Saxion Kenniscentrum Leefomgeving

Chapter 3:

Development Midstream processing of Flax-fiber

Final Report

Investing in Opportunities

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Summary

Stricter environmental policies, increased energy prices and depletion of resources are forcing industries to look for bio-based and low carbon footprint products. For industries, flax is interesting resource since it is light, strong, environmental friendly and renewable. From flax plant to fiber products involves biochemical and mechanical processes. Moreover, production and processing costs have to compete with other products, like petroleum based materials. This research focusses on sustainable process improvement from flax plant to fiber production.

Flax retting is a biological process at which mainly pectin is removed. Without retting, the desired fibre remains attached to the wooden core of the flax stem. As a result, the flax fibres cannot be gained, or have a lows quality. After retting, the fibers are released from the wooden core. Furthermore, machines have been introduced in the flax production process, but the best quality fibers are still produced manually. Due to the high labor intensity the process is too expensive and the process needs to be economical optimized. Since the retting process determines all other downstream processes, retting is the first step to focus on.

Lab-scale experiments were performed to investigate the retting process. Factors that were researched were low cost processing conditions like, temperature, pH, dew retting and water retting. The retting rate was low, around three weeks for complete retting. The best retting conditions were at 20°C with water and any addition of chemicals. The process could be shortened to two weeks by recycling the water phase.

In a scale-up experiment, a rotating drum was used at the optimal conditions from the lab-experiment (20°C and water). First the flax did not mix with the water content in the rotating drum. The flax was too rigid and did not tumble. Therefore, bundles of flax plants were used. The inner core of the bundle seemed to be protected and the retting rate was less compared to the flax on the surface of the flax bundle. This implies that mechanical impact increased retting in the rotating drum, however heterogeneous retting should be avoided.

To overcome the heterogeneous retting problem, a water column was used to improve heterogeneous retting. Retting was performed in a water column and mixing was accomplished by bubbling air. As a result of the mixing, the flax bundle was retted homogenously. And after drying, it was possible to separate the fibers from the wooden flax core. Retting with a bubble column can overcome this problem and seems to be a usable retting process step.

Water samples of the lab-scale experiments, the rotating drum and the bubble column showed a chemical oxygen demand (COD) content up to 4 g/L. Overall, 1 kg Flax resulted in 40 g COD. This indicates the possibility to produce biogas that can be used for generating heat and electricity, to make the process sustainable.

Around 50% of the weight consists of wooden shives. The shives can be used for pyrolysis and it was possible to produce around 30% coal and 20% oil. These compounds can be used as building blocks, but also to generate heat and electricity. Heat and electricity can be used for the flax processing. Shives were only dried for 1 day at 105°C and slow pyrolysis was used. This indicates that a higher yield can be expected at fast pyrolysis.

Overall, the reported implicates that quality fiber production from flax plant can be a feasible, sustainable and a renewable production process. Feasibility of the process can be obtained by, (1) retting at low-cost process conditions of 20°C and using water without any addition of chemicals, (2) with increased flax retting rate by recycling water, (3) with increased flax retting rate by introducing mixing forces, and the ability to lower the energy consumption of the overall process, (4) producing biogas from the COD with anaerobic digestion and (5) producing pyrolysis oil and pyrolysis charcoal.

Contents

3.1 Introduction

Flax (*Linum usitatissimum*) has been used in linen for centuries especially in the Netherlands, northern France and Belgium, (Naik et al., 2014). After the 1950's, the production of linen from flax declined due to cheaper alternative technologies in textile industries like cotton and plastic (fibers). Nowadays, flax has the attention again from several industries like the auto industry (Khalfallah et al., 2014) and textile industry, since they are looking for the production of strong, light weight, renewable composite materials. Flax fibers are biodegradable and fully recyclable , (Deyholos and Potter, 2014).

Flax fibers can also be integrated with other natural or artificial materials and form composites. For example: flax fibers/polypropylene composites (Martin et al., 2013). These composites lower the impact on the environment (e.g. carbon footprint) with keeping quality compared to 100% artificial products. Moreover, new products can be composed with special properties. The combination of the matrix and the fibers results in a composite that can be stronger than the separate components. The SchwinnVestige flax fiber reinforced composite bicycle frame is an example. The composite exists of 80% flax, 20% carbon fiber, (Deyholos and Potter, 2014)

Disadvantages of flax fibers compared to synthetic fibers are the absorption of moisture, odor and lower fire resistance. Because flax fiber is a natural product, it has probably a higher natural environmental degradation. Also, fibers have a poor adhesion on composite material and currently this is still an issue (Deyholous and Potter, 2014)

Industrial processing to obtain several flax fiber properties (yield, strength, elastic modus) are obtained by biochemical plant processes. The mechanical properties of the fibers are unaffected by the flax plant size, (Bourmaud et al., 2015) .The quality of flax can only be decreased in quality by wrong processing and treatment. The companies demand for a certain quality has to be fulfilled by the final product after the processing steps, (Lammers 2014)

The production chain from plant to flax products is still economically limited caused by several factors:

1. With many natural products a large variation of the quality of the Flax plant can occur due to age and field exposure, (Foulk et al, 2001).

2. Variation in several types of the Flax plant.

3. A poor adhesion with conventional resins

4. Moisture absorption

5. High process cost . (Deyholos and Potter, 2014).

At the moment the major flax production areas are France and China and mostly for clothing and domestic textiles. (Deyholos and Potter, 2014) Whether the flax fiber properties have the same quality each year is still a debate, but results show that fiber quality could be constant regardless the year of flax cultivation (Lefeuvre et al., 2013).

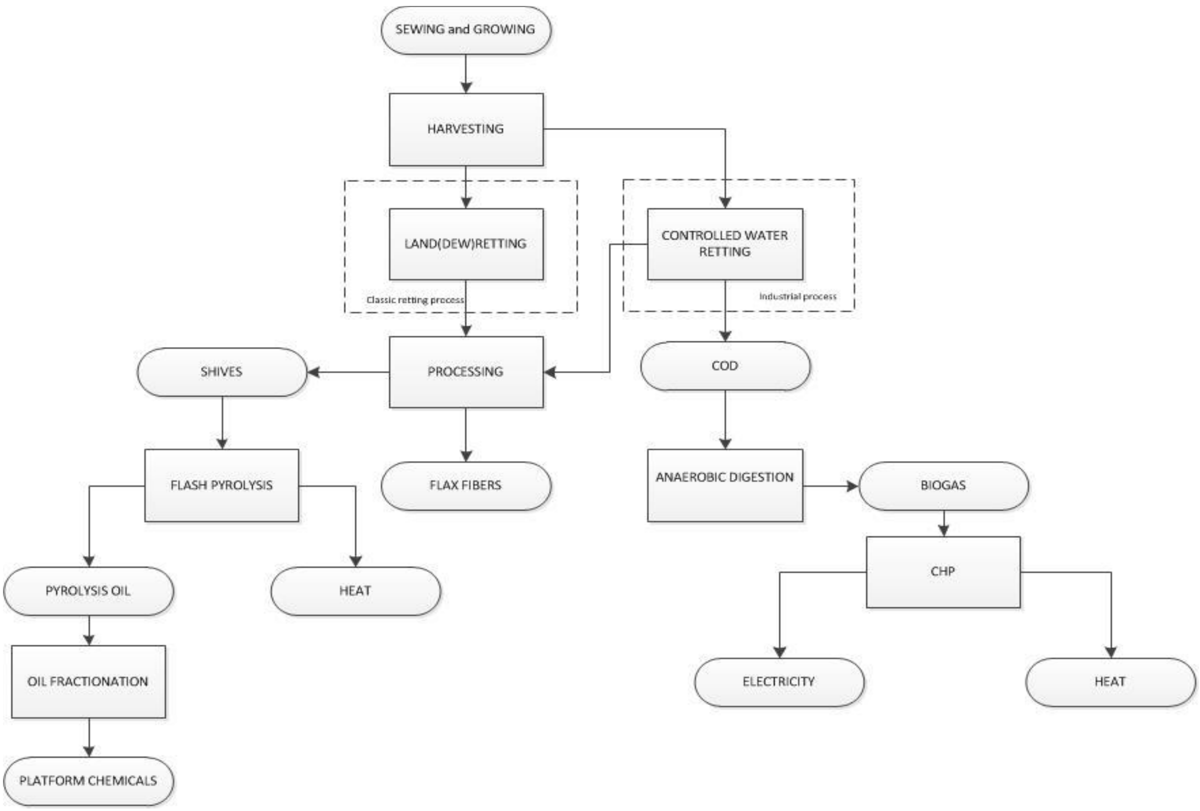
For the Twente region several barriers have to be taken to boost flax economy, among them are the price, quality and quantity of the flax production, and more research and development (Lammers 2014). Improvement of the retting process is the key step in processing from flax to product.

Flax fibres are sustainable and have a low impact on the environment, however also production needs to be sustainable to claim sustainability of flax fibres. Knowledge of, sustainable (bio)processing, conversion of biomass to high value building blocks, cradle to cradle processes, and knowledge of bio-energy and bio-based economy are needed to develop a suitable production process.

The retting process is researched, since this process is the first step and affects the further downstream processing. Other parts of the flax plant are secondary products, but the mount makes these composites useful. Shives are the major part of trash components of flax retting (Akin et al., 2005) This report investigated also shives pyrolysis to improve the total flax production chain. The pyrolysis proses can help to make the process energy neutral. The challenge is to design a sustainable, independent energy and CO_2 neutral processing system to contribute to the flax process (Scheme 3-1).

Objective of the Grow to Build retting research :
The effectiveness of the biochemical retting is very important but the knowledge and the efficiencies of existing chemical and mechanical processes are incomplete (Hoondal et al, 2002). A better understanding of process conditions on the biochemical activities is required to produce economically fibers with the desired properties.

First a brief process from seeds to fibers is described. Subsequently, the retting processes are discussed since this is a crucial step. In lab scale experiment some low cost processes are researched to enable retting. Next, the lab-scale processes are scaled up and a rotating drum is used. Later also a bubble column is used to improve the homogenous retting quality. Then, a side step has been made to research the use of shives in pyrolysis processes. Finally, conclusions and recommendation are given to improve the sustainability and economics of the flax production chain. The overall aim is to produce flax fibers at low costs, good quality and also sustainable. The latter is important since the main reason to use flax composites is the fact that is renewable. This means that the process should also be sustainable.



Scheme 3-1: Simplified overall process of the flax pyrolysis in this research.

3.2 The overall process: from flax to yarn

3.2.1 Simplified process: from plant to fiber

A simplified process scheme of flax processing is given below. All process steps are briefly described to understand the total flax processing. Retting is described in more detail at the end of this paragraph.

The flax plant

The flax plant (*Linum usitatissimum* Translated: very useful) is an annual plant with blue and white flowers. Flax can be grown for seeds or for fibers. For seed production, the plant has more branches to produce a maximum amount of seeds per plant. The seeds are mainly used for linseed production. For fiber production, the plant is more tall and less branches are needed to produce long fibers (Engelen 2015).

Harvesting

Flax can be harvested with the roots attached to the plant or without the roots. Also machines are available for harvesting, however the best flax quality fibers are obtained from manual harvesting. (Decktowel, 2014). For economical harvesting, quality versus labor is important. In some countries the land cost is too high to produce flax, even when the best quality of flax has been grown,(Lammers 2014).

Retting is the process in which the bast fiber (the desired flax fiber) is released from the wooden parts of the flax plant. This retting process is described in more detail in **3.2.2.**

Scutching the flax

After the retting process, flax needs to be dried before breaking and removing the wooden core pieces. This process is called scutching. Scutching can be performed on a wooden knife. This process can be performed by machines, but also manually. The manual process results in the best quality fibers but is labor expensive. The better flax is retted, the higher the fiber yield. Else the fiber is still connected to the wooden part and will be vanished, (Decktowel, 2014). If the fibers are over retted, then a greater loss of fiber will be present in the wooden pieces fraction, that is the shive fraction, (Pallesen, 1993). The wooden pieces can be used as construction material, filter material, floor material and bioenergy production (Engelen 2015). Shives and other parts of the flax might be treated with ethanol to recover wax or lipids, (Holser et al., 2008).

Heckled

The separated flax fibers are further processed by heckling. Heckling is combing the fibers by a bed of nails. In this process, the shorter fibers (tow) will be separated from the longer fibers. The shorter fibers can be used in rough yarn, low quality linen, isolation material. The longer fibers are used high quality linen, (Decktowel, 2014).

Spinning – Weaving

Log thin flax fibers are used to produce very fine yarn. Fine yarn can be combined to produce thicker and stronger yarn. Before bleaching and dyeing, flax is frequently treated by different processes: damping to adjust moisture content, beetling and stenting. Different treatments result in different linen products, for example dress linen (Hoondal et al, 2002)

Flax properties for yarn

Advantages of natural structures above synthetic glass fiber structures is that natural fibers are less abrasive and therefor more friendly to processing machines and employees, (Deyholos and Potter, 2014). Compared to cotton yarn, flax is smooth, a little more brittle and less flexible. On the other hand, flax has a higher tensile strength.

Flax fibers are relatively smooth, straight and lustrous. They are more brittle and less flexible than those of cotton. The yarn produced, however, has a higher tensile strength (Hoondal et al, 2002). For yarn industry, thin and well separated flax fibers are needed to process the fibers to thin yarns (Martin et al, 2013). Thin and well separated fiber bundles can be used as selection criteria for process optimization.

3.2.2 Retting processes

General process description

Retting is the process at which the bast fiber (the desired flax fiber) can be released from the wooden parts of the flax plant. Pectin and calcium bind the bast fibers to other parts of the flax. By removing the pectin the bast fiber is disconnected and bast fibers can be separated. Pectine removal is a natural process in which pectine is degradeded by microbial processes. The retting process results in a decay of the pectine and microbes generated energy for their metabolism. After retting, fibers are dried, broken, scuthced and hackled to obtain fibers without impurities, (Nair et al., 2014)

Retting processes affect also the quality of the desired bast fibers. Microbes have the ability to decay bast fibers as well. It is important to be sure that the microbes selectively remove pectin and leave the bast fibers unaffected. This retting selection can be accomplished since bast-fibers consist of cellulose compounds. This means that cellulose decaying micro-organisms should be avoided. The selective capacity of pectin removal enzymes (pectinase) can be used in this process. Pectinase is designed to degraded pectin, and will prefer the degradation of pectin instead of the bast fibers. This implies also the risk of chemical retting, since both, the pectin and the bast fibers, are subjected to the same chemical processes.

Well retted flax is easier to process further by scotching and hackling than unretted flax (Lefeuvre et al, 2013). The retting progress decreases the bast fiber percentage present in the raw flax material (Pallesen, 1996) and also the fiber quality is affected Overall, the retting process is a crucial important economical factor in the overall production process. Therefore , several retting processes are compared to find the best cost effective retting process.

On laboratory scale, the effluent stream from retting can have a chemical oxygen demand (COD) range to over 19 g/L,(Naik et al., 2014). This means that a part of the COD can be used as renewable energy if this is converted to biogas (around 7L biogas per liter effluent of 19 COD g/L) Water retting (see below) in stagnant waters takes less time compared to water retting in streaming waters perhaps since microbes can develop in the water. In streaming water retting the produced fibers are cleaner, perhaps due to the dilution effect. Water retting in the river Lys in Belgium gives a very high quality of fibers

Dew retting

In dew retting the harvested flax stems are laid on an open field and microbes are allowed to start the retting process (Deyholos and Potter, 2014). The retting time varies between 10 days to 6 weeks, (Naik et al, 2014). However, night dews and warmer day temperatures are needed for an effective retting process. (Decktowel, 2014) .

The **advantage** of dew retting is its minimal investments and easy to operate, (Hoondal et al, 2002).

An **disadvantage is** the chance on variable weather conditions. This can give a poor quality and yield of the fibers, (Naik et al, 2014).

The quality decrease can directly be affected by the weather conditions, (Deyholos and Potter, 2014), but also some weather conditions stimulate the formation of cellulose degrading bacteria and this results in fiber quality reduction as well

(Hoondal et al., 2002). In general, the quality of the fibers is uncertain due to the variable process conditions in the open field.

Wet or water retting

Water retting can be performed with anaerobic bacteria, (Hoondal et al, 2002). Long exposure of flax stalks to water results in osmotic pressure in the plant cells. This osmotic pressure results in damaged cells and nutrients are released in the water and are now available for the present microbes. Also, the damaged cells in the stalks resulted in open places for microbes to enter the stalks further. The retting process is accelerated. Water retting is performed in natural waters.

An **advantage** of this wet or water retting is the shortened time period for the whole process compared to dew retting. Water retting duration is up to 10 days, (Naik et al., 2014). The temperature buffer capacity of the water results in a more controlled temperature. On one hand, water retting is common practice, on the other hand it is more expensive compared to dew retting.

The main **disadvantages** of water retting is the high labor intensive process, and also extreme water pollution (Naik et al., 2014; Deyholos and Potter, 2014) is a drawback of this process.

Tank retting

Tank retting is water retting, but the water pond is artificial. During the retting process acids might be released in the water, these acids are corrosive and therefor tank retting is performed in non-metallic reactors, like concrete. Often, first the stalks are washed. Then the actual retting starts with fresh water in around 4 to 6 days, (Akin 2013). **Advantages:** The retting process is controlled and uses relatively simple equipment.

Disadvantages: The amount of water that is used during the water retting process.

Enzymatic retting

In enzymatic retting the straws are in water. The enzymes are added to accelerate the processes. Different enzymes have been described by several researchers (Table 3.1). Because of these different enzymes several optimum process conditions for enzymatic conversion are known and given. Optimum process conditions for enzymatic conversion can have a negative impact on the fiber quality. An example can be given with Scourzyme L, were detected optimum of 60°C for the retting process (quality), but the optimum conditions for Scourzym-L are pH=8 and T=40°C, (Table 3.1).

Advantage is that the retting process time is shortened. For example in 9 to 15 hours with Scourzyme-L, Another advantage is that the enzyme in principal can be recycled to reduce enzyme costs, (Naik et al., 2014).

The main **disadvantage** is the high cost of the enzymes.

Table 3.1: Flax retting enzymes

Enzyme	Remarks	Source
Pectinase	Separate the fibers and removes pectin	(Hoondal et al., 2002)
cellulases	Improvement fibers flexibility in combination with preservation of tensile strength	(Hoondal et al., 2002)
Viscozyme L (Novo Nordisk, Franklinton, NC, USA)	0.05% v/v Enzyme solution : 50 mM EDTA in water at pH=5 40°C for three days	(Foulk et al., 2001)
Polygalacturonase (Pgase from *Rhizopus sp.* , *Viscozyme L*, and *A.niger*)	Several PGases, and Pgase from *A.niger* is a better retting agent at 40° for 20 hours	(Evans et al., 2002)
Scourzym L	60° for 9 hours (high optimum temperature) Scourzyme-L. from Novozyme, Denmark Enzyme conditions: pH =8 and T=40°C 275U/ml	(Naik et al., 2014)
Flaxzyme	Culture filtrate from *Aspergillus*	(Henriksson et al., 1999)
Pectinase activity	Pectinase from *Rhizomucor pusillus*, pH=6 40°C	(Henriksson et al., 1999)

Microbial retting

Microbial retting has similarities with water retting or dew retting, this paragraph focusses on the microbial community. Microbial retting involves often a mixture of several microbial populations (Table 3.2) . First these microbial mixtures have to develop from the start of the retting process. These microbial mixtures deliver the enzymes that catalyses the retting process. The main difference with dew or water retting is that the microbial population can be added to start the retting process.

Advantage: the microbial population provides the enzymes that break down the pectin bonds, development of the microbial community is a spontaneous process and therefore, costs are reduced. Microbial communities might be cultivated to speed up the retting process and to create a microbial population for retting.

Disadvantage: also unwanted enzymes can be released from the micro-organisms to the stalks. It is suggested that non-cellulolytic enzyme solutions produces the strongest fibers, (Evans et al, 2002). Control of microbial activity, or enzyme activity is possible by treating the catalysts with ammonia, addition of ammonium stops the retting process, (Pallesen, 1996) For effective retting, hemicellulases or cellulases are not essential (Henriksson et al., 1999)

Table 3.2: Microbial communities

Microbial population	Remarks	Source
Soft-rot bacteria	Microbial pectinase (pectin-depolymerizing enzymes) release cellulosic fibers from fiber bundles	(Hoondal et al., 2002)
Rhizomucor pumilis	High levels of pectinase activity, cultivation up to 54°C.	(Henriksson et al., 1999)

Chemical retting combined with heating

Chemical retting is also a wet retting process and an chemical is added to the liquid phase to accelerate in combination with heating the retting process. An example is base, 12–20% NaOH and boiling (Hoondal et al., 2002).

The **advantage** is the shortening of the process time since this is a relative quick process. After around 24 hours soaking and 1 to 4 hours boiling, the retted flax stems can be processed further.

The **disadvantage** are the use of chemicals that are costly. Often, these chemicals have to be neutralized and removed from the fibers after use. With rising energy costs and depletion of chemical sources and the impact on the environment (waste production) chemical retting seems to be an unattractive process. The effluent water stream from chemical retting can contain toxic, non-biodegradable products. Moreover, chemical treatment is non-selective. This means that a part of the chemical activity or treatment power is lost to unwanted side-reactions.

Other retting processes

Flax can be crimped **by rollers**. The crimping process generates space in the stalks for enzymes. As a result the retting process could be accelerated, (Foulk et al., 2001). Treatment with **vacuum processes** effect on the retting process of untreated flax stems. However, the effect of vacuum on retting process time of stems that were treated with rollers was limited (Foulk et al., 2001). **Micro-wave assisted** retting affected the retting process, but in total also other parameters as temperature and time are still of significant influence (Nair et al., 2014).

Retting procedure	Electrical energy usage	Chemical added	Process time	Water use	Fiber quality	Biogas production possibility	Shives quality	Environmental impact	Investment costs	Specific costs (consume)	Sum +	Sum-
Dew retting	+ +	+ +	- -	+ +	- -	- -	+	+ +	+ +	+ +	13	6
Water retting (open water)	+ +	+ +	0	- -	+	-	+ +	- -	+	+ +	10	5
Tank retting (closed water)	+	+ +	0	-	+ +	+ +	+ +	0	0	+ +	10	1
Enzymatic retting	0	- -	+ +	+	+ +	- -	+	- -	0	- -	6	8
Microbial retting (added microbes)	0	+ +	+	0	+	+ +	+ +	0	0	0	8	0
Retting and Pre-processing (rollers or vacuum)	-	+ +	+	N/A	N/A	N/A	+	N/A	- -	N/A	4	3
Micro-wave assisted retting	- -	+ +	+	N/A	+	-	+	0	-	-	5	5
Chemical retting	+	- -	+	- -	+	- -	-	- -	0	- -	3	11

Matrix 3-1: *Overview of the retting procedure and the positive (+) or negative (-) effect on: electrical energy usage, addition of chemicals, water use, fiber quality, biogas production quality, shives quality, environmental impact, investment costs, and specific costs (consumables).*

Overall, fiber quality is limited for dew retting (Matrix 3-1). However, the dew, water and microbial retting processes have overall a high positive result; Sum + > 7 (Matrix 3-1). Enzymatic retting is quick and result in good fibers, however the process has also negative impact on side processes (Matrix 3-1).

3.2.3 sub-conclusion and recommendations (retting)

Variation in the quality of the yearly flax harvest results in difficulties to produce flax fibers with constant quality. The best quality fibers are still produced with labor intensive work. The labor intensive work results in relatively high costs. Flax worker experience cannot be replaced by machines. As a result: Costly processing results in high quality fibers and cheap processing (machines) results often in poor quality fibers. The main crucial step in the production of fibers from flax is the retting process.

Controlled retting in the lab might reduce the variation in quality and time can be saved due to optimization of the process. Currently, dew and water retting are the most applied retting processes. This means that natural processes have an advantage above artificial retting proceedings: low impact on the environment and no costs for additions.

Active biomass can be reused by filtrating the biomass from the effluent. Perhaps not all biomass can be re-used, for example cultivation of cellulose degrading biomass should be avoided. The high COD in the effluent can be bio-converted to biogas (methane) for energy production. By recycling the biomass, microbes can be cultivated with ideal properties for retting conditions.

An option for retting is to use of mechanical interaction (for example a rotating drum reactor or a bubble column). The stalks are weakened by the mechanical forces and opens the route for the micro-organisms and enzymes to reach the pectin.

3. 3 Lab-Scale Retting Experiments

3.3.1 Introduction

Retting is a crucial step in the flax processing chain from flax plant to flax fiber. This crucial step has an enormous diversity in retting methods(see chapter 3.2). The main varieties are:

(1) dew retting on the field with uncontrolled weather conditions;

(2) more process controlled water retting ;

(3) enzymatic retting

(4) chemical retting

(5) biotechnology retting with selected enzymes from micro-organisms.

A lot of research has be done in enzymatic retting, however since 2012 the amount of enzymatic research declined. The reason for this decline is the lack of support from industry and the relative high prices of enzymes, (Atkin et al., 2012). Dew retting is currently common and provides retting with low costs, on the other hand retting conditions are uncontrolled and this can result in fiber quality losses. Altogether, natural retting (dew or water retting) processes are the most common retting methods at this moment.

To overcome quality losses by natural retting a more controlled retting would be beneficial for the fiber quality. On the other hand, the investments costs that enables controlled retting should be as low as possible. Therefore in this research, the focus is on process control (Temperature and pH) that enables spontaneous retting. That is, the microorganism responsible for retting are already on the flax stems. These microbes are responsible for retting and need a controlled climate to develop (microbial growth). This includes too that the process remains natural and this reduces probably waste stream treatment costs after the retting process. The controlled natural retting process needs to compete with enzymatic retting (enzymatic retting results in high quality fibers but the process is expensive) and dew retting (possible losses of fiber quality but is a relatively cheap process). Therefor the benchmarks are set on: Production as cheap as possible (dew retting), production as fast as possible (4 hours for enzymatic retting). Moreover, production of long flax fiber is desired.

To define the best conditions for natural retting at controlled process conditions, small scale experiments need to be performed. These process conditions are unknown for the flax used and also for the water (tap water Enschede) used. The obtained process conditions can be used in the scale-up experiments. Further, limitations in the retting process can be detected. This is important for long term experiments, for example the risk of acidification in water recycling experiments.

In this research process conditions like, temperature and the pH of the water are varied to test the effect on retting. Besides water retting it is also tested if dew retting could be simulated in the lab at 20°C and 40°C. To accelerate the retting process, it could be beneficial to reuse the water with the microbes and the enzymes from previous batch-experiment. Micro-organisms that provide retting have been grown in the medium in a previous batch experiment. By recycling the water, the start of the microbial growth phase can be shortened. All experiments are performed in bottles and can be compared with tank retting.

3.3.2 Material and Methods

Flax, *Podium late*, was used in the batch-experiments. This flax was stored at room temperature (19.9°C-21.2°C) and the relative humidity varied between 26%-39%. The roots and the top of the flax plant were removed and 17 cm long mid-stems were used for all batch-retting tests. Both, water retting and dew retting, were simulated in the batch bottles.

Batch bottles

Unless stated otherwise, all batch bottles were prepared in 500 mL glass bottles and a total of 10 g mid-stems flax was added (see also figure 3-1). The bottles were prepared in open air, but during incubation the bottles were closed and an anaerobic environment could be created. During sampling, the bottles were opened at atmospheric conditions. The gas phase could be filled with oxygen air again. Bottles were place on an orbital shaker at 100 rpm.

The effect of temperature was studied at T=20°C and T=40°C. The effect of pH during the water retting was studied at both temperatures with the addition of 0.1 M HCl or 0.1M NaOH. A total of 5 mL base or acid was added and labelled as "minimum". A total of 21 mL base or acid was added and labelled as "strong". Dew retting was studied by starting the experiments with the addition of a minor amount of water (10 mL). Finally, for water retting, the liquid of some bottles were re-used to see if the retting could be accelerated. Control experiments were performed with 10 gram mid-stems in a closed empty bottle.

Measurements

The rate of retting was analyzed by a modified Fried test. A sample of 4 stems was transferred to a Fried tube. This tube was 20 cm long and 7 cm in diameter (770 mL). Subsequently, around 150 mL just boiled water was added and the tube was closed. After 10 second vertical shaking the samples were analyzed for a Fried test value. The fraction of fiber that was free from the flax stems was reported as retting: 0=No retting, 0.5 = initial retting, 1= between initial retting and 50% retting, 1.5 = 50% retting, 2=between 50% and almost totally retted, 2.5=almost totally retted and 3=totally retted.

After retting, the flax stems were dried at room temperature. The dried retted stems were manually processed and the fibers were separated from shives. The quality of processing was valued: 0= not , 1= bad, 2=good, 3=very good. Also the quality of the fibers were classified as soft and thin (value 3), intermediate (value 2) and rough and brittle (value 1). An example of good processing (value 3) and soft and thin fibres (value 3) is given in figure 3-12B.

Figure 3-1, batch retting experiments. The bottles#1 and #2 are blanc. The bottles #3, #4 and #13 are dew-retting experiments. All other bottles contain water.

3.3.3 Results and discussion

Control experiments

Control experiment were without water and showed almost no retting degree in three weeks. For both temperatures the Fried factor was 0 at the end of the incubation time (Figure 3-2).

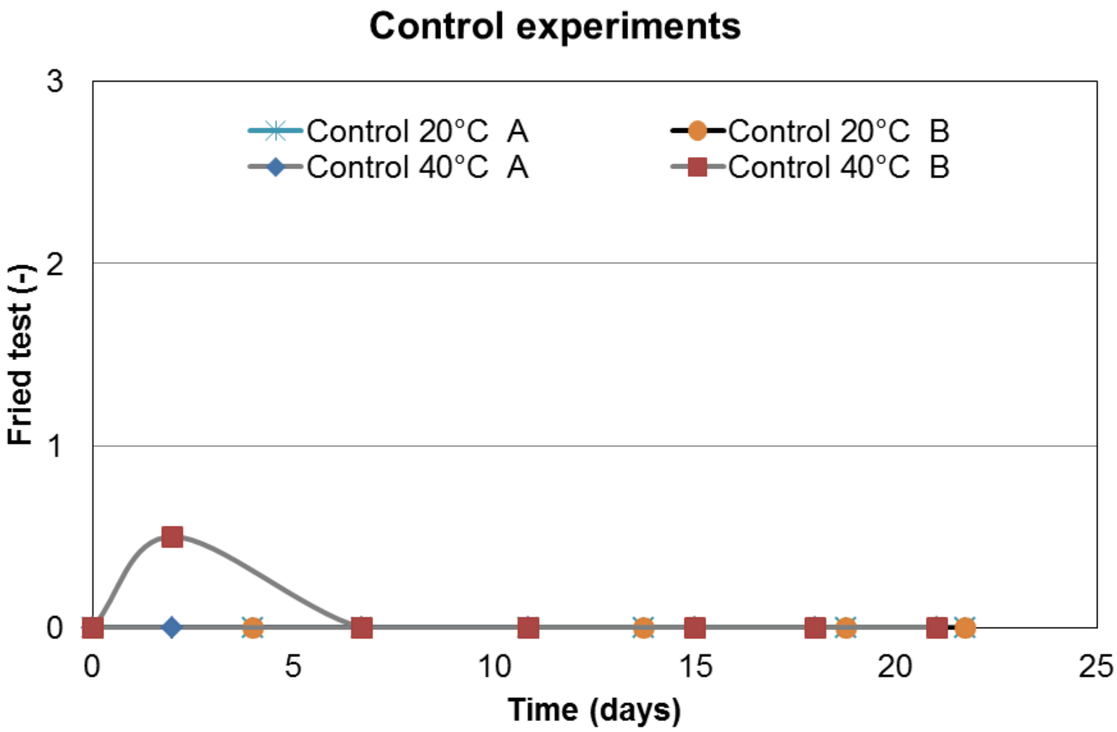


Figure 3-2, control experiment at 20°C and 40°C. Control experiments did not result in retting according the Fried test.

Dew Retting

Dew retting was simulated by the addition of a 10 mL water. Dew retting resulted in a Fried test values of 2.5 and 3 in three weeks. Literature indicate dew retting times of 10 days to 6 weeks (Niralli et al., 2012). However, in this experiment the temperature was kept at a constant value, in dew retting on the land fluctuations in temperature might have accelerated the retting rate. Overall, the addition of a small amount of water resulted in fully retted flax in three weeks, (Figure 3-3).

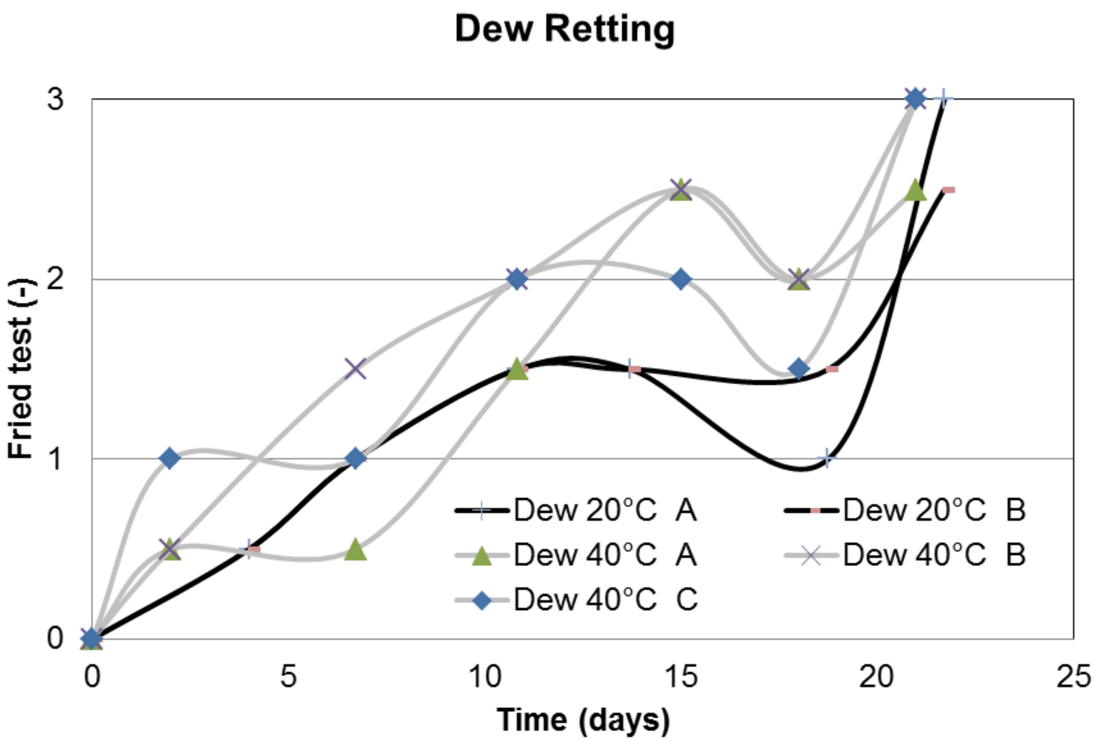


Figure 3-3, dew retting experiments at 20°C and 40°C. Dew retting resulted in retting and a Fried test resulted to 2.5-3.0 in 21 days.

Water retting (Neutral)

Water retting at 20°C resulted in comparable retting performance as the dew retting experiments at 20°C. Demi or tap water resulted in comparable retting rates (Compare figure 3-4 with figure 3-3). The final pH range in the 20°C experiments was 5.1-5.3.

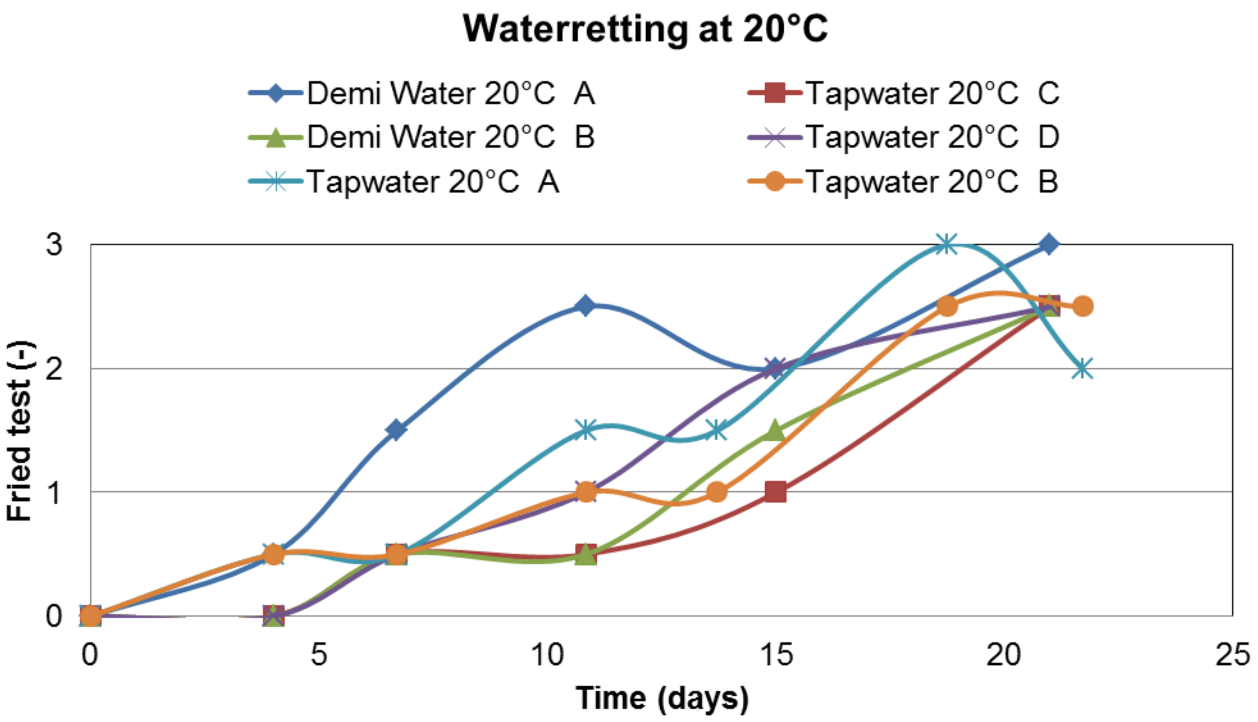


Figure 3-4: Water-retting at 20°C with tap and demi-water.

At 40°C the water retting was less compared to the dew retting at 40°C and the water retting at 20°C (compare figure 3.5 with figure 3.4 and figure 3.3). The temperature could have been too high for water retting micro-organisms. The pH ranged varied between 4.7-5.0 in the 40°C experiments. The difference in pH is small. Probably the pH is not limiting the retting at 40°C, as is explained in the next paragraph.

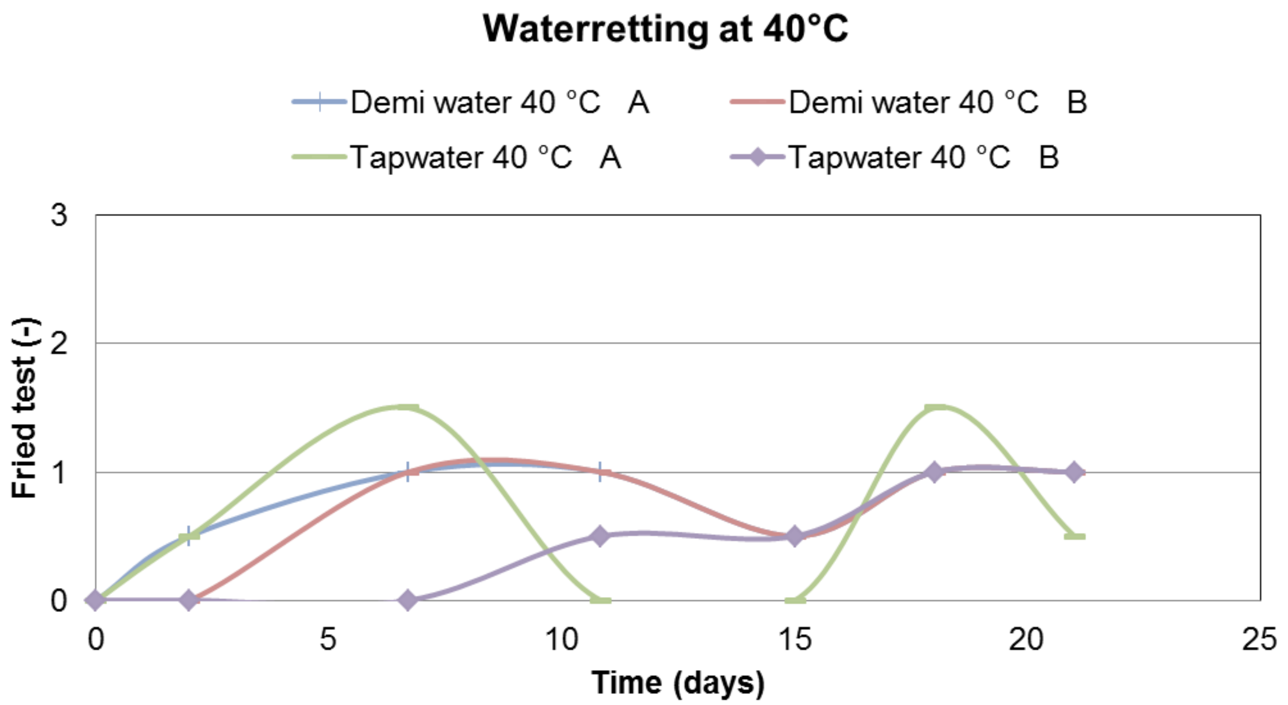


Figure 3-5 :Water-retting at 40°C with tap and demi-water

Water retting (pH-effect)

The pH in the water retting experiments were controlled by the addition of 0.1M NaOH or 0.1M HCl. Directly after the addition of the base and acid, pH-measurements indicated a pH as low as 2.8 and as high as 11.5. This range was already smaller, 3.5 and 5.1 at 7 days of incubation. Probably the biological compounds from the flax or compounds produced by the micro-organisms are able to buffer the pH to a certain value. This means that the pH value is a result of natural control and that pH control by acid and base is deactivated by the natural system. The pH, with no base or acid additions, has a value of around pH=5. And this value is a little bit higher with tap water instead of demi water. Finally, the pH was lower with acid addition and the pH was higher with base addition. See also figure 3-6 for the pH stability during incubation at 40°C.

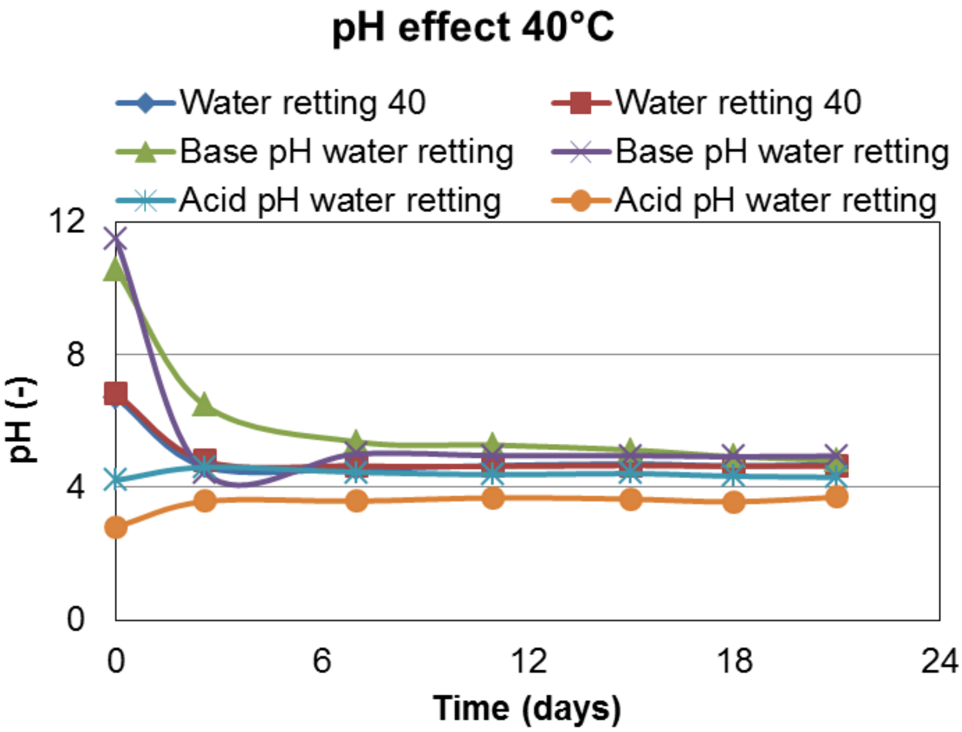


Figure 3-6 : The pH stability during the retting process at 40°C.

At 20°C the "strong" addition of acid resulted in a reduced retting degree (compare figure 3-7 with figure 3-8). The addition of base did not seem to have an effect on the retting rate. At 40°C the same result regarding the pH is detected, the addition of acid resulted in a lower retting rate (compare figure 3-9 with figure 3-10). The addition of base did not seem to effect the retting rate too much. Concluding, the retting process is limited by the addition of acid. The pH below 5 seems to limited the retting process with tap water. However with demi water and non-acid addition the pH was around 4.7 and retting seems to be complete after three weeks of incubation. Perhaps the addition of Cl^- (chloride) could have limited the retting process as well.

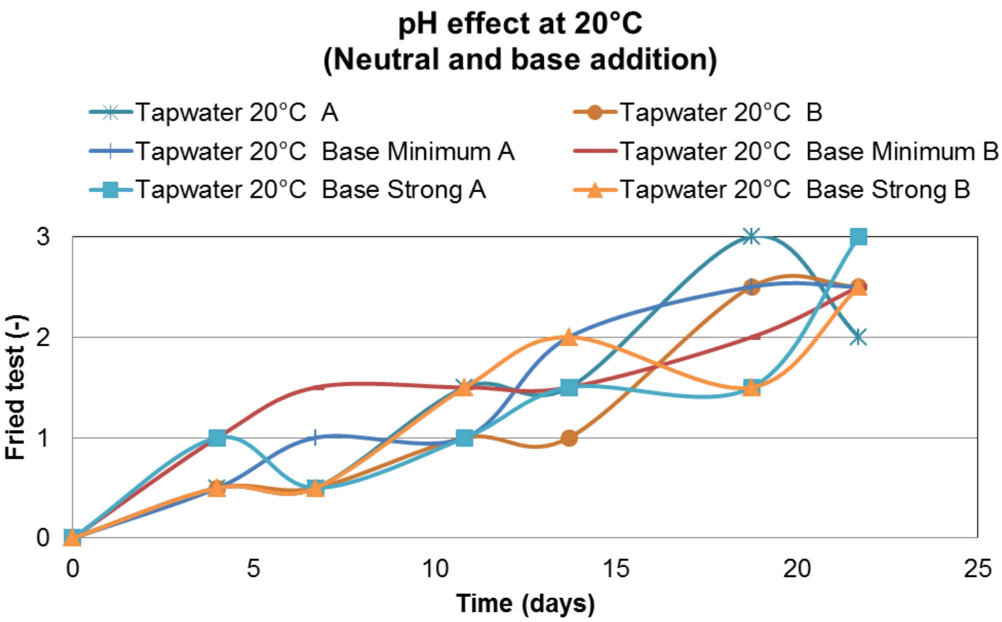


Figure 3-7, Neutral and base retting at 20°C

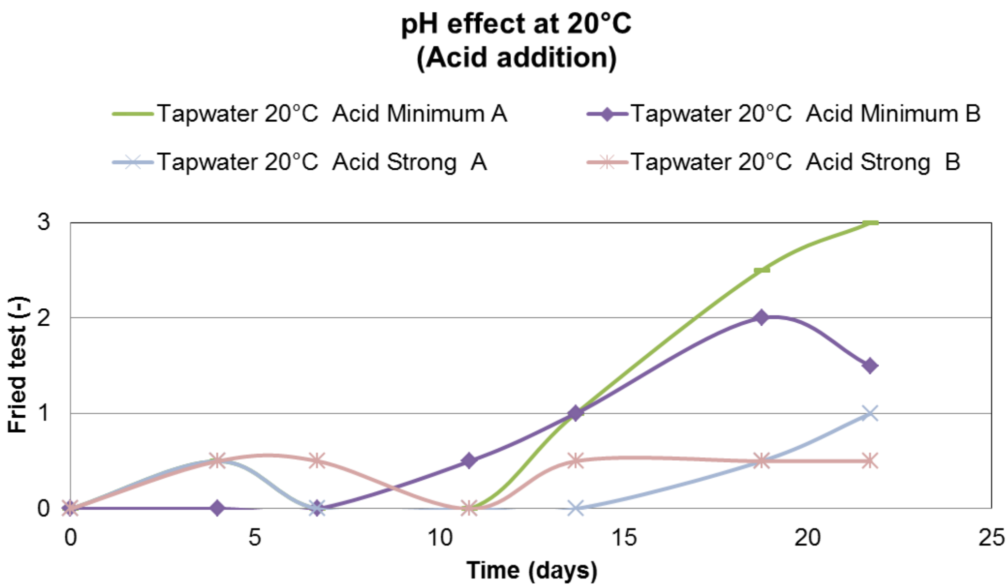


Figure 3-8 : Acid retting at 20°C

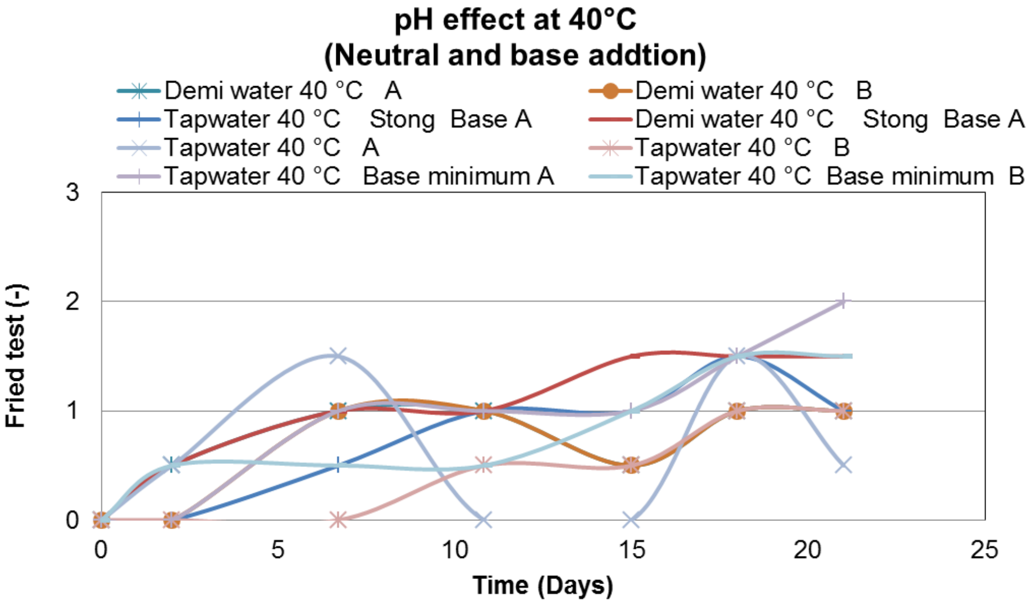


Figure 3-9: Neutral pH and base addition effect on the retting rate at 40°C

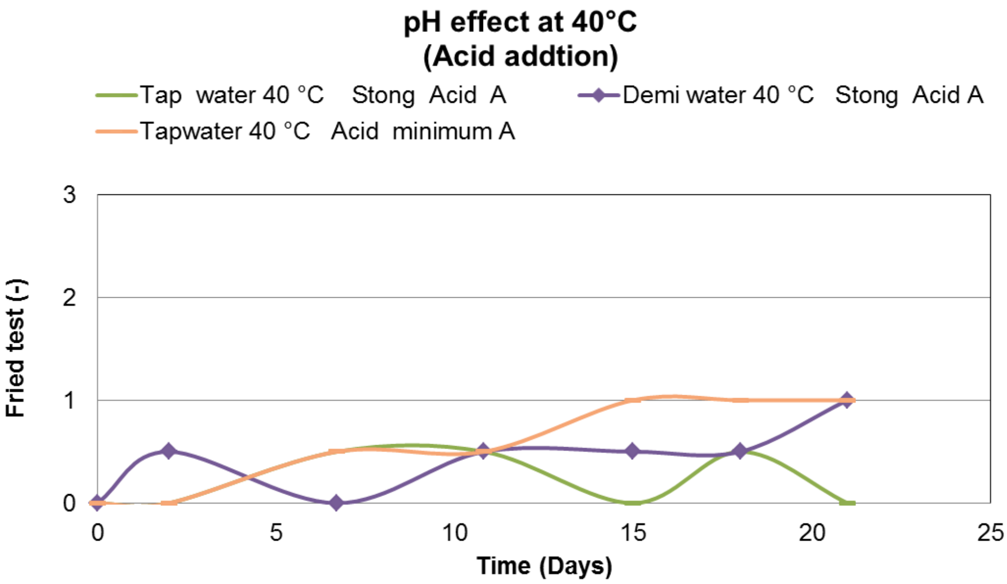


Figure 3-10 Acid effect on the retting rate at 40°C

Water retting recycled water

Micro-organisms that enhance the retting process have to develop in the first phase of the incubation. It was tested to reuse the water from previous incubation tests.

Water that was used for retting processes at 20°C without any base or acid addition was tested for retting at 20°C and 40°C. From figure 3-11 it can be concluded that the retting process time could be shortened from three weeks to two weeks. The higher retting rate is explained by the fact the micro-organisms need less time to develop and the enzymes responsible for the retting process were already present in the medium.

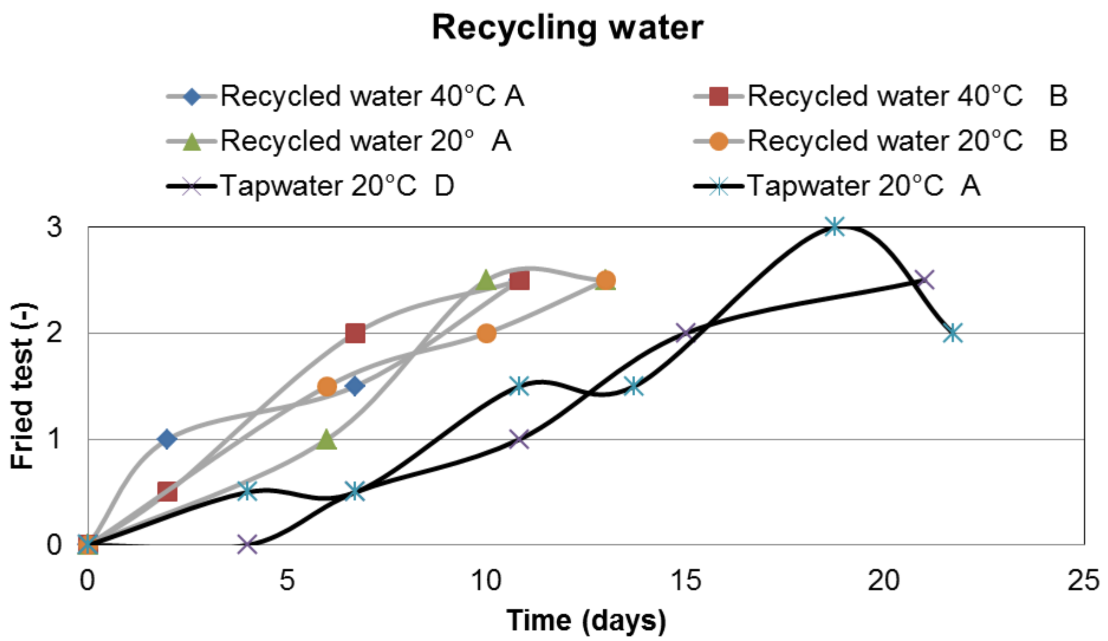


Figure 3-11 : Retting with recycled water and with fresh water. Dark lines fresh water, other lines recycled water.

3.3.2 Processing after retting and fiber quality

After the retting process step (Figure 3-12A), the flax straws need to be processed further. The processing was tested manually by separation of the fibers from the wooden core (figure 3-12B). This process is given a processing quality number. The processing number and the Fried test number was correlated for 20°C and 40°C experiments (figure 3-13 and figure 3-14). For both temperatures, the retting degree is correlated with processing degree and with the fiber quality.

At 20°C, higher values for processing and flax fiber qualities were obtained at low retting degree 1.5-2 (see figure 3-13). Perhaps the relative warm water of 40°C affected the Fried test (soaking at 40°C). This high temperature weakened the connection of the flax fiber with the wooden part of the flax. This could affect the Fried test, and the Fried test results are higher at 40°C than at 20°C. As a result, less fiber quality and less processing degree could be obtained at higher retting degree at 40°C (compare figure 3-13 with figure 3-14).

Figure 3-12A , Retted flax from several conditions.

Figure 3-12B , Lab-scale experiments and processing after retting. From left to right: (1) Water-retted flax stems. (2) Flax stems that were partly water-retted and partly dew retted. The upper darker part is dew retted. (3) Short fibers. (4) Corresponding retted and dried stems. (5) corresponding shives. The shives are clean and regarding the size of the shives (2 cm long) it was separated easily from the fibers. (5) A cloud of short fibers that was obtained during the separation of the shives and the fibers.

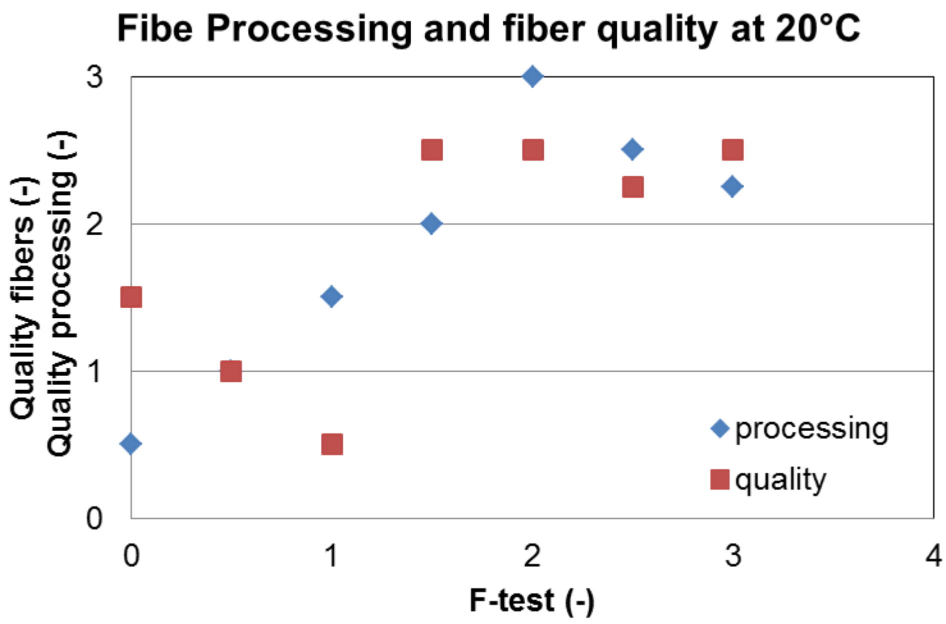


Figure 3-13: Relation between processing and quality of retting with the retting degree at 20°C.

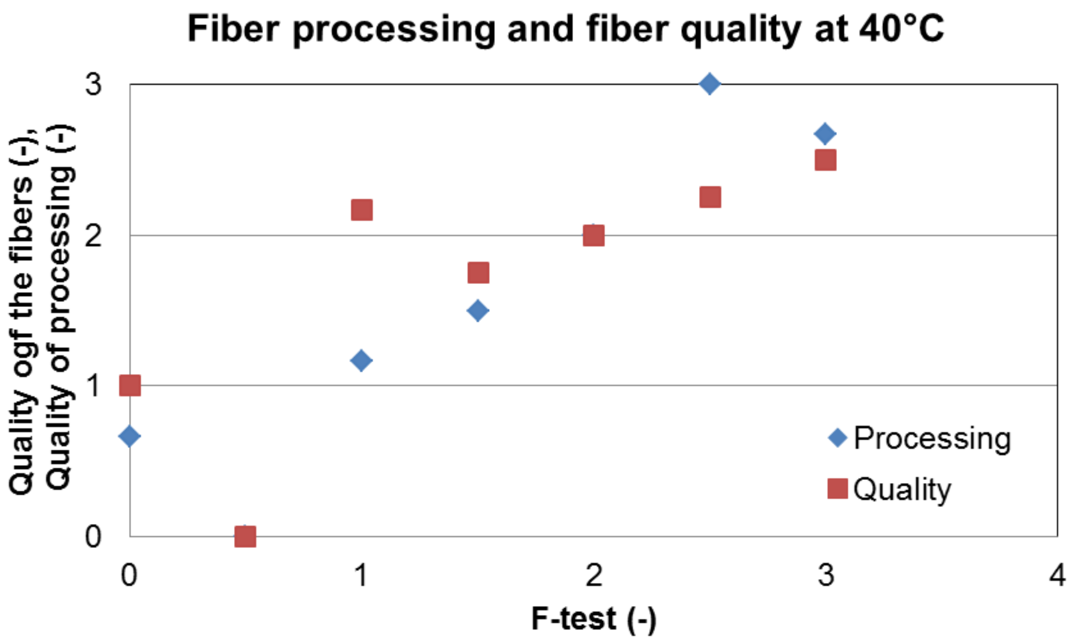


Figure 3-14: Relation between processing and quality of retting with the retting degree at 40°C

The quality of the flax fiber was correlated with the Fried test number. A relatively high Fried test number resulted in easy separable and to handle fibers. Most of the fiber properties were comparable at high retting degree. However, several fibers were really soft and thin and very easy in processing. These fibers were produced at 20°C with tap water and no added chemicals. Unretted flax resulted in poor fiber yield, broken fibers and damaged fibers. The dew retting flax stems resulted in darker coloured fibers. The shives produced consists of bigger pieces when the flax has a higher retting. The shives size can be used as a measurement for processing.

3.3.4 Sub-conclusion and recommendations (lab retting)

Water retting with recycled water at 20°C and a pH>5 resulted are the optimum retting conditions in these tests. Water retting should be performed at the lower temperature range of 20°C instead of the higher temperature range around 40°C. Both temperatures didn't seem to affect the dew retting rate. However, to accelerate the process, only water retting can be recycled for a next retting batch. Acidification of the process water should be avoided, since a lower pH (below 5), seems to have a negative effect on the retting rate. The water retting time is three weeks, and this is long compared to current water retting processes of 6-10 days. In the scale-up experiment 20°C and tap water will be used.

The process conditions (20°C and water) have a low impact on the environment. For dew retting, no recyclable possibilities are available. With water retting the process time can be shortened. This means that the process from flax to fibre can be made sustainable and CO_2 neutral and this could result in a stronger position for flax products in the bio-based economy.

3.4 Scale-up Experiments (Rotating Drum and Bubble Column)

3.4.1 Introduction

It is concluded to recycle water and to use controlled water (tank) retting for scale-up experiments. To make the total midstream sustainable, research was performed to use shives for the production of pyrolysis oil and coal (3.5). Finally, a preliminary midstream process is developed and presented in (3.6).

In previous experiments flax was retted as 17 cm samples. The production of long fibers (up to 90 cm) is desired since these fibers are valuable. The top of the flax plant is thin compared to the stem near the root. It is important however to produce long flax fibers, from top to root, with the same quality. Therefore a scale-up machine is build and experiments are performed with conditions from the lab scale experiments (20°C and tap water without any chemical addition).

3.4.2 Material and Methods

Figure 3-15: Set-up of the rotating drum for flax. The van is needed to cool the motor. The motor is equipped with an internal cooling system, but this internal cooling system is only effective at a high rotation speed. Flax and water can be added via the side of the drum. During the retting process, the water takes up natural compounds from the flax stems (on the right).

A rotating flax drum was built to scale-up the retting process (figure 3-15). Around 250 gram flax and 10 L water was used for the retting process. The rotation speed of the drum was around 40 RPM.

Analytical methods

Occasionally, the chemical oxygen demand (COD) was measured with a HachLange Kit and the results were achieved with a spectrophotometer.

3.4.3 Results and discussion

Several rotating barrel batch experiments were performed. Due to technical issues, climate control or uncertainties not all the batch experiments were useful. Here four batch experiments that represent the mixing problem in the rotating drum are presented.

First rotating batch

The first amount of flax were loosely placed in the rotating drum. The drum rotated, but the flax did not move. The inner surface of the drum was really smooth, and the dry flax was gliding along the inner surface of the drum. After 3 days, the flax had become wet and started to tumble. After one week turning the flax was retted. The fibers were damaged and could simply be demolished to smaller pieces. Moreover, the flax was mixed and if the fibers were still in one piece, resulted in a mixture and clothes of the flax. Probably no further processing could be obtained. As a results, in the next experiments, the flax stems were bundled first to 250 gram units (see figure 3-16A).

Figure 3-16: Long flax-fiber production with rotating drum and bubble column. (A) Flax bundle from rotating drum (B) Close-up of retted flax in a bundle (C) opened flax bundle (D) Long flax fiber production from bubble column.

Second rotating batch

A bundle of 250 gram flax was placed in the rotating drum. Again the flax didn't tumble during the first day of the experiment. After 24 hours, the flax was soaked with water and the tumbling started. According the Fried-tests, the flax was retted in 5 days. The COD obtained was 852 mg per liter and the pH was 7.0.

Third rotating batch

Water from the second batch was recycled. A new flax bundle of 250 gram was used for the retting process. Within three days, the fried test indicated that the flax was fully retted. The COD level was raised further to 1860 mg per liter and the pH was 7.7.

Fourth rotating batch

Now the flax bundle was increased in mass to 450 gram of flax. Fresh water was used and the flax was retted in 4 days. The pH was 7.99.

Discussion rotating drum results

According the Fried tests, the retting process in the rotating drum is faster than in the lab-scale experiments. This is explained by the fact the mixing energy and shear stress positively affected the retting. No acidification of the process water (means: pH < 5) could be detected.

However, after the retting steps in the rotating drum the processing and fiber quality was poor. First of all, a close up of the retted flax bundle (figure 3-16B) showed some heterogeneous retting pattern. At the tips of the bundle, the retting seems to be further then in the middle of the bundle. Also the fibers, at the tips of the flax bundle, were mixed and this is a draw-back for further processing. After opening the flax bundle (figure 3-16C) it was observed that the core of the flax bundle was less retted.

Processing of the rotating drum flax fibers (numbers two, three and four) resulted finally in flax fibers (First three fiber bundles in figure 3-16D). But the quality was poor (brittle fibers) and also some shives were still present. It is unclear why the Fried test resulted in positive retting values, because a part of the flax bundles were retted poorly. Perhaps the flax stems that were unretted were also very tightly connected in the flax bundle. During sampling, these flax stems could have been deselected and the more loose flax stems were easily to obtain for sampling. As a result, the more retted stems were used in the Fried test.

3.4.4 Conclusions rotating barrel

The rotating barrel results indicate that flax retting can be accelerated by increasing mixing capacity. The temperature and the pH were not limiting. Heterogeneous retting should be avoided for further processing. The rotating drum seems not to be the most efficient process for retting. To overcome heterogeneous retting and introduce mixing capacity to a water retting process, a bubble column can be used.

3.4.5 Flax retting in a bubble column

Experimental setup

The bubble column is constructed in order to determine if aeration and mixing will improve the retting process. A flax bundle of 450 grams is put in the top of the column. The bottom of the column consists of a conical nozzle to disperse the compressed air. Above the nozzle, a sieve plate is constructed with holes in it of 6 mm each in diameter. This plate is used to equally distribute the bubbles over the perpendicular surface in the column. The top of the column contains an overflow head for the produced foam and possible excess water. A photo of the bubble column is given in figure 3-18. Operating conditions: gauge pressure 2 bar, temperature 20 °C, residence time 4 days.

Figure 3-18: The bubble column used with flax retting experiments.

3.4.6 Concluding results flax retting in a bubble column

The air bubbles were equally distributed in the bubble column. The water phase around the flax bundle was mixed by the up-flow of the gas bubbles. Fried tests resulted in 2.5 , indicating a high retting rate of the flax stems. The pH was 8.1. After 4 days, the flax from the bubble column was washed and air dried.

After drying, the flax was further processed. Shives could be removed and flax fibers were obtained. Compared to the rotating drum experiment, it was easier to remove the shives and the color of the fibers was more grey (figure 3-16D). Not all the shives could be removed easily.

Based on the tests with the scale up experiments, the bubble column seems to give the most promising results. The advantage is that the flax is more evenly retted along the whole bundle and in contradiction to the drum retting scale up, the fibers are not entangled, which makes further processing easier.

3.5 Pyrolysis Experiments

3.5.1 Introduction

Wooden shives are a byproduct from processing flax stems to fibers. Around 50% of the flax stems weight are processed to shives. Currently these raw shives are used for bedding in cow, pig and horse stables. But shives can also be processed and used to fabric building-materials like isolation material. Here we investigate the possibility to pyrolyse these shives to oil and other products.

Pyrolysis is anaerobic conversion at high temperature. Mostly woody materials are converted to gasses, oil and ash. The woody chemical compounds, cellulose and hemicellulose (pallesen, 1996) as well as lignin and pectin, (Nair et al., 2013) can be measured in the shives. An example of a pyrolysis reaction equation is given by Tushar et al, 2012 .

$CH_{1.5}O_{0.62} \rightarrow CO + CH_4 + H_2 + Char + Tar\ (CH_nO)_x$ (Tushar et al., 2012)

The stoichiometry of the reaction can be changed by altering the process conditions like temperature, size of the shives, water content and residence time. Char can be used as a fertilizer and as a fuel. Tar is condensed to oil during the pyrolysis process. Pyrolysis oil can be used as fuel for heating, as building blocks for chemicals, and as preservation chemical to extend for example the lifetime of wood. By replacing petroleum based products by pyrolysis based products, CO_2 reduction can be achieved since the flax is a natural product. A combination of the pyrolysis oil and the natural flax fiber could also result in new renewable, sustainable and only flax based materials.

Pyrolysis oil and char can compete with fossil fuels on the energy market. The production cost of pyrolysis oil is estimated to be 75 to 300 Euro per ton oil (Eubia 2015). The heating value of pyrolysis oil is around 16-19 GJ per Tonne. The heating value for char is 28-30 GJ per Tonne, (Bradley et al., 2006). Selling prices for conventional coal is 1.52 Euro per GJ, and for petroleum based oil ranging between 5.53 to 9.08 Euro per GJ, (Bradley et al., 2006). Production costs of pyrolysis oil is 4 to 18 Euro per GJ, (Eubia, 2015). Overall, this means that in some cases production of pyrolysis oil for combustion is economically effective compared to oil or natural gas (€ 7-8 per GJ, July 2015). Costs for pyrolysis oil production in the above mentioned calculations are mainly caused by transport (shipping costs). For flax processing, flax is already transported to factories. This will benefit the pyrolysis oil costs and pyrolysis oil from flax shives will probably be cheaper than the 4 to 18 Euro per GJ.

The last option for biomaterials is combustion (when a biomaterial is useless, burning for energy is always an option). This means that the paragraph above discussed the worst case scenario for flax pyrolysis oil and charcoal (**Table 3.3**). Char and pyrolysis oil have a higher value besides their energy content. They are renewable and can be used in bio-based and renewable products (or processes). Produced pyrolysis oil and charcoal, can be used to generated energy in the total flax processing chain to keep the production process sustainable.

Table 3.3: Energy production from pyrolysis

Mass	Costs	Selling (Energy)
1000 kg (Shives)		
Products (slow pyrolysis, over 100 seconds residence time)		
Gas (20%)	(Flammable gas can be used for heating)	Heat
Pyrolysis – oil (50%)	500 kg (equivalent 9 GJ)	70 Euro
Coal (30%)	300 kg (equivalent 9 GJ)	15 Euro
Total	**10 to 150 Euro**	**85 Euro**

Overall, it seems economical feasible to produce oil and coal from flax shives. It is tested in an experiment to produce flax oil and coal with the lowest pyrolysis production cost possible: almost no shives pre-treatment costs and a simple pyrolysis system are used. It is aimed to produce as much charcoal and oil with low processing costs. Two temperature settings 400°C and 500°C and two nitrogen flow settings were used to affect the yield on oil and coal from shives.

3.5.2 Material and Methods

Flax shives were dried in a 105°C oven for 24 hours to remove possible water content. The average particle size of the flax shives was around 1 cm long. The experiments performed are summarized in table 3.4. The pyrolyse reactor (fig. 3-19) was pre-heated to 255°C. Around 80 g of flax shives were added to the reactor under nitrogen conditions. Subsequently the temperature was raised to 400°C or 500°C. The nitrogen flow was 0.6 or 0.8L per min. Sand was used to improve the heat transfer to the shives. Vapor flows from the reactor through a filter to a condenser where it is cooled using cooling water. Condenser 1 (C1) was 11°C and condenser 2 (C2) was 0°C (ice-water). Uncondensed gases are being vented. Char remained in the reactor.

Table 3.4: Shives pyrolysis experiments.

Experiment number	1	2	3	4	5	6	7
T_{end} (°C)	400	400	500	500	400	400	500
T_{start} (°C)	253	259	258	260	260	265	261
ϕN_2 (L/min)	0.6	0.8	0.8	0.6	0.6	0.8	0.8
Mass flax (g)	74.1	87.3	76	75.8	76.3	75.2	70.7
Mass sand (g)	520	514	518	505	502	506	508

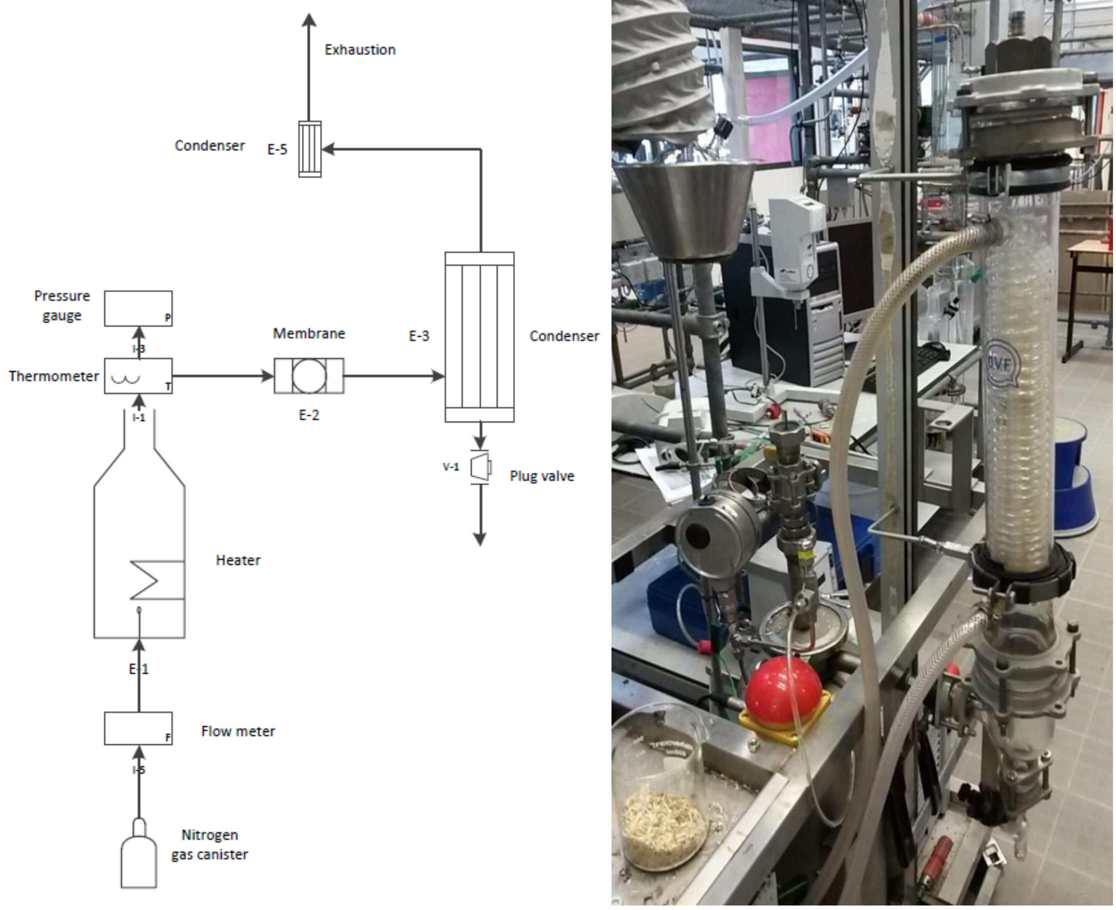


Figure 3-19, Schematic overview and photo of the pyrolysis system used. The pressure valve is to avoid overpressure of 0.5 bar in the reactor.

3.5.3 Results and discussion

Pyrolysis oil (figure 3-21) could be produced in all the seven experiments. The majority of the oil was obtained from the first condenser. The first condenser yielded 10.5 to 18.5 gram pyrolysis oil and the second condenser yielded 1.5 to 3.4 gram pyrolysis oil. The average total oil of both condensers yielded around 20% weight percentage of the shives. This amount is low compared to other pyrolysis experiments at 400°C to 500°C, and around 50% oil would be expected (Eubia website, 2015). Two main reasons for this low yield could be given: (1) the small scale experimental set-up used are sensitive for small disturbances like oxygen interference that might have accessed the system. (2) The condensers were not capable of condensing all the smoke, since a lot of smoke was leaving the system via the exhaustion (fig 1).

Table 3.5. Results of the flax pyrolysis experiments

	1	2	3	4	5	6	7
T_{end} (°C)	400	400	500	500	400	400	500
T_{start} (°C)	253	259	258	260	260	265	261
ϕN_2 (L/min)	0.6	0.8	0.8	0.6	0.6	0.8	0.8
Mass flax (g)	74.1	87.3	76	75.8	76.3	75.2	70.7
Mass sand (g)	520	514	518	505	502	506	508
Mass oil C1 (g)	10.7	12.6	10.5	18.5	14.5	12.5	10.8
Mass oil C2 (g)	1.5	3	3.4	1.5	1.8	2	1.5
Mass char (g)	22.9	25.2	21.9	20.5	33.7	22.2	18.9
pH value oil C1	2.47	2.58	2.53	2.77	2.68	2.66	2.76
Density oil (g/cm^3)	1.00	1.02	1.07	1.04	0.96	1.03	1.03
Heating time (min)	-	5.2	5.5	6.3	4.2	5.0	6.0
Cooling time (min)	-	20	35	32	20	20	35
Reaction time (min)	-	35	25	20	20	35	25

Charcoal (figure 3-21) could also be produced in all the seven experiments. The average total charcoal yielded around 31% weight percentage of the shives. This amount is comparable to other pyrolysis experiments at 400°C to 500°C. (Eubia website, 2015). The charcoal does not leave the pyrolysis reactor, and this makes it easier to obtain comparable yield values of 30% reported in the literature. The higher charcoal yield in experiment 5, (table 3.5 and figure 2) could not be explained.

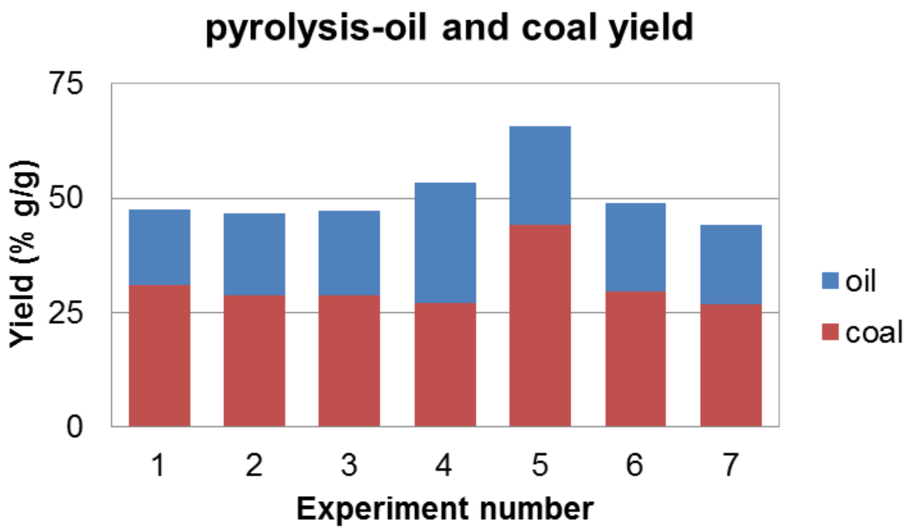


Figure 3-20: graphical result of the coal and oil yield of pyrolysis.

Interestingly, in the membrane (figure 3-19) some black particles could be detected. These particles resembles black crystalline carbon particles (figure 3-21), and could also be seen as an usable product in for example: inks, plastic and paints.

Figure 3-21: Photo of the pyrolysis products: 1. Charcoal 2. Probably carbon crystals 3. Pyrolysis-oil

As last result of the pyrolysis experiments, the pyrolysis oil was combined with produced flax fiber. Pyrolysis can be used as conservator for word, but perhaps also for flax fibers. The flax fiber can be treated with the pyrolysis oil and the fiber absorbs the oil. This means that it is possible to conserve flax fibers with flax pyrolysis oil, (see also figure 3-22).

Figure 3-22: Flax fiber obtained during lab-scale retting processes with pyrolysis oil as conservator.

3.5.4 Conclusion and recommendations pyrolysis

Conclusion

The slow flax shives pyrolysis resulted in oil and charcoal. The minimum pretreatment step of only drying resulted in comparable charcoal yield compared to other pretreatment steps (smaller shives) from experiments performed by other researchers. The oil yield was low compared to the yield reported in other literature studies, this was probably the result of the small scale pyrolysis equipment used in our experiments.

Recommendations

The oil yield can be increased by using other equipment. Probably the condenser capacity or the heat transfer in this system was not capable enough to enable a fast pyrolysis process. For growth to build, the combination of the oil with flax fibers offers opportunities to produce new flax-based products.

3.6 Preliminary process design

For the full scale process, the target is to produce 1000 metric tons per year of yarn for the utilization in composite materials for production of non-critical car parts. This is considered to be the main end material of the mid-stream process. This chapter is a preliminary design for screening purposes. The amount per hour produced, based on full continuous process operation, which will be 8000 h/year, is then 125 kg/h. For the spinning of yarn, only long flax fibers are of interest and the assumption is made that due to processing(mechanical shearing e.g.) also short fibers will be present. These fibers are of no interest for Ten Cate. The assumption is made that approx. half of the total fiber biomass will be short fibers. This results in a total fiber output of 250 kg/h out of the retting process. The short fibers can be used f.i. as backbone material for building purposes like insulation plates, ceiling plates and other building composite materials. This will be an amount of 125 kg/h. Based on experiments half of the total biomass(raw flax plant) consist of chives. The total amount of chives will be then also 250 kg/h. All this results in a total raw flax plant input of 500 kg/h or 4000 metric tons per year.

The controlled water retting process consists of an aerated bubble column. Based on the experiments with a bubble column in the lab at Saxion, this gives an equally distributed retting over the total surface area of the flax plants instead of a rotating drum, therefore the choice will be an aerated bubble column. The aeration will be generated by a compressor which force the air through the column. To estimate the power demand of this compressor, the oxygen transfer rate(OTR) is needed in order to determinate the volumetric flowrate of the compressor. In the lab size bubble column a gauge pressure of 2 bar is used to get sufficient air bubbles for the retting. In fermentation industry bubble columns are common practice and at normal operation the OTR is approx. 19 kg O_2 per m^3 Reactor volume per day(BSDL, 2015) based on a superficial gas velocity of 5 cm/s and an average bubble diameter of 6 mm. For the retting process approx. 10 liter of water is needed per kg of flax plant, therefore the total mass of water needed in the retting columns will be 5000 kg/h or 5 m^3/h. Based on the lab experiments the retention time of the flax is 4 days. In order to get continuous production five reactors of 120 m^3 are needed in batch mode. Four of them are in aeration modus and one will be unloaded for further processing. The density of oxygen in pressurized air of 2 bar gauge at 20 ^oC is 2,45 kg/m^3(www.engineeringtoolbox.com) which gives a total volume of oxygen of 19/2,45 = 7,76 m^3 O_2 per m^3 Reactor volume per day. Since there is 21% vol. of oxygen present in air the total amount of air needed will be 37 m^3 air per m^3 reactor volume per day which gives a volumetric air flow rate of $4 \cdot 120 \cdot 37/24$ = 740 m^3/h. At 2 bar gauge pressure the amount of power consumed by the compressor will be $2 \cdot 10^{5} \cdot 740/3600 = 41$ kW.

After the controlled water retting the retting water needs to be separated from the retted flax, this can be done by a simple separator with a moving grate which allows the water to be collected at the bottom of the apparatus. From the measurements in the lab the amount of water uptake in the retted flax is in the order of magnitude 5 liters per kg retted flax. This gives a retted water outflow from the separation apparatus of 2500 kg/h containing 40 mg COD per kg flax input. Based on the throughput of flax this is 20 kg COD/h available for anaerobic digestion.

For the anaerobic digestion the amount of biogas can be calculated with the following formula(Metcalf & eddy, 2004):

$$C_vH_wO_xN_yS_z + \left(v - \frac{w}{4} + \frac{x}{2} + \frac{3y}{4} + \frac{z}{2}\right)H_2O \rightarrow \left(\frac{v}{2} + \frac{w}{8} + \frac{x}{4} + \frac{3y}{8} + \frac{z}{4}\right)CH_4 + \left(\frac{v}{2} - \frac{w}{8} + \frac{x}{4} + \frac{3y}{8} + \frac{z}{4}\right)CO_2 + yHH_3 + zH_2S$$

The approximation formula of biomass is(BSDL,2015):

$C_1H_{1,8}O_{0,5}N_{0,2}$, based on the stoichiometric reaction formula this gives:

$$C_1H_{1,8}O_{0,5}N_{0,2} + 0{,}95H_2O \rightarrow 0{,}925CH_4 + 0{,}475CO_2 + 0{,}2NH_3$$

Based on the molar fractions the amount of methane can be calculated on COD input of 20 kg COD/h. It assumed that due to the low content of COD (< 50 mg/L) that all the COD can be converted into biogas(Metcalf & Eddy,2002). The ratio CH_4/biomass on kg basis is $0{,}925 \cdot 16/24{,}6 = 0{,}6$. This results in a methane production of 0,6.20 = 12 kg CH_4/h. The lower heating value of methane is 50 MJ/kg(www.engineeringtoolbox.com) which gives a chemical power input for the combined heat and power, CHP of 166,7 kW. As a rule of thumb 40 % percent of the chemical power input is converted into electricity and 40 % is converted in to usable heat, the remaining 20% will be losses(internal friction of moving parts in the engine and radiation losses). This gives an available electric power output of 67 kW and 67 kW of usable heat(exhaust gases mainly).

The retted flax contains 5 kg water per kg flax. The most efficient method is to use a filter press to get rid of the mayor amount of water but the downside of this principle will be that the flax gets severely damaged and cannot be used anymore for spinning yarn. Therefore a band dryer is chosen to get rid of excess water in the retted flax. The process conditions will be around 80 oC of heated air blown through via a fan at atmospheric pressure. The latent heat of the water will be around 3000 kJ/kg. The amount of thermal energy needed for this process step is in the order of magnitude of 2,1 MW. After this drying step the flax contains around 30% of water.

After drying, the next process step will be combing the flax. This will be done by a heckling comb and is a mechanical process. The electrical power input is assumed to be 15 kW. The final process step is Spinning the long fibers into yarn and it is assumed that the electrical power input is around 10 kW.

The chives released by the combing process can be further processed to pyrolysis oil via the pyrolysis process developed by BTG(Biomass Technology Group). This process consists of a Rotating Cone reactor in which the dry shives will be converted in a very short time(2 seconds) into mainly pyrolysis oil(75% mass basis). Syngas(10% mass basis), ash(10% mass basis) and Char coal(5 % mass basis) at a temperature of 500^{o}C at atmospheric pressure in the absence of oxygen. The amount of energy needed for the pyrolysis will be generated via combustion of the syngas and the char coal and is self-sustaining proven technology. Before the chives can enter the rotating cone reactor, they need to be dried further to 10% weight basis moisture content. An extra drying step is needed to reduce the moist content of 30% weight basis in the chives to 10%. This drying process is done in the lab at a temperature of 105^{o}C. the latent heat will be around the magnitude of 4000 kJ/kg. The amount of shives is 250 kg/h, this gives a thermal power demand of 278 kW. The amount of pyrolysis oil will be around 187 kg/hr.

Schematically, the mid-stream process can be visualized in the following way (see figure 3-23). In table 3-6 shows the numerical data of the mass flows of the process.

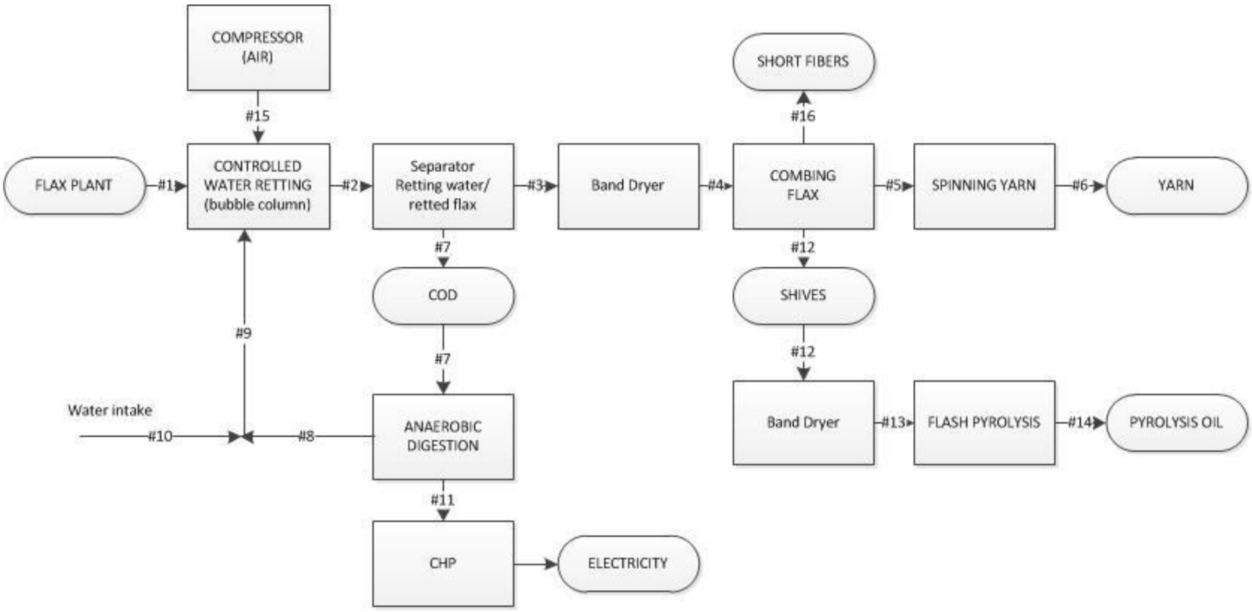


Figure 3-23 schematic of the mid-stream processing of flax plant to yarn.

Table 3-6, mass flows mid-stream process.

#	Description	Amount(kg/hr)
1	Flax plant	500
2	Retted flax and retting water	500(flax) 5000(water total)
3	Retted flax and water uptake	500(flax) 2500(water uptake)
4	Dried retted flax and remaining water	500(flax) 150(remaining water)
5	Long Fibers	125
6	Yarn	125
7	Retting water	2500(water) 20 (COD)
8	Effluent digestion	2500 (water)
9	Water intake bubble columns	5000
10	Water input	2500
11	Biogas	12(CH_4) 7,6(CO_2) 0,4(NH_3)
12	Shives	250(chives) 75(remaining water)
13	Dried shives	250(shives) 25(remaining water)
14	Pyrolysis oil	187
15	Compressed air	1776
16	Short Fibers	125

Conclusion mid-stream preliminary process design.

Based on the calculations on energy demand it appears that the amount of electricity produced from the CHP will be sufficient enough to maintain the compressor, combing and spinning step. The amount of heat needed for drying can be extracted from the pyrolysis process, since these process conditions for drying are much less in energy demand then generated in pyrolysis process self.

3.6 Overall conclusions and recommendations

Conclusions:

This research demonstrates that flax is retted with water at 20°C without the addition of any chemicals. The process time can be shortened from three to two weeks by recycling water. The process was pH-stable and retting sustained.

Upscaling of retting

Upscaling with a rotating drum resulted in a water retting processing time of days. Unfortunately the retting was not equally distributed over the flax bundle. After retting, further processing of the flax fibres was unsuccessfully. Probably the mixing energy caused by the rotating drum resulted in damaged flax stems. These damaged flax stems resulted in shorter retting time. This damage effect was stronger on the surface of the flax bundle used. The flax stems in the core of the flax bundle were more protected.

Upscaling In a bubble column, the flax was successfully homogenous retted. After retting, this sample could easily be further processed resulting in flax fibers.

Pyrolysis of flax shives

Flax shives were successfully converted to pyrolysis oil and charcoal, these products can be used as energy supply to keep the flax process renewable and CO_2 neutral.

Mid-stream preliminary process design

According the midstream calculations, the whole process can be made sustainable and CO_2 neutral. The process from flax plant to flax fiber can be sustainable and self-supporting by using energy from the flax plant itself: biogas from anaerobic digestion and pyrolysis oil from shives.

Recommendations:

- Controlled flax retting is possible at simple process conditions of 20°C and water. However, the retting time is relatively long (two weeks with recycled water) compared with water retting at higher temperatures (6 to 10 days). It is strongly recommended to shorten the retting time or to design a process in which time and retting space is not limiting.

- Active stimulation of the water with the flax stimulates retting processes. This is observed during the experiments in the rotating drum. Retting processes should always result in homogenous retting.
- Retting in a water column showed steady results. Perhaps, the flax water retting system could be integrated in a waste water system to lower process costs.
- Pyrolysis oil can be used to conserve wood. Since pyrolysis oil is renewable, it could be an option to use this oil from flax in other Growth to Build cases.
- The pyrolysis equipment used was a small scale set-up. Yield of pyrolysis-oil could be improved by scaling-up and to use a flash-pyrolysis system (short retention time)
- Pyrolysis resulted in several products, among them is possibly crystalline carbon. It would be beneficial for the overall flax process to research on the possibilities to use this carbon in other products.

3.7 Literature

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