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Lignocellulosic and microalgal biofuels are poised to play a significant role in the future energy matrix. This article reviews recent developments related to lignocellulosic and microalgal biofuels from a chemical engineering perspective. The major challenges in the production of these biofuels are identified and potential solutions are discussed. For lignocellulosic biofuels, development of efficient pre-treatment methods and improved enzymes is a key challenge. For microalgal biofuels, increasing culture density and reducing dewatering requirement require immediate efforts. The issues associated with practical implementation and scale-up as well as the value of an integrated biorefinery are discussed. Finally, sustainability related challenges are presented and the value of a systems approach is highlighted.

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Introduction

In the drive towards developing sustainable sources of energy, biomass based energy holds a distinct advantage. It can produce liquid fuels as direct substitutes for gasoline and diesel. Moreover, biomass availability is relatively stable throughout the year [1,2]. Additionally, biomass can be converted to heat, power, and other low volume, high value products and chemicals [3••,4]. The biomass based energy can also potentially be carbon neutral and can be produced locally, thereby meeting the key sustainability criteria.

The first generation biofuels are produced from food based sources such as corn, soybean, sugarcane, and palm oil. Although they are techno-economically feasible, they are controversial due to the potential conflict with food sources [5]. The fuels from non-food lignocellulosic sources such as agricultural residue, dedicated energy grasses, forestry residue, and short rotation woody biomass constitute the second generation of biofuels. They are expected to satisfy a significant fraction of the total future energy demand [6]. The third generation biofuels are those produced from micro and macro algae [7•]. They can be produced on marginal and degraded land as well as in coastal regions using saline water. Additionally, microalgae cultivation can be used to treat waste water and sequester carbon from flue gases [8]. Therefore, there is considerable excitement towards third generation biofuels in land starved countries such as India and China.

Substantial research is currently being undertaken globally to make the second and third generation biofuels feasible. This article reviews some of the major research activities over the last decade in the field of chemical engineering. Considering the vast amount of literature available, this review is certainly not exhaustive. Instead, it reports certain key developments in the field. Moreover, critical aspects that are often ignored by scientists are also highlighted.

Lignocellulosic biofuels

Lignocellulosic biomass consists mainly of lignin, cellulose, and hemicellulose. The lignocellulosic biomass can be converted to fuels and value added products via two different routes, biochemical and thermochemical [9]. The biochemical route is mainly used to produce ethanol or butanol, and is typically more feasible at smaller scales. The thermochemical route, which includes options such as gasification, pyrolysis or torrefaction, can produce liquid and gaseous fuels as well as power. Both the routes can also be used to produce chemical building blocks in addition to fuels [3••].

For biochemical processing, pre-treatment and enzymatic hydrolysis are the key steps in the overall economic feasibility of the process [10–12]. Bhutto *et al.* [13••] have recently provided an excellent summary of various pre-treatment methods and their applicability to different feedstock based on process chemistry as well as experience at existing plants. The pretreatment performance depends on feedstock type and form. Different pre-treatment methods are often compared based on cost, energy requirement, cellulose and hemicellulose recovery fraction, degradation product formation, and washing requirement. For example, ionic liquid pre-treatment is expensive but does not produce degradation products. Acid pre-treatment, in contrast, is relatively cheaper but suffers from degradation product formation. Similar trade-offs exist for other pre-treatment methods [14,15]. Enzymatic hydrolysis is a slow process carried out under mild conditions to reduce the formation of fermentation inhibitors. It is sensitive to several factors, including solid loading, inhibitor concentration, product inhibition, and temperature [16•]. The major research focus is currently on developing effective and cheaper enzymes through genetic engineering and strain optimization [17]. Single pot hydrolysis and fermentation, where both C-5 and C-6 sugars are fermented, is the ideal scenario being targeted.

The experimental studies are often conducted using ideal feedstock, such as finely ground biomass for pretreatment or pure cellulose for hydrolysis. However, size reduction below a certain level can be highly energy and cost intensive [18•,19•]. Recently, DeMartini *et al.* [20] studied the impact of particle size on steam pre-treatment of poplar wood, and concluded that rapid decompression treatments such as steam explosion work better for larger particle sizes. Khullar *et al.* [21] also studied the impact of particle size on hydrolysis efficiency. Greater focus on understanding this aspect is required in the near future. Seasonal and distributed availability and low bulk density make feedstock procurement challenging, thereby affecting the scale and design of the biorefinery. Some recent studies have provided insights into these supply chain aspects [22–25]. The impact of different storage methods on the processing efficiencies has also been studied [26–30].

For thermochemical processing route, one of the key challenges is to understand the process chemistry and kinetics [31•]. Gasification and pyrolysis give a range of products through several parallel reactions at the temperatures and pressures employed for these processes [3••,32]. Improving the selectivity and stability of the oil [33], reducing tar formation [34], and making these processes energy efficient are some of the major challenges. Development of novel catalysts to achieve these goals is a very active area of research [35].

Microalgal biofuels

The production of fuels and value added products from microalgae is economically non-profitable [7•]. The environmental benefits of algal biofuels as compared to fossil fuels vary based on the region, source of energy, and impact category [36,37••]. Wijffels and Barbosa [7•] proposed that the cost of algal biofuel must reduce by a factor of 10 and its scale of production must increase by three orders of magnitude to make it cost-competitive with other energy sources.

One of the important challenges is increasing the concentration of the growth culture [38]. The productivity of microalgae in open pond cultivation is often limited to 10–20 g/m^2 day [39], which means that a biorefinery producing 30 tonnes per day of biodiesel would require about 30 000 ha of open pond cultivation [40]. One approach to address this limitation is to develop better strains using genetic engineering tools to achieve higher yield, better light tolerance, or higher percentage of the desired component (e.g. lipids). On the other hand, photobioreactors (PBRs) that provide higher productivity of up to 20–45 g/m^2 day [39] can be used. However, PBRs are costlier and energy intensive [38]. Consequently, novel photobioreactors that reduce cost and provide higher productivity have been developed [41,42]. These designs aim to improve light availability, avoid dead zones, and reduce energy requirement and cost of construction. After the design has been developed, computational fluid dynamics (CFD) modelling has often been used for benchmarking and design optimization of PBRs [43–46]. Bigot *et al.* [47•] provided a comprehensive review of the various CFD studies reported in the literature. Incorporating microalgal growth kinetics into the hydrodynamic CFD simulations is one of the challenges that require more research.

Harvesting and drying of the microalgal culture and removal of large quantities of water contributes up to 20–30% of net biomass production cost [48]. Therefore, developing novel alternatives for wet-processing of microalgae that can work with high water content are very attractive. Hydrothermal liquefaction (HTL), which can be carried out with up to 80% water content in the biomass, is one of the promising alternatives [49••,50,51]. In HTL, wet microalgal slurry is subjected to sub-critical temperatures (200–350 °C), where water acts as a solvent as well as a catalyst. The HTL process yields biocrude as the primary product, which can be processed and upgraded to produce refined transportation fuel. Although HTL reduces water removal requirement, the process itself is very energy intensive. Energy requirement can be reduced using heat integration [51]. Additionally, since the process is conducted under extreme conditions, the material of construction of the reactor needs to be corrosion resistant, which further increases the costs. Therefore, this option is currently economically infeasible [52].

Challenges and opportunities

This section discusses some of the key challenges and opportunities that are common to both second and third generation biofuels.

Integrated biorefinery

Integrated biorefineries (Figure 1) that produce biofuels as well as other value added products may be required to achieve economic feasibility [53]. Such a biorefinery will produce multiple fuels such as ethanol, biodiesel, and methane, as well as food supplements, specialty chemicals, chemical building blocks, fine chemicals of medicinal importance (e.g., omega-3 fatty acids, chlorophyll), and livestock feed [53]. For both lignocellulosic and microalgal systems, the potential value added chemicals have been identified in literature [4,54–56]. Efficient processes for producing these chemicals need to be developed, and the impact of co-production on individual process yields needs to be quantified.