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MINI-REVIEW

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Principles of biorefineries

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Abstract Sustainable economic growth requires safe, sustainable resources for industrial production. For the future re-arrangement of a substantial economy to biological raw materials, completely new approaches in research and development, production and economy are necessary. Biorefineries combine the necessary technologies between biological raw materials and industrial intermediates and final products. The principal goal in the development of biorefineries is defined by the following: (biomass) feedstock-mix + process-mix → product-mix. Here, particularly the combination between biotechnological and chemical conversion of substances will play an important role. Currently the "whole-crop biorefinery", "green biorefinery" and "lignocellulose-feedstock biorefinery" systems are favored in research and development.

Introduction

The maintenance and management of resources are the fundamental political areas within sustainable development passed in Agenda 21 as an action program for the twenty-first century by more than 170 states in June 1992 in Rio de Janeiro and updated at the world summit in September 2002 in Johannesburg. This requires first of all a search for new solutions to decrease today's rapid consumption of fossil, non-renewable resources (petroleum, natural gas, coal, minerals). While the economy of energy can be based on various alternative raw materials (wind, sun, water, biomass, nuclear fission and fusion), the material economy of substances mainly depends on biomass, in particular plant biomass.

Success will depend on how far it is possible to change today's production of goods and services gradually from fossil to biological raw materials. The re-arrangement of whole economies to biological raw materials as a source for increased value requires completely new approaches in research and development. On the one hand, biological and chemical sciences will play a leading role in the generation of future industries in the twenty-first century. On the other hand, new synergies between biological, physical, chemical and technical sciences have to be elaborated and established. This will be combined with new traffic technologies, media and information technologies and economic and social sciences. Special requirements will be placed on both the substantial converting industry and on research and development with regard to raw material and production line efficiency and sustainability.

The development of substance-converting basic product systems and polyproduct systems, such as biorefineries, will be "the key for the access to an integrated production of food, feed, chemicals, materials, goods, and fuels of the future" (National Research Council 2000).

Initial situation

The dimension of the influences of this fundamental change in industrial raw material on the economy is gigantic. Fossil carbon sources, such as petroleum and natural gas, should be replaced by a renewable raw material: biomass, particularly plant biomass. The corresponding products will be called "biobased products" and "bioenergy". The fundamental basic technology which will be introduced in "biorefineries", which as new production plants will replace petroleum-based refineries. In fact, the term "bioeconomy" will be used.

Whereas great successes regarding research and development in the young field of biorefinery system research

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are most notable in Europe (Kamm et al. 1998, 2000; Narodoslawsky 1999), significant industrial developments are being pushed in the United States by both President (U.S. President 1999) and Congress (U.S. Congress 2000). In the United States, it is expected by 2020 to provide at least 25% (compared with 1994) of organic carbon-based industrial feedstock chemicals and 10% of liquid fuels from a biobased product industry. This would mean that more than 90% of the consumption of organic chemicals in the United States and up to 50% of liquid fuel needs would be covered by biobased products (National Research Council 2000).

The Biomass Technical Advisory Committee (BTAC) of the United States, in which leading representatives of industrial companies, such as Dow Chemical, E.I. du Pont de Nemours, Cargill Dow LLC, Genecor International and corn growers' associations and the Natural Resources Defense Council are involved and which acts as advisor to the Government, has a detailed step-plan for targets in 2030 regarding bioenergy, biofuels and bioproducts (Biomass R&D Technical Advisory Committee 2002a).

The BTAC national vision goals for biomass technologies in the United States

Simultaneously, the roadmap "Biomass technology in the United States" has been published by the Biomass R&D Technical Advisory Committee (2002b) in which research, development and the construction of biorefinery demonstration plants are set out (Table 1).

Research and development are necessary to: (1) increase the scientific understanding of biomass resources and improve the tailoring of those resources, (2) improve sustainable systems to develop, harvest and process biomass resources, (3) improve efficiency and performance in conversion and distribution processes and technologies for a host of products development biobased products and (4) create the regulatory and market environment necessary for increased development and use of biobased products.

BTAC has established specific research and development objectives for feedstock production research. Target crops should include oil- and cellulose-producing crops that can provide optimal energy content and usable plant components. Currently, however, there is a lack of understanding of plant biochemistry and inadequate genomic and metabolic data for many potential crops. Specifically, research to produce enhanced enzymes and chemical catalysts could advance biotechnology capabilities.

In Europe there are current regulations regarding the substitution of non-renewable resources by biomass in the area of biofuels for transportation (European Parliament and Council 2003) and the "Renewable energy law" passed in the year 2000 (Gesetz für den Vorrang erneuerbarer Energien 2000). According to the European Community Directive "On the promotion of the use of biofuels" the following products are considered biofuels: (a) bioethanol, (b) biodiesel, (c) biogas, (d) biomethanol, (e) biodimethylether, (f) bioethyltertiarybutylether, based on bioethanol, (g)'biomethyltertiarybutylether, based on biomethanol, (h) synthetic biofuels, (i) biohydrogen and (j) pure vegetable oil.

Member States of the European Union (EU) are requested to define national guidelines for a minimal amount of biofuels and other renewable fuels (with a reference value of 2% by 2005 and 5.75% by 2010, calculated on the basis of the energy content of all petrol and diesel fuels for transport purposes). Table 2 summarizes this EU goal, together with those of Germany regarding the establishment of renewable energy and biofuel (European Parliament and Council 2002; Umweltbundesamt 2000).

Targets of EU and Germany concerning the introduction of technologies based on renewable resources

Until today, there have been no guidelines concerning biobased products in the EU or in Germany. However, after passing directives for bioenergy and biofuels, such a decision is now on the political agenda. The directive for biofuels already includes ethanol, methanol, dimethylether, hydrogen and biomass pyrolysis, which form the fundamental product lines for the future biobased chemical industry.

Table 1 The United States national vision goals for biomass technologies. *quads* Energy units, based on the British thermal unit (BTU, 1 kW h^{-1}): 1 quad = 1 quadrillion BTU (10^{24} BTU)

Technology	Year			
	Current	2010	2020	2030
Biopower				
Biomass share of electricity and heat demand in utilities and industry	2.8% (2.7 quads)	4% (3.2 quads)	5% (4.0 quads)	5% (5.0 quads)
Biofuels				
Biomass share of demand for transportation fuels	0.5% (0.15 quads)	4% (1.3 quads)	10% (4.0 quads)	20% (9.5 quads)
Bioproducts				
Share of target chemicals that are biobased	5%	12%	18%	25%

Table 2 Targets of the European Union and Germany concerning the introduction of technologies based on renewable resources

Technology	Year			
	2001	2005	2010	2020–2050
Bioenergy				
Share of wind power, photovoltaics, biomass and geothermal electricity and heat demand in utilities and industry	7.5%	–	12.5%	26% (2030) 58% (2050)
Biofuels				
Biomass share of demand in transportation fuels (petrol and diesel fuels)	1.4%	2.8%	5.75%	20% (2020)
Biobased products				
Share of target chemicals that are biobased	8–10%	–	–	–

Currently, there are strong activities to establish an appropriate political framework for the market introduction of biologically degradable polymers, in particular those produced from biological raw materials, such as polylactide (PLA), which is based on lactic acid from plant raw material (Industrial Association of Biodegradable Polymers and Bioplastics Manufacturers and Processors 2003). The company Cargill Dow has built a commercial production facility for PLA in Blair, Neb., USA. The Blair facility started operations in late 2002 and has a maximum capacity of 140,000 t of PLA/year (http://www.cargilldow.com, data for 2003). The establishment of further capacity within the company will follow within the next 10 years, up to a capacity of 450,000 t in Asia and Europe. So, it is expected that the price will decrease within the next years to the level of petrochemical-based thermoplastics (Cargill Dow 2001).

Many of the currently used, industrially made biobased products are the result of a direct physical or chemical treatment and processing of biomass, such as cellulose, starch, oil, protein, lignin and terpene. By biotechnological processes and methods, feedstock chemicals are produced, such as ethanol, butanol, acetone, lactic acid and itaconic acid, and amino acids, e.g. glutaminic acid, lysine and tryptophan. In contrast, currently only 6×10^9 t of the 170×10^9 t of biomass produced yearly by photosynthesis are used; in addition, only 3% of this is in the non-food area, such as chemistry (Zoebelin 2001). Today's product lines in the chemical industry produce a few basic chemicals from petrochemical raw materials, which represent the basis for the synthesis of a wide product palette for nearly all areas of life.

The development of comparable biorefineries—however not in the sense of a direct copy—is necessary to produce a broad variety of biobased products in an efficient construction-set system. Each biorefinery refines and converts its corresponding biological raw materials into a multitude of valuable products. The product palette of a biorefinery includes not only the products produced in a petroleum refinery, but also in particular products that are not accessible in petroleum refineries. Therefore, it is necessary to develop new biorefinery-basic technologies, such as: (1) the lignocellulosic feedstock biorefinery (LCF), including LCF pre-treatment and effective separation into lignin, cellulose and hemicellulose, (2) further development of thermal, chemical and mechanical processes, such as extractive methods, gasification (syngas) and liquefaction of biomass, (3) further development of

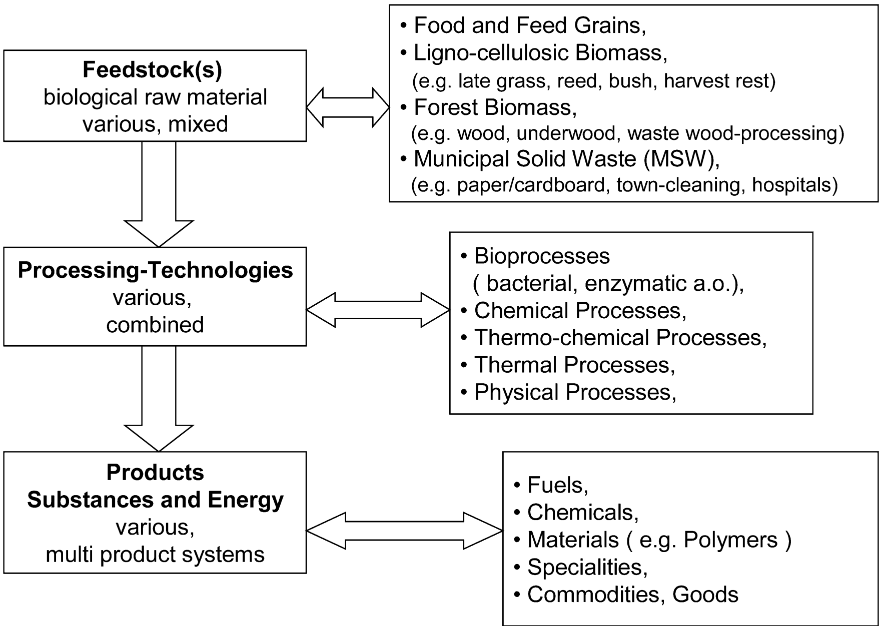

Fig. 1 Basic principles of a biorefinery (phase III biorefinery)

biological processes (biosynthesis, bacteria for the degradation of starch and cellulose, etc.), (4) combinations of substantial conversions, such as biotechnological and chemical processes, (5) corn biorefinery concepts, (6) green biorefinery concepts and (7) the promotion of research and development into phase III biorefinery (biomass feedstock-mix + process-mix → product-mix; Kamm et al. 2000; National Research Council 2000; Fig. 1)

Biological raw material, biomass and precursors

Biomass for the above-cited United States program is defined as follows (U.S. President 1999; U.S. Congress 2000): “The term biomass means any organic matter that is available on a renewable or recurring basis (excluding old-growth timber), including dedicated energy crops and trees, agricultural food and feed crop residues, aquatic plants, wood and wood residues, animal wastes, and other waste materials.”

The majority of biological raw materials are produced in agriculture, forestry and by microbial systems. Forestry plants are excellent raw materials for the paper and cardboard industry and for the construction and chemical industries. Field fruits represent an organic chemical pool, from which fuels, chemicals and chemical products are produced, as well as biomaterials (Fig. 2; Morris and Ahmed 1992). Waste biomass and biomass from nature and landscape cultivation are valuable organic reservoirs of raw material and must be used in accordance to their organic composition. During the development of biorefinery systems, the term waste biomass will become obsolete in the medium term (Kamm et al. 2000).

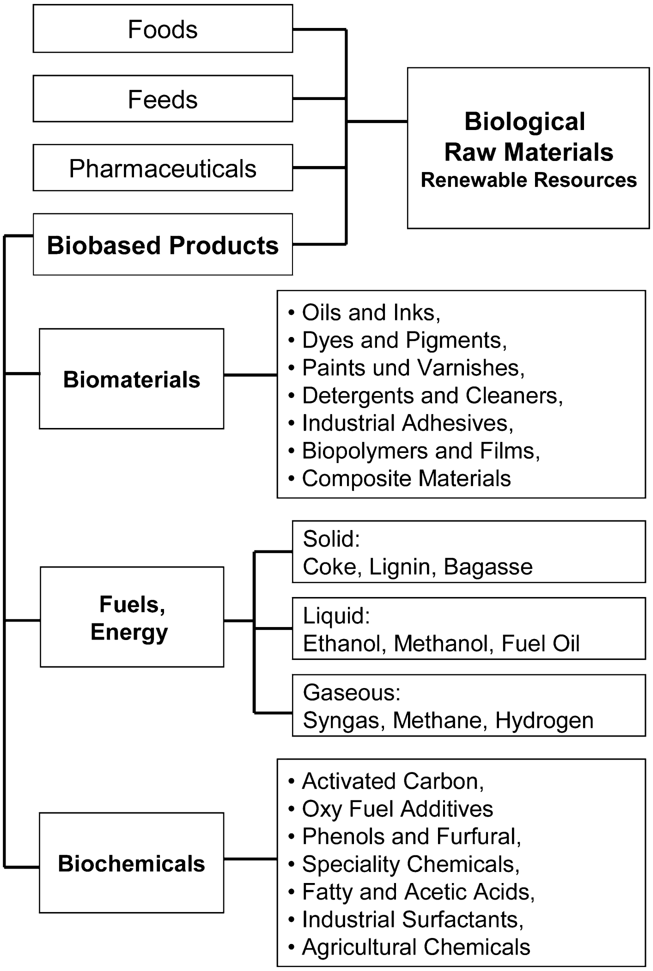

Fig. 2 Products and product classes based on biological raw materials

Biomass has a complex composition, similar to petroleum. Its primary separation into the main groups of substances is appropriate. Subsequent treatment and processing of those substances leads to a whole palette of products. An important difference is that petroleum is obtained by extraction, whereas biomass already exists as product—mostly that of an agricultural substantial conversion process. Thus, biomass can be modified within the actual process of genesis, in such a way that it is suitably adapted for subsequent processing, as particular target products have already been formed. For those products, the term precursors is used (Ringpfeil 2002).

Plant biomass always consists of the basic products carbohydrates, lignin, proteins and fats, beside various substances, such as vitamins, dyes, flavors and aromatic essences, which have very different chemical structures. Biorefineries combine the essential technologies to transform biological raw materials into industrial intermediates and final products (Fig. 3).

Biorefinery general scheme

A technically feasible separation operation, which would allow the separate use or subsequent processing of all these basic compounds, has existed until now only in the form of initial attempts. Assuming that, of the estimated annual biosynthesis production of biomass of 170×10^9 t, 75% is carbohydrate, mainly in the form of cellulose, starch and saccharose, 20% is lignin and only 5% is other natural compounds, such as fats (oils), proteins and various substances (Röper 2001), the main attention should first be focused on an efficient access to carbohydrates, their subsequent conversion to chemical bulk products and the corresponding final products. Glucose, accessible by microbial or chemical methods from starch, sugar or cellulose, is among other things predestined for a key position as a basic chemical, because a broad palette of biotechnological or chemical products is accessible from glucose. In the case of starch, the advantage of enzymatic compared with chemical hydrolysis has already been realized (Kamm et al.1997).

In the case of cellulose, this is not yet realized. Cellulose-hydrolyzing enzymes can only act effectively after pre-treatment to break up the very stable lignin/cellulose/hemicellulose composites. These treatments are still mostly thermal, thermo-mechanical or thermo-chemical and require a considerable input of energy. Once in

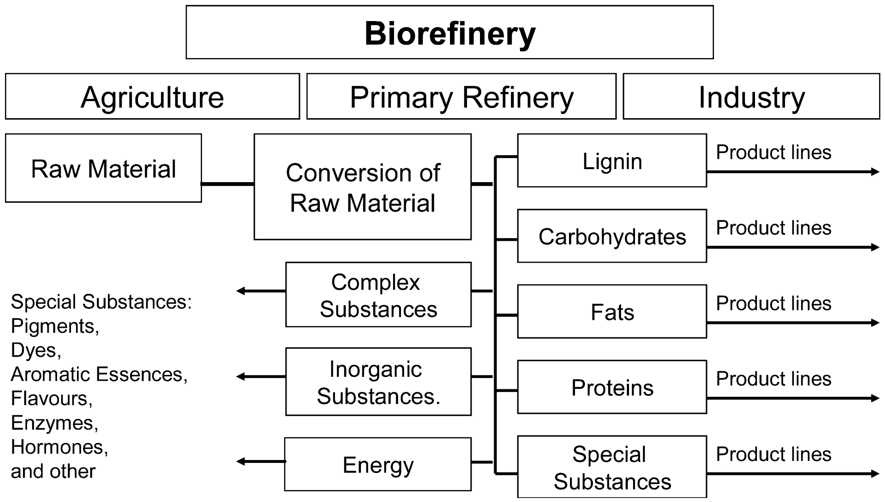

Fig. 3 Code-defined basic substances produced (via fractionation) for the development of industrially relevant product family trees

this state, the acidic hydrolysis can be finished, although only low yields of glucose can then be achieved, compared with treatments using enzyme combinations. There is the question whether recent developments, such as the use of expansins (Cosgrove 2000), will bring a breakthrough regarding low-energy break-up processes. The arsenal for the microbial conversion of substances out of glucose is large and the reactions are energetically profitable. It is necessary to combine the degradation processes from glucose to bulk chemicals with the building processes that give their subsequent products and materials (Fig. 4).

Among the variety of possible products from glucose which are accessible microbially and chemically , lactic acid, ethanol, acetic acid and levulinic acid are particularly favorable intermediates for the generation of industrially relevant product family trees. Here, two potential strategies are considered: either the development of new (possibly biologically degradable) products (e.g. follow-up products of lactic and levulinic acid) or the entry as intermediates into the conventional product lines (e.g. acrylic acid, 2,3-pentandion) of petrochemical refineries.

Definitions of the term biorefinery

The young working field of biorefinery systems, in combination with biobased industrial products, is in various respects still an open field of knowledge. This is also reflected in the search for an appropriate description,

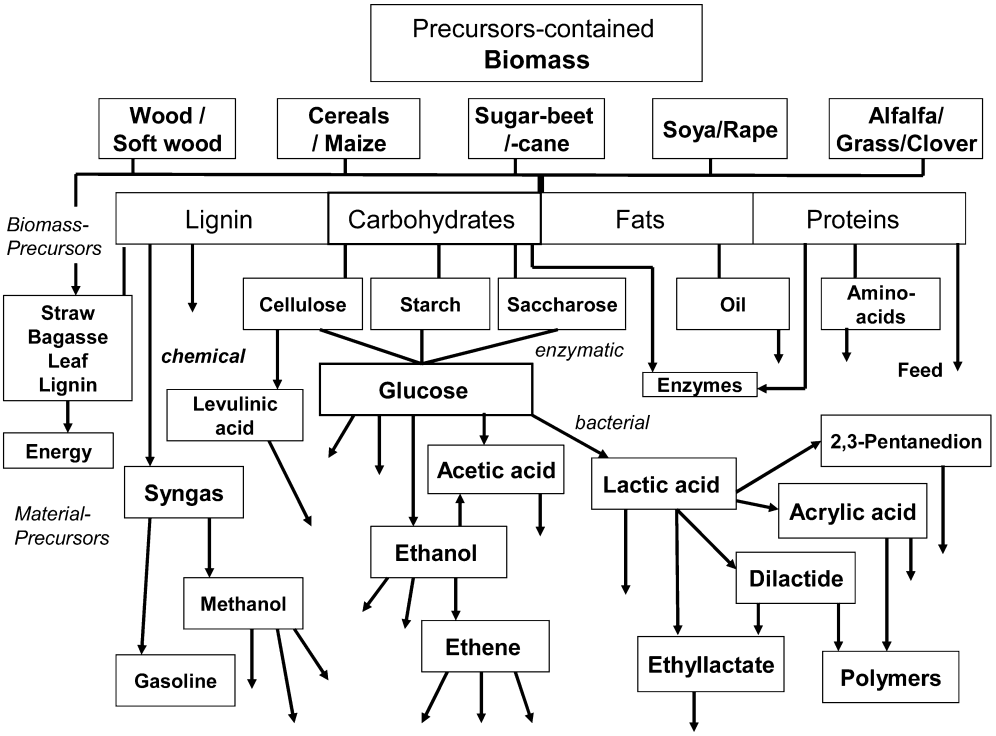

Fig. 4 Biorefinery general scheme for precursor-containing biomass, with preference for a carbohydrate line

the content of which is in parts controversial. A selection of descriptions is given below.

The term green biorefinery was defined in the year 1997 as follows: green biorefineries represent complex (to fully integrated) systems of sustainable, environment- and resource-friendly technologies for the comprehensive (holistic) utilization and the exploitation of biological raw materials in the form of green and residue biomass from a targeted sustainable regional land utilization (Kamm et al.1998). The term used originally in Germany “complex construction and systems” was substituted by “fully integrated systems”.

The United States Department of Energy, in its Energy, environmental, and economics (E3) handbook, uses the following definition (U.S. Department of Energy 1997): a biorefinery is an overall concept of a processing plant where biomass feedstocks are converted and extracted into a spectrum of valuable products, based on the petrochemical refinery.

There is (more or less) agreement about the *goal*, which is briefly defined as: developed biorefineries, so-called phase III biorefineries, start with a biomass feedstock-mix to produce a multiplicity of products by a technology-mix (Van Dyne et al. 1999; Fig. 1). An example of the phase I biorefinery is a dry-milling ethanol plant. It uses grain as a feedstock, has a fixed processing capability and produces a fixed amount of ethanol, feed co-products and carbon dioxide. It has almost no flexibility in processing. Therefore, this type can only be used for comparable purposes. An example of the phase II biorefinery is the current wet-milling technology. This technology uses grain feedstocks, yet has the capability of producing various end-products, depending on product demand. Such products include starch, high fructose corn syrup, ethanol, corn oil and corn gluten feed and meal. This type opens numerous possibilities to connect industrial product lines with existing agricultural production units. Thus, the integrated production of biodegradable plastics, such as poly-3-hydroxybutyrate, sugar and ethanol in a conventional sugar plant is an example of a phase II biorefinery (Nonato et al. 2001).

A phase III biorefinery is not only able to produce a variety of chemicals, fuels and intermediates or end-products, but can also use various types of feedstocks and processing methods to produce products for the industrial market. The flexibility of its feedstock use is the factor of first priority for adaptability towards changes in demand and supply for feed, food and industrial commodities.

Some aspects towards biorefinery technologies

In the first step, the precursor-containing biomass is separated by physical methods. The main products and the by-products are subsequently subjected to microbiological or chemical methods. The follow-up products of the by- and main products can furthermore be converted or enter a conventional refinery. Therefore, the term biorefinery receives a double importance, on the one hand because of the biological genesis of the corresponding raw material and on the other hand because of the rising biological character of selected treatment and processing methods.

Currently, three biorefinery systems are favored in research and development. First, the whole-crop biorefinery, which uses raw materials such as cereals or maize. Second, the green biorefinery, which uses naturally wet biomass, such as green grass, lucerne, clover, or immature cereal. Third, the lignocellulose feedstock (LCF) biorefinery, which uses naturally dry raw materials such as cellulose-containing biomass and wastes.

The LCF biorefinery

Among the potential large-scale industrial biorefineries, the LCF biorefinery will most probably be pushed through with highest success. On the one hand, the raw material situation is optimal (straw, reed, grass, wood, paper-waste, etc.) and, on the other hand, conversion products have a good position within both the traditional petrochemical and the future biobased product markets. An important point for the utilization of biomass as a chemical raw material is the cost of raw materials. Currently, the costs are U.S. $ 30/t for corn stover or straw and U.S. $ 110/t for corn (U.S. $ 3/bushel; Dale 2002).

Lignocellulose materials consist of three primary chemical fractions or precursors: (a) hemicellulose/polyoses, sugar polymer of predominantly pentoses, (b) cellulose, a glucose polymer and (c) lignin, a polymer of phenols (Fig. 5).

An overview of the potential products of a LCF biorefinery is shown in Fig. 6. In particular, furfural and hydroxymethylfurfural are interesting products. Furfural is the starting material for the production of Nylon 6,6 and Nylon 6. The original process for the production of Nylon 6,6 was based on furfural. The last of these production plants was closed in 1961 in the United States due to economic reasons (the artificially low price of petroleum). Nevertheless, the market for Nylon 6 is huge.

However, there are still some unsatisfactory parts within the LCF, such as the utilization of lignin as fuel, adhesive or binder. It is unsatisfactory because the lignin scaffold contains considerable amounts of mono-aromatic hydrocarbons which, if isolated in an economically efficient way, could add a significant value increase to the primary processes. It should be noticed that there are no obvious, natural enzymes to split the naturally formed lignin into basic monomers as easily as is possible for naturally formed polymeric carbohydrates or proteins (Ringpfeil 2002).

$$\text{Lignocellulose} + H_2O \rightarrow \text{Lignin} + \text{Cellulose} + \text{Hemicellulose}$$

$$\text{Hemicellulose} + H_2O \rightarrow \text{Xylose}$$

$$\text{Xylose } (C_5H_{10}O_5) + \text{acid Catalyst} \rightarrow \text{Furfural } (C_5H_4O_2) + 3H_20$$

$$\text{Cellulose}(C_6H_{10}O_5) + H_2O \rightarrow \text{Glucose } (C_6H_{12}O_6)$$

Fig. 5 General equation of the conversion of precursors of the lignocellulosic feedstock (LCF) biorefinery

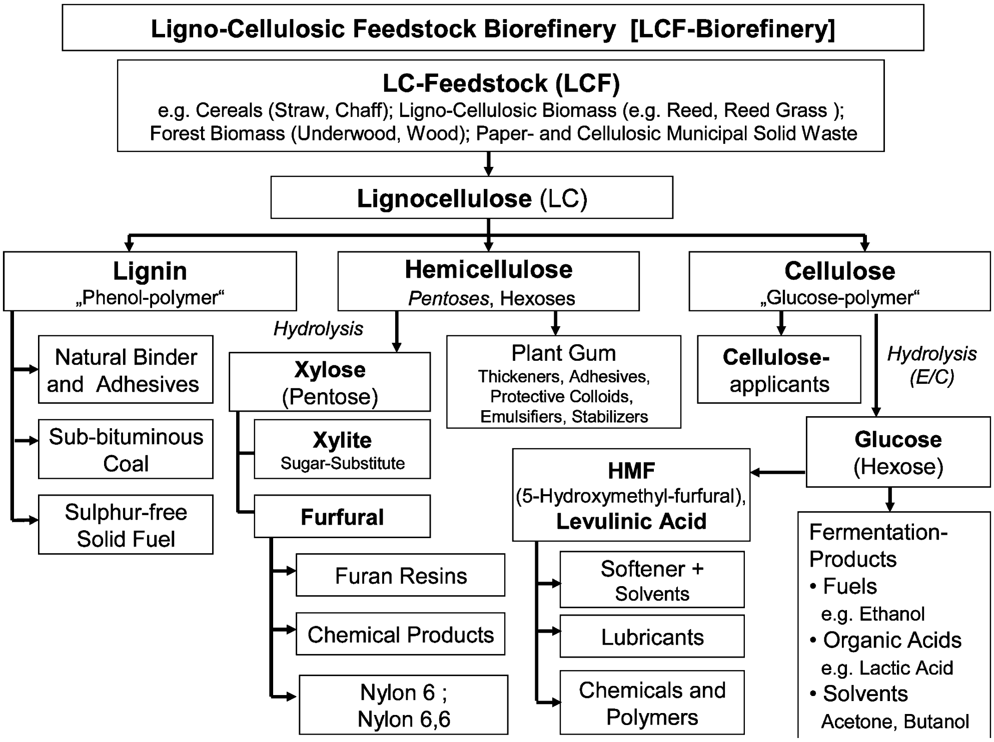

Fig. 6 Lignocellulosic Feedstock Biorefinery (LCF-Biorefinery, Phase III)

An attractive accompanying process to the biomass–Nylon process is the already mentioned hydrolysis of cellulose to glucose, with the production of ethanol. Certain yeasts give a disproportionation of the glucose molecule during their generation of ethanol which practically shifts the entire reduction process towards ethanol and makes the latter obtainable in a 90% yield (w/w, regarding the formula turnover).

Based on recent technologies, a plant was conceived for the production of the main products furfural and ethanol from LCF for west-central Missouri (USA). Optimal profitability can be reached with a daily consumption of about 4,400 t of feedstock. Annually, the plant produces

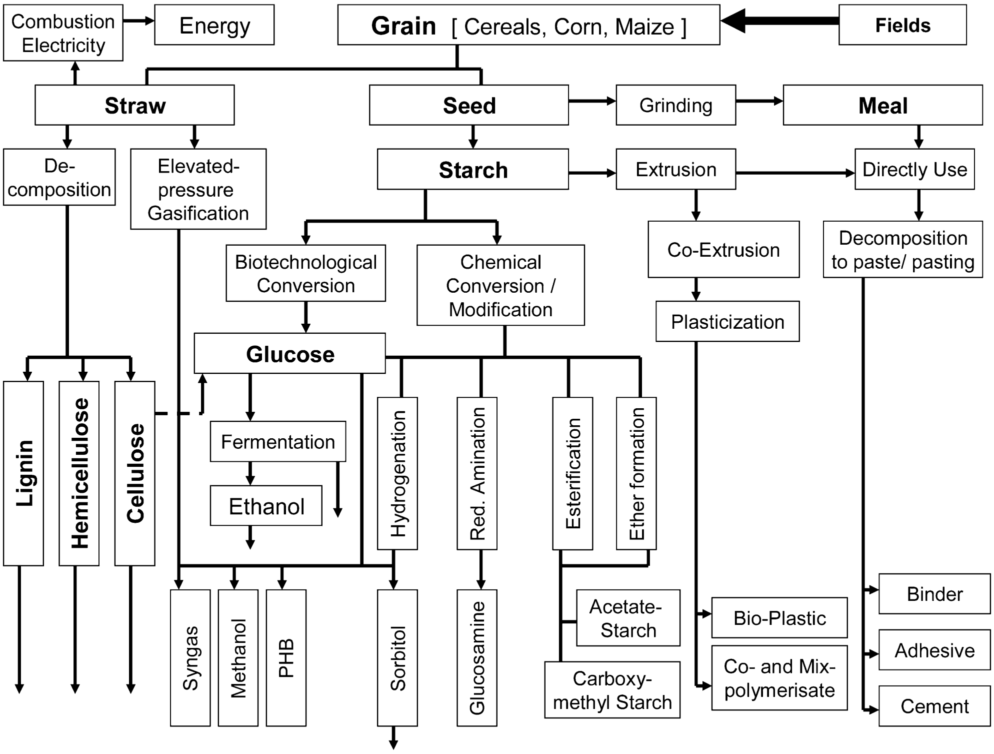

Fig. 7 General scheme for a whole-crop biorefinery

180×10^6 l of ethanol and 323×10^3 t of furfural (Van Dyne et al.1999).

Ethanol may be used as a fuel additive. Ethanol is also a connecting product for a petrochemical refinery. Ethanol can be converted into ethene by chemical methods. As is well known for petrochemically produced ethene, today it starts a whole series of large-scale technical chemical syntheses for the production of important commodities, such as polyethylene and polyvinylacetate. Further petrochemically produced substances can similarly be manufactured by substantial microbial conversion of glucose, such as hydrogen, methane, propanol, acetone, butanol, butandiol, itaconic acid and succinic acid (Zeikus et al. 1999; Vorlop and Willke 2003).

The whole-crop biorefinery

The raw materials for the whole-crop biorefinery are cereals, such as rye, wheat, triticale, and maize. The first step is mechanical separation into corn and straw, which are obtained in almost the same amount. The straw represents a LCF and may further be processed in a LCF biorefinery. There is the possibility of separation into cellulose, hemicellulose and lignin and their further conversion within the separate product lines shown in the LCF biorefinery (Fig. 6). Furthermore, the straw is a starting material for the production of syngas via pyrolysis technologies. Syngas is the basic material for the synthesis of fuels and methanol (Fig. 7).

The corn may either be converted into starch or directly used after grinding to meal. Further processing may be carried out in four directions: (a) breaking-up, (b) plasticization, (c) chemical modification or (d) biotechnological conversion via glucose. The meal can be treated and finished by extrusion into binder, adhesives and filler. Starch can be finished via plasticization (co-, mix-polymerization, compounding with other polymers), chemical modification (etherification into carboxymethyl starch, esterification, re-esterification into fatty acid esters via acetic starch, splitting reductive amination into ethylene diamine, etc., hydrogenative splitting into sorbitol, ethyleneglycol, propyleneglycol, glycerine) and biotechnological conversion (poly-3-hydroxybutyric acid; Nonato et al. 2001).

The green biorefinery

Green biorefineries are multi-product systems which handle their refinery cuts, fractions and products in accordance with the physiology of the corresponding plant material, i.e. the maintenance and utilization of the diversity of syntheses achieved by nature.

Green biomass for example includes grass from the cultivation of permanent grassland, closure fields, nature preserves and green crops, such as lucerne, clover and immature cereals from extensive land cultivation. Thus, green plants represent a natural chemical factory and food plant. Careful wet-fractionation technology is used as the first step (primary refinery) to isolate the green biomass substances in their natural form. Thus, green crop goods (or humid organic waste goods) are separated into a fiber-rich press cake and a nutrient-rich green juice (Fig. 8).

Beside cellulose and starch, the press cake contains valuable dyes and pigments, crude drugs and other organics. The green juice contains proteins, free amino acids, organic acids, dyes, enzymes, hormones, other organic substances and minerals. In particular, the application of biotechnological methods is predestined for conversions, because the plant water can simultaneously be used for further treatments. In addition, the

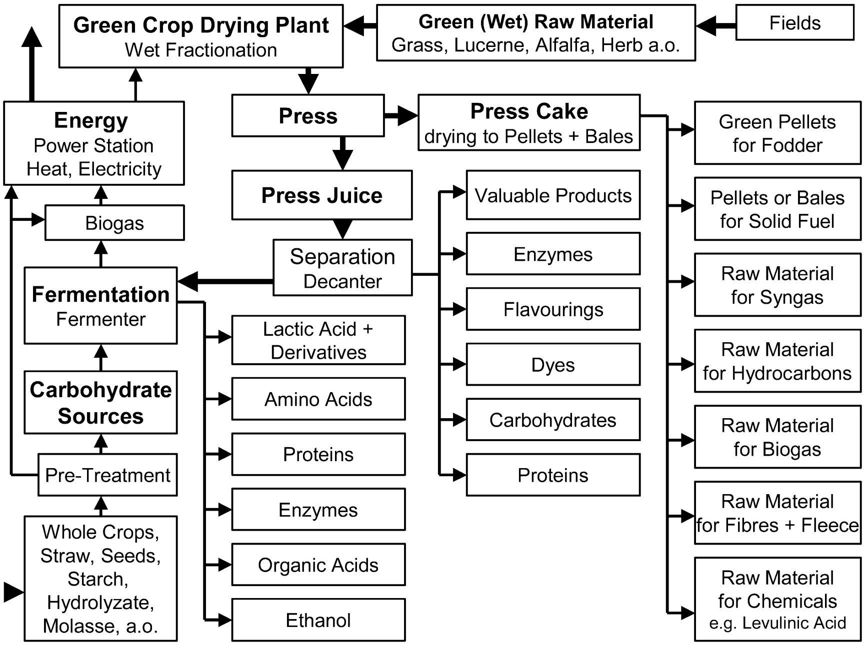

Fig. 8 A green biorefinery system combined with a green crop-drying plant. *a.o.* And others

lignin–cellulose composites are not so strong as those in LCF materials. Starting from green juice, the main focus is directed to products such as lactic acid and the corresponding derivatives, amino acids, ethanol and proteins. The press cake can be used for the production of green feed pellets, as a raw material for the production of chemicals, such as levulinic acid, and for conversion to syngas and hydrocarbons (synthetic biofuels). The residues of a substantial conversion are suitable for the production of biogas, combined with the generation of heat and electricity. Reviews of the green biorefinery concepts, contents and goals are available (Kamm et al. 1998, 2000; Narodoslawsky 1999).

Outlook

There are several requirements for entering the development of industrial biorefinery technologies and biobased products. It is necessary, on the one hand, to increase the production of substances (cellulose, starch, sugar, oil) from a base of biogenic raw materials and, on the other hand, to push the introduction and establishment of biorefinery demonstration plants. Important points are to get the commitment of the chemistry, particularly the organic chemistry, needed for the concept of biobased products and biorefinery systems and to force the combination of the biotechnological and chemical conversion of substances. Last but not least, the development of systematic approaches to new syntheses and technologies is required to meet the sustainable principles of ideal synthesis (Clark 1999).

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