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Review of US and EU initiatives toward development, demonstration, and commercialization of lignocellulosic biofuels

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Abstract: Advanced biofuels produced from lignocellulosic biomass offer an exciting opportunity to produce renewable liquid transportation fuels, biochemicals, and electricity from locally available agriculture and forest residues. The growing interest in biofuels from lignocellulosic feedstock in the United States (US) and the European Union (EU) can provide a path forward toward replacing petroleum-based fuels with sustainable biofuels which have the potential to lower greenhouse gas (GHG) emissions. The selection of biomass conversion technologies along with feedstock development plays a crucial role in the commercialization of next-generation biofuels. There has been synergy and, even with similar basic process routes, diversity in the conversion technologies chosen for commercialization in the EU and the US. The conversion technologies for lignocellulosic biomass to advanced biofuels can be broadly classified in three major categories: biochemical, thermochemical, and hybrid conversions. The objective of this review is to discuss the US and EU biofuel initiatives, feedstock availability, and the state-of-art conversion technologies that are potentially ready or are already being deployed for large-scale applications. The review covers and compares the developments in these areas in the EU and the USA and provides a comprehensive list of the most relevant ongoing development, demonstration, and commercialization activities in various companies, along with the different processing strategies adopted by these projects. © 2013 The Authors. *Biofuels, Bioproducts, Biorefining* published by Society of Chemical Industry and John Wiley & Sons, Ltd.

Keywords: biofuels; thermochemical; biochemical; commercialization; lignocellulosic biomass

Introduction

732

Efforts are underway to transform the petroleum-
based economy to a bio-based economy.^{1,2} As the
name implies, a bio-based economy is focused based economy to a bio-based economy.^{1,2} As the name implies, a bio-based economy is focused

on deriving fuels and chemicals from renewable plant-, algal-, or microbial-based materials such as lignocellulosic biomass. The development of new processes for fuels and chemicals from lignocellulosic feedstocks represents an extremely important field for R&D and

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industrial innovation within the bioenergy sector today. While the fundamental and applied research for technology development is carried out in research institutions, companies are using those technologies to actively scale up to demonstration- and commercial-scale activities. In general, major motivations to launch second-generation technologies into full-scale commercial applications will increase the sustainability of biofuel production (compared to first-generation biofuels that are produced from food-grade materials). At the same time, venture capital and government funds are available and have been used by innovative companies working on biotech, biochemical, and thermochemical processes to demonstrate that the processes are reasonable at a large scale. Several companies around the world are currently setting up state-of-the-art technologies that produce advanced biofuels from lignocellulosic biomass. Among them, companies in United States (US) and the European Union (EU) are actively involved, since the basic policy framework for producing biofuels and biochemicals is favorable in these regions.

A definition for the term 'advanced biofuels' is not yet clearly agreed. In the Renewable Fuels Standard of 2010, advanced biofuels were defined as 'non-grain' based fuels³ (other than corn-based biofuels). In 2011, International Energy Agency (IEA) gave the following definition for advanced biofuel technologies:⁴ *'*Conversion technologies which are still in the research and development (R&D), pilot or demonstration phase, commonly referred to as second- or third-generation. This category includes hydro treated vegetable oil (HVO), which is based on animal fat and plant oil, as well as biofuels based on lignocellulosic biomass, such as cellulosic-ethanol, biomass-to-liquids (BtL)-diesel and bio-synthetic gas (bio-SG). The category also includes novel technologies that are mainly in the R&D and pilot stage, such as algae-based biofuels and the conversion of sugar into diesel-type biofuels using biological or chemical catalysts.' Thus, the focus is more on the technology rather than on selecting the feedstock.

The definition of advanced biofuels in the European context is instead still under discussion. The European Commission (EC), for instance, in its recent proposal of revision of the Renewable Energy Directive (RED) ,⁵ defined advanced biofuels⁶ as biofuels that 'provide high greenhouse gas savings with low risk of causing indirect land use change (ILUC) and do not compete directly for agricultural land for the food and feed markets'. Recently, the leaders of Sustainable Biofuels Group, the group merging the major EU industries working exclusively on second-generation biofuels, proposed the following definition⁷: '(1) produced from lignocellulosic feedstocks

(i.e. straw, bagasse, empty fruit bunch, forestry residues, lignocellulosic energy crops, crude tall oil & tall oil pitch), non-food crops (i.e. grasses, miscanthus, algae), or industrial waste and residue streams or manufactured from the biomass fraction of municipal wastes, (2) having low $CO₂$ emission or high GHG reduction, and (3) reaching zero or low ILUC impact.'

The key element in the debate on defining advanced biofuels remains their sustainability and their conflict with food crops. In our opinion, advanced biofuels are any fuels that use advanced technologies to deal with lignocellulosic materials or other unconventional feedstocks that are cultivated on marginal land or that use agricultural/forestry residues. The efficient integration of energy flows in the process makes the overall greenhouse gas emissions and environmental balance of advanced biofuels very favorable and largely superior to most of the socalled first-generation biofuels (excluding the sugarcaneto-ethanol case).

Following the Energy Independence and Security Act of 2007,⁸ the US set a target of 36 million gallons per year (MGPY) advanced biofuels by 2022,⁹ thus forecasting that non-grain-based biofuels (according to the RFS reported above, this includes sugarcane ethanol, lignocellulosic and algal biofuels, etc., but excludes cornstarch-based fuels,) will enter the marketplace at a higher volume. In February 2012, the US Department of Energy (DOE) invested more than US\$1 billion in 29 integrated biorefinery projects to produce advanced biofuels, including ethanol, butanol, gasoline, diesel, and jet fuels; chemicals; and power. Out of the 29 projects, the DOE supported 16 cellulosic ethanol projects with US\$766 million support, 11 hydrocarbon fuel projects with US\$326 million support, 1 butanol project with US\$30 million support, and one succinic acid production facility with US\$50 million support. Among these projects there were two R&D bench-scale demonstration facilities, 12 pilot-scale demonstration facilities, 9 full-scale demonstration plants, and 6 commercial scale plants.

Also in 2007, the EU set its 20-20-20 targets, referring to the goals of increasing the share of renewable energy to 20% (with 10% contribution of renewable alternatives in transportation fuels), improving energy efficiency by 20%, and reducing greenhouse gas (GHG) emissions by 20%, all by 2020, as well as a number of other policies that were also developed and put in place. Among these policies, sustainability criteria where set for biofuels in the Renewable Energy Directive (RED), which mainly address minimum GHG saving requirements, and protection of land with high biodiversity or carbon stock.

More recently, the EC issued a proposal for amending 'the directive 98/70/EC' and 'the directive 2009/28/EC'.⁶ This proposed revised directive, also known as the ILUC directive, better specifies the conditions and the targets for biofuel production in the EU under the light of ILUC considerations. The key issues in the Commission's proposal are the following: (i) 5% limit to the amount of firstgeneration biofuels that can count toward the RED targets, (ii) enhanced incentives for advanced non-land using biofuels (quadruple accounting), (iii) increase to 60% GHG savings requirement for new installations, and (iv) ILUC factors included in the reporting of GHG savings in both directives.

In addition, an explicit list of feedstocks counting between two and four times is given in Annex IX of the document. The consultation with the European Parliament, the council member states and the stakeholders is ongoing, and a decision will be reached soon. The discussion about the future policy framework in the EU (beyond 2020) has also started, with the very recent Green Paper by the EC.¹⁰ Here the EC calls for another consultation (open until 2nd July 2013) focused on addressing targets, the coherence of policy instruments, the competitiveness of the EU economy, and the different capacity of the member states.

The major EC programs¹¹ supporting the development of R&D and demonstration in the field of biofuels are the 7th Framework Program (7FP), the European Industrial Bioenergy Initiative (EIBI) (which addresses only largescale industry-led projects), and the Intelligent Energy Program (not supporting concrete implementation, but market, barrier removal, information and dissemination actions).

In regards to lignocellulosic ethanol production programs, the EC supported 7 industrial demonstration projects through the 7FP for a total of more than €70 million. Recently (December 2012), the EC awarded over €1.2 billion to 23 highly innovative renewable energy demonstration projects under the first call for proposals for the NER300 funding program. Among these, a considerable amount of resources (~€630 million) was allocated to advanced biofuels, with $\sim \epsilon$ 82 million for biochemical routes and the rest (~€548 million) for thermochemical.

With respect to projected production costs of lignocellulosic ethanol, recent communications by major EU industries involved in the construction or operation of industrial demo plant seems to converge around a cashcost target of $1.5-2$ US\$/gal.^{12,13} This cost estimate is very competitive with projected costs for other advanced biofuels production chains, as estimated by the DOE.¹⁴ On the

other hand, the cost of biodiesel from algae were instead estimated at 10.66–19.89 US\$/gal, (one order of magnitude higher than the options previously reported).

It is widely believed that the biofuel process cost will come down as the biorefining technology matures, as it has always happened in the past for new technologies entering the market. A good example is Brazil, where the cost of sugarcane ethanol was substantially reduced mainly due to (i) learning effect, (ii) large-scale operations, and (iii) efficient system integration (including the whole of the supply chain): this was well represented by the well known 'Goldemberg curve', that reported the reduction of ethanol costs in Brazil during the years. In the case of highly innovative technologies, it is reasonable to expect a significant learning factor, which will drive downwards the production costs quite rapidly compared to more mature/less innovative solutions.

Commercial R&D and scale-up activities in the US and EU

The assessment of most relevant EU and US initiatives in the field of lignocellulosic fuels was carried out though the analysis of R&D projects, literature,¹⁵ data sources, $16-18$ other similar work,¹⁹ company websites and personal contacts with several of the companies listed in Tables 1 and 2.

US projects

In the US, the National Advanced Biofuels Consortium (NABC) is a major research initiative and partnership of 17 industry, national laboratory, and university members. The goal of the NABC is the development of technologies to convert lignocellulosic biomass feedstocks to advanced biofuels. Led by the National Renewable Energy Laboratory (NREL) and Pacific Northwest National Laboratory (PNNL) and supported with US\$35 million of American Recovery and Reinvestment Act (ARRA) funding from the DOE and US\$14.5 million of partner funds, NABC is investigating six process strategies including (i) fermentation of lignocellulosic sugars, (ii) catalysis of lignocellulosic sugars, (iii) catalytic fast pyrolysis, (iv) hydrothermal pyrolysis, (v) hydrothermal liquefaction, and (vi) syngas to distillates for converting lignocellulosic biomass feedstock to advanced biofuels.

At the industrial level, 31 US projects are currently involved with the development of advanced biofuels from lignocellulosic biomass (Table 1). With respect to the different biomass conversion routes shown in Fig. 1, 17

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736

					technology and enzymatic hydrolysis process, pilot plant is running since Advanced Steam-Ex pre-treatment http://www.poet.com/biofuels
		biocatalyst			2008
duction in the first Commercial pro- half of 2012	commercial oper- ation in 2013	start-up of Florida Facility on Sept. demonstration the successful Announced 10, 2012	Commercial oper- ation in 2015	Lansing, Mich. in BioAcrylic at the Institute (MBI) in 3,000-liter scale tion process for at the Michigan Biotechnology demonstrated its fermenta- Successfully Oct 2012.	Commercial oper- ation by the end of 2013
feedstocks Multiple	Forest Residue	Sweet sorghum syrup, Biomass hydrolysate	Wood pulp and chips	Sugars	Corn crop residue
\bullet Bioethanol 83.28 ML/y Isobutanol 68.14 ML/y (22 MGPY) $(18$ MGPY) \bullet	\bullet 15.14 ML/y (4 MGPY)	37.85 ML/y (10 MGPY) \bullet	Commercial 75.71 ML/y Pilot 0.757 ML/y (20 MGPY) (0.2MGPY) \bullet \bullet	n.a	\bullet Commercial 75.71 ML/y Pilot 0.076 ML/y (0.02 (20 MGPY) MGPY) \bullet
Isobutanol	nicals Bioethanol Biochem	Fatty alcohols	\overline{Q} Bioethar	Electrofuel, BioAcrylic Bio-based chemicals and fuels e.g. diesel	Bioethanol, Biogas
(Biochemical) Demo	(Hybrid) Demo	(Biochemical) Demo	(Biochemical) Pilot/Demo	(Biochemical) Pilot	(Biochemical) Pilot/Demo
St. Joseph, MO (Commercial) Luverne, MN (Demo)	Soperton, GA	Okeechobee, FL	Rome, NY (pilot/ (commercial) Kinross, MI demo)	Boulder, CO	Emmetsburg, IA Scotland, SD (commercial) (pilot)
Gevo	LanzaTech	L39	Mascoma	Biotechnologies OPX Inc.	Poet-DSM
	The project would combine Beta's mentation process for biobutanol PROESA pre-treatment technol- ogy and Gevo's proprietary fer- http://gevo.com/ production.	Gas fermentation, acquisition of the former Range Fuels biorefinery on http://www.lanzatech.com/ January 3, 2012	Single-step fermentation, proprietary http://www.ls9.com/	Proprietary consolidated bioprocess- http://www.mascoma.com/ ing (CBP) technology	bioprocesses to efficiently produce enables rapid, rational, and robust Engineering) technology platform Dow Chemical Company to bring optimization of microbes and Efficiency Directed Genome OPXBIO is working with The Proprietary, leading EDGETM fatty acids and manufacture BioAcrylic into the market. http://www.opxbio.com/ bioproducts,

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Figure 1. Different biomass conversion routes used in the industry. Here, I, Thermochemical and Hybrid Conversion; II, Biochemical and Hybrid Conversion and III, Hybrid conversion are given.

industrial projects have adopted biochemical conversion methods. The biochemical route is followed mainly for the production of bioethanol using pre-treatment of biomass followed by fermentation. Some of the projects are also pursuing other advanced biofuels such as long chain liquid hydrocarbons (Amyris) and biobutanol (Butamax, Cobalt, and Gevo) using their innovative and proprietary technologies.

Intermediate to the research and industrial initiatives, Michigan Biotechnology Institute (MBI), which is a part of Michigan State University (MSU), is working toward scaling up and commercializing ammonia fiber expansion $(AFEX^{TM^*})$ pre-treatment through a US\$4.3 million grant from the DOE. A one ton-per-day pilot AFEX reactor is

currently being installed. In 2013 another US\$2.5 million DOE grant was awarded to Novozymes and MBI in partnership, to examine the use of AFEX-pre-treated biomass as a feedstock for enzyme production.

Thermochemical routes include pyrolysis, liquefaction, and gasification, and are used to produce long chain liquid hydrocarbons (Fig. 1). Hybrid routes (i.e. combined thermochemical and biochemical) are used for producing both bioethanol and long chain liquid hydrocarbons. As shown in Table 1, the thermochemical platform has been adopted by 14 industries, 5 of which are pursuing hybrid routes. Swedish Biofuels' approach is interesting in that it first produces bioethanol via the conventional biochemical route and then catalytically upgrades it to 'drop-in' biofuels. Similarly, Zeachem's approach is to produce lactic *AFEXTM is a registered trademark of MBI International, Lansing, MI. acid though fermentation and subsequently upgrade it to

bioethanol via hydrogenation. Coskata, Ineos Bio, and Lanza Tech's process strategies depend on syngas (CO + $H₂$) fermentation to bioethanol using their proprietary micro-organisms. The projects reported in Table 1 are not exhaustive and include only those industries whose project details are publicly available. There are several other US projects that are developing some innovative technologies to produce advanced biofuels but are maintaining a very low profile or operating in stealth mode because of their business strategy.

In addition to the single company commercial ventures listed above, technology evaluations are often done through industrial partnerships. A number of partnerships currently exist between Beta/Chemtex/M&G and Genomatica (renewable chemicals, as bio-butadiene BD and bio-butanediol BDO), Gevo (integrated process for bio-isobutanol production), Amyris (renewable fuels and chemicals, as bio-farnasene/farnasano) and Codexis (second-generation detergents from cellulosic biomass), in which the pre-treatment process is combined with various technologies and know-how provided by the partners.

EU projects

With regard to EU initiatives in the field of lignocellulosic biofuels, out of the 40 EU projects reported in Table 2, 17 are based on the thermochemical process, 22 on the biochemical process, and 1 is based on a chemical approach (we identified a total of 5 projects for the chemical route, but only one from a lignocellulosic feedstock). This includes the new projects, either thermochemical or biochemical, recently selected for support by the EC through the NER300 program, 5 of which were for lignocellulosic liquid fuels, and the remaining on lignocellulose-derived biomethane/syngas or intermediate energy liquid carrier (pyrolysis oil, so far targeting district heating). No project was identified in EU as hybrid process technology.

In the field of biochemical conversion, several plants with the capacity to generate thousands or tens of thousands of tons of product per year exist or are under development in the EU. One of the very first EU industrial demonstration initiatives (by Sekab) has been interrupted, but several other processes have been successfully developed into demonstration scale plants. Among these, the largest industrial scale-up efforts are being carried out by Abengoa, Biogasol, Borregaard, Chempolis, Chemtex/M&G (licensed by Beta Renewables), Clariant, Dong Inbicon, Clariant, IMECAL, Inbicon/Dong, Schweighofer Fiber, and UPM.

The situation for thermochemical technologies appears to be slightly different. The largest EU projects aimed

at Fischer-Tropsch (FT) products from lignocellulosic biomass (such as Choren or Neste StoraEnso) have been abandoned or interrupted for various reasons. Today the most relevant initiative is one by Metso/Fortum, a demo project which mainly aims at producing energy rather than a second-generation transport fuel from lignocellulosic biomass. However, the number of initiatives in the thermochemical area focused on generation of transportation fuels could significantly expand if the BTG/Empyro, UPM/Stracel/Btl, VAPO/Ajos-Forest Btl, Billerud/Pyrogrot, CEG plant Coswinowice/Bioagra, BioMCN/Woodspirit, Goteborg AB/Gobigas2, Chemrec and KIT Bioliq projects move toward demonstration-scale. The recent NER300 decision allocated ~ $€457$ million to liquid biofuels produced by the thermochemical route and ~€59 million to the biochemical route, corresponding to only three projects: two using hydrolysis and fermentation and one using anaerobic digestion. This is expected to give a considerable jumpstart to thermochemical pathway technologies. Other than FT-liquids (especially diesel), DME is a major product addressed through the thermochemical pathway. Conversion of biomass to other energy sources such as gasoline (MTG), hydrogen, and natural gas are also under investigation. Synthetic natural gas is another area of fast growth and innovation in the EU and was developed as a method for upgrading $CO₂$ and $H₂$ to synthetic $CH₄$ using energy from fluctuating sources (photovoltaic PW, wind). Goteborg AB GoBiGas project is one example of a demo SNG project of a relatively large size.

Several of the EU-based conversion processes are also going to be implemented in the US or outside the EU, either as first installments or as replications or extensions of an EU demo unit. This is the case of Abengoa, M&G/Chemtex, Swedish Biofuels, and British Airways/Solena. This confirms that industrial development of second-generation biofuels in a given region can have wide-ranging global impacts.

A total of 31 and 35 biofuels projects using lignocellulosic biomass as a feedstock are listed in Table 1 (US) and Table 2 (EU), respectively. It appears that the biochemical conversion platform dominates (18 projects) the commercialization activities in the US and the majority (10 projects) of these projects are aimed toward commercial production of bioethanol by the year 2015. There are seven ongoing projects in the US that are mainly focused on producing liquid hydrocarbon fuels. It is interesting to note that four US projects have adopted a hybrid route whereas there are no active projects in the EU that use this pathway to produce biofuels from lignocellulosic biomass.

The EU projects are almost equally distributed between thermochemical (17 projects) and biochemical (18 projects) conversion platforms. This shows that the biochemical pathway and bioethanol production may be the preferred route in the US, but EU commercialization activities do not show an obvious preference.

Lignocellulosic feedstock for the biorefinery

Available biomass in the US

North America is comprised of 23 countries with roughly 16.5% of the global land area. The USA is one of the biggest countries in North America with an area of 3.79 million square miles. $(9.83 \text{ million km}^2)$, or nearly 2263 million acres of which the composition is 33% forest land, 26% pasture grassland, 20% crop land, 8% parks and recreation area used by public, and 13% urban areas, swamp and desert. Of the total available land, nearly 60% of the land has the potential to grow different biomass depending on the soil conditions. Both the DOE and the US Department of Agriculture (USDA) are developing and funding biomass-to-energy programs. By doing this, it is widely believed that the twenty-first century will see several biorefineries that produce a variety of fuels and chemicals

Figure 2. Current and future biomass available in the US Here, (A) breakdown of total available forest residue by 2030 based on 2005 study; 21 (B) breakdown of total available Agricultural residue by 2030 ;²¹ and (C) summary of current use and future total potential biomass based on baseline assumptions and high yield assumptions based on 2011 study.22 There are subtle differences in the assumptions between the 2005 *Billion Ton Study* and 2011 *Son of Billion Ton Study*. The 2011 study did include county-level analysis with aggregation to state, regional, and national levels that include 2009 USDA agricultural projections and 2007 forestry RPA/TPO 2012–2030 timeline. Biomass annual projections are based on a continuation of baseline trends (USDA projections) and changes in crop productivity, tillage, and land use.

using biomass from agricultural and forest residues. Development of clean, reliable, and affordable energy technologies will strengthen the nation's energy security (less dependence on foreign oil), have positive environmental benefits (reduced GHGs) and strengthen the economy (by generating jobs in the rural sector). $5,20$

The Energy Independence and Security Act (EISA) of 2007 set up a mandatory Renewable Fuels Standard (RFS) to achieve 36 billion gallons per year (BGY) of biofuels by 2022. Only 15 billion gallons can come from corn ethanol and the remaining 21 billion gallons of advanced biofuels should come from non-corn starch based feed stocks (e.g. sugars or cellulose). To meet the targets set by the mandate, not only do sufficient production facilities need to be constructed, but also sufficient quantities of biomass need to be generated and available. The DOE Office of the Biomass Program and Oak Ridge National Laboratory attempted to answer the question of how much biomass was available and where was it located with a report in 2005,²¹ often called the *Billion Ton Study*, and later with an update report in 2011.²² These reports estimated that there is ~1.3 billion tons of biomass/year available in US alone by 2030 based on reasonable assumptions. Of this, 368 million dry tons will come from forest resources

including: (i) fuel wood harvested from forest (52 million), (ii) wood process mill residues and pulp and paper mill waste (145 million), (iii) urban wood waste from construction and demolition debris (47 million), (iv) residues from logging and site cleaning operations (64 million), and (v) biomass that could be harvested to reduce fire (60 million) (Fig. 2(a)). The remaining 998 million tons will come from agricultural resources that include: (i) annual group residues (428 million), (ii) perennial crops (377 million), (iii) grains used for biofuels (87 million), and (iv) animal manure, process residues and other feedstock's (106 million) (Fig. 2(b)). In order to estimate the amount of biomass that will be available in 2030, we need to consider two different assumptions: (i) with moderate crop yields and (ii) with high crop yields (Fig. $3(c)$). In both assumptions, energy crops that are currently being developed by several biotech companies in the US (Ceres, Thousand Oaks, CA; Mendel, Hayward, CA; Monsanto, St Louis, MO) will play an important role in meeting the projected estimates. Energy crops will be made available only if the state or federal government give incentives to farmers to grow them or the companies have a buy back guarantee contract with the farmers or group of farmers (co-op). The biomass residues coming from the agriculture sector are

Figure 3. (A) EU27: Share of biomass in total final energy consumption and (B) Current and 2020–2030 potential for reference scenario.²⁴

about three-quarters of the total available resources in the US. These have high potential for improvement by using advanced farm management technologies, using superior plant breeds, and by adopting best agricultural practices (growing cover crops, crop rotation, growing perennial crops on marginal land, etc). Removal of agricultural residues from the field could vary depending on the soil condition, as the removal rate must maintain soil quality. Agricultural residue availability has been calculated based on five different scenarios, each with a different assumption (low/high crop yield and with/without land use change).²² These scenarios include: (i) currently available from agricultural lands, (ii) under moderate crop yield increase without land use change, (iii) under high crop

yield increase without land-use change; (iv) under moderate crop yield increase with land-use change, and (v) under high crop yield increases with land-use change (Table 3). Dedicated energy crops (switchgrass, Miscanthus, energy cane, forage sorghum, *Erianthus*, Napier grass, etc.,) will contribute significantly to satisfy the growing demand of agricultural residues. Many companies are taking a leading role in establishing businesses in these sectors.

Biomass available in Europe

Based on Fig. 3(a), from the 27 EU member states National Renewable Energy Action Plans (NREAPs), biomass is expected to play a major role in achieving EU targets on renewable energies. It has been projected that 12% of total

S1 - Current availability of biomass from agricultural lands.

750

S2 - Biomass from agricultural lands under moderate crop yield in crease without land use change.

S3 - Biomass from agricultural lands under high crop yield in crease without land use change.

S4 - Biomass from agricultural lands under moderate crop yield in crease with land use change.

S5 - Biomass from agricultural lands under high crop yield increases with land use change.

gross energy demand in the EU will be met using renewable energy in 2020, rising from a total of 85 million tons of oil equivalents (MTOE) in 2010 to 134 MTOE in 2020.²³ The estimation of EU biomass availability in 2012 was around 314 MTOE, expected to grow to 429 MTOE and then set at 411 MTOE in 2020 and 2030, respectively.²⁴ The different biomass resources that are available in EU are shown in Fig. 3(b).

The analysis of biomass availability shows that both in the EU and US the potential for the most sustainable biomass (i.e. wastes and residues), is considerable and represents the largest amount of the total. The EC defines residues as 'no land using crop', to indicate that their sustainable use ensures no additional pressure on land use. Nevertheless, it is always necessary to evaluate case by case the amount of residue that can be removed from the field without impoverishing the land. In the US, the potential for agricultural residues at 2030 is more than the double that of forest residues. In the EU, agricultural residues, wastes, and forestry residues also cover the largest share of the potential. Thus, from a sustainability point of view, the focus in the coming years will be on sustainably managed forestry, agricultural, and agro-industrial lignocellulosic residues, where the ILUC factor is less important than in the case of forestry/agricultural products.

The EU Intelligent Energy Biomass Futures project (www.biomassfutures.org) reported that the share of EU biodiesel on global demand will rise from 42% in 2010 to 74% in 2020, while bioethanol share will also rise to 13% in 2030. It must also be observed that meeting 2020 and 2030 EU biomass targets will require a significant import of feedstock from different parts of the world. Implications on direct and ILUC are currently under evaluation and discussion in Europe.

Biomass logistics

The bulk density of biomass is relatively low and occupies a larger volume compared to other solid materials used for energy such as corn grain or coal. As such, the bulk density significantly influences the transportation and storage of biofuel feedstocks, and becomes a major limiting factor with regard to the size of the biorefinery. A common estimate for feedstock consumption by the biorefinery is 2000 tons of lignocellulosic biomass/day or 7 to 8 million tons of biomass/year. In order to satisfy the biomass demand, yet limit transportation costs and associated GHG emissions, the transportation radius for the biorefinery is commonly set at 50 miles. Development of the biomass supply chain (harvest, collection, storage, preprocessing, handling, and

transportation) is of critical importance if lignocellulosic biofuels are ever to be successfully produced.

Biomass processing

Biomass has low bulk densities, $80-150$ kg/m³ (for herbaceous) and $150-200$ kg/m³ (woody biomass). Current biomass harvesting and bailing machinery produce either rectangular (130-200 kg/m³) or round bales (60-100 kg/m³). These materials should be densified to increase the bulk density and that will help in storage, loading, and transportation. A detailed study conducted by the Idaho National Laboratory (INL) transformed biomass bales into pellets (560–640 kg/m³ with 8–10% moisture) or briquettes $(320-545 \text{ kg/m}^3 \text{ with } 10-12\%$ moisture) through a combination of milling and grinding followed by extrusion based densification. Binding agents (proteins or lignosulfonates) are usually used to hold biomass together. Pre-treatment processes (steam explosion, AFEX, and pre-heating) can relocate lignin to the biomass surface and improve the binding characteristics. Though there are several advantages of biomass densification, it comes with added capital for machinery/ energy cost (milling, briquetting, and cooling units) and requires additional safety measures including dust control systems and spark detection and fire protection systems. $24,26$

Biomass transportation and storage

For transportation purposes, both unit density (kg/m^3) and bulk density $(kg/m³)$ are important parameters. Biomass pellets and briquettes are preferred for biomass conversion due to high energy content per unit volume. Average pellet size (1/4 to 5/16 inches in diameter and up to 11/2 inch long) can be handled just like corn grain (45 lb/ft^3) by truck and railroad, using the existing infrastructure.27 On the other hand, special infrastructure is needed to handle and transport briquettes depending on their shape (pucks, logs of varying diameter and thickness). Moisture content of the biomass needs to be less than 10% moisture if they are to be stored for long periods of time without microbial degradation of biomass sugars. Another approach to reduce the biomass transportation and storage costs is to deploy Regional Biomass Processing Depots (RBPD) that can pre-treat and densify 100–200 tons of biomass per day that can then be transported to a centralized biorefinery.^{28,29} Several thousand RBPDs can be set up around the country in a co-op fashion (involving several farmers) establishing a sustainable biomass supply chain.

Thermochemical and hybrid routes

The production of liquid and gaseous fuels from lignocellulosic feedstocks can also be carried out through thermochemical (or hybrid) approaches (Fig. 1). Thermochemical processes convert the organic matter into a mixture of liquid, gaseous, and solid products whose characteristics depend on the pre-treatment conditions, types of feedstocks, and downstream processing conditions.

In literature, the main biomass thermochemical conversion processes are often classified as torrefaction, (fastintermediate-slow) pyrolysis, hydrothermal liquefaction and gasification. Torrefaction³⁰ is a biomass upgrading and energy-densifying pre-treatment step in which the lignocellulosic biomass is kept for sufficient time at temperatures between approximately 200 and 300 °C in the absence of oxygen. Biomass is thus converted into a hydrophobic product with an increased energy density and more favorable grind-ability (i.e. less energy is necessary to grind the biomass into small particles).

Pyrolysis 31 is a process that decomposes biomass in the absence of oxygen at temperatures between 300 to 550–600 °C. Lower process temperatures and longer vapor residence times increase the production of charcoal, the pyrolysis solid product, while higher temperatures and longer residence times favor the gas phase production. Thus, depending on the process conditions (including the downstream steps such as vapor condensation), the relative amount of solid (char), liquid (pyrolysis oil) and gaseous products can vary considerably, as well as the pyrolysis oil properties. Also, the feedstock characteristics play an important role in the process. Fast pyrolysis maximizes the oil yield, a highly oxygenated acidic and viscous liquid, while slow pyrolysis, also named carbonization, has char is the main product. Both torrefaction and pyrolysis are more and more seen as possible pre-treatment steps before further conversion into liquid products or energy. In case of pyrolysis, it is also possible to upgrade the fuel through catalytic or hydro-de-oxygenation steps into a transport fuel.

Hydrothermal liquefaction is a thermochemical conversion process in which organic material is fed in a wet form to a high pressure (order of hundred bars) and temperature (typically 300–400 $^{\circ}$ C) reactor. The product contains less oxygen than pyrolysis oil and shows more favorable characteristics for downstream processing and use either as fuel or chemicals, but process conditions are very severe and represent a technological challenge.

Gasification occurs when, at higher temperature than pyrolysis or HTL, i.e. around 800–1500 °C or above), the biomass is converted into a CO and H_2 rich gaseous product. The producer gas composition depends on the reactor configuration, process conditions and gasification agent: different reactors should be chosen depending on the final destination. Depending on the final application, it can be necessary to convert the producer gas into a syngas fuel whose composition (e.g. H_2 –CO ratio) is suitable for downstream processing (as FT reactions): this is always needed in the case of synthetic liquid production. The production of liquid fuels from biomass is possible based on the above mentioned processes.

Thermochemical conversion can effectively be used. For instance, catalytic reactors, as Fischer-Tropsch reactors, are used to convert a synthesis gas (syngas) consisting of a mixture of CO and H_2 into hydrocarbons over a catalyst. Other possible process routes convert syngas to methanol, DME, hydrogen, and gasoline. Since, these are mostly catalytic processes, the removal of tar from syngas is a fundamental condition to allow proper operation and avoid catalyst poisoning.

Finally, regarding the hybrid process, some companies like Lanzatech and Coskata are first thermo chemically converting biomass to syngas via gasification and then converting them into liquid fuels by means of a microbial conversion process. Now several industrial initiatives, especially in the US, are testing this process route at demo scale. The other possible hybrid route includes companies like Byogy, CA, that converts ethanol produced using the biochemical route into jet fuel using a proprietary catalyst. Other companies, like Zeachem, produce acetic acid using fermentation route and hydrogenate them into ethanol using a catalytic route.

Biochemical and hybrid routes

Three different conversion scenarios are possible in a biorefinery (Fig. 1). They are:

- (i) Biological conversion, where biomass will be preprocessed by size reducing using milling, followed by chemical pre-treatment. Then, hydrolyzed to fermentable sugars both using acids or commercial enzymes and fermented to fuel molecules of different choices either using bacteria or yeast. In a few cases, the sugars producers are catalytically transformed to fuel molecules. Fuels molecules produced using fermentation or through a catalytic route are further distilled or separated to biofuels.
- (ii) Thermochemical conversion, where the processed biomass is either pyrolyzed to bio-oil/charcoal and

catalytically upgraded to different fuel molecules or gasified to syngas/ash and processed through FT synthesis or microbial fermentation.

(iii) Hybrid route, where fuels are chemically produced using a biological route and then further transformed by thermochemical/catalytic conversion (hybrid route) to another fuel molecule.

Biomass pre-treatment

In the biochemical conversion route, pre-treatment is one of the important processing steps, where different industries adopt different technologies. Pre-treatment can be classified into (i) physical pre-treatment (e.g. extrusion), (ii) chemical pre-treatment (e.g. using acid or base as a catalyst), (iii) physiochemical pre-treatment (e.g. wet oxidation, steam explosion), and (iv) biological pre-treatment (e.g. using microbes). Except for the biological pre-treatment process, which is time consuming, all are used in the industry. Several excellent review articles have been published in the past which provide more detailed information about these pre-treatment processes.³²⁻³⁵ Some details about six well-established pre-treatment technologies that are used in the pilot plants in US and EU are given below.

Wet oxidation

Wet oxidation is an oxidative pre-treatment process where the biomass is wetted with water followed by passing oxygen/air (10–12 bar) at elevated temperatures (170–200 $^{\circ}$ C).³⁶ Since this reaction is an exothermic reaction, the energy needed to heat up the reactor is relatively lower. Though this process solubilizes hemicellulose, most of them are present in an oligomeric form. Phenolic acids are the major degradation products produced during this pre-treatment, which are then degraded into other small organic acids like formic acid. Carbonates ($Na₂CO₃$) are usually added during the process, which elevate the pH to an alkaline condition. Several degradation products that are produced during wet oxidation are toxic for downstream processing. However, highly toxic compounds like hydroxyl methyl furfural (HMF) and furfural are produced in lower amounts. The high costs of carbonate and oxygen are the main bottleneck for this process.

Dilute acid

Cellulose present in biomass is more inert to acid when compared to hemicellulose and lignin. Almost 70–85% of hemicellulose in biomass could be solubilized depending on the pre-treatment conditions, which helps to hydrolyze

cellulose to glucose more efficiently when commercial enzymes are added. Acids are usually used either in dilute or concentrated forms. Companies like Virdia (Dansville, Virginia) use concentrated HCl (1–40%), as they have developed a patented process of efficient recovery and re-use of the catalyst. There is no need to add enzyme to hydrolyze the cellulose to monomeric sugars. However, the hydrolyzed sugars need to undergo a detoxification step prior to fermentation. Most other processes use dilute sulfuric acid (0.22–0.98%). Pre-treatment conditions include 140-180 °C, 15-60 minutes resident time. Most of the hemicellulose is hydrolyzed to xylose³⁷ which has to be either fermented separately or catalytically converted to other high value chemicals. Even at controlled conditions, xylose is further degraded into toxic inhibitory compounds like furfural. In addition to these compounds, several other phenolic degradation compounds are produced. 38 These degradation products have higher inhibitory effects when compared to alkaline pre-treatment processes and have a much lower inhibitory effect when compared to concentrated acids. NREL (Golden, CO) has pioneered this technology and has commissioned a pilot plant to study this process.

Steam explosion

This technology has been in existence since 1920, where it was used to make wood particle board. High pressure stream (280 °C, 1000 psi) was used in those processes. In a biorefinery process, biomass is subjected to a typical temperature range (160-260 °C) for several seconds and then discharged to a cyclone and collected in a different vessel.³⁹ During the pre-treatment, the fibers are mechanically disrupted, thereby increasing the surface area for easy enzyme access and producing a high sugar yield during hydrolysis. Several degradation products, like acetic, formic and levulinic acids, are produced in the process and are inhibitory to the microbes that are used in fermentation. Lignin melts at elevated temperatures and is re-polymerized and re-distributed to different parts of the plant cell wall. Recently dilute sulfuric acid or $SO₂$ impregnated hardwoods are used which reduces the pretreatment temperature and time to produce fewer degradation products.⁴⁰

Ammonia based

Most of the alkali (KOH, NaOH, Ca(OH)) solvents available in the market are strong in nature and are soluble in water. Ammonia is a weak alkali and is volatile which provides an opportunity to recover and reuse it in the

pre-treatment process. It can be used as a gas, liquid ammonia41 or as ammonium hydroxide. MBI and MSU together have developed a pre-treatment process called AFEX that uses either gaseous or anhydrous ammonia in the process. The pre-treatment is done at 100-140 °C using 1:1–3:1 ammonia to biomass ratio for a residence time (of $10-60$ min).⁴¹ Only 3% of ammonia equivalent to biomass is consumed during pre-treatment, producing various nitrogenous compounds like amides (acetamide, feruloyl amide, cumaryl amide), 38 and the remaining ammonia can recovered and reused. DuPont uses dilute ammonium hydroxide, which does not need an expensive recovery step. However, the residence time is longer and the process requires a neutralization step prior to hydrolysis and fermentation.

Mechanical extrusion

Almost all the pre-treatment processes required size reduced biomass. Size reduction includes chipping, milling (Hammer and knife) and grinding. Moisture content, rate of feeding and physical properties of biomass (hard wood or grasses) will influence the energy requirement for size reduction. For particle size reduction to 3–6 mm require about 11 kWh/ton of biomass (agricultural residues).42 However, switch grass, which has a higher silica content, requires about 30 kWh/ton, which corresponds to ~1% of the total energy content in biomass. For hard woods, size reduction to 0.2–0.6 mm requires require kWh/tonne and to 0.15–0.3 mm requires 100–200 kWh/ tonne. Other methods used for size reduction include mechanical extrusion process,⁴³ which helps to disrupt the biomass structure, causing defibrillation and reduced fiber length. Typical conditions used for this process include: screw speed 350 rpm, maximum barrel temperature 80 °C and in-barrel moisture content 40% (wet basis). Though this process is environmentally friendly when compared to thermochemical pre-treatment processes, dust pollution and high energy requirements are major concerns.

Hydrothermolysis/liquid hot water (LHW)

At super critical conditions (>320 °C), water loses its hydrogen bonding and becomes a weakly polar solvent that produces H⁺ and OH⁻ ions. When biomass is subjected to a super critical pre-treatment process, it gets solubilized and hydrolyzed.⁴⁴ The high energy requirements needed for this process was one of the discouraging factors for this technology to become commercialized. However, some companies have started using this technology at pilot scale with improved process development. Other researchers have demonstrated that LHW at controlled pH and milder conditions (190 $\,^{\circ}$ C, 15 min) efficiently pre-treats biomass that could provide a 90% sugar yield using 15 FPU of enzymes.⁴⁵

Other pre-treatments

In addition to the above-mentioned well-established pre-treatment processes, other pre-treatments like lime, ionic liquids and organic solvents (e.g. ethanol) are also being used in commercial scale; their process details are reported elsewhere.35 In particular, the successes of ionic liquid pre-treatment processes developed by companies like SuGanit and Hyrax (US) depend on the efficiency at which the ionic liquid can be recovered and re-used in the subsequent cycles due to high cost of catalyst.

After the biomass is subjected to pre-treatment using one of the above-mentioned process technologies, they undergo enzyme hydrolysis using commercial enzymes and are then subjected to microbial fermentation to produce biofuels. The details about the downstream processing steps are given below.

Enzyme Hydrolysis

For carrying out enzyme hydrolysis a commercial enzyme cocktail is used which consists of 40–50 enzymes with specific activities that are broadly classified into two classes of enzymes: (i) cellulase (that degrade cellulose) and (ii) hemicellulase (that degrade hemicellulose).⁴⁶ Companies like Novozyme, Genencore, Dyadic, DSM, and Iogen are commercial producers of these enzymes using different fungal strains. In the beginning, one cocktail of enzymes (comprising of cellululases and hemicellulases) was sold for hydrolyzing biomass. However, due to variation in the composition of the pre-treated biomass (e.g. dilute acid pre-treatment results is biomass comprising of higher cellulose content and lower hemicellulose content when compared to native feed stock, while ammonia pretreatment like AFEX does not change any composition after pre-treatment) the companies now sell two cocktails of enzymes to hydrolyze cellulose and hemicellulose. These enzymes can be mixed in different ratios depending on the feedstock composition. Most of the enzymes operate at 50 °C, while some of them originated from thermophile microbes and can operate between $60-65$ °C. Many biofuel companies team up with enzyme producers to supply enzymes from centralized production facilities, or in some cases enzymes are produced on the site of a

biorefinery to overcome the cost issues (associated with concentrating the enzymes three-fold) and logistical issues (related to enzyme transportation cot).⁴⁷ Cost of enzymes is one of the key factors that significantly influence the biofuel processing cost and companies are looking at innovative ideas to reduce the enzyme loading and recycle the enzymes over several batches of hydrolysis. After biomass is hydrolyzed into fermentable sugars it is fermented to different fuel molecules using microbes like bacteria or yeast, or in some cases chemically modified using catalysts.

Microbial fermentation

In some processes, the glucose and xylose stream are found together after hydrolysis (e.g. AFEX). While in others, the clean xylose sugar streams that are generated during pre-treatment (dilute acid or steam explosion) can either be combined with the glucose/xylose stream after hydrolysis or processed into chemicals using a biochemical or catalytic route. Separate hydrolysis and fermentation (SHF) is a time-consuming process (3–5 day hydrolysis and 3-day fermentation). However, SHF has some advantages: the microbes can be recycled for the subsequent fermentation cycles or can be processed and sold in the market as animal feed supplements. To overcome the processing time, simultaneous saccharification and co-fermentation (SSF/SSCF) is an option.⁴⁸ Here, the hydrolysis is kick-started at 50 °C for a period of 6 to 12 h. Then, the temperature is brought down to 30 °C and microbe seed cultures are added. Though the efficiency of enzymes (operating at low temperature) is sacrificed, there is some significant time savings. Also, there is some capital cost savings by performing hydrolysis and fermentation in one tank when compared to doing in two separate tanks. Some companies like Virent, Madison are catalytically converting these sugars into long chain alkanes (hybrid route). The process strategy of Mascoma Corporation is based on an innovative consolidated bioprocessing (CBP) approach. The CBP platform utilizes genetically modified yeast or bacteria to convert cellulosic biomass into bioethanol in a single step that combines enzyme production, enzymatic hydrolysis and fermentation.⁴⁸

Biofuel processing

Biofuel processing is dependent on the type of biofuels produced in the industry.⁹ For example, in the case of ethanol (which is miscible in water) distillation is the

preferred option, followed by passage through molecular sieves (to remove residual water). In some cases perevaporation technology (separation of mixtures of liquids by partial vaporization through a non-porous or porous membrane) is also followed. If the biofuel is immiscible in water (such as long chain alkanes and lipids), they separate out on the surface of the water and can be siphoned away. In the few cases where the biofuel produced is toxic to the microbes (e.g. butanol/iosbutanol), they are separated using affinity based separation techniques and further purified. In some cases (e.g. fatty alcohols) reactive distillation during fermentation is also used.

Comparing the Policy Framework in the EU and the US

After the current demonstration phase, the deployment of second-generation technologies in the EU and the US will probably move forward differently according to the Policy frameworks that is in place in each region. In the EU, major EU industries investing in the development of these processes and technologies clearly stated that:⁷ (i) secondgeneration advanced biofuel technologies are ready to compete with conventional biofuels, with EU companies keen to invest in commercial projects given appropriate conditions; and (ii) a stable long-term investment condition is needed, which will encourage investment while at the same time promote true advanced biofuels. This will have a positive economic as well as ecological impact on the EU. Other recent statements from the EU industry were given at the Third International Conference on Lignocellulosic Ethanol held in Madrid (June 2013).⁵⁰

Companies are asking for mandates for advanced biofuels, a clear growing pathway to 2030 and sustainability as reference criteria to evaluate any biofuel production. However, given the peculiarities of lignocellulosic fuels, certification schemes should also be further developed, harmonized among Member States and adapted to respond to the specific characteristics of lignocellulosic fuel chains, particularly when produced from agricultural and forestry residues and wastes (so-called 'no landconsuming feedstocks'). The current certification system in place in the EU is in fact very complex when applied to lignocellulosic residues from agriculture, and difficult to be implemented on an industrial scale on agricultural wastes.

Thus, the main concern from a technological and industrial point of view is the policy framework (including the agricultural policy) in place and its long term stability,

which is needed to secure investments and make projects become bankable in order to make them a reality. According to the EU industries, another urgent need is the development of suitable financing schemes to cover risks and provide guarantees for these very innovative technologies. The situation in the US (and Brazil, even if not discussed here) looks instead very different, with the industrial activities on advanced biofuels and biorefineries supported by the DOE (and BNDES, in Brazil) not only through various forms of grants but also risk covering measures. A number of demo plants are being built in the US, as reported in this work, as the proposed projects gets implemented, conditions could be even more favorable for further commercialization and large scale deployment.

As of today, the EU is in a well-advanced stage of technology development when compared to the US. Given the existing policy framework in the US, it is most likely that the commercial deployment of advanced biofuel generation technology will take place at a faster rate in the US, if no specific measures are taken in Europe. The result of this unclear policy and financial framework is that the EU industries, leading today the technological global competition on advanced biofuels, after having developed their demo plant in the EU, will invest abroad due to less complex and more stable and favorable conditions. This is the case of M&G, partnering with Graalbio in Brazil, where a plant similar to the demo plant in Crescentino is already under construction and new ones will follow, or Abengoa, which is constructing a large industrial demo plant in Hugoton (KS), USA.

Conclusion

A complete summary of biofuels demonstration and commercialization activity in the US and in the EU are presented in this review. A majority of the projects in the US and the EU are either at pilot/demonstration scale or under advance stages of construction of commercial plants. Presently, bioethanol via a biochemical route is the leading process strategy in the US and in EU. The US EISA, 2007 mandates 36 billion gallons of advanced biofuels production per year by 2022 from non-cornstarch-based biomass (sugars or cellulose); whereas the EU's initiative is guided by its 2007 climate and energy 20-20-20 targets with 10% contribution of renewable fuels in transport. With respect to biomass availability, it is projected that about 1.3 billion tons of lignocellulosic biomass per year can be available in the US to meet the advanced biofuels objectives. The biomass resources in the EU may not be adequate for meeting the 2020 and 2030 EU biofuels targets and it may require a significant import of biomass feedstock from different parts of the world. In view of upcoming processing strategies, thermochemical and hybrid routes provide potential to produce 'drop in' biofuels that are compatible with the existing transportation infrastructure.

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Disclaimer

Authors presented data collected through review of available literature, analysis of publications, press and personal contacts. Information here given is to the best of their knowledge, but not necessarily totally exhaustive, complete, or updated. Some deviations from factual situation may be presented. The presentation does not claim to completely cover the given topic.

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758

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