

Lignocellulosic Feedstock Biorefinery – Combination of technologies of agroforestry and a biobased substance and energy economy

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Kurzfassung

Die Entwicklung von Bioraffinerien präsentieren den Schlüssel für eine integrierte Produktion von Nahrungsmitteln, Futtermitteln, Chemikalien, Materialien, Gebrauchsgütern und Kraftstoffen der Zukunft (NATIONAL RESEARCH COUNCIL, USA, 2000). Bioraffinerien kombinieren die notwendigen Technologien für die Verarbeitung von biogenen Rohstoffen zu Zwischen- und Finalprodukten. Das Hauptaugenmerk ist im gleichen Maße auf neue Landnutzungsformen zur Biomasseproduktion, die Prinzipien der Bioraffinerien, die Lignocellulosic-Feedstock Bioraffinerie, die Plattform-Chemikalien sowie die direkte Nutzung des Polymers Cellulose gerichtet.

Abstract

The development of biorefineries represents the key for the access to an integrated production of food, feed, chemicals, materials, goods, and fuels of the future (NATIONAL RESEARCH COUNCIL, USA, 2000). Biorefineries combine the necessary technologies of the biogenic raw materials with those of intermediates and final products. The main focus is directed on the new land use forms for biomass production, the principles of biorefineries, the lignocellulosic feedstock biorefinery, the platform chemicals as well as the direct polymer cellulose.

1 Introduction – Raw material change in the substantial converting industry and chemical industry

One-hundred-and-fifty years after the beginning of coal-based chemistry and 50 years after the beginning of the petroleum-based chemistry industrial chemistry is now entering a new era. In the twenty-first century utilization of renewable raw material will gain importance in the chemical conversion of substances in industry. Partial or even complete re-adjustment of whole economics to renewable raw materials

will require completely new approaches in research, development and production (KAMM et al., 2006a). One approach is the development of biorefinery technologies and systems. Biorefining is the transfer of the efficiency and logic of the fossil-based chemistry and substantial converting industry as well as the energy production on to the biomass industry (KAMM et al., 2006b).

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 of biobased products and (4) create the regulatory and market environment necessary for increased development and use of biobased products (BIOMASS R & D TECHNICAL ADVISORY COMMITTEE, 2002a, b).

Chemistry foresee up to 30 % of raw materials for the chemical industry coming from renewable sources by 2025 (INDUSTRIAL BIOTECHNOLOGY SECTION, 2005). Lately European Commission and U S Department of Energy are come to an agreement for cooperation on this field (US DEPARTMENT OF ENERGY, 2005).

Because of low cost, plentiful supply, and amenability to biotechnology, carbohydrates appear likely to be the dominant source of feedstocks for biocommodity processing. Starch-rich and cellulosic materials each have important advantages in this context. Corn is by far the dominant feedstock for biological production of commodity products today. Advantages to cellulosic materials include much larger ultimate supply, lower purchase cost and lower anticipated transfer cost, less erosivity, and lower inputs of chemicals and energy required for production (LYND et al., 1999). Recently the goal of the US Department of agriculture and the US Department of energy is the additional supply of 1 billion ton biomass for a prize of 35 US-Dollar per tons per year for the industrial chemical and biotechnological utilization, without restriction of today's applications of biomass from agriculture and forestry (US DEPARTMENT OF AGRICULTURE (USDA) and US DEPARTMENT OF ENERGY (DOE) 2005).

2 New land use forms for biomass production in the temperate zone

In the temperate zone there is an increasing awareness for the need to substitute fossil raw materials by regenerative resources, particularly lignocellulosic rich biomass. This recent development calls for an adaptation of traditional forest and agricultural landuse systems in order to satisfy the upcoming demand for biomass. The production of biomass instead of food crops and fodder requires new production systems in order to cope with quality standards and quantities needed. Therefore, new forms of landuse have to be tested and implemented which allow farmers to flexibly respond to the requirements of the market, to secure a permanent product provision and to be compatible with regard to traditional crop production systems. At the same time new approaches in landuse should consider upcoming risks such as climate change, extended summer drought, the ongoing loss of biodiversity, and both groundwater formation and quality.

Such paradigm change in traditional landuse gains even more relevance with regard to the reformation of the agricultural policy at the EU level (CAP reform) which will entail a change from product to area based subsidies. Particularly for agricultural set-aside areas, for marginal land and for decontaminated sites landowners will be challenged in future to apply alternative landuse concepts in order to re-validate such low productive areas without being subsidised. As to the high areal potential of such areas, the production of biomass for biorefinery purposes would not conflict with other demands.

In this context agroforestry systems come increasingly into focus as they offer an integrated approach for the production of both crops and non-crop biomass, thus diversifying the agricultural production at both the enterprise level and for regional economies (ONG 1997, WALLACE 1996, YOUNG 1997). They are highly suitable to re-validate agricultural set aside areas, to reclaim mining areas or to restore contaminated sites when placing less emphasis on crop but rather on biomass production for chemical or thermo-physical processing.

Under tropical and subtropical conditions numerous studies have pinpointed the economic, social and environmental benefits evolving from agroforestry systems (CANNELL et al. 1996, VAN NOORDWIJK et al. 1998, ONG et al. 1996, HUXLEY 1996, BRENNER 1995, SCHROTH 1999, HERZOG 1997). Integrating elements of forest and agricultural production within one system, agroforestry offers the opportunity to diversify and to adapt the production and to minimize management efforts and input of costs. The economic efficiency of such management systems highly depends on synergisms resulting from the interaction between trees and crops. Trees improve the microclimate by giving shadow and by decreasing the wind speed, thus, increasing the formation and persistency of dew water (HEISLER und DEWALL 1988, ONG and SINCLAIR 1997; YOUNG 1997, KIEPE 1995, McNAUGHTON 1988, FRIDAY and FOWNES 2001, SCHROTH 1994, SWAMY et al. 2006, PINTO et al. 2005, POWELL and BORK 2004, MUTHURI et al. 2005). Agroforestry systems are, therefore, well-known to be adaptive to changing climatic conditions at regional scales and may help to secure the provision of crops and woody biomass even under extreme climatic conditions such as extended drought. They minimize wind and water erosion,

and the tree litter provides an additional nutrient input to crops in the intercroppings, thus stimulating the formation and accumulation of organic matter and increasing soil fertility even without external input (YOUNG 1997, KIEPE 1995, BANZHAF 1988, McNAUGHTON 1988, FRIDAY und FOWNES 2001, SCHROTH 1994, SWAMY et al. 2006, PINTO et al. 2005, POWELL und BORK 2004, MUTHURI et al. 2005). The same factors form a prerequisite for the biological diversity of such systems (SCHROTH et al., 2004) which is further enhanced by the structural variability and the extensive management of such systems, and, particularly, by the absence or limited application of pesticides and fertilizer.

However, a successful implementation of an agroforest management system was always depending on a careful adaptation of such approach to the specific edaphic and climatic conditions and to the local socioeconomic context. Generally, agroforestry practices can be subdivided into alley cropping, silvopasturing, shelterbelt systems, river bank replantations and forest farming, including the production of secondary forest products. This clearly reflects a demand driven choice of options. Therefore, prior to an implementation of agroforestry systems at the European level it will be crucial to carefully verify those objectives being generally attributed to an agroforest management and also to consider the specific social, ecological and economic environment of the different European regions.

In 1996 the Chair of Soil Protection and Recultivation established an agroforestry system on reclaimed mine sites of the Lusatian mining districts near the town of Jänschwalde (NE GERMANY, GRÜNEWALD 2005). For this particular study an alley-cropping system was chosen which was composed of intercroppings (18 m width) and tree hedges (rows of 6 m width) in between (Fig. 1). Besides assessing the yields of different clones of poplar (*Populus* spp.), willow (*Salix viminalis* L.), and black locust (*Robinia pseudoacacia* L.) at 3-, 6- and 9-year-rotation (s. *Woldt* et al.; this journal) the study also included an assessment of environmentally relevant parameters, i. e. the seepage water rate, microclimatic effects of tree hedges, and the monitoring of the soil carbon contents.



Figure 1:
Scenic view of the alley-cropping system at the post-mining reclamation site near the town of Jänschwalde (Lower Lusatia)

In the topsoil layers of the overburden sediment almost no carbon could be detected one year after the alley cropping systems had been established (Tab. 1). Already in 2005 the carbon concentrations in the intercroppings were 2-fold and in tree rows 3-fold higher as compared to the initial state. This finding reflects the potential of such extensive-

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ly managed agroforestry system to sequester carbon. The results also indicate the need for a long-term monitoring of the soil carbon pool as well as for a comparative analysis of carbon sequestration potentials of different landuse systems. In the future such information may help to reward the benefits that arise from extensive vs. intensive management approaches with regard to climate and soil protection.

Table 1:
Soil carbon contents (%) in tree hedges and intercroppings one year and seven years after the establishment of the alley-cropping system

| Depth (cm) | 1997 | 2005 Tree Alleys | 2005 Transition: Tree-Intercropping | 2005 Intercropping |
|------------|-----------------|------------------|-------------------------------------|--------------------|
| 0-10 | 0,45 (0,26)* | 1,55 (0,64)* | 1,13 (0,25) | 1,04 (0,24) |
| 10-30 | | 0,85 (0,28) | 1,03 (0,34) | 0,99 (0,28) |

* standard deviation

Calculating the water flow for intercroppings and tree hedges (Fig. 2) it became evident that the rate of seepage water under trees was only slightly lower than under the intercroppings. Therefore, no negative impact would be expected for the water balance at the landscape level. Due to the restricted fertilisation regime and as to the fact that no pesticides had been used in the study period it may be assumed that the quality of the seepage water exceeded that known from intensive-ly managed agricultural land.

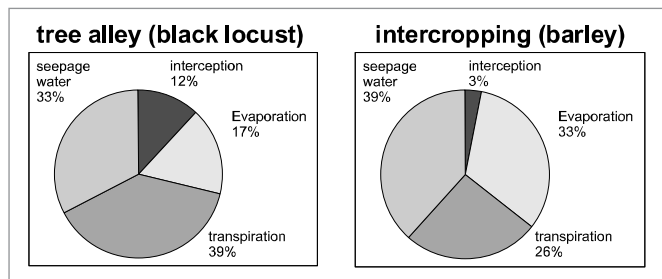


Figure 2:
Evapotranspiration, interception and seepage water under fast growing tree species and intercroppings

A further approach was dedicated to the analysis of microclimatic effects comparing the change of the soil water contents at high temporal resolution in 0-10 cm and 10-30 cm soil depth in the slipstream of tree hedges and on an adjacent traditionally managed agricultural field without a shelterbelt (Fig. 3). The water content was always significantly higher on the lee side of the hedges than on the plane field. It was noteworthy that behind the hedges the water content remained at nearly the same level in both soil depths whereas the evaporation losses on the plane field were much higher and even more pronounced in the top 10 cm of the soil.

The present findings highlight the potential ecological benefits of agroforestry systems and underline their suitability as an alternative form of landuse in the temperate zone for the production of biomass for chemical and thermal processing, particularly, on degraded land and in agricultural set-aside areas. Combining tree rows and intercrop-

pings will help to increase the structural richness of landscapes, but will also extend the number of management options for farmers in order to achieve flexibility in adapting and securing the production of biomass with regard to a highly varying demand in the biorefinery sector and to an increasing production risk arising from a changing climate.

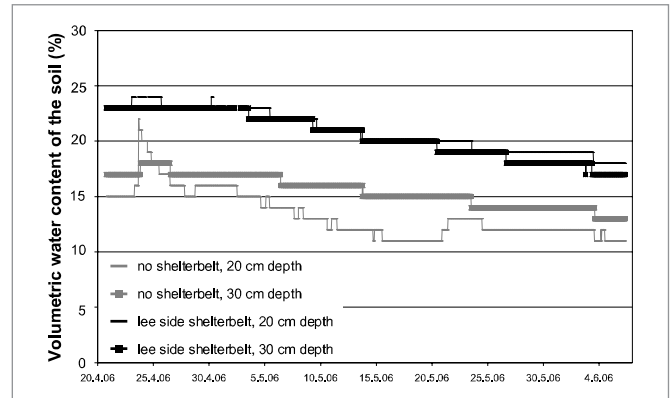


Figure 3:
Change of the soil water contents in intercroppings on the lee side of tree hedges and on adjacent traditionally managed agricultural land without tree shelterbelts

3 Principles of biorefineries

Biomass has similar to petroleum a complex composition. Its primary separation into main groups of substances is appropriate. Subsequent treatment and processing of those substances lead to a whole palette of products. Petrol-chemistry is based on the principle to generate from hydrocarbons simply to handle and well defined chemically pure elements in refineries. In efficient product lines, a system based on family trees has been built, in which basic chemicals, intermediate products and sophisticated products are produced. This principle of petroleum refineries must be transferred to Biorefineries. Biomass contains the synthesis performance of the nature and has another C:H:O:N-ratio than petroleum. The biotechnological conversion will become, beside the chemical, a big player in the future Fig. 4.

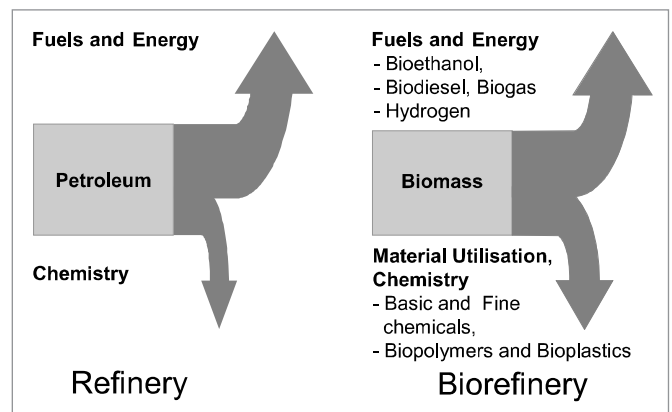


Figure 4:
Comparison of the basic-principles of petroleum-refinery and biorefinery (acc. to KAMM, 2006a)

Thus biomass can already be modified within the process of genesis in such a way, that it is adapted to the purpose of subsequent processing and particular target products already have been formed. For those products the term “precursors” is used.

Plant biomass always consists of the basic products carbohydrates, lignin, proteins and fats, beside various substances such as vitamins, dyes, flavours, aromatic essences of most different chemical structure. Biorefineries combine the essential technologies between biological raw materials and the industrial intermediates and final products (Fig. 5).

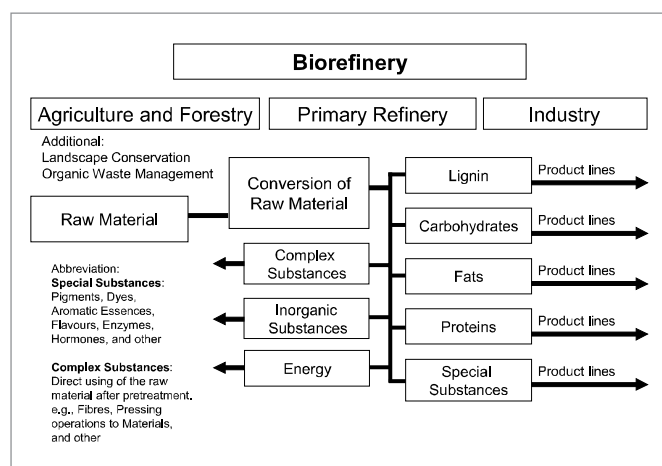


Figure 5: Providing code-defined basic substances via fractionation for the development of relevant industrial product family trees (acc. to KAMM, 2006b)

A technically feasible separation operation, which would allow a separate use or subsequent processing of all these basic compounds, exists up to now only in form of an initial attempt. Assuming that out of the estimated annual production of biomass by biosynthesis of 170 billion tons 75 percent are carbohydrates, mainly in form of cellulose, starch and saccharose, 20 percent lignin and only 5 percent other nature compounds such as fats (oils), proteins and various substances (RÖPER, 2001), the main attention firstly should be focused on an efficient access to carbohydrates, their subsequent conversion to chemical bulk products and 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 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 to chemical hydrolysis is today already realized (LINKO et al., 1996; ZIELINSKA et al., 2000).

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 (KAMM et al., 2006c). These treatments are still mostly thermal, thermo-mechanical or thermo-chemical and require a considerable input of energy. The arsenal for microbial conversion of substances out of glucose is large, the reactions are energetically profitable. It is necessary to combine the degradation processes via glucose to bulk chemicals with the building processes to their subsequent products and materials (Fig. 6).

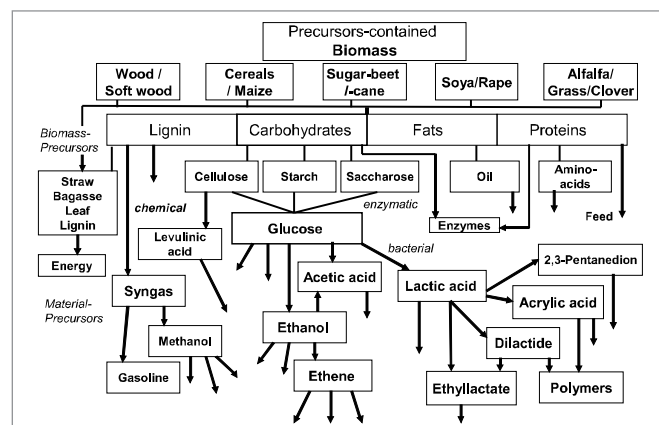


Figure 6: A possible Biorefinery rough-scheme for precursors-containing biomass with preference of carbohydrate line (acc. to Kamm, 2004a, b)

Among the variety of possible from glucose accessible microbial and chemical products, in particular lactic acid, ethanol, acetic acid and levulinic acid are favorable intermediates for the generation of industrially relevant product family trees. Here, to potential strategies are considered: first, the development of new, possibly biologically degradable products (follow-up products of lactic and levulinic acid) or secondly, the entry as intermediates into conventional product lines (acrylic acid, 2,3-pentandion) of petrochemical refineries (KAMM, 2004a).

4 Lignocellulosic Feedstock Biorefinery

Currently four complex biorefinery systems are forced in research and development:

1. the “Lignocellulosic Feedstock Biorefinery” using “nature-dry” raw material such as cellulose-containing biomass and wastes.
2. the “Whole Crop Biorefinery” uses raw material such as cereals or maize.
3. the “Green Biorefineries” using “nature-wet” biomasses such as green grass, alfalfa, clover, or immature cereal (KAMM, 2004a,b).
4. the “Biorefinery two platforms concept” includes the sugar platform and the syngas platform (WERPY et al., 2004).

Among the potential large-scale industrial biorefineries the Lignocellulosic Feedstock (LCF)-Biorefinery will most probably be pushed through with highest success. On the one side the raw material situation is optimal (straw, reed, grass, wood, paper-waste etc.), on the other side conversion products have a good position on the traditional petrochemical as well as on the future biobased product market. An important point for utilization of biomass as chemical raw material is the cost of raw material. Currently the costs for corn stover or straw are: 30 USD/ton; for corn 110 USD/ton (3 USD/bushel) (DALE, 2002).

Lignocellulosic materials consist of three primary chemical fractions or precursors: a) hemicellulose/polyoses, a sugar-polymer of predominantly pentoses; b) cellulose, a glucose-polymer; and c) lignin, a polymer of phenols (Fig. 7).

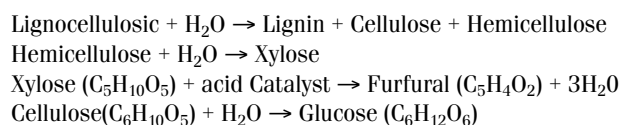


Figure 7:

A possible general equation of conversion at the LCF-Biorefinery

The Lignocellulosic biorefinery-regime has a distinct ability for genealogical trees. Main advantage of this method is the fact that the natural structures and structure elements are preserved, the raw materials have also low price, and large product varieties are possible (Fig. 8). Nevertheless there is still development and optimization demand for these technologies, e.g. in the field of separation of Cellulose, Hemicellulose and Lignin as well as the Lignin utilization in the chemical industry.

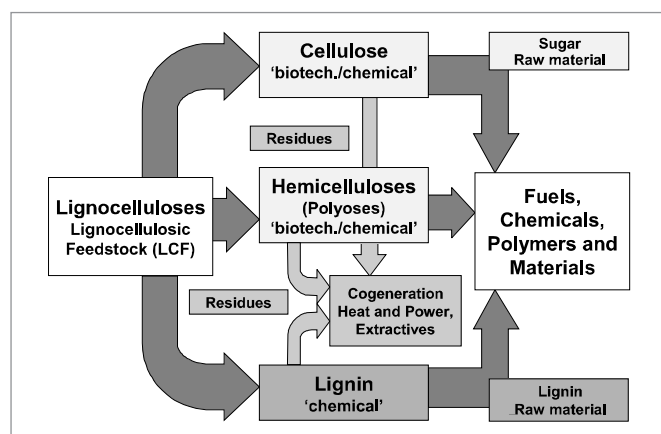


Figure 8:

Lignocellulosic feedstock biorefinery (acc. to KAMM, 2006a)

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 U.S.A. due to economical reasons (the artificial low prize of petroleum). Nevertheless the market for Nylon 6 is huge.

However, there are still some unsatisfactory parts within the LCF, such as utilization of lignin as fuel, adhesive or binder. Unsatisfactory because the lignin scaffold contains considerable amounts of monoaromatic 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 obviously no natural enzymes to split the naturally formed lignin into basic monomers as easy as this is possible for the also naturally formed polymeric carbohydrates or proteins (RINGPFEIL, 2001).

An attractive accompanying process to the biomass-nylon-process is the already mentioned hydrolysis of the cellulose to glucose and the production of ethanol. Certain yeasts give a disproportionation of the glucose-molecule during their generation of ethanol to glucose which practically shifts its entire reduction ability into the ethanol and makes the last one obtainable in 90 % yield (w/w; regarding to the formula turnover).

Based on recent technologies a plant was conceived for the production of the main products furfural and ethanol from LC-feedstock for the area West Central Missouri (U.S.A.). Optimal profitability can be reached with a daily consumption of about 4,360 tons of feedstock. Annually the plant produces 47.5 million gallon of ethanol and 323 000 tons of furfural (VAN DYNE et al., 1999).

Ethanol may be used as fuel additive. Ethanol is also a connecting product for a petrochemical refinery. Ethanol can be converted into ethene by chemical methods. As it is well-known from petrochemically produced ethene, it starts today a whole series of large-scale technical chemical syntheses for the production of important commodities, such as polyethylen, or polyvinylacetate. Further petrochemically produced substances can similarly be manufactured by microbial substantial conversion of glucose, such as hydrogen, methan, propanol, acetone, butanol, butandiol, itaconic acid, succinic acid (ZEIKUS et al., 1999, VORLOP et al., 2005, WERPY et al., 2005). DuPont has entered a 6-year alliance with Diversa in a biorefinery to produce sugar from husks, straw, stovers and develop a processes to co-produce bioethanol and value-added chemicals, such as 1,3-propandiol (Chem World (2003). Through metabolic engineering an Escheria coli K12 microorganism produces 1,3-propandiol (PDO), in a simple glucose fermentation process developed by DuPont and Genencor. In a pilot plant operated by Tate & Lyle, the PDO yield reaches 135 g l⁻¹ at the rate of 4 g l⁻¹ h⁻¹. PDO is used for the production of PTT (polytrimehtylen-terephthalate), a new polymer which is used for the production of high quality fibres branded Sorona (DUPONT 2004). Production is predicted to reach 500 kt per year by 2010.

5 Building blocks, Chemicals and Potential screening

A team from PNNL and NREL submitted a list of 12 potential biobased chemicals (WERPY, 2004). Key area of the investigation have been Biomasse-precursors, platforms, building block, secondary chemicals, intermediates, products and uses (Fig. 9).

The final selection of 12 building blocks began with a list of more than 300 candidates. The shorter list of 30 potential candidates was selected using iterative review process based on the petrochemical model of building blocks, chemical data, known market data, properties, performance of the potential candidates and the prior industry experience of the team at PNNL and NREL. This list of 30 was ultimately reduced to 12 by examining the potential markets for the building blocks and their derivatives and the technical complexity of the synthesis pathways.

The reported block chemicals can be produced out of sugar via biological and chemical conversions. The building blocks can be subsequently converted to a number of high value biobased chemicals or materials. Building block chemicals, as considered for this analysis are molecules with multiple functional groups that possess the potential to be transformed into new families of useful molecules. The twelve sugar based building blocks are 1,4-diacids (succinic, fumaric and malic), 2,5-furan dicarboxylic acid, 3-hydroxy propionic acid, aspartic acid, glucaric acid, glutamic acid, itaconic acid, levulinic acid, 3-hydroxybutyrolactone, glycerol, sorbitol, and xylitol/arabinitol (WERPY et al., 2004).

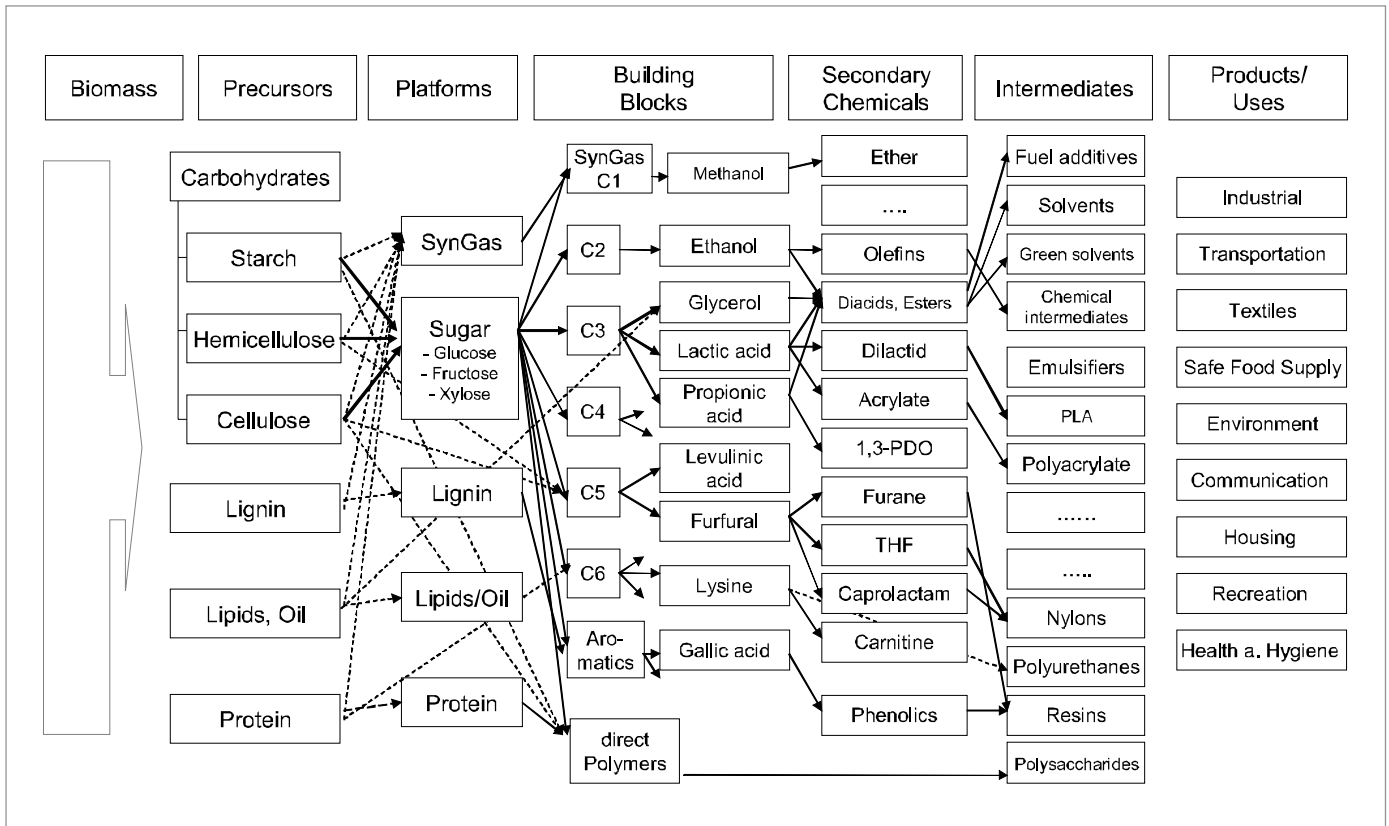


Figure 9: Model of a Biobased Product Flow-chart for Biomass Feedstock, cut out (acc. to WERPY et al., 2004)

A second-tier group of building blocks was also identified as viable candidates. These include gluconic acid, lactic acid, malonic acid, propionic acid, the triacids, citric and aconitic; xylonic acid, acetoin, furfural, levoglucosan, lysine, serine and threonine. Recommendations for moving forward include examining top value products from biomass components such as aromatics, polysaccharides, and oils; evaluating technical challenges in more detail related to chemicals and biologicals conversions; and increasing the suites of potential pathways to these candidates. From Syngas no further down select products was undertaken. For the purposes of this study hydrogen and methanol comprise the best near-term prospects for biobased commodity chemical production because obtaining simple alcohols, aldehydes, mixed alcohols and Fischer-Tropsch liquids from biomass are not economically viable and require additional development (WERPY et al., 2004). Actually a new international research approach to convert lignocellulosic biomass by pyrolysis procedures is prepared at Chair of Mineral Processing (CAO et al., 2006). Main goals are the fractionation of biomass and the supply of homogenous, nearly pure, high quality products by systematic modification of the processing procedures.

6 Direct Polymers – Example Cellulose

Based on share of natural occurrence especially the fibrous, polymeric components of hemicellulose, cellulose, and lignins, collective-

ly named as “lignocellulosics” are a main group of substances for further processing of biomass feedstocks from biorefinery systems.

Particularly cellulose has significant advantages in this context: (1) Cheap and almost everywhere available renewable bio product, (2) Insoluble, mechanically and thermally stable, non abrasive, (3) Easily derivatized to ionic forms, (4) Fibrous morphology, anhydrous crystal structure (Fig. 10).

These characteristics enable the application of cellulose in a multitude of processes and products in its natural form.

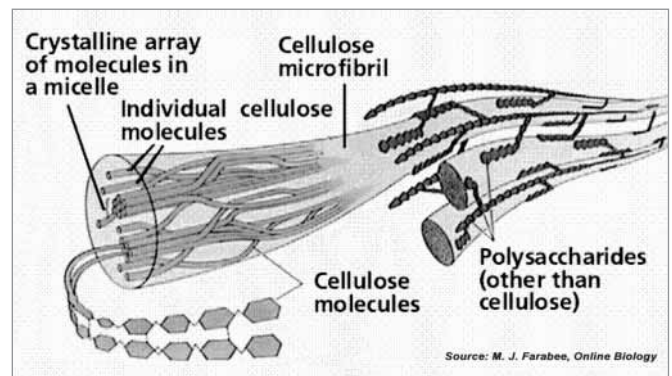


Figure 10: Appearance and structure of cellulose

Already for hundreds of years cellulose fibres resp. fibrils are used in its natural form after mechanical separation from woody or herbaceous biomass. Cellulose and hemicellulose as main components of plant fibres from timber, flax, hemp or similar, are responsible for their special characteristics and make them useful for many textile or technical products. The more or less unique characteristic of crystalline cellulose structure gives plant fibres a lot of advantages such as strength and durability compared to some other man made fibres. Furthermore their (in comparison to glass fibre) lower density favours the use of plant fibres as reinforcement for (bio-)plastics. Several advantages of such composites can be emphasized:

(1) They are environmentally friendly at levels of production, processing and waste, (2) properties comparable to those of materials reinforced with glass fibre, (3) better elasticity of polymer composites reinforced with natural fibres, ability to absorb vibrations and large amounts of energy in case of mechanical impact, (4) Good sound proofing properties, (5) given ability for “energetic recycling”. In particular the Chair of Mineral Processing is engaged in several projects to develop better processing as well as characterization procedures for plant based fibres (GUSOVIUS et al., 2006). As part of a research group new innovative processing and cleaning technologies for native bast fibres like hemp or flax were developed and investigated both in lab as well as semi-industrial scale (GUSOVIUS et al., 2003a, GUSOVIUS et al., 2003b, FÜRLI et al., 2006). New procedure principles for natural fibre agglomeration could be patented as results of further research activities (AY et al., 2005, HILDEBRANDT et al., 2005).

One of today's main markets for native cellulose is the pulp/paper and cardboard industry. Next to the application of wood cellulose in bulk products bast fibre cellulose is employed also to the production of durable special papers.

High quality products like rayon fibres or cellophane can be processed by chemical modification due to solution, too. However, cellulose is always molecularly decomposed by dissolution and reprecipitation (regeneration) in specific scale and furthermore the regenerate distinguishes in crystalline modification and fibre morphology. By handling solid cellulose with not too concentrated acids, first the amorphous, easy accessible areas are decomposed by hydrolysis of the glycosidic ether bonds. The resulting cellulose powder is a high crystalline product and is called microcrystalline cellulose. It can be utilized in medicine (for example as a tablet basic element that acts as a filling or lacing agent) or as an organic filling material.

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Biomass production

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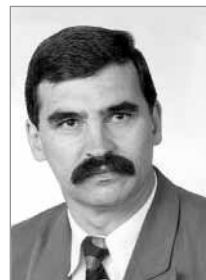
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