Research School
Sustainable Management and Utilization of Forests

Lodgepole pine (*Pinus contorta*) for bio-refineries

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1 Introduction

1.1 An increased need for biomass – a substitute for fossil fuels and materials

Trying to make the society less dependent on fossil oil is currently a major environmental and political issue. Oil is a limited resource that only a few countries can extract. The use of oil for fuel and the production of other chemicals for more than a century has led to huge releases of carbon dioxide (CO_2 – a greenhouse gas); this is one of the main reasons for the climate change that we are currently experiencing. Global warming is leading to floods, droughts, storms and other weather-related natural catastrophes. This, in turn, can lead to loss of agricultural land, conflicts about resources, diseases, migration and loss of species and habitat diversity (European Commission, 2008).

The transport sector consumes the greatest part of the oil in many countries including Sweden, followed by using in heat production. The industry is the third largest consumer of oil in Sweden (McCormick et al., 2006). Thus, alternative fuels for the transport sector are urgently required. Much effort has already been dedicated to finding alternatives, and Sweden has great potential for wood-based bio-refineries to produce fuels and other important chemicals and materials which could have both environmental advantages and may become important exports.

The demand for forest-based biofuels is also encouraged by public policy measures that increase the cost of fossil fuels in favor of biofuels. Such measures are effectively, carbon dioxide taxes. Thus, biofuels can currently compete financially with fossil fuels. The promotion of renewable energy can, therefore, also be regarded as an insurance against high oil prices in the future (Söderholm & Lundmark, 2009).

"*A bio-refinery is a facility that integrates biomass conversion processes to produce fuels, power, and chemicals from biomass. Biomass upgrading processes include fractionation, densification (e.g. pellets), liquefaction, pyrolysis, hydrolysis, fermentation and gasification. The bio-refinery concept attempts to apply to biomass conversion, the methods that have been applied to the refining of petroleum. Bio-refineries would simultaneously produce biofuels as well as bio-based chemicals, heat and power.*" (Demirbas, 2009)

1.2 Pinus contorta – a tree with potential for use in bio-refineries

Lodgepole pine (*Pinus contorta*) is a species with an initially rapid growth rate and high biomass production compared to other conifers in boreal forests; this increases the chances of an early harvest. By direct seeding it may also be possible to produce more stable trees in dense stands at a low cost. The wood quality is similar to that of Scots pine; however, the wood density is slightly lower, the fibers longer and the proportion of heart wood greater (Skogforsk, 1999; Skogforsk, 2006). Pines also contain more extractives than spruce; these can be valuable when producing liquid biofuels (Heinze & Liebert, 2001; Anon., 2009m). All these characteristics make lodgepole pine a tree with potential for use in bio-refineries.

1.3 The aim of this report

The aim of this introductory paper is to examine the following objectives:

- What are the specific characteristics of lodgepole pine?
- What was the purpose of introducing lodgepole pine into Sweden?
- Which wood types and extractives can be used in bio-refineries?
- Historically, what have been the products of bio-refineries, and what products may be valuable in the future?

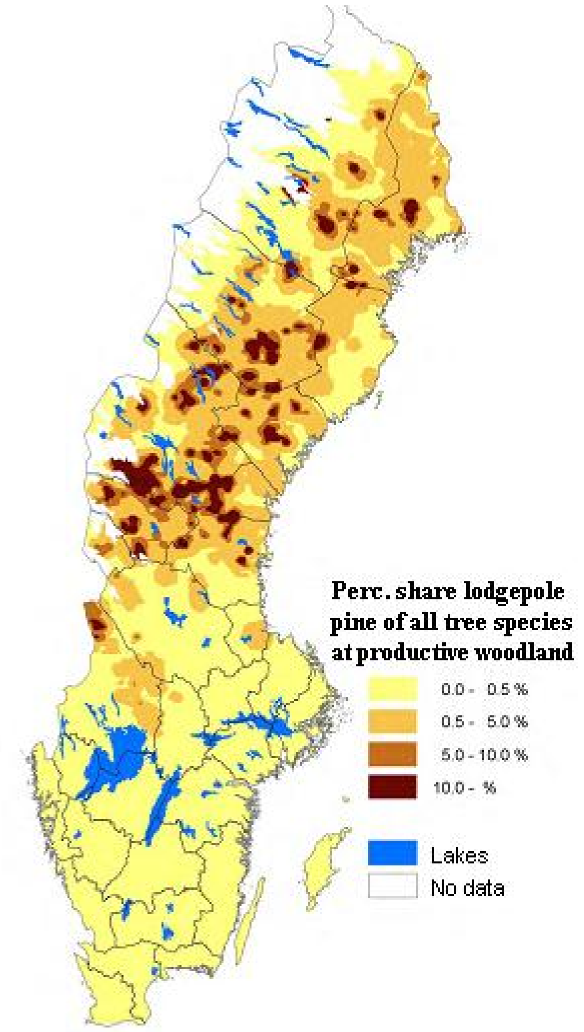
- What biomass products can be produced from lodgepole pine in bio-refineries?

2 The characteristics of lodgepole pine

2.1 Lodgepole pine in North America

Lodgepole pine (*Pinus contorta)* grows between latitudes 30 and 64°N in western North America, from the Yukon Territory and southeastern Alaska as far south as Baja California, and from the Pacific coast eastwards to South Dakota (Elfving et al., 2001). Lodgepole pine occurs as four subspecies, each of which has different morphological characteristics and a different geographical distribution. Subspecies *bolanderi* grows in a small area of highly acid soils in northern California. This subspecies is smaller than the other subspecies of *P. contorta*.

Subspecies *contorta* (shore pine) is found in harsh conditions along the Pacific coast. Subspecies *murrayana* is distributed from the mountains of Baja California in Mexico up along the Sierra Nevada Mountains. Subspecies *latifolia* has the most northerly range of the four, and its habitat extends from northern New Mexico in the USA, to the Yukon Territory in Canada (Despain, 2001; Hagner, 1983).



The *P. contorta* subspecies found to be most suitable for introduction into Sweden was *latifolia*. Subspecies *murrayana* and *latifolia* are specialists in becoming established in recently burnt woodlands. The seeds are borne in serotinous cones that require a temperature of c. 50°C to open (Engelmark et al., 2001). The cones containing the seeds can, therefore, last for several years until a fire provides the high temperatures necessary for them to open. Thus, lodgepole pine has an advantage over other species, and dense stands of it can develop rapidly after a fire (Hagner, 1983). Seeds can remain in the serotinous cones for 30-40 years (Despain, 2001). Young trees up to 30 years of age mainly produce cones that are non-serotinous (Engelmark et al., 2001). Lodgepole pine can reproduce, therefore, even in the absence of fire.

In addition, the species exhibits very fast initial growth, thus reducing the period when the tree is vulnerable due to its small size. The American name lodgepole pine comes from the fact that the trees grow tall and slender, making them perfect material for the lodgepoles that the native American Indians used to build their tepees of (Earle, 2009).

Figure 1: Lodgepole pine, proportion of total productive woodland in Sweden (Riksskogstaxeringen, 2009)

2.2 The introduction of lodgepole pine into Sweden

Lodgepole pine was first introduced as small plantations in Sweden in 1928 (Elfving et al., 2001). Due to its rapid growth, combined with its hardiness and ability to grow in many different climates and on many types of soil, the species started to be studied in Sweden in the late 1950s (Hagner, 1983). At the time, a foreseen lack of spruce- and pine timber was expected at the beginning of the 21st century, and lodgepole pine was considered to have the potential to fill this gap.

The forestry directors of two of the major pulp mill companies, Stig Hagner at SCA and Roland Nellbeck at AB Iggesundsbruk, led the work to introduce lodgepole pine more widely across Sweden in the 1970s (Elfving et al., 2001). Hagner went to British Columbia, Alberta and The Yukon territory in Canada in 1963 to collect seeds, and these became the foundation for some comparative experiments in Sweden. It was found that seeds from northern British Columbia and the Yukon, the northern boundary of the species' natural occurrence in North America, survived best in the Swedish climate (Hagner, 1983). These plants put on most growth at the beginning of summer and were, therefore, better prepared for the early onset of winter, particularly with respect to surviving damage caused by climate and pathogenic fungi (Hagner, 2005, pp 157-158).

By the late 1960s, Iggesundsbruk had begun planting lodgepole pine to reach their goal of the species accounting for 11.5 percent of their total woodland area. The health of the coastal plantations suffered as a result of the material originating from latitudes 50 to 51°N in North America; this southern stock was not suited to Swedish conditions. Iggesundsbruk's plantations located in the inlands of Härjedalen were more successful (Hagner, 2005, pp 155-156).

SCA began planting on a large scale in 1973, and a period of quite intensive lodgepole pine-planting followed from the end of the 1970s until the late 1980s (Lidström, 2008). Because of the risks associated with the introduction of a foreign species, the new Forestry Act specified that lodgepole pine should be restricted to the area north of latitude 60°N, except in the counties of Värmland and Örebro, where lodgepole pine could be planted down to latitude 50°30'N. The act also specified that lodgepole pine should only be planted where domestic species did not regenerate satisfactorily.

Private forest owners followed the lead of the forest companies, culminating in peak planting in 1984-85, when almost 40 000 ha were converted to lodgepole pine plantations annually in Sweden (Loman, 2008). One predicted advantage of lodgepole pine was that the need for pre-commercial thinning would diminish as a result of the rapid juvenile growth, which makes it difficult for other vegetation to compete with the species (Hagner, 2005, p 161). Private forest owners continued planting about 5000 ha/year until 1988, when planting the species declined

Figure 2: Red pine infected by Gremmeniella abietina (Government of Canada, 2009)

rapidly (Hagner, 2005, p 161).

Lodgepole pine seedlings used to be grown in paper pots, and because of the rapid growth of the fine roots combined with the impenetrable paper, the roots became twisted. Therefore, some lodgepole pines grew rapidly but their root system was unable to ensure stability of the above-ground biomass. Such problems were most severe in fine grained dense soils. The lower part of the stem also became bent, due to the root problems (Skogforsk, 2006). The roots of lodgepole- and Scots pine develop at the end of the root laces. Unlike pine, spruce is able to produce new roots anywhere on the original root, making spruce less vulnerable to root deformation caused by it being grown in pots (Hagner, 2005, p 222).

The years 1986-89 had very cold winters with warm and damp intervals combined with cold summers; these were perfect conditions for the pathogenic fungi *Gremmeniella abietina.* Lodgepole pines already weakened by their unstable root systems and stands in harsh areas were most susceptible to the fungi. Shoots infected by gremmeniella often die and become rust-colored. Because of the gremmeniella outbreak, the planting of lodgepole pine drastically decreased, especially among the private forest owners. However, the damage mostly occurred in areas with severe winters.

2.3 Lodgepole pine in Sweden today

Today, SCA has a total area of 280 000 ha of lodgepole pine stands (Lidström, 2008), which is about half of the total area of lodgepole pine in Sweden, amounting to 576 000 ha (Elfving, 2010). Lodgepole pine accounts for 2 percent of the total woodlands in Sweden today, and about 2500 ha are planted annually (Loman, 2008). This is much lower than the Swedish Forest Agency's principle that no more than 14 000 ha should be planted each year.

Lodgepole pine was introduced in Sweden mainly for pulp wood production from rather short rotations. Experience of Swedish lodgepole pine stands has shown that timber production is also possible with longer rotations, since lodgepole pine produces more than 30 % more stem volume than Scots pine (Skogforsk, 1999; Norgren, 1995a). The wood quality also much resembles Scots pine. However, the wood density is slightly lower, the fibers longer and the proportion of heart wood greater (Skogforsk, 1999). Lodgepole pine has a thinner bark, but more branches than Scots pine (Skogsstyrelsen, 1992).

Sown lodgepole pine-stands have been found to develop stable trees in dense stands at a much lower cost than for planted saplings. Thus, lodgepole pine can either be managed like we manage Scots pine today but in rotations 10-15 years shorter; or as high density-stands for biomass production with much shorter rotations. Lodgepole pine is more suitable for the latter type of management because of its greater survival, earlier exposure of new needles, longer needles with a bigger surface area that absorbs a large quantity of light, fast root development and a higher productivity per unit of nitrogen due to the lower nitrogen content of the needles (Skogforsk, 1999; Norgren & Elfving, 1995b). In combination, these features result in higher biomass production.

Lodgepole pine is also more seldom grazed by moose (*Alces alces*), but is more sensitive to *Gremmeniella abietina* and voles (Arvicolinae) than Scots pine. Lodgepole pine has slower self pruning than Scots pine, and more trees are lost after thinning due to snow- and wind-damage. Therefore, more trees may be damaged but more light may also reach the ground in old lodgepole pine stands than in old Scots pine stands (Skogforsk, 1999). Lodgepole pine should not be planted in very windy or snowy locations, and thinning should be undertaken with care to reduce damage caused by wind and snow (Norgren, 1995a).

2.4 The chemical properties of lodgepole pine

The chemical profile of lodgepole pine differs from Scots pine. Lodgepole pine has a lower nitrogen content but higher detergent fiber- and lignin contents (Table 1).

Table 1. Chemical composition of Pinus sylvestris, Pinus contorta and Picea abies (g kg^{-1} dry wt.), n=4 per species (After Stolter, et al., 2009).

Compound	***Pinus sylvestris***	***Pinus contorta***	***Picea abies***
Organic matter	968.41±2.51	984.75±4.09	971.44±1.20
Nitrogen	13.68±0.95	9.89±0.82	9.54±0.63
Neutral Detergent Fiber	556.59±11.99	570.59±10.52	577.06±18.87
Acid Detergent Fiber	412.62±21.10	429.94±10.04	427.83±12.51
Lignin	113.85±8.67	180.87±7.92	155.03±4.00

As can be seen in Table 2, the amounts of condensed tannins and total phenolics are higher in lodgepole than Scots pine. Ruminants like moose often avoid plants which are rich in tannins, but tannins can also be of nutritional benefit for them, depending on the specific tannin composition (Foley & Moore, 2005). The higher tannin content of lodgepole pine may explain why the species is browsed by moose less frequently than is Scots pine. Lodgepole pine, like tea, has a high concentration of the polyphenolic antioxidant (+)catechin. In addition, lodgepole pine also contains high levels of DHPPG (3,4-Dihydoxypropiophenone-3-glucoside), a phenolic compound absent from Scots pine. Overall, lodgepole pine has a lower nitrogen content but more extractives than Scots pine (Stolter, et al., 2009).

Table 2. Concentration of condensed tannins (%), total phenolics (%) and individual phenolics (mg g^{-1} dry wt.) (After Stolter et al., 2009).

Compound	***Pinus sylvestris* (*n*=8)**	***Pinus contorta* (*n*=6)**	***Picea abies* (*n*=8)**
Condensed tannins	1.10±0.11	2.54±0.72	1.83±0.11
Total phenolics	1.17±0.17	2.04±0.48	2.01±0.22
(+)catechin	1.14±0.15	2.27±0.81	26.58±3.74
3,4-Dihydoxypropiophenone-3-glucoside (DHPPG)		59.10±27.76	

3 What chemical constituents of a tree can be used in a bio-refinery?

Wood composition:

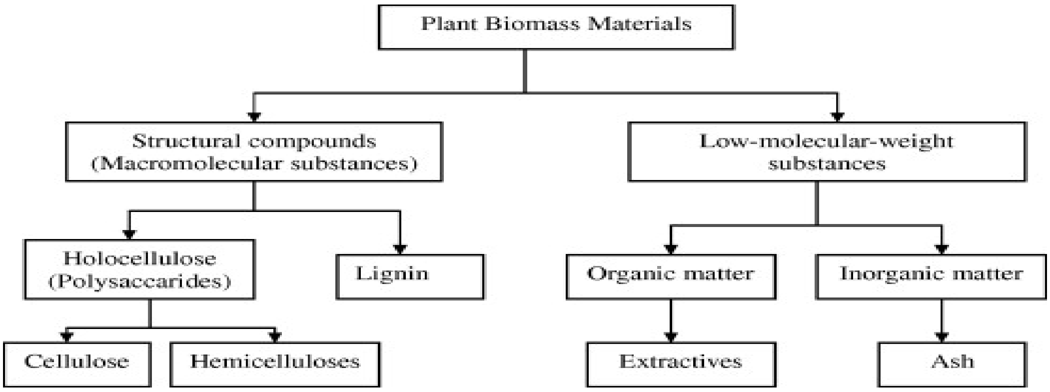


Figure 3. Plant constituents (Demirbas, 2009).

3.1 Cellulose

Cellulose $(C_6H_{10}O_5)_n$ is the main constituent of a tree; it is a linear polysaccharide built up by D-glucose units attached to each other by a 4-hydroxyl group (β-1,4 glucosidic bonds) (Nationalencyklopedin, 2009; Shen & Gu, 2009). The chain length of cellulose varies between 100 and 14 000 units. These chains are linked to other chains by intermolecular hydrogen bonds and to other units in the same chain by intramolecular hydrogen bonds. The cellulose chains held together by hydrogen bonds are called fibrils. These bonds and the linear form of the cellulose result in there being crystalline parts of the molecule; these are very stable and difficult to breakdown by, for example, hydrolysis (Träkemi & Naturproduktkemi). Cellulose is the most abundant organic substance (polymer) on earth, and a conifer consists of 40-45% cellulose.

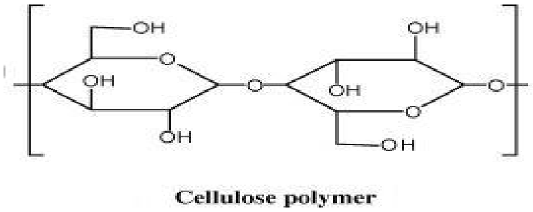


Figure 4. The chemical structure of cellulose. (Shen & Gu, 2009)

3.2 Hemi-celluloses

Hemi-celluloses are a group of branched polymer carbohydrates built up of poly-saccharides. The most common ones in trees are constructed of glucose and mannose (glucomannan) or xylose. These polymers are much shorter than those of cellulose. Hemi-celluloses represent about 20-25% of the tree, and are bonded to cellulose by hydrogen bonds (Anon., 2009g; Träkemi & Naturproduktkemi). However, the hemi-cellulose polymer chain is branched and hydrogen bonds like those found in cellulose cannot occur. Therefore, hydrolysis of hemi-celluloses is relatively easy. The hemi-celluloses fulfill the same role as cellulose, mainly as structural components of the tree.

3.3 Lignin

Lignin is a polymer consisting of aromatic units; it is the second most abundant polymer on earth. Lignin is a constituent of all plants except the most primitive ones. Lignin occurs in tissues specializing in fluid transport and gives the plant physical strength. Conifers contain about 25-30% lignin, which is connected to the hemi-celluloses of the tree by covalent bonds (Träkemi & Naturproduktkemi). Conifer lignin is composed of coumaryl-, cinamyl- and sinapyl-units, in different proportions in different species.

3.4 Extractives

Extractives, also known as secondary metabolites, are unlike cellulose, hemi-celluloses and lignin in that they are not a part of the cell wall and are, therefore, nonstructural constituents of wood. They are most abundant in the external heartwood and in damaged parts of the tree, as they provide resistance to insects, fungi and rot. Resins, fats, inorganic salts, some sugars and phenols are all classed as extractives, though salts and minerals are sometimes referred to as ash. Hardwoods contain more extractives than softwoods, and pine contains more extractives than spruce: 6% compared to 3% (Heinze & Liebert, 2001; Anon., 2009m).The extractives in softwoods consist of 40-45% resin acids, 40-60% fatty acids and a small amount of monoterpenes and phenolics. Extractives are relatively small molecules ($C<40$) that can be extracted using a variety of organic solvents (polar or non-polar) from wood, bark or foliage. The content of extractives varies greatly between species, and between individual trees (Cole, 2006). In the pulping processes extractives can affect the pulp strength and other properties, which in turn can lead to a reduction in yield (Anon., 2010).

3.5 Ashes and remaining parts of the tree

The remaining constituents can be used for heat- and electricity production, and the ash can also be valuable. The ash content differs between different parts of the tree. Based on dry weight, the stem wood has an ash content of 0.4-0.6 %, the stem bark 2-5 %, the branches 1-2 % and the needles 2-6 %. Stabilized ash consists of carbonates, oxides, hydroxides, sulfates, chlorides and silicates of base cat-ions and trace elements, mainly calcium, potassium, silicon, magnesium and phosphorus (Emilsson, 2006). The ash can be used as a fertilizer or for building roads or other surfaces. Thus, all parts of a tree can be used in a bio-refinery.

4 Historically, what has been produced in bio-refineries?

4.1 From the 19th century: Timbers and pulps

Many sawmills were built in Sweden in the mid-19th century, among them Korsnäs in Falun, Östrand in Timrå and Högland in Örnsköldsvik. A sawmill had already been built in Håknäs, Västerbotten by 1761 (Anon., 2009f). In the 1880s, sulfite pulp mills were being constructed, based on a method developed by Carl Daniel Ekman in 1871. Prior to this, paper was mainly produced from rags. The technique of producing paper from wood was poorly developed; the paper was fragile and easily discolored. By using the sulfite method, stronger white paper could be produced. Many bio-refineries started out as sulfite pulp mills. The Norwegian bio-refinery group Borregaard started as a pulp mill in 1889. Domsjö started to produce sulfite pulp in 1903. The bio-refineries Nippon Paper Chemicals in Japan and Lenzing in Austria also started out as sulfite pulp mills (Anon., 2009c; Anon., 2009k; Larsson & Ståhl, 2009).

The sulfate pulp process was invented by the German Carl F. Dahl in 1882. Mills based on this process were built around Sweden at the beginning of the 20th century, and the sulfate process dominates the pulping industry today (Nationalencyklopedin, 2009).

4.2 From the 1930s: Viscose cloth

In the 1930s, Domsjö started to produce viscose pulp from cellulose, at first for making diapers and later for clothes such as uniforms for soldiers in the 1940s. Forage for cattle was also produced from cellulose. The technique of manufacturing viscose (a.k.a. Rayon or cellulose-xantogenat) has been known since 1892. To produce it, cellulose is mixed with sodium hydroxide, which produces an ester, cellulose xanthate. After being dissolved in water, carbon disulfide is added and cellulose-xantogenat forms. Today, viscose can also be manufactured without the addition of sulfur, in which case it is known as lyocell. Until the late 1960s, viscose was the most common synthetic fiber used to make clothes. Viscose in various forms is still used frequently in clothing, and is nowadays presented as an environmentally friendly alternative to cotton and polyester (Nationalencyklopedin, 2009; Larsson & Ståhl, 2009).

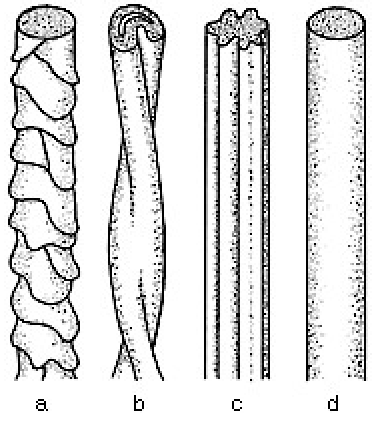


Figure 5. Different textile-fibers: a) wool b) cotton c) viscose d) polyester (Nationalencyklopedin, 2009)

4.3 From the 1940s: Chemicals

Because of trade restrictions during the Second World War, production of both chlorine and ethanol began at the Domsjö sulfite pulp mill in the 1940s. Ethanol can be produced from brown liquor, a residue produced by sulfite pulp mills. The process had been known since the beginning of the 20th century. In the 1980s, the ethanol production at Domsjö became incorporated into SEKAB, Swedish Ethanol Chemistry. In the 1940s, Akzo Nobel started to make thickening agents out of wood. Today, the thickening agent Bermocoll (ethylhydroxiethylcellulose) is produced in Örnsköldsvik from cellulose, ethylene oxide and ethyl chloride. Since 2000, Domsjö has been committed to becoming a complete bio-refinery, with products including special cellulose and lignosulfonates (Anon., 2009c; Assarsson & Blomqvist, 2005).

5 Bio-refinery products with future potential

5.1 Biofuels for use in vehicles

Biofuels can be divided in three general groups: solid, liquid and gaseous. Examples include wood and pellets; ethanol and biodiesel; and biogas and hydrogen, respectively. It is also possible to separate bio-fuels according to the processes used in their production from biomass, e.g. biological and chemical processes, thermochemical or physical upgrading processes (Arshadi & Sellstedt, 2008).

Currently in Sweden, the three types of biofuel that are most commonly used for vehicle fuel are ethanol, biogas and different types of biodiesel (Anon., 2009a). High expectations have also been placed on hydrogen as the fuel of future, since it is possible to produce from infinite resources and the only exhaust emission is steam.

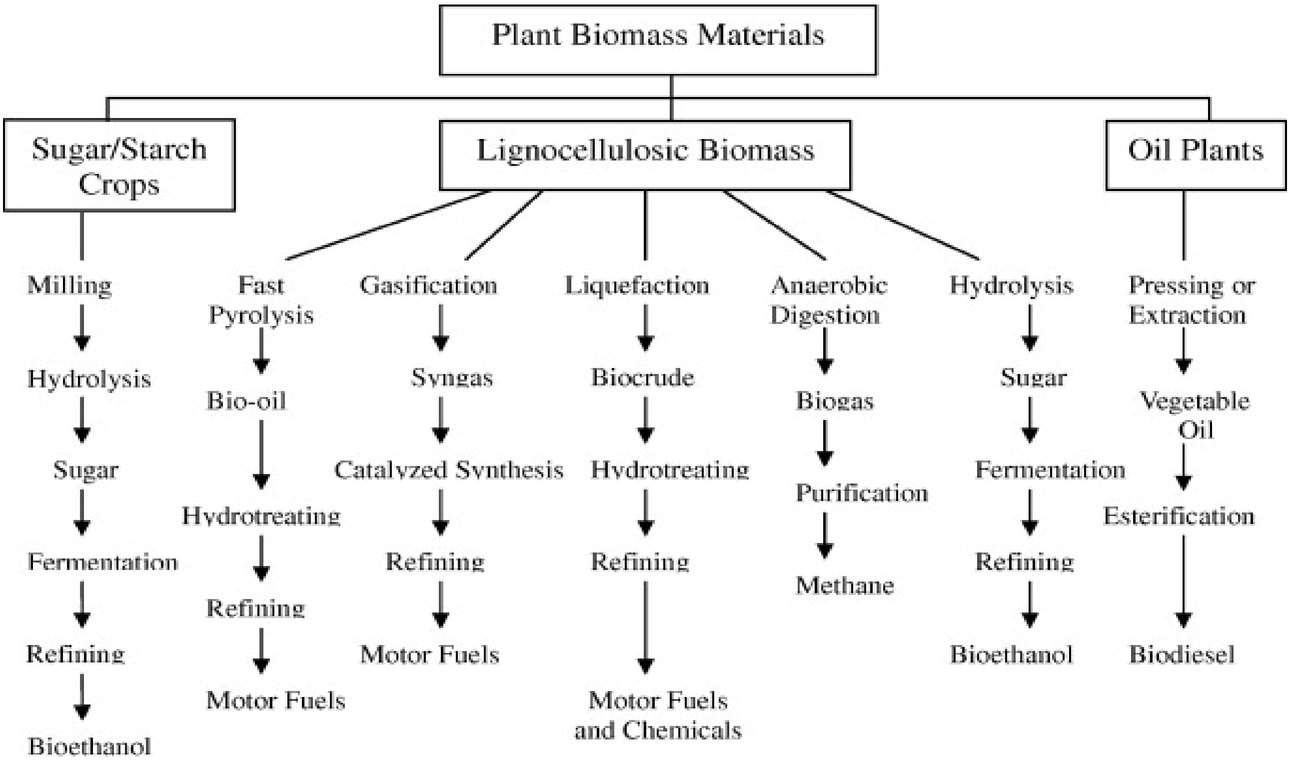


Figure 6. Overview of the processes for converting plant materials to biofuels (Demirbas, 2009).

5.1.1 Alcohols

The two alcohols that are mainly used as vehicle fuels are methanol and ethanol.

Methanol

After the first oil crisis, the Swedish car maker Volvo promoted methanol, CH_3OH, as the preferred alternative choice for vehicle fuel, because it was cheap, easy to produce from natural gas and had lower emissions than ethanol. The Swedish Methanol Development Company (SMAB), was founded by Volvo in 1975 in cooperation with the Swedish government. SMAB received a large part of the R&D-funding for alternative fuels during the 1970s. At first, a blend of gasoline with 15% methanol seemed to be most interesting. By 1981 the focus had changed to pure methanol, with wood instead of natural gas as the raw material. This was opposed in some quarters, because wood is more efficiently used for the production of electricity and heat. In the 1980s, overproduction of sugar beet and wheat in Sweden was one of the reasons that focus changed to ethanol. Ethanol could be produced from domestic renewable materials such as these two crops. Ethanol is also less toxic than methanol and less corrosive (Ulmanen et al., 2009).

By the end of the 1990s, a new dawn for methanol was breaking, due to the possibility of gasifying black liquor to form methanol. Black liquor is a residue from the sulfate pulp industry that has traditionally been burnt for energy use in recovery boilers at the pulp mills. In 2004 the company Chemrec commenced production at a pilot plant in Piteå, using the gasification process (Ulmanen et al., 2009). The technique will be used in a project at Domsjö Industries in Örnsköldsvik, producing the motor fuels Biomethanol and BioDME (Anon., 2009j).

Ethanol

Ethanol, C_2H_5OH, can be produced from glucose, starch or cellulose. In Sweden most ethanol has been made of wheat and barley, but ethanol production from cellulose – the second generation of ethanol – is on its way (Anon., 2009a). At Agroetanol at Händelö in Norrköping, ethanol is made from grains such as wheat, triticale and barley. The grain starch is converted by enzymes into sugar and thereafter is fermented to ethanol and carbon dioxide; 2.7 kg of grain produces 1 l of ethanol (Haaker, 2003).

$$C_6H_{10}O_5 + H_2O \rightarrow C_6H_{12}O_6 \rightarrow 2C_2H_5OH + 2CO_2$$

The fermentation process has been known for a very long time. Sugarcane has been used to produce ethanol since 6000 BC (Demirbas, 2009). Making ethanol out of cellulose is a much more recent development. SEKAB (Swedish Ethanol Chemistry AB), together with Domsjö Fabriker AB in Örnsköldsvik, have been refining ethanol from hemi-cellulose for 100 years, and since 2004 they have been working on producing ethanol from cellulose. About half of a tree is made up of cellulose, compared to 25-30 % hemi-cellulose. Thus, a larger proportion of the tree can be used for ethanol production if cellulose is the raw material. The most difficult part of ethanol production from cellulose is breaking down the cellulose chains to sugar that can be fermented (Anon., 2009l). Large volumes of ethanol are imported into Sweden, mostly sugar cane ethanol from Brazil and other tropical regions and wine ethanol from Southern Europe. Almost all petrol in Sweden contains at least 5 % ethanol, while the fuel E85 contains 85% ethanol and 15% petrol (Anon., 2009a).

5.1.2 Biogas

Biogas consists of methane, CH_4, which is the same gas that is the main constituent of fossil natural gas. Methane is produced during anaerobic digestion of organic compounds. Organic waste or sewage sludge is often used as the raw material for the production of biogas. Organic waste from abattoirs, farms and food industries is also a suitable raw material (Anon., 2009a). There are huge amounts of waste all over the world that could be used for the production of biogas. When such organic material is digested, methane and carbon dioxide are produced. A purification process results in almost 100 % pure methane, which can be used as fuel. One cubic meter of biogas contains as much energy as 1.1 l of petrol. Natural gas has a higher energy value than biogas because it contains other gases such as propane and butane (Werner, 2009).

Today there are 100 filling stations selling biogas in Sweden, but only four of them are located north of Uppsala (Anon., 2009e). Biogas production from wood is still at the development stage. Because of the potential to produce biogas from many types of waste, wood is probably not the most cost-efficient raw material for biogas production. However, Domsjö is currently Sweden's largest producer of biogas as a byproduct of its purification process. The biogas is, among other things, used to dry lignosulfonates in the factory (Thorén, 2009).

5.1.3 Biodiesel

Different fuels produced from renewable resources go under the name biodiesel. Their common feature is that they can all be used in diesel engines with no or minor engine adjustments. The three most common types of biodiesel are FAME (Fatty Acid Methyl Ester), DME (Dimethyl Ether) and FT-diesel (Fischer-Tropsch diesel).

FAME – Fatty Acid Methyl Ester

FAME (Fatty Acid Methyl Ester) consists of mono-alkyl esters of long-chain fatty acids that can be made from various vegetable oils and waste oils such as those previously used for frying. The vegetable oils that are most commonly used are oils from soybean, rapeseed, palm and sunflower. In Sweden, rapeseed methyl ester, known as RME is the most frequently used. Oils consist of triglycerides, comprising one glycerine molecule and three long-chained fatty acids. The fatty acid composition differs between oils, and each fatty acid has a unique chain length and number of double bonds. Thus, each vegetable oil has its own physical characteristics (Anon., 2000; Ramos et al., 2009).

To produce FAME, oils are transesterified with methanol. In a transesterification reaction, one mole of triglyceride reacts with three moles of alcohol to form one mole of glycerol and three moles of fatty acid alkyl esters (Ramos et al., 2009). In this case the alcohol used is methanol, and the product is therefore fatty acid methyl esters, FAME. Ethanol can be used instead of methanol in the reaction, the product thus formed is FAEE, fatty acid ethyl esters. The main reason that methanol is used is that it is cheaper than ethanol, since it is a by-product of the organic-synthesis industry. The properties of FAME and FAEE are similar (Zajac et al., 2008).

Table 3. Fatty acid compositions (wt.%) of some vegetable oils. The numbers in the second column refer to chain length and number of double bonds. Thus, "C18:2" indicates that the fatty acid has 18 carbon atoms and two double bonds in its hydrocarbon-chain. (Ramos et al., 2009; Altiparmak et al., 2007; Hopkins & Hüner, 2004).

Fatty acid		**Palm**	**Soybean**	**Sunflower**	**Rapeseed**	**Pine**
Saturated						
Palmitic	C16:0	36.7	11.3	6.2	4.9	-
Stearic	C18:0	6.6	3.6	3.7	1.6	2.1
Myristic	C14:0	0.7	-	-	-	-
Arachidic	C20:0	0.4	0.3	0.3	-	-
Behenic	C22:0	0.1	-	0.7	-	-
Lignoceric	C24:0	0.1	0.1	0.2	-	-
Lauric	C12:0	0.1	-	-		-
Total saturated	**-**	**44.7**	**15.3**	**11.1**	**6.5**	**2.1**
Monounsaturated						
Oleic	C18:1	46.1	24.9	25.2	33.0	52.7
Gadoleic	C20:1	0.2	0.3	0.2	9.3	-
Palmitoleic	C16:1	0.1	0.1	0.1	-	-
Erucic	C22:1	-	0.3	0.1	23.0	-
Total monounsaturated	**-**	**46.4**	**25.6**	**25.6**	**65.3**	**52.7**
Polyunsaturated						
Linoleic	C18:2	8.6	53.0	63.1	20.3	38.3
Linolenic	C18:3	0.3	6.1	0.2	7.9	6.9
Total polyunsaturated	**-**	**8.9**	**59.1**	**63.3**	**28.2**	**45.2**

The properties of FAME improve the combustion efficiency and emission profile of a diesel engine, compared to diesel derived from fossil crude oil. FAME-biodiesel and blends of biodiesel with fossil oil diesel have reduced carbon monoxide, sulfur oxides and particle emissions. Nitrogen oxide emissions are often increased when using biodiesel, because there are more nitrogen compounds in the fuel and better combustion of the fuel, leading to increased maximum temperature in the engine. However, temperature characteristics such as the pour point (freezing point) are different in fossil fuel diesel than some types of biodiesel, making the latter unsuitable for use during cold winters. The calorific value of biodiesel is also lower than that of fossil oil diesel, which leads to a slightly higher fuel consumption, especially at low engine speeds (Altiparmak, 2007).

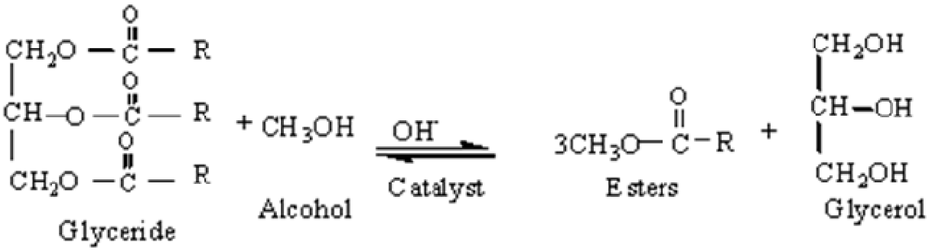


Figure 7. Transesterification of glycerides (Anon., 2000).

- ***RME – Rapeseed Methyl Ester***

RME is produced by extracting the oil from oilseed rape either by mechanical processes or by solvent extraction, followed by purification and transesterification with methanol. Current research is focused on selective breeding to achieve a better blend of fatty acids in oilseed rape for the purpose of fuel production. RME has a pour point of -9^{o}C, compared to -29^{o}C for fossil oil diesel. Without additives to reduce the cloud and pour-points, RME can cause problems in diesel engines at temperatures below -10^{o}C (Williamson & Badr, 1998).

- ***Tall oil-FAME a.k.a. TOME (Tall Oil Methyl Ester)***

Tall oil is an extractive of pines and a by-product of pulp-manufacturing by craft or sulfate pulping process (Keskin, et al., 2007). When producing paper pulp, cellulose fibers are separated and black liquor is left as a residue. Black liquor is a solution of lignin residues, hemi-celluloses, extractives and inorganic chemicals used in the process. If some of the pulpwood comes from pine or birch, tall oil can be extracted from the black liquor. Today, most of Sweden's production of crude tall oil, 200 000 tons a year, is used for the production of soft soap and other chemicals (Hultén, 2008).

Tall oil-FAME consists of hydrocarbon chains with 16-20 carbon atoms and a carboxylic acid group at the end. Most of the hydrocarbon chains are 18 carbons long. Such chains closely resemble fossil oil diesel. Thus, the characteristics of tall oil-FAME are similar to fossil oil diesel, with similar energy- and temperature characteristics, making it possible to use tall oil-FAME as diesel fuel even in cold winters in Sweden. This is not possible with RME, because it freezes at higher temperatures than fossil oil diesel (Hultén, 2008). Both rape seed oil and tall oil are highly unsaturated, although rape seed oil contains more saturated fatty esters than tall oil. This means that RME has a higher cloud point than tall oil-FAME (Ramos et al., 2009; Imahara et al., 2006).

Other important properties of diesel fuel are the cetane number, which indicates how easily the fuel will ignite; and the oxidation stability. A large proportion of polyunsaturated fatty acids results in a lower cetane number and decreased oxidation stability. Accordingly, RME has slightly better properties than tall oil-FAME (Table 3). However, tall oil is sometimes used to improve cetane number in petrol diesel fuel. The oxidation stability can be improved by adding antioxidants (Ramos et al., 2009; Imahara et al., 2006; Altiparmak et al., 2007). The chemical composition of tall oil differs slightly according to tree age, species, geographical location and pulping process. Tall oil is generally cheaper than other vegetable oils (Altiparmak, et al., 2007, Keskin et al., 2007).

A factory for the production of tall oil-FAME, SunPine, is under construction in Piteå in northern Sweden, and production is planned to start early in 2010. The factory is owned by Sveaskog, Södra, Preem and Kiram. The latter belongs to the inventor of the manufacturing process, Lars Stigsson, who also founded Chemrec. Sveaskog and Södra will contribute raw materials while Preem will blend its fossil diesel with 10 percent tall-oil diesel at their refinery in Gothenburg. A total of 100 000 m^3 of tall oil-diesel will be manufactured annually; this is equivalent to the fuel requirement for 100 000 diesel vehicles that are each driven 10 000 km per year, and equates to reduced discharges of carbon dioxide of 250 000 tons a year (Anon., 2008). The quantity of raw tall oil that will be used almost corresponds to Sweden's annual production, today used for making oils, soaps and resins. Thus, there may be a shortage of tall oil for the industry in the future if no efforts are made to increase production (Hultén, 2008).

DME – Dimethyl Ether

DME (dimethyl ether, C_2H_6O) is the simplest of the ethers. DME is an easily ignitable gas that becomes liquid at a pressure of more than 5 bar. It can be produced by condensation of methanol with a sulfuric acid catalyst at a high temperature. The energy content of DME is low compared to fossil diesel. As a pure fuel it would require more than twice the injected volume, compared to fossil diesel, due to the DME's lower density and specific combustion enthalpy. The low self-ignition temperature and high cetane number, however, make it interesting as an alternative to fossil diesel. In comparison with fossil diesel, DME has lower NO_x- and smoke-emissions but higher CO-emissions (Östman, 2007; Crookes & Bob-Manuel, 2007). The technique developed by Chemrec, as used at Domsjö Industries to produce Biomethanol and BioDME from black liquor, will also be used at the Smurfit Kappa pulp mill in Piteå to produce BioDME. A pilot plant will be in production by July 2010 (Anon., 2009i).

FT-diesel – Fischer-Tropsch diesel

FT-diesel can be produced from natural gas, coal or biomass. The raw material is first gasified by the addition of oxygen and steam. The gas produced, which contains contaminants like H_2S, CO_2 and NH_3, is then purified to synthetic gas (syngas), a mixture of H_2 and CO. After adjusting the H_2/CO-ratio, the syngas is introduced into a reactor where hydrocarbons of different lengths are produced under the influence of high pressure, high temperature and a catalyst such as iron or cobalt. The resultant hydrocarbons can be hydrocracked to form diesel. The most complicated step is the gasification of the biomass (Tijmensen, 2002; Anon., 2009d). FT-diesel has the advantage that it is fully compatible with existing vehicles. The fuel has a high cetane-number, does not contain nitrogen or sulfur and has very low levels of aromatics (Van Vliet, et al., 2009; Tijmensen, et al., 2002).

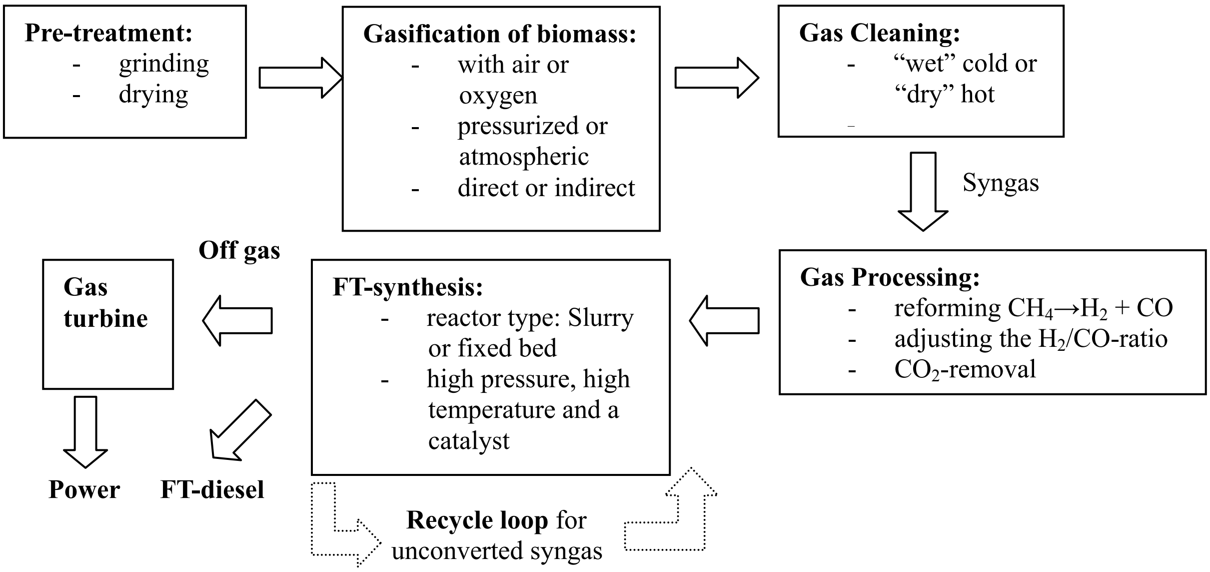


Figure 8. Schematic view of key components for converting biomass to FT-liquids combined with gas turbine-power generation. (After Tijmensen, et al., 2002).

The first Fischer-Tropsch fuel-plants were coal-to-liquid CTL)-plants, built in Germany in 1935. The nine plants were closed at the end of World War II, in 1945. In the 1970s and 80s, South Africa produced FT-fuels made from coal, in order to reduce its dependence on foreign oil, a tactic required because of the sanctions against South Africa at that time. Gas-to-liquid-plants (GTL) have been being tested since the 1990s. Today, large GTL-plants are planned in Qatar, and CTL-plants are planned in China and India. In Germany, experiments on BTL (biomass-to liquids) are on-going, and the building of a full-scale BTL plant is planned for 2011. Biomass from Scandinavia and the Baltic States will be used as the raw material (Van Vliet, et al., 2009; Anon., 2009b). Thus BTL, unlike the other types of FT-diesel presented here, is made from renewable resources. BTL-diesel improves combustion efficiency and the emission profile of diesel engines in a similar way to FAME.

5.2 Other chemicals and materials that can be produced in a bio-refinery

5.2.1 Cellulose based products

At Domsjö Fabriker AB, a bio-refinery in Örnsköldsvik, Sweden, special cellulose is produced for uses as diverse as viscose clothing and textiles, hygiene products including diapers and sanitary towels, binding agents in medicines and sausage-skins, washing detergents and dish cloths, as well as a thickening agent for paint (Anon., 2009c; Arshadi 2007). New biomass-based materials are under development. With nanotechnology it is possible to create materials from biomass that are simultaneously very strong, light and environmentally friendly (Larsson & Ståhl, 2009).

5.2.2 Hemi-cellulose based products

Ethanol derivates made from ethanol and oxygen, also known as denatured spirits, are used as solvents and can be found in paints, perfumes, cleaning products and pharmaceuticals (Anon., 2009l). Acetic acid (CH_3COOH) can also be produced from hemi-celluloses, i.e. ethanol. The water retaining capacity of glucomannan can be utilized in a range of products including diet-foods, packaging and agricultural needs (Assarsson & Blomqvist, 2005; Larsson & Ståhl, 2009).

5.2.3 Lignin based products

Lignosulfonates are used as cost-effective additives, because they are efficient dispersants and binding agents. They can be used for water reduction in concrete, in the production of bricks and in dye pigments (Anon., 2009c). Lignosulfonates have been produced on a large-scale since 1934 when production started at Marathon's in Rothschild, Wisconsin, USA (Anon., 2009h). Borregaard in Norway is one of the world's largest suppliers of wood-based chemicals and, in addition to special cellulose, lignosulfonates and ethanol, it also produces lignin-based vanillin, yeast and omega-3 products (Anon., 2009k).

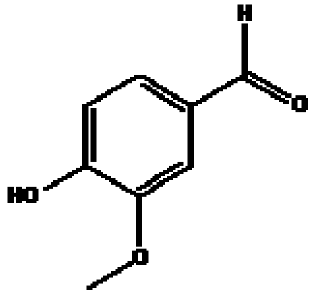


Figure 9. The chemical structure of vanillin.

5.2.4 Extractive-based products

Soft soap has historically been made from either animal fats or vegetable oils that have been boiled with potassium hydroxide. Today, most commercial soft soap in Sweden is made from tall oil, a byproduct of the sulfate pulping process. Esters and resins can also be made by distillation of tall oil (Hultén, 2008). Phytosterol from extractives can be used to reduce cholesterol in margarine and other foods, while terpenes are used as solvents and in paints (Assarsson & Blomqvist, 2005). Lignans from spruce are used as antioxidants.

5.3 Heat and electricity

Two important products that can be produced at the same time as other products in a bio-refinery are heat and electricity. Traditionally, residues from pulp manufacture have been burnt in recovery boilers to produce steam that can be used for internal processes as well as for electricity production in the pulp mill. Many pulp mills produce energy as a byproduct. In a bio-refinery there is the potential for making both electricity and heat for district heating. Gas turbines can produce electricity at gasification plants. At SEKAB, for example, heat and power are important products alongside ethanol, in making the company financially viable (Larsson & Ståhl, 2009).

6 Discussion

6.1 Which biomass products are suitable for production in bio-refineries using lodgepole pine as a raw material?

The properties of lodgepole pine, with its rapid growth and high biomass production compared to other Swedish conifers, make it suitable for use in bio-refineries. Lodgepole pine can be used for gasification, for example, but what is special about lodgepole pine is the large amounts of extractives combined with the potential for tall oil production.

Today, most of Sweden's production of crude tall oil, 200 000 tons a year, is used for the production of soft soap and other chemicals such as esters and resins. However, tall oil can also be used for the production of biodiesel. Biodiesel made from tall oil exhibits desirable fuel properties and is very similar to fossil diesel. Currently, there are plans to produce 100 000 m^3 of biodiesel from tall oil each year beginning in 2010; this represents the majority of Sweden's current yearly production of tall oil.

Blending fossil diesel with tall oil-FAME will be one viable aspect of achieving the goal of using a higher proportion of renewable fuels. In the near future, it will be possible to drive diesel vehicles solely on tall oil-FAME, even in cold winters. The main problem is scarcity of raw materials. There will probably be a shortage of tall oil in the future, if no efforts are made to increase its production. The high proportion of extractives in lodgepole pine, combined with its hardiness and rapid growth rate, make it a suitable species for production of pulp and tall oil. Combinations with later harvest of timber wood with a low content of juvenile wood can be possible.

Other uses of tall oil could be developed simultaneously. "Natural products" like soft soap from tall oil are becoming more popular as we become more aware of how our daily habits affect the environment. The development of new textiles, chemicals and other products from biomass that can replace oil-based products is occurring rapidly and may be one of Sweden's most lucrative industries in the future. It may also be possible to extract the antioxidant (+)catechin from lodgepole pine for use in health products. In addition, a use may be found for DHPPG, the phenolic compound that is abundant in lodgepole pine but is absent from Scots pine.

It is also interesting that bio-refineries can make use of the whole tree. Any remaining parts of the tree can be used for heat- and power production. Combined heat- and power plants could, conversely, become producers of ethanol and other bio-based products.

In conclusion, there is great potential for using lodgepole pine in bio-refineries. In the future, we will probably see a wide spectrum of bio-refinery products and changes in the silvicultural practices associated with lodgepole pine that make it more suited for use in bio-refineries.

6.2 Need for further research

The main problem with bio-refineries in the future will probably be a shortage of biomass. The competition for biomass is increasing. The production of crude tall oil is likely to be insufficient if the production of biodiesel from tall oil begins. Lodgepole pine may be the perfect species for the production of tall oil. Most of Sweden's existing lodgepole pine-stands were planted in the 1970s and 1980s, and are, therefore, fairly even-aged. Research is needed to determine how to produce biomass from lodgepole pine optimally. The economics of such silviculture are important subjects for research, as is the detailed chemistry of lodgepole pine. Thus, investigations are needed to examine the suitability of lodgepole pine for use in bio-refineries and to determine the silvicultural methods necessary to produce an appropriate raw material.

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