



The cellulose resource matrix

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ABSTRACT

The emerging biobased economy is causing shifts from mineral fossil oil based resources towards renewable resources. Because of market mechanisms, current and new industries utilising renewable commodities, will attempt to secure their supply of resources. Cellulose is among these commodities, where large scale competition can be expected and already is observed for the traditional industries such as the paper industry.

Cellulose and lignocellulosic raw materials (like wood and non-wood fibre crops) are being utilised in many industrial sectors. Due to the initiated transition towards biobased economy, these raw materials are intensively investigated also for new applications such as 2nd generation biofuels and 'green' chemicals and materials production (Clark, 2007; Lange, 2007; Petrus & Noordermeer, 2006; Ragauskas et al., 2006; Regalbuto, 2009). As lignocellulosic raw materials are available in variable quantities and qualities, unnecessary competition can be avoided via the choice of suitable raw materials for a target application. For example, utilisation of cellulose as carbohydrate source for ethanol production (Kabir Kazi et al., 2010) avoids the discussed competition with easier digestible carbohydrates (sugars, starch) deprived from the food supply chain. Also for cellulose use as a biopolymer several different competing markets can be distinguished. It is clear that these applications and markets will be influenced by large volume shifts. The world will have to reckon with the increase of competition and feedstock shortage (land use/biodiversity) (van Dam, de Klerk-Engels, Struik, & Rabbinge, 2005).

It is of interest – in the context of sustainable development of the bioeconomy – to categorize the already available and emerging lignocellulosic resources in a matrix structure. When composing such "cellulose resource matrix" attention should be given to the quality aspects as well as to the available quantities and practical possibilities of processing the feedstock and the performance in the end-application.

The cellulose resource matrix should become a practical tool for stakeholders to make choices regarding raw materials, process or market. Although there is a vast amount of scientific and economic information available on cellulose and lignocellulosic resources, the accessibility for the interested layman or entrepreneur is very difficult and the relevance of the numerous details in the larger context is limited. Translation of science to practical accessible information with modern data management and data integration tools is a challenge.

Therefore, a detailed matrix structure was composed in which the different elements or entries of the matrix were identified and a tentative rough set up was made. The inventory includes current commodities and new cellulose containing and raw materials as well as exotic sources and specialties. Important chemical and physical properties of the different raw materials were identified for the use in processes and products. When available, the market data such as price and availability were recorded. Established and innovative cellulose extraction and refining processes were reviewed. The demands on the raw material for suitable processing were collected. Processing parameters known to affect the cellulose properties were listed. Current and expected emerging markets were surveyed as well as their different demands on cellulose raw materials and processes.

The setting up of the cellulose matrix as a practical tool requires two steps. Firstly, the reduction of the needed data by clustering of the characteristics of raw materials, processes and markets and secondly, the building of a database that can provide the answers to the questions from stakeholders with an indicative character. This paper describes the steps taken to achieve the defined clusters of most relevant and characteristic properties. These data can be expanded where required. More detailed specification can be obtained from the background literature and handbooks. Where gaps of information are identified, the research questions can be defined that will require further investigation.

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1. Introduction (why a cellulose resource matrix?)

The emerging bioeconomy is causing a shift, regarding carbon feedstock, from petrochemical to renewable raw materials (OECD, 2009). Established and novel industries that are utilising biomass will require security of their supply, which will be influenced by the market. Cellulose is one of those markets among these commodities, where large scale competition can be expected and already is observed for the traditional industries, such as the paper industry.

Cellulose is one of the most abundant available natural polymers occurring in structural cell wall tissues of all higher plants and some algae (Heinze, Petzold-Welcke, & van Dam, 2012, chap. 2). Also some bacteria are capable of excreting cellulose. Annually many billions of tons of cellulose are produced in nature through photosynthesis from atmospheric CO₂. Only a small part of the total global biological production of woody biomass is utilised for industrial conversion for human consumption. Sustainable production of timber and pulp wood is promoted world-wide and millions of farmers grow fibre commodity crops like cotton, flax, jute, hemp, sisal, coconut coir, etc. Also different recycled cellulose streams are processed on industrial scales (paper, demolition wood, cotton and jute rags). Moreover, there is cellulose containing residues or side streams which are not yet utilised on industrial scale. These low valued streams are equally relevant in the emerging biobased economy as resources. The increasing demand for lignocellulosic raw materials as feedstock coincides with large volumes of underutilised biomass that are available from agro-food industry. It is very important that the stakeholders in industries aiming at utilising alternative cellulose containing raw materials are aware of the price and performance value and volumes available. This information can speed up new developments and support the shift towards renewable resources. Moreover, suitable match of raw materials with the right industry and application, will lead to improvement in the overall sustainability as raw material and logistics efficiency is promoted and value addition is stimulated.

Cellulose is a worldwide important commodity and is utilised in various industries. Specific cellulose containing raw materials are utilised for products like paper, textiles, construction, composites and plastics, but also more and more to produce 'green energy'. Driven by the bioeconomic development, lignocellulosics are currently investigated for their suitability as raw materials for the production of biofuels, 'green' chemicals and materials (Clark, 2007; Lange, 2007; Petrus & Noordermeer, 2006; Ragauskas et al., 2006; Regalbuto, 2009). The use of cellulose as raw material provides an alternative source of carbohydrates and prevents the debated competition with food production (corn starch, sugar cane, which are easier fermentable to bioethanol) (Kabir Kazi et al., 2010). There are also different competing bulk production markets for celluloses that are sensitive to effects of declining resource supplies. Bioeconomic issues like competing land use, water claims, nutrients scarcity, progressing deforestation and its effects on biodiversity are critical for developments of sustainable production. Therefore, the quantitative ecological footprint or whole life cycle impact of cellulosic products on the environment need to be considered in this context (Wellisch, Jungmeier, Karbowski, Patel, & Rogulska, 2010). Besides collection of data on the techno-economics of crop production, processing and use also the sustainability criteria for the production and consumption chain of the different ligno-cellulosic commodities are to be transparently compared (van Dam, 2008; van Dam & Bos, 2004).

Potentially interesting lignocellulosic resources are diverse and of varying properties. Therefore it is needed to categorize these raw materials to identify the most appropriate utilisation route and application. In order to compose such "cellulose resource matrix" attention needs to be paid both to qualitative aspects (purity, contaminants, crystallinity) and quantitative data of the biomass

source (production volume, current use). Moreover, the practical processability of the biomass determines its suitability for a certain end-use.

Therefore a matrix structure is composed to deliver this information.

The objective of the cellulose resource matrix is to serve as a practical tool that allows stakeholders to make a founded selection regarding certain lignocellulose raw material options, its processing or market. The detailed objectives include;

- Allowing the raw material producer to select the most suitable process to supply a certain market based on the matrix data providing information on the existing processes and competing raw materials
- Allowing the owner of a process – the raw material processing industry – to determine which alternative raw materials can be processed and identify which potential new markets he can supply
- Allowing the producer of cellulose based end-products to determine which raw materials and process combinations are most suitable or more competitive for their products
- Allowing the cellulose trade to supply alternative (local) products of similar quality, enabling a better logistic organisation and reduction of transportation distances
- Helping governments to obtain background information about the possibilities of stimulation of sustainable developments

A detailed matrix structure containing entries of functions and products was composed. Underlying existing documentation was converted and structured and was placed according the various defined elements.

2. Method

The amount of literature and information on cellulose is almost inexhaustible. For the decision making process it is very relevant to arrange and cluster this vast amount of information. It was decided to arrange the information in three clusters. Primarily, in the first column A information is collected about the various raw materials that contain cellulose and are currently industrially applied, or have potential as new raw materials (Fig. 1). Secondly, the information was clustered on the processing technologies in column B for (ligno)cellulose extraction and conversion, that are used on industrial and pilot scales, or are expected in the near future. Thirdly, the current and new application areas and markets in column C for cellulose and cellulose containing products were classified. These clusters of information are logically linked. In the processing column (Fig. 1) the information is given on demands for the input of feedstock for each process. In the market column the demands are listed on the required product performance. From such data arrangements the cellulose resource matrix is created. The cellulose matrix provides links between raw materials, processing and end-use markets. In Fig. 1 the matrix is represented graphically.

The data inventories and overviews of raw materials, conversion processes and uses are the core of the cellulose resource matrix. Linking arrows between entries show that different biomass feedstock may be used for the same process and different processes may yield similar products.

The raw materials column A is characterized by two data sets. The first contains entries with technical information on the chemical and physical properties of specific (ligno)cellulosic source (literature references). The second data set contains entries of economic information on the availability (production volume, price ranges, etc.).

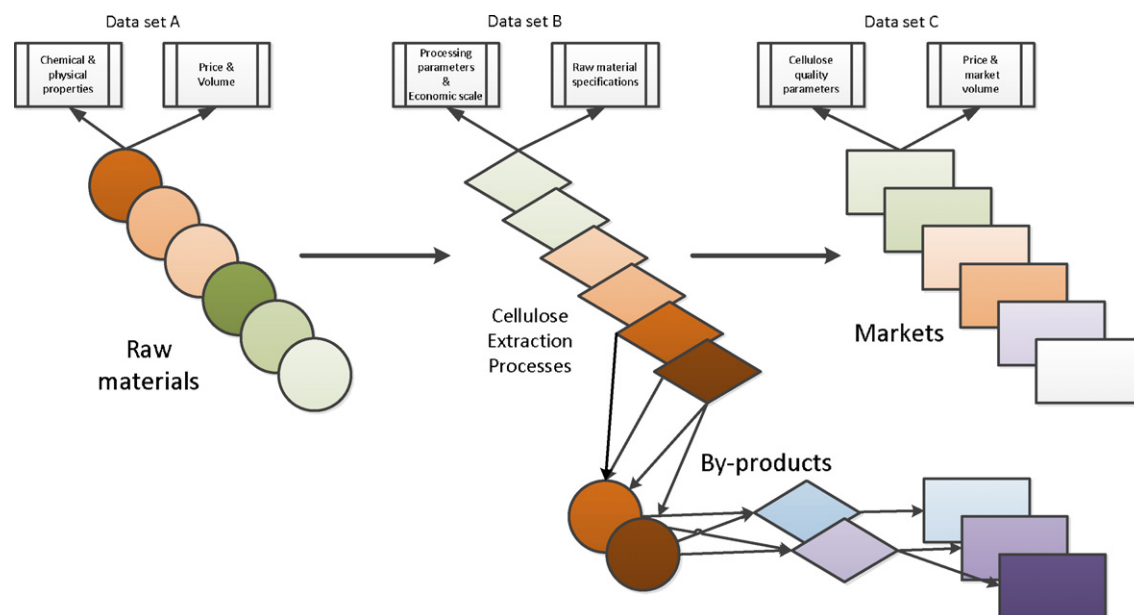


Fig. 1. Graphical reproduction of the cellulose matrix structure.

The cellulose processing column B also is characterized by two data sets. The entries in the first data set are the technical information on the processing parameters, yields and operational scales. The second set provides entries with information on the technical and economic demands of the process on raw materials. The combination of raw material of column A and cellulose extraction/conversion processes of column B leads to non-cellulosic by-products which further are not taken into consideration here in the matrix. The column C describes the market data in two data sets. The first technical data set describes the quality demands on cellulose by the market. The second data set provides entries of market information on the market prices and the volume of the various cellulose applications.

3. Inventory raw materials, processes and markets

3.1. Inventory raw materials

Cellulose containing raw materials are subdivided into primary, secondary and tertiary raw materials. Primary raw materials are the cultivated raw materials for the cellulose fibre production purpose. Secondary raw materials include by-products of (food) crop cultivation. Tertiary raw materials are released from conversion processing and recycling of biomass products. So, besides wood and the well-known fibre crops (van Dam, van Vilsteren, Zomers, Hamilton, & Shannon, 1994) also the residues and cellulose containing recycling streams are incorporated. It should be noted that this inventory is not intended to be complete. The inventory will encompass new crops or industrial side streams, that may appear with changing cellulose compositions. Residues and by-products often vary strongly in composition and quality depending on changes in raw material and the processing conditions.

The raw materials categorizing is relevant because, in case of lack of detailed information on a certain raw material, the properties of a related product can be considered. Unfortunately, most often the differences between raw materials are too big for using the data specifications of one raw material simply for the other. For many processes and products it is the small differences for the layman that are critical and make distinctive difference. In Table 1 an overview is given of the classification of lignocellulosic raw materials.

3.2. Inventory cellulose processing

The cellulose extraction and refining processes include the current paper pulping processes as well as processes applied in bio-refineries. Although these processes are often suitable for conversion of different raw materials, the processing parameters are optimised for just one feedstock. Change of feedstock often requires adaptation, for example: change of temperature, pressure or residence time or change of chemical loads. In Table 2 the different industrial and laboratory processes are listed.

3.3. Inventory of markets

The overview of cellulose markets is very diverse and it comprises all possible uses such as building applications, textiles, non-wovens, paper and board, but also dissolving cellulose, fibre reinforced composites and cellulosic films. Moreover, the growing demand for cellulose based fuels and green chemicals need to be anticipated. In Table 3 the various cellulose applications and commercialized products are classified. These markets are subdivided according product groups. The different demands on cellulose specifications within a product group are comparable, but may differ on detail level.

4. Characterization of raw materials, processes and markets

The raw materials are characterized by two different data sets. The entries of chemical and physical specifications are 'fixed' properties of the raw materials. Economic data set on price and availability may be variable or sometimes even not available.

4.1. Data set (A1): chemical and physical properties of raw materials

This data set of chemical and physical properties of lignocellulosic raw materials contains the cellulose content percentage and other accompanying components (hemicellulose, pectins, lignin, extractives, ash) in the raw material (Thomas, Paul, Pothan, & Deepa, 2011, chap. 1). The physical properties are related to the dimensions of the cellulose fibres. The data collected in this set, are directly linked to the raw material demands and the cellulose

Table 1
Classification of cellulose raw materials (A).

			Examples	
Primary cellulose sources				
1	Softwoods (Gymnospermae)	Pine Spruce Fir	<i>Picea abies</i> <i>Pinus sylvestris</i> <i>Abies alba</i>	Pinaceae Pinaceae Pinaceae
2.1	Temperate hardwoods (Angiospermae)	Birch Beech Oak Aspen	<i>Betula pendula</i> <i>Fagus sylvatica</i> <i>Quercus rubra</i> , <i>Q. robur</i> <i>Populus tremuloides</i>	Betulaceae Fagaceae Fagaceae Salicaceae
2.2	Tropical hardwoods	Eucalyptus Azobé Ipé Meranti, Bankirai Merbau Purperhart	<i>Eucalyptus globulus</i> <i>Lophira alata</i> <i>Tabebuia</i> sp <i>Shorea</i> sp <i>Intsia bijuga</i> <i>Peltogyne</i> sp	Myrtaceae Ochnaceae Bignoniaceae Dipterocarpaceae Fabaceae Fabaceae
3	Non-wood fibre	(Dicotyledons)		
3.1	Seed hairs/fluff or floss fibres	Cotton	<i>Gossypium</i> sp	Malvaceae
3.2	Bast fibres	Jute Kenaf Hemp Ramie Flax	<i>Corchorus capsularis</i> , <i>C. olitorius</i> <i>Hibiscus cannabinus</i> <i>Cannabis sativa</i> <i>Boehmeria nivea</i> <i>Linum usitatissimum</i>	Tiliaceae Malvaceae Cannabaceae Urticaceae Linaceae
3.3	Hardfibres	(Monocotyledons)		
	Leaf fibres	Sisal	<i>Agave sisalana</i>	Agavaceae
	Stem fibres	Abaca	<i>Musa textilis</i>	Musaceae
	Seed hull fibre	Coir	<i>Cocos nucifera</i>	Arecaceae (Palmae)
3.4	Grasses and reeds	(Monocotyledons)		Poaceae (Graminea)
	Stem fibres	Esparto Bamboo Miscanthus	<i>Stipa tenacissima</i> <i>Phyllostachys heterocycla</i> <i>Miscanthus sinensis</i>	
3.5	Brush/piassava fibres	Raphia	<i>Raphia hookeri</i>	Arecaceae (Palmae)
3.6	Miscellaneous	Viscan Mulberry	<i>Viscum album</i> <i>Brousonetia papyrifera</i>	Santalaceae Moraceae
3.7	Specialties	Bacterial cellulose	<i>Enterobacter aerogenes</i> <i>Acetobacter xylinum</i>	Enterobacteriaceae Acetoacteriaceae
		Algal cellulose	<i>Valonia</i> sp	Valoniaceae
		Tunicates	<i>Halocynthia roretzi</i>	Pyruridae
Secondary cellulose sources				
4	Forestry residues			
4.1	Softwood	Bark, branches, needles, cones, sawdust		
4.2	Hard woods	Bark, branches, leaves, seeds, sawdust		
5	Agro-residues/agri-food residues			
5.1	Cereal grain straws and	Wheat straw, chaff Rice straw, husk Corn stover	<i>Triticum vulgare</i> <i>Oryza sativa</i> <i>Zea mays</i>	Poaceae (Graminea)
5.2	Starch (tuber) crop residues	Potato stalks and peelings	<i>Solanum tuberosum</i>	Solanaceae
5.3	Sugar crop residues	Sugar cane bagasse Beet pulp	<i>Saccharum officinarum</i> <i>Beta vulgaris</i>	Poaceae (Graminea)
5.4	Oil crop residues	Oil palm hulls, stems, stalks, fruit bunch Soybean stem hulls Linseed straw	<i>Elaeis guineensis</i> <i>Glycine max</i> <i>Linum usitatissimum</i>	Arecaceae (Palmae) Fabaceae (Papilionaceae) Linaceae
5.5	Fruits and nuts, prunings and residues			
5.6	Flowers and gardening, byproducts and residues			
5.7	Nature management and conservation biomass waste and verge grasses			
Tertiary cellulose sources				
6.1	Recollected textile waste			
6.2	Primary pulp mill residues	Paper sludge, deinking sludge		
6.3	Municipal solid waste		Organic waste	
6.4	Building demolition waste		Timber and wood working residues	
6.5	Manure		Organic waste	
6.6	Compost/dredging's		Organic waste	

contained by the processing conditions and the end product (Sections 4.3.4 and 4.4.3).

The chemical composition of a raw material is not just a simple listing of the components contained. For the cellulose extraction process the ratio and interaction between the various components are just as relevant. The exact constituents and linkages between cellulose fibrils, hemicellulose, lignin and the other components

determine the suitability of a lignocellulosic raw material for a certain extraction process. Because of this, very different requirements for the lignocellulosic raw materials are defined by processes and markets.

The physical properties of cellulosic fibres need to be established in different dimensional scales. For different markets the properties of fibre bundles (cm), elementary fibre cells (mm),

Table 2

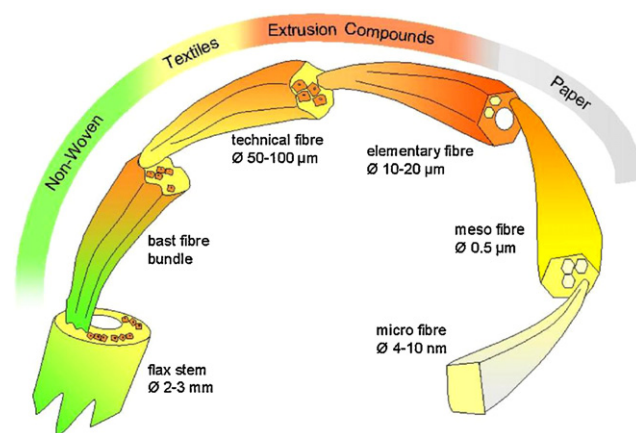
Classification of cellulose extraction and conversion processes (B).

1	Cellulose primary extraction	
1.1	Harvesting	
	Wood	Felling, logging, debarking Storage and transport, drying sawing, chipping, pelletizing, Picking, pulling, cutting or mowing
	Fibre crops	
1.2	Postharvest fibre crop processes	
	Cotton	Ginning, baling, Retting, scutching, hackling Stripping, decorticating, drying
	Bast fibres	
	Leave fibres	
2	Textile processes	
2.1	Yarn formation	Carding, drawing, combing, roving
2.2	Yarn spinning	Ring spinning; open end (rotor) spinning; air jet (vortex) spinning
2.3	Fabric formation	Weaving/knitting
2.4	Non-woven processing	Needle punching; wet laid or hydro-entanglement; air laid; chemical bonding; Spinning, twining
2.5	Rope making	
3	Pulping processes	
3.1	Mechanical pulp (groundwood pulp)	
3.2	Thermo-mechanical pulp (TMP)	
3.3	Chemi-thermomechanical pulp (cTMP)	
3.4	Kraft pulping	Sulphate process (90%)
3.5	Sulfite pulping	Acid pulping (10%)
3.6	Alkaline pulping	Soda pulping
3.7	Organosolv pulping	Ethanol, methanol pulping
3.8	Acetosolv pulping	Acetic acid/formic acid pulping
4	Biorefinery and lignocellulose processing	
4.1	Steam explo- sion/ultrasonication/hydrothermal microwave	
4.2	Hydrolysis (enzymes/acid)	
4.3	Pyrolysis/hydrothermal liquefaction/hydrogenation (HTU)/syngas	
4.4	Carbonization	
4.5	Biopulping/white rot fungi/aerobic & anaerobic fermentation	
5	Dissolving cellulose	
5.1	Viscose process	
5.2	Lyocell process (NNMO)	
5.3	Other processes e.g. (ammonia/phosphoric acid)	
5.4	Ionic liquids	
5.5	Nanocellulose	
5.6	Cellulose derivatives	

Table 3

Classification of cellulose markets (C).

Market	Products
1 Textiles	Fabrics of cotton, linen, viscose, lyocel, acetate
2 Non-woven	Needle punched, wet laid, air laid, hygienic tissues, diapers, wipes, filters
3 Wood and timber	Building and construction, furniture, carving and wood working
4 Pulp, paper and board	Newsprint, writing, specialty paper, corrugated boards
5 Cellulose dissolving pulp	Viscose, lyocel, cellulose derivatives, CA, CMC, HEC
6 Cellulosic films	Packaging, membranes,
7 Building materials	Veneer, plywood, fibre boards, insulation
8 Cellulosic fibre composites	Moulded compounds (Sheet, compression, or injection compounds), laminates, mineral matrix fibre composites
9 Green chemicals	Bioethanol, organic acids, furans, biopolymers

**Fig. 2.** Graphical representation of the different fibre dimensions of a bast fibre (flax) cellulose.

or micro- (μm), or nano-fibrillar (nm) building elements are relevant.

4.1.1. Overview of essential data

Chemical data, commonly provided in literature of lignocellulosic raw material compositions, firstly consist of its relative contents of carbohydrates, extractives and minerals:

- Polysaccharides and sugars
- Cellulose
- Hemicellulose
- Pectins
- Lignin
- Other plant components

The lignocellulose raw material is furthermore characterized by a number of other variables:

A. Fibre dimensions

- Fibre length
- Diameter
- Lumen
- Cell wall thickness

B. Cellulose properties

- Cellulose fibre strength properties
- Microfibril orientation
- Density
- Polymerisation degree molecular mass distribution
- Swelling
- Solubility in alkali/ionic liquids

C. Cellulose quality parameters

- Purity
- DP
- Crystallinity/amorphous phase

A. Fibre dimensions: Cellulosic fibres from plants are widely diverse (van Dam & Gorshkova, 2003, chap. MS 46; Willför, Alén, van Dam, Liu, & Tähtinen, 2011, chap. 1). One can consider the fibre bundle, well known from bast fibre crops, the elementary fibre cells, microfibrils and nano-fibres (Fig. 2). Fibre bundles are composed of aligned bundled elongated plant cells. The elementary fibre cell is a highly specialized cell with thickened cell walls with deposited

differently oriented cellulose microfibril layers. The microfibrils are composed of cellulose crystallites which are of nano-scale dimensions. The cellulose polymerisation degree and the ratio crystalline and amorphous cellulose are relevant for the fibre strength properties, and its susceptibility for chemical or enzymatic hydrolysis. Fibre length and diameter ratios can be important and are determined for each magnification scale. The lumen is the hollow space inside an elementary cell, while also some fibre bundles may contain vacuoles. The size of the lumen is determined in transverse direction so the total diameter of the fibre is composed of the lumen and twice the cell wall thickness. Fibres with high length to diameter ratio for example can be especially suitable for the reinforcement of matrix composite materials. Fibres with a small lumen and thus a relatively thick cell wall are commonly more stiff.

B. Cellulose properties: Cellulose fibre strength properties can be determined by specially designed tensile tests. The tensile strength will be affected by the various processing steps. These may be of both chemical and mechanical nature.

Microfibril orientation in the cell walls of the elementary fibres commonly are not parallel to the fibre direction. The angle in which the fibres are deposited is of influence on the elongation and elasticity (Nishiyama, 2009; Wang, Drummond, Reath, Hurst, & Watson, 2001). When a strain is put on the fibre, the microfibrils are dislocated relative to each other. The maximum elongation is often small for fibres with a small fibre axis angle (5–10°). Coir fibres for example have a larger fibrillar angle (ca. 45°) explaining the typical high elongation of coir (van Dam et al., 2006).

The density of pure cellulose is almost 1.5 kg/dm³. Due to the presence of pores, lumen and vacuoles in the fibres the density of the various fibres are substantially lower (Mwaikambo & Ansell, 2001).

The polymerisation degree and molar mass distribution of cellulose fibres are of high influence on the fibre mechanical properties (Nishiyama, 2009). The number of glucose units building the cellulose differs significantly for each plant and the plant tissue where the cellulose is extracted from. During the chemical and mechanical processes for cellulose extraction the bonds may be broken. This results in a decrease of the polymerisation degree (DP). For high quality dissolving cellulose demands on the remaining DP are critical.

Swelling and dissolution behaviour of a cellulose fibre (Cuissinat & Navard, 2008) is strongly affected by the composition of the fibre and the presence of pores, and by the pore size and crystallinity degree of the cellulose (Leppänen et al., 2009). Fibres that show easy swelling are more susceptible for chemical treatments like dyeing and depolymerisation, but have lower dimensional stability.

The solubility of cellulose is very poor in most solvents. But for several chemical processes and derivatization reactions the solubility of the polymeric structure is desired. For example complete solubility is important in the regenerated cellulose production (Jin, Zha, & Gu, 2006) or cellulose acetate and carboxymethylation reactions (Heinze, Schwikal, & Barthel, 2005).

In Table 4 the chemical and physical values of different lignocellulosic fibres are given.

4.2. Data set (A2): price and availability of the lignocellulosic raw materials

The price and availability of the various raw materials depend on location and moment. In this data set the local influences and access to transportation are to be considered besides the global market values and the volumes of feedstock production. The actual availability of a commodity raw material is affected by many factors, including for example politics and stock market speculations (van Dam et al., 2005). The completion of such data set and keeping it updated is almost impossible. However, it is possible to provide

basic information on main market trends and to illustrate those by focussing on specific topics.

4.2.1. Overview of most relevant economic data

The price and availability of various raw materials is very much dependent on region or country. Besides the world production data, as provided for example by the FAO statistics (FAO, 2011) or CEPI (CEPI, 2011), a regional actual data set may be needed. This data set for the primary lignocellulosics consists in:

- Production area (ha)
- Production volume (ton)
- Production yields (ton/ha)
- Dry matter/cellulose content (ton/ton)
- Price (€(\$)/ton)

For the secondary and tertiary resources similar data may be provided in the completed data set. In Table 5 some examples on the price and availability of lignocellulosic feedstocks are given.

4.3. Classification of cellulose extraction processes

Classification of the various cellulose extraction processes is complex because of that each process can be performed in a range of different ways. Optimization of the processing conditions, introduction of new catalysts or enzymes may change the way raw materials are converted to products. Current optimised production scale or yield maximized extraction processes may change due to shifting economic circumstances or alliances with other industrial sectors.

4.3.1. Data set (B1): processing parameters and economic scale of operation

The cellulose extraction processes are characterized by the technical processing parameters applied (temperature, pressure, chemicals, time) for efficient conversion but also by the economic scale of operation (Kaylen, Van Dyne, Choi, & Blase, 2000). These processing conditions largely determine the suitability of a raw material for a certain use. This data set does not provide all details of the technical process but focuses on the processing parameters that will affect the cellulose properties.

4.3.2. Overview relevant data

Important parameters that are included in this data set are:

- Maximum and minimum pressure
- Maximum and minimum temperature
- Processing time
- Chemical load
- Mechanical impact
- Economy of scale
- Energy consumption

In this data set (B1) the connection can be made with released by-products and residues, that may compete or influence the selection of process conditions and economic value of the whole crop. Also the quantitative technical data of energy and chemicals consumption, waste disposal and emissions can be included here. In Table 6 the most important processing parameters are given.

4.3.3. Data set (B2): raw material specifications for cellulose processing

The different processes for cellulose production have different requirements for the raw materials to comply with. When a lignocellulosic raw material does not comply these requirements, it is impossible to produce an interesting product for the market with

Table 4

Data set (A1): chemical and physical properties of selected lignocellulose raw materials.

Raw material	Chemical composition					Fibre dimensions		Cellulose properties	Cellulose quality	
	Cellulose	Hemicell	Lignin	Extr	Ash	L (mm)	D (μm)		DP	Crystallinity (CrI)
Primary Cellulose sources^a										
Softwood	43–45	20–30	25–30	2–9	0.4	2.7–4.5	20–45	Long tracheid fibre	10.000	50–55%
Hardwood	40–55	25–30	16–24	2–5	0.2	0.7–2.5	20–30	Short parenchyma and vessel cells	10.000	40–60%
Cotton	85–90	0.1–3	0.4	2.8	1.6	18–25	20	Highly pure, twisted	3000–15.000	60–80%
Bast and leaf fibres (jute, ramie, flax, hemp, sisal)	55–75	7–15	3–15	8–10	1–4	2.5–6.0 (>30)	20–50	Long aligned bundles of strong fibre cells	6500–8000 (>15.000)	55–90%
Grasses and reeds	40–45	20–30	20–30	2–5	5–12	1.0–2.7	10–20	High content of fines, silica	1500–8000	40–50%
Secondary Cellulose sources										
Mixed wood (forestry/saw dust)	20–50	2–25	–	–	0.2–20	0.7–4.5		Heterogeneous		
Straws (Agro residue)	35–45	15–30	10–20	3–17	5–15	0.5–1.5	8–15	High content of fines and silica		
Tertiary cellulose sources^b										
Recollected textile waste	Depending on recollection and sorting process of cotton, viscose and acetate fabrics from synthetics							Heterogeneous mixed with synthetic en semi-synthetic fibres	Mixed cellulose I (cotton) and II (viscose) and derivatives (CDA); bleached dyed and finished	
Primary pulp mill residues/deinking sludge	35–45% cellulose in wet fraction (50% dm) with high ash (15–25%) and lignin (20–30%) content							Mechanical damaged/mainly short fibers		
Municipal solid waste	30–35% cellulose containing biomass							Cellulose content variable mixed in organic fractions with plastics,		
Building demolition waste	ca. 5% weight non-mineral residues composed of synthetic polymers, wood and fibre boards							Heterogenic fraction of wood and wood fibre products with paint/glue and metal contaminants		
Manure	Cattle manure (<30% dm) contains ca 27% cellulose/17% hemicellulose/9% lignin dm basis (Chen et al., 2003)							Heterogenic fraction of fibre/partly decomposed		
Compost/dredgings	nd							Heterogenic fraction of fibre/partly decomposed		

nd: no data available.

^a See reviews and handbooks e.g. Willför et al. (2011), chap. 1, van Dam and Gorshkova (2003), chap. MS 46.^b See: Broder, Harris, and Ranney (2001), Chalmin and Gaillochet (2009), Chen et al. (2003), IEA Bioenergy (2003), Shi, Ebrik, Yang, and Wyman (2009), Werner (2008).

Table 5

Data set (A2): price and volume of the raw materials (estimate 2011).

Raw material	Total production (ton/year)	Price range €/t	
Primary cellulose sources			
Softwood + hardwood	1200–1500 × 10 ⁶	Pulp	400–450
Cotton	27 × 10 ⁶	Pulp	2500–4000
Jute	2.5 × 10 ⁶	Raw jute	350–700
Other fibre crops (flax hemp, kenaf, ramie, sisal, coir)	Each between 90 and 500 × 10 ³	Flax/hemp/sisal	300–600
		Flax/hemp linen	700–1000 (short)
		Textile quality	1600–2000 (long)
		Abaca pulp	1500–2500
		Baled	40–150
Grasses and reeds	— ^a		
Secondary cellulose sources			
Mixed wood (forestry)	— ^a	Mixed wood	10–20
		Pallets	90
		Saw dust	200
Straws (Agro residue)	350–730 × 10 ⁶	Baled	40–150
Tertiary cellulose sources			
Recollected textile waste	550–900 × 10 ³ (UK)/10 × 10 ⁶ (USA)/75 × 10 ³ (NL)	— ^a	
Primary pulp mill residues	100.000(?) ^a	— ^a	
Municipal solid waste	243 × 10 ⁶ (USA); 8 × 10 ⁶ (NL)	–31 (cost of disposal)	
Building demolition waste	25 × 10 ⁶ (NL)/3% wood	10	
Manure	— ^a	— ^a	
Compost/dredging	— ^a	— ^a	

^a No data available.

the chosen process. This data set contains the current processing demands. Further development and adaptation of the biorefineries or cellulose extraction processes may change the feasibility of using a particular raw material.

4.3.4. Overview of most relevant data

The requirements that are dictated by a process are linked to the cellulose quality that needs to be produced. The demands on raw materials can be clustered around a number of specific aspects:

- Physical properties e.g. size, dry matter
- Chemical composition e.g. interaction between cellulose/hemicellulose and lignin
- Unwanted components e.g. extractives and minerals (ash/silica)
- Prior treatments of raw material
- Physical, chemical, biological stability of the cellulose

In Table 7 an overview is prepared of the most relevant parameters for cellulose processing.

Table 6

Processing parameters (B1) and economic scale of cellulose extraction.

Process	Scale	Yield	Process conditions			
			Pressure	Temp (°C)	Chemicals	Mechanical impact
1 Cellulose extraction						
1.1 Harvesting/storage and transport	Large		Ambient	Ambient	Low	Low
2 Textile processing	Large		Ambient	Moderate	High	Low
3 Pulping processes^a						
3.1 Mechanical pulp (groundwood pulp)	Medium	95%	Ambient	Moderate	n.a.	High
3.2 Thermo-mechanical pulp (TMP)	Medium	93%	Moderate	Moderate	Low	High
3.3 Chemithermomechanical pulp (cTMP)	Small	90%	Moderate	120–140	Moderate	High
3.4 Kraft pulping 90% (sulphate process)	Large	40–55%	High	150–170	High	Low
3.5 Sulfite pulping 10% (acid pulping)	Large	40%	High	125–140	High	Low
3.6 Alkaline pulping	Large	40–50%	High	120–165	High	Low
3.7 Organosolv pulping (Alcell/ASAM/Organocell)	Small	50%	High	180–190	High	Low
3.8 Acetosolv pulping (Acetocell/Formacell/MILOX)	Small	50%	Moderate (low)	150–200 (80–100)	High	Low
4 Biorefinery and lignocellulose processing						
4.1 Steam explosion/ultrasonication	Small	60%	High	180–220	Low	High
4.2 Hydrolysis (enzyme)	Small		Ambient	Moderate	Low-enzymatic	Low
4.3 Pyrolysis/hydrothermal liquefaction/hydrogenation (HTU)/syngas	Small	— ^b	High	400–600	Low	Low
4.4 Activated carbon	Small		High	600–900	n.a.	Low
4.5 Biopulping/white rot fungi	Small		Ambient	Moderate	Low-enzymatic	Low
5 Dissolving cellulose						
5.1 Viscose process	Small		Low	20–30	High	Low
5.2 Lyocell process NNMO	Small		Vacuum	<100	High	Low
5.3 Other processes; ammonia/phosphoric acid	Small	— ^b	Low	45	High	Low
5.4 Ionic liquids (IL)	Small		Low	70–140	High	Low
5.5 Nanocellulose	Small		High		High	High
5.6 Cellulose derivatives	Medium		High	High	High	Low

^a Willför et al. (2011), chap. 1.^b Data not available.

Table 7

Overview of major demands on feedstock per cellulose process (B2).

		Cellulose properties	Contaminants and residues	Pretreatments
1	Cellulose extraction			
1.1	Harvest storage and transport	Mature crop/dried (<10% moisture)/compressed bales	Wood particles/lignin/tannin/resin	Retting/breaking/scutching debarking/hackling
2	Textile processing	Cleaned (bast) fibres: (A) Long fibre bundles Wet spinning length > 30 cm); strength > 50 cN/tex; fineness > 40 Shirley; Dry spinning length > 8 cm (B) Short fibres (cottonised) (ring spinning) (2.5–4.5 cm) (C) Fibre blends (D) Solvent spinning	> 95% pure (wood free), colour homogeneity, fineness ca. 50–80% cellulose Highly pure dissolving cellulose	Carding/bleaching/twining/
3	Pulping processes^a			
3.1	Mechanical pulp (groundwood pulp)	Unpurified wood particles chips	Compression wood/bark	Debarked logs/chips. Milling at high moisture >30–45%
3.2	Thermo-mechanical pulp (TMP)	High lignin % fibres	Mix of long and short fibres; rosin	Chips steam pressure refining
3.3	Chemithermomechanical pulp (cTMP)	High yield long fibres	Rosin and small amount black liquor	Chips pretreatment with NaOH/Na ₂ CO ₃
3.4	Kraft pulping (sulphate process)	Strong fibres	Rejects coarse particles/compression wood; yields black liquor, tall oil	Chips Alkaline (NaOH/Na ₂ S) sulfate cooking
3.5	Sulfite pulping (acid pulping)	Weaker fibres, simple bleaching	Yields lignosulphonates	Acidic (Na ₂ SO ₂) sulfite cooking
3.6	Alkaline pulping	Non-wood pulps	Yields soda lignin	Alkaline cooking
3.7	Organosolv pulping	Lower pulp quality, suitable for hardwood and non-wood pulps	Yields pure organosolv lignin	Methanol/ethanol cooking
3.8	Acetosolv pulping/MILOX	Hydrolysis of hemicellulose	Formation of furfural	Acetic/formic acid cooking
4	Biorefinery and lignocellulose processing^b			
4.1	Steam explosion/ultrasonication	Mechanical damage/higher porosity/low hemicellulose %	Compression wood/butts	Steam pressure impregnation
4.2	Hydrolysis (chemical/enzymatic)	Chemical or enzymatic depolymerisation to cellobiose/glucose	Pentose/lignin	Lignin extraction
4.3	Pyrolysis/hydrothermal liquefaction/hydrogenation (HTU)/syngas	Thermal conversion/degradation cellulose/hemicellulose and lignin	Char/ash	Whole biomass conversion
4.4	Activated carbon	Thermal conversion	Non-cellulose	Microcrystalline cellulose 800–950 °C
4.5	Biopulping/white rot fungi	Partial digested	Fungal biomass	Inoculation wetted biomass
5	Dissolving cellulose			
5.1	Viscose process	Cellulose (II)	None	Dissolving cellulose of highest purity NaOH, H ₂ S, Xanthate
5.2	Lyocell process NMMO	Cellulose (II)	Minimal	Dissolving cellulose of less purity; N-methylmorpholine-N-oxide (NMMO)
5.3	Other processes ammonia/phosphoric acid	Cellulose (III)/preserved morphology	None	Dissolving cellulose of high purity in Liquid ammonia or Super phosphoric acid (74%)
5.4	Ionic liquids (DMAc/LiC–BIMiM and other)	Cellulose (II)	None	Dissolving cellulose of highest purity
5.5	Nanocellulose	10–30 nm fibril diameter 100–1000 nm length	None	Dissolving cellulose of highest purity/high pressure/temperature homogenisation desintegration/microfluidizing

^a Willför et al. (2011), chap. 1.^b Reith et al. (2010).

4.4. Identification and characterization of markets

The cellulose markets are agile and developing permanently, influenced by global economic trends and shifts in supply and demand (Rustemeyer, 2004). Niche products like specialty cellulose have a higher price than commodity bulk products (Fig. 3), but may be less influenced by cyclic fluctuations in supplies.

Substitution of one raw material with an alternative, more environmental sustainable source, evokes changes in prices and demands. This is certainly the case when a cellulose based product is replacing a fossil carbon based product. A change in colour, texture or quality of a market product may restrict the commercialization. However, the basic quality aspects will remain valid for cellulose based raw materials and products.

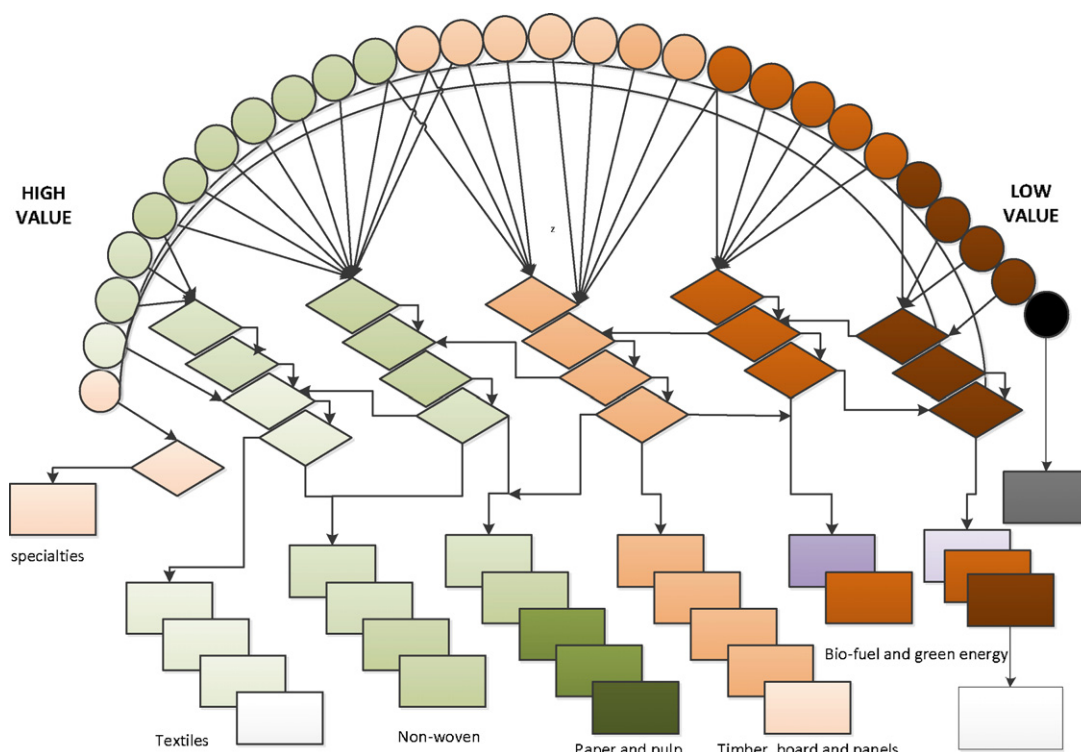


Fig. 3. Schematic representation of the different lignocellulosic raw materials and the flow scheme of their conversion processes to market products (circle: lignocellulosic raw material; lozenge: process; rectangle: market).

4.4.1. Data set (C1): market volume and price of cellulotics

This data set comprises the current market price developments and the current market volume prospects. Price and volume differ for each region. Local trade and competition dictates the value of the commodities. This is especially true for traditional fibre applications of cellulotics and possibly to a lesser extend for the chemical and biofuel applications (Kabir Kazi et al., 2010).

4.4.2. Overview most relevant economic data

This data set comprises the market prices and volumes listed according the country/region. This is a very dynamic and interdependent data set, which needs constantly updating in order not to become obsolete very fast. Historical import and export economic statistical data (CEPI, 2011; EUROSTAT, 2011; FAO, 2011) give indication on the available capacity per region. The most important data in this data set are:

- Current price (per region)
- Price development statistics
- Current market volumes
- Market developments

In Table 8 global estimated market prices and market volumes of some examples are given. Besides the techno-economic figures of the different cellulosic market chains also information on the socio-environmental impacts of the different supply chain scenarios can be included for comparison of their sustainability.

4.4.3. Data set (C2): market demands on purified cellulose

This data set contains the market defined requirements for purified celluloses. This includes the all the different requirements for cellulose of biofuel producers, paper makers and dissolving pulp processors as well as textile manufacturers. The data set does not include the paper properties, for example, but contains

the requirements of the pulp factory on its raw materials. This means that the requirements on cellulose are described, and not the requirements on the properties of the end product.

4.4.4. Overview most relevant data

The market requirements for a certain lignocellulosic raw material are often very specific for the selected end use. End-uses for cellulose as a fibre are focussed on purity but also on fibre dimensions and mechanical strength. End-uses applying cellulose as a polymer or source as carbohydrate will have higher chemical purity requirements on polymeric cellulose.

Table 8
Data set (C1): market volume and market price.

Cellulose market		Price range (raw material price €/ton)	Market volume Global estimation (Mt/y)
1	Textiles	1200–1900	70 (fibre)
2	Non-woven	200–400	0.6
3	Wood, timber	450–600 €/m ³	1200–1500
	Tropical hardwoods (sawn)	950–1500 €/m ³	
4	Firewood	75–220 €/m ³	
	Pulp, paper and board	450–650	328 paper
5	Cellulose	1600–2000	186 pulp (2003)
6	dissolving pulp		19–21 (non-wood pulp)
7	Cellulosic Films	3000–3500	4.5
8	Building materials	– ^a	0.10
9	Cellulosic fibre	200–400	0.07–0.8 (automotive)
	Composites		– ^a
	Green chemicals	50–100	– ^a

^a No data available.

5. Discussion

5.1. Availability of data

It is impossible and also an unnecessary exercise to complete the proposed cellulose matrix with all data on each raw material, all processes and markets. Instead, a logical frame is set up to fit the continuous growth of data on diverse cellulose containing raw materials and the continuously changing and developing markets. In Fig. 3 a schematic process flow scheme is given for the different ligno-cellulosic resources and their market applications. In Fig. 4 two examples are highlighted. One is cotton at the high value-end of the spectrum and the other is miscanthus at the low value-end. Both crops are converted by various steps into a range of products and potentially they may be both converted into pulp and dissolving cellulose. The number of processing steps from raw cotton to produce pure cellulose is substantially lower than for miscanthus and competition may only occur when the cotton prices are increasing. The dissolving celluloses from various sources may be used for further conversion to cellulose derivatives (Barba, Montané, Rinaudo, & Farriol, 2002). The cellulose resource matrix is set up as a directive guide for stakeholders and cannot provide the final solution for its users. Primarily, a reduction of the quantity of data is possible for each data set to include generic data that comprise a cluster of raw materials, processes, and markets. These clustered data are already shown in the inventories in Sections 3.1, 3.2 and 3.3. In each cluster, the most important raw materials, processes and markets can be filled in, in as much detail as required. Furthermore, there is a restriction possible in a number of data sets:

- Data set (A1): chemical and physical properties of the raw material. The number of data can be reduced by definition of a limited number of key parameters. These key parameters are the absolute precondition for processes or products. This can be the intrinsic fibre length for example in textile processing or the degree of polymerisation for manufacturing of cellulose derivatives. When the fibre length is too short for spinning there is no need to further evaluate the textile industry specifications.
- Data set (A2): price and availability of raw materials. The amount of data collected can be reduced by defining the required minimum amount for commercial processing in relation to the available quantities of raw material. Also a minimum feedstock production price may eliminate the feasibility of a certain ligno-cellulosic material as raw material.
- Data set (B1): processing parameters and economic scale of operation. The exact processing parameters need to be established separately for each raw material and each market. These data can be categorized according the market specifications.

In Tables 4–9 the various data sets already have been limited according these principles. In this way it is possible to obtain an insight and quick overview of the matrix, but for a more detailed overview and questions answered, the exploration of the background information is necessary.

In other data sets it is not the vast quantity of data that is the issue, but the availability of accurate information:

- Data set (B2): demands made on raw material properties for the cellulose processing steps. Current industrial processes and new developments are concentrating on a certain group of easily accessible raw materials. Therefore, it is not often known if the alternative raw material can be applied. Additional research is needed to determine if underutilised lignocellulosic raw materials can be made suitable for manufacturing processes, whether or not involving extra processing steps.

Table 9

Data set (C2): market requirements for extracted cellulose.

Market	Market requirements
1 Textiles	Purity/colour/fibre length distribution/lustre/softness/hygienic
2 Non-woven	Purity/fibre length distribution/absorbency/
3 Wood, timber	Density/strength and modulus/durability/hardness/colour
4 Pulp, paper and board	Brightness, tensile and tear, freeness, writeability
5 Cellulose dissolving pulp	α -Cellulose %, DP
6 Cellulosic Films	α -Cellulose %, DP
7 Building materials	Strength, moisture absorbency, fire retardancy
8 Cellulosic fibre Composites	Compatibility,
9 Green chemicals	Glucose yield/extractability

- Data set (C1): market requirements for purified cellulose. The specifications which end-users apply for purified cellulose, have often evolved from properties that are known from current products available on the market. Other raw materials may have deviating properties, that may require adaptation of the end-user's requirements, when these are to be processed. The strict fixation to current specifications and requirements limits the innovation towards new products. In this data set the potential modification of processes and product parameters need to be taken into account besides the current common industrial specifications.

In Tables 7 and 9 a cursory overview of the demands on lignocellulosic raw materials for cellulose extraction and market specifications is given.

5.2. Operating instructions for the cellulose resource matrix

The cellulose resource matrix should become a useful instrument for different stakeholders in selecting the most suitable options for raw materials and processes for end-product manufacturing. The information contained in the cellulose matrix can be made available as a data base software/internet site where the user has access to the information through searching his query and evaluate the outcome by himself. When a cellulose raw material supplier wants to identify an application for his product, an overview of potential markets, possible conversion processes and any competing cellulose feedstock can be found. Besides that, he will be provided with an overview of background information about the quality of his specific raw material in certain processes and end-use. Besides the further elaboration and expansion of the data sets, development of the software is required before such cellulose matrix can be utilised. The structure of the matrix makes it possible to compose a database in which the various data sets can be further specified and elaborated. With the help of specified searches, answers can be supplied from this database.

When the stakeholder is informed accordingly, it is possible to provide in-depth know-how and expertise. This may be provided by including a literature/patent database to the cellulose matrix for self-gaining in-depth background information, or by directed approach of the relevant experts.

5.3. Completeness of the cellulose resource matrix

An important condition for the usefulness of the cellulose resource matrix is that the available information should be sufficiently accurate to provide direct answers or direction. The biggest challenge for the knowledge management is to obtain sufficient overview of all relevant aspects and convert scientific data into

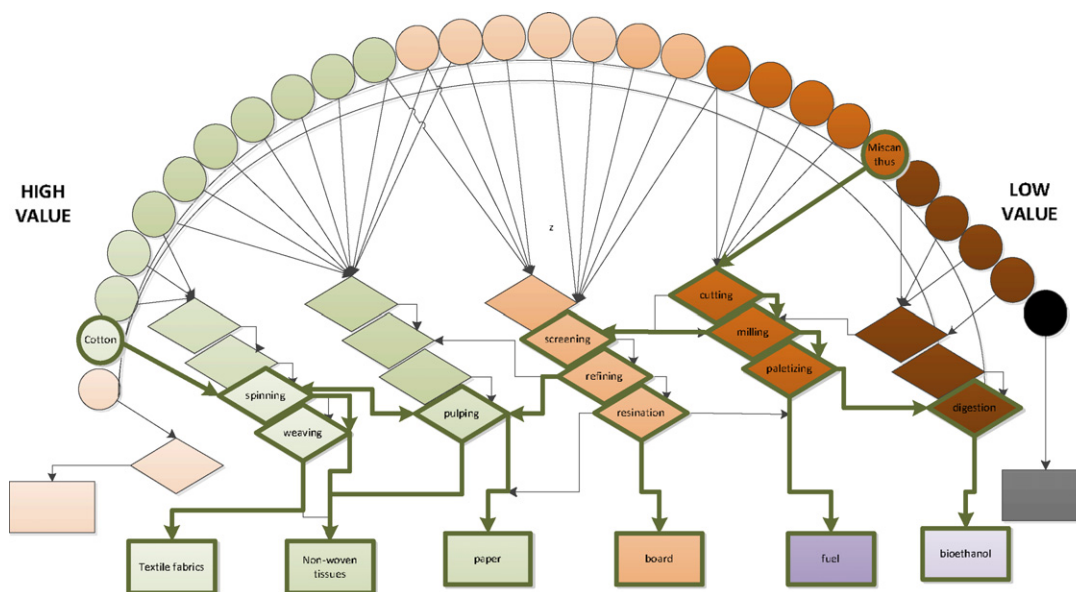


Fig. 4. Conversion process flow scheme of examples of high value (cotton) and low value (miscanthus) cellulosic feedstock.

easily understandable practical and technical solutions for value added applications. Moreover, the economic feasibility of a process is not only determined by the quality of the contained cellulose but also by the total composition of the biomass resource and all the by-products that can be produced from that specific raw material. The expansion of the cellulose resource matrix by including such residue valorisation scheme is, due to substantially increasing complexity.

5.4. Further development of the cellulose resource matrix

The collected and clustered data in this survey, need further elaboration. Due to the complexity of cellulose and the many different processes that are used in the various industries, the literature data require conversion and interpretation to be fitted in the cellulose matrix. A user friendly cellulose resource matrix can be designed by combining the ICT expertise of data management, data integration with the essential cellulose expertise and understanding of the stakeholders needs. For this purpose the suitable standardized ontologies need to be defined, that are most relevant for information accessibility and linking of data on crop production with (bio)economic values and end product quality.

6. Conclusions

The cellulose resource matrix includes elaborated a logical frame of data sets that are relevant for the various stakeholders, such as raw material producers, processors and end-users.

These structured data sets contain classification of current and new lignocellulosic raw materials most important chemical and physical parameters as well as price and availability. Furthermore, current and emerging markets for cellulose based products were identified and described. The technical and economical requirements of the markets for cellulose products, obtained by the various cellulose purification processes, were investigated.

A preliminary design of these relational databases was made. The presented data already may give the various stakeholders a broad distinction of the possible choices. This was achieved by condensing the amount of information, by grouping the parameters of raw materials, processing and market data. In order to make the cellulose resource matrix available as a practical tool for obtaining

information, data management and decision making, the relational databases need further elaboration and extension.

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