates and process stability. If the production cost is very high, the biogas production cost is increased. Companies aim to develop enzymes with a wider range of applications and better activity performance during enzymatic hydrolysis. Thus, recent research initiatives focus on the development of microorganisms and/or biocatalysts with a wide range of applications, better characteristics, and low production cost [104,105]. AD technology also requires utilities such as electrical power and heat. The optimal application of utilities is an engineering issue that can be improved in pilot facilities and that can shift process efficiency. In addition, the conversion of lignocellulosic waste into biogas can be combined with fertilizer production, which improves market competitiveness through by-
product (i.e., digestate) revenues. Recent studies focus on combining processing technologies such as multiple-stage or high-pressure technologies [106].

The production of biogas includes technical and economic parameters such as microorganism species, pretreatment and purification technologies, substrate properties, and optimal reactor conditions. Optimizing the combination of these parameters is the key to cost-effective biogas production. Research can play a catalytic role in filling the gap between engineering and biology/biotechnology (Table 5) in order to provide innovative and sustainable technological alternatives for the biogas sector [107,108].

5.2. The future of biogas in a circular/green economy

The biogas economy is related to factors such as waste availability and logistics, process efficiency, and end-product properties. AD technology has been demonstrated, and has robust commercial availability. There is a wide variety of lignocellulosic waste with low cost and high availability that can be treated for biogas production. Another important issue in a green economy is the registration of cellulosic gaseous fuels (i.e., biomethane) for sale and use under the renewable fuel standard. Industry stakeholders have predicted that a combination of federal programs for research funds and private industrial investments could accelerate the introduction of these fuels to the market at a competitive cost. However, biogas-based engines are not yet developed enough to deal with the technical issues of biogas use; thus, the need to modify engines for biogas combustion must be taken into account [108].

Biogas is a key player in the European bio-based economy because it provides strategic perspectives for global producers, especially when the price of oil is reduced. The EU’s Renewable Energy Directive calls for a 10% increase in the use of green vehicle fuels by 2020. European policy is aiming to establish environmental sustainability criteria for green gaseous fuels, and European countries are encouraged to invest in biogas installations [109].

According to the recently published EBA Biogas Report, there are already more than 15,000 biogas plants in Europe (Fig. 9) [110], and this number is continuing to grow. Table 6 [110] shows the number of biogas plants in the main European biogas-producing countries.

During the last decade, the biogas sector has grown within Europe, driven by different parameters such as the feed-in tariffs in Germany, the obligation certification for energy renewability in the UK, and the tax policy (i.e., economic exemptions) in Sweden [111]. A high share of the electric power in Germany comes from biogas as a result of governmental initiatives promoting power generation from wastes. Most biogas production is currently based on sewage sludge; however, it is estimated that by 2030, an increasing amount of biogas (about 224 TW-h) will be produced from wet manure, landfill, undigested sewage sludge, and food-processing residues [112].

6. Conclusion

Investments in AD are expected to succeed due to the low cost of available feedstocks and the wide range of uses for biogas (i.e., for heating, electricity, and fuel). Many lignocellulosic sources such as manure, fruit, and vegetable wastes can be used for biogas produc-

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Table 5

<table>
<thead>
<tr>
<th>Issues</th>
<th>Focus of R&amp;D efforts</th>
</tr>
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| Use of enzymes, bacteria, or catalysts | • Increased range of applications  
• High production cost                                      |
| Utility requirements          | • Consumption of electrical power  
• Surplus of oxygen and hydrogen  
• High pressure and heat                                    |
| Technology                    | • Pretreatment  
• Multiple-stage technology  
• Advanced techniques (high pressure)  
• Microscale technology                                         |
| Fuel properties               | • Enriched-methane biogas  
• Less hydrogen sulfide                                         |

Table 6

<table>
<thead>
<tr>
<th>Country</th>
<th>Number of biogas plants</th>
</tr>
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<tbody>
<tr>
<td>Germany</td>
<td>~8000</td>
</tr>
<tr>
<td>Italy</td>
<td>1491</td>
</tr>
<tr>
<td>UK</td>
<td>813</td>
</tr>
<tr>
<td>France</td>
<td>736</td>
</tr>
<tr>
<td>Switzerland</td>
<td>633</td>
</tr>
</tbody>
</table>
tion, and AD can be applied on a small or large scale. This flexibility allows the production of biogas anywhere in the world. Current research initiatives aim to improve AD control, and thus its efficiency. Microbial activity during AD is a crucial parameter for process stability and biogas yield, and thus requires further investigation. Biogas production is growing in the European energy market; in a few decades, it will offer an economical alternative for the production of bioenergy.

**Compliance with ethics guidelines**

Spyridon Achinas, Vasileios Achinas, and Gerrit Jan Willem Euverink declare that they have no conflict of interest or financial conflicts to disclose.

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