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Separation of Cellulose Fibres from Pulp Suspension by Froth Flotation Fractionation

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Abstract

Flotation of cellulose pulp suspension in paper industry is primarily used for separation of ink particles from cellulose fibres. Entrainment, an unwanted phenomena well described in the field of mineral flotation, also leads to a removal of fibres with the flotation froth. We find that the entrainment phenomena can be used for the separation of long fibres from a fibre pulp suspension, and hence for pulp fractionation. Specifically, we use a 2D bubble column to investigate the influence of (i) bubble size, (ii) wash rate and (iii) stirring on the separation of long fibres from cellulose pulp suspension. Separation of fibres from fibre pulp suspension is tested for mechanical pulp and chemical pulp. We find that size selective recovery yields best result for (i) large bubbles, and (ii) additional washing due to the increase of small particle drainage. However, both strategies lead to a reduction of the total recovery rate. Stirring significantly improved the total recovery and benefited the selective separation. Best results are achieved with small bubbles for chemical pulp. For mechanical pulp, fractionation is more challenging due to lower froth stability, but still fibres with a reduced amount of smaller fraction can be recovered.

1. Introduction

In paper and pulp production, effluent streams containing cellulose fibres, fibre fines, and inorganic fines accrue. Common strategy to reduce waste in paper and pulp production is to recover fibre and fibre-fines and reuse them in paper and/or paper board production [1,2]. However selectively recovered cellulose fibres could also be used as raw material for plastic compounds [3–8]. Classical separation of cellulose fibres and fibre-fines by size are mechanical screening and separation technologies [9]. Pulp is typically fed at concentration of several percent. For those obviously the process energy demand increases with decrease of concentration as more water has to be pumped. Flotation can serve as alternative process for recovering cellulose fibres, especially from thin suspension.

Flotation of cellulose fibres typically occurs as unwanted effect in de-inking of recycled paper. Along with the hydrophobic ink particles, fibres are removed with the froth. In the past decade, researcher could show, that the removal of fibres and fibre-fines which are hydrophilic, occurs rather due to entrainment than due to true flotation [10–18]. Ajersch [11] reported, that gas bubbles do not adhere or form at the fibre surface. Deng et.al. [13,18] performed experiments with surface modified fibres. The amount of fibres removed by flotation did increase for modified fibres with a hydrophobic surface (contact angle of 39°). But even for hydrophobic fibres 67% to 75% had been removed due to entrainment than by true flotation. Untreated cellulose fibres do not adhere to the ascending gas bubbles but are removed by physical entrainment into the flotation froth. A result which was also shown experimental by Turvey [19,20], however without a mechanistic explanation.

Being entrained into the flotation froth, particles can move freely in the suspension filling the void between the bubbles. Also, they are subject to be removed with the draining suspension water. However, removal rate differ and it is found, that the fibre fraction increases in the froth [21–26].

In this study we perform experiments based on literature suggestion to exploit the capability of flotation froth fractionation on the separation of fibres from thin fibre and fibre-fine suspension. Increased knowledge on the topic however will also benefit de-inking flotation where loss of organic material is to be prevented [27], recovery of fibres from effluent streams [28,29], and/or controlled to set certain sludge properties [30]. Additionally,

capabilities of controlling fibre to fibre-fine ratio also allows to produce paper of differing properties and qualities [31,32].

To highlight the importance of process parameters on the result of froth fractionation we first reviewed literature on entrainment of particles in mineral flotation and de-inking flotation. A special focus is given to the change of fibre length distribution in the flotation froth.

1.1. Particle Entrainment in Flotation Froth

A recent review paper by Wang et.al. discusses the mechanism of particle entrainment in mineral flotation, and the impact of process parameters [33]. The entrainment mechanism is characterized by particles being trapped in the froth independent on their chemical properties, i.e., wettability. In literature the following phenomena are used to explain the entrainment mechanism: (i) particle transport in the liquid film around the bubble, (ii) particle transport in the wake of rising bubbles, and (iii) bubble swarm theory. The bubble swarm theory after Smith and Warren [34] is illustrated in Figure 1, Step 1 to Step 2. It is assumed, that particles at the suspension froth interface (Step 1) are pushed in the froth (Step 2). This might be especially important for fibrous material forming coherent networks. For such it is noticed, that small bubbles accumulate beneath the flock leading to a rise of the flock as a whole [11,17,35,36].

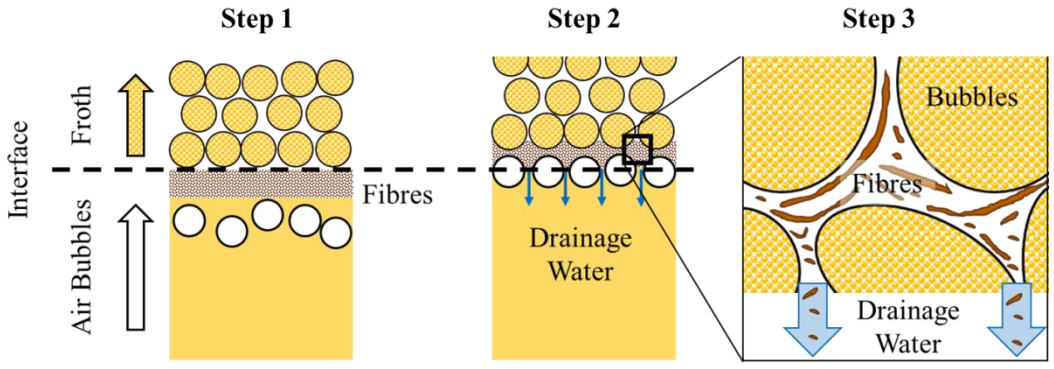

Figure 1: Step 1 to Step 2 illustrate the entrainment of fibres after bubble swarm theory by Smith and Warren [34]**. Fibres are pushed into the froth by ascending bubbles. Step 3 illustrated the reduction of fibre-fines after Eckert et.al.** [25]**. Draining water washed fibre-fines from the froth.**

Due to differences in the density (and the build-up of a hydrostatic pressure gradient), liquid drains from the froth to the suspension through the plateau borders between the bubbles (Figure 1, Step3). Collapse of bubbles results in an increased transport of liquid. Entrained

particles, suspended in the liquid, are removed likewise. Thus, the rate of particle drainage is related to the rate of liquid drainage. However, the intensity of both phenomena can be different [21,37,38]. Factors influencing the recovery of hydrophilic material includes (i) solid concentration in the suspension, (ii) retention time, (iii) froth structure, and (iv) particle size [39]. Important influencing parameters will be explained in detail to understand their importance on the froth fractionation performance:

- The transfer rate of (solely) entrained particles from the suspension into the froth correlates directly with the **particle concentration** in the suspension. A higher particle concentration in the suspension results in a higher particle concentration in the froth [11].
- **Retention time** is the ratio of **froth height** to superficial **gas velocity**. Increasing the froth height results in a larger drainage time, consequently the froth will be drier, and the concentration of entrained particles decreases. For an increase in the superficial gas velocity, which describes an increase in gas flow rate, the carryover of liquid into the froth is larger. Consequently more particles are entrained in the froth. Summing up, an increase in retention time yields a reduction of entrained particles in the froth [16,21]. The limiting factor is the water content of the froth: for a froth being too dry, particle transport stops and entrained particles are permanently captured in the froth. Additional top spraying of liquid prevents the froth from drying out, and increases the drainage rate [21,40–42]. The result is the desired reduction of entrained (small) particles from the froth.
- The **froth structure** is of most importance to the froth stability and hence to the capability of froth containing solids and to the drainage. However the froth structure is complicated to describe and depends on several variables including the water content, aeration rate, bubble size, type of frother (which affect the thickness of the lamella), and the length of the plateau borders. In general, a froth of low coalescence rate consisting of small bubbles promotes high recovery of entrained particles [13].
- Differences in the effect of **particle size** are found for mineral flotation and flotation of cellulose fibres. In mineral flotation, recovery of solids by entrainment reduces with the size of the solids [37,38]. It is argued, that larger particles settle faster towards the suspension in the liquid between the plateau borders. In flotation of cellulose fibres however, an increase of fibre length with the froth height and drainage time is found [21–

26]. Those findings are the basis to use flotation froth for length based fractionation of fibres from the suspension. Results of previous studies will be discussed following.

1.2. Fractionation of Cellulose Fibre Fines in Flotation Froth

Eckert et.al. [24,25] used lab-scale copy of a conventional flotation cell to fractionate fibre pulp suspension. Their findings of the influence of operational parameters on the fractionation performance are summarized in Table 1. They noticed a strong correlation of the average length of recovered fibres on the bubble size. However in their study, they could not control the bubble size and noted the bubble size resulting from changing the other process parameters. Zhu and Tan [21–23] looked closer on the dynamics in the fibre laden froth. They found for steady froth, that fibre length evolved with froth height and drainage time. Time evolution of length-based fibre length reached a plateau level. They concluded that a minimum of water flux is needed for fibres to be washed from the froth. For a dry froth, fibres remain permanently trapped. Results show, that mechanical pulped fibres (TMP process) needed nearly twice the time of chemical pulped fibres (Kraft process) to reach the plateau level. Both cases then showed a decrease in length-based fibre length population for fibres smaller 0.8 mm. The fibre consistency in the froth increased, due to drainage of water whilst fibres remained trapped in the froth. Figure 1, Step 3 sketches the increase of fibre consistency and increase in fibre length. With the draining water, small fibres and fibre-fines are removed whilst the long fibres remain permanently trapped in the froth.

Table 1: Impact of flotation operation parameters on the fibre recovery and fibre/fines fractionation capabilities [24]**. Qualitative change upon an increase of the operation parameter.**

Increase in Operating Parameter	Total recovery	Fibre/Fines fractionation
Impeller speed	No significant change	No significant change
Surfactant concentration	Increased	Decreased
Feed fibre concentration	Increased	Decreased
Feed flow rate	Decreased slightly	Decreased
Air flow rate	Increased	Decreased

Bubble size	Decreased	Increased

1.3. Objective of this Paper

Literature describes the mechanism of particle and liquid transport in the flotation froth, and suggests the ability of froth flotation to recover fibre selectively by their length. Important parameters had been reviewed and experiments are planned accordingly. The goals of the experimental study are:

- ✓ to show the importance of bubble size on the fractionation performance by changing the bubble size as only operational parameter,
- ✓ to highlight differences in the fractionation performance for different types of pulp,
- ✓ to investigate changes in the flotation process by adding a stirrer and top-spray,
- ✓ to finally highlight the capability of froth flotation fractionation to separate cellulose fibres selectively by their size.

To achieve those goals, we modified a quasi-2D bubble column to serve as flotation cell. Bubbles are generated by two different bottom sparger (aerator) yielding different bubble sizes. To check the influence of the internal flow structure in the flotation cell, baffles had been added later which change the profile.

2. Materials

2.1. Flotation Cell and Aeration System

The flotation cell is based on a quasi-2D bubble column presented by Becker et.al. [43], and has the inner dimension of 900 x 250 x 80 mm. Two spargers, a needle sparger (NS) system and a frit sparger (FS) system had been designed for bubble generation. Bubble sizes were measured to 3.3 mm to 3.5 mm for NS and 0.47 mm to 1.11 mm for FS. The bubble size was determined at the sparger in a clear liquid. Unfortunately, it was not possible to perform image analysis of bubbles near the froth layer, since suspended fibres disturbed our optical technique. However, it is safe to assume that the bubble size does not change due to changes in the hydrostatic pressure, and also coalescence events should by extremely rare in our bubble column.

In the second set of experiments we modified the flotation cell to function as air loop flotation cell [44]. Baffle elements had been included promoting circulation of pulp suspension in the cell similar to an air-lift reactor. Furthermore, stirrer and washing utilities were used, specifically: (i) a 4 blade radial stirrer with diameter of 50 mm (Lactan, Austria), (ii) a one-component spray nozzle (Danfoss A30-256.10, Austria) yielding a wash rate of 1 L/h and, (iii) a pressurized hand sprayer ("Gloria Drucksprüher" Hobby 100, Germany) yielding a wash rate of 5.5 L/h. Detail description of the equipment including a sketch of the flotation cell can be found in the Appendix.

2.2. Pulp and Frother Material

The chemical pulp (CP) used was a mixture of unrefined 85% spruce and 15% beech, provided by Sappi Gratkorn (Austria). Mechanical pulp (MP) was 100% spruce, provided by Norske Bruck (Austria). Figure 2A presents the volume based fibre length distribution q_3. Due to differences in production, MP contained larger amount of fines, and no fibres larger 4 mm. CP contained smaller amount of fines and also consisted of relative long fibres up to a length of 5 mm to 6 mm. Also the surface of the fibre differed. MP fibres were more fibrillated (Figure 2B). CP fibres were less fibrillated, the surface was smoother (Figure 2C).

The frother solution was prepared separately and was added to the pulp. Following the work of Eckert [24], we used tall oil (Zellstoff Pöls, Austria) which is similar to pine oil as frother. Tall oil was saponified with sodium hydroxide at a mass ratio of 2:1. The required frother concentration was determined in pre-experiments. Frother was added to the pulp aerated by a NS until a stable froth was formed. In the flotation experiments, frother was added in excess. Based on the mass of dry fibre 0.7 g tall oil per g mechanical pulped fibres, and 0.45 g tall oil per g chemical pulped fibres is used. The optimization of the frother concentration was no topic of this work, however, would be an interesting topic to reduce the amount of chemical used.

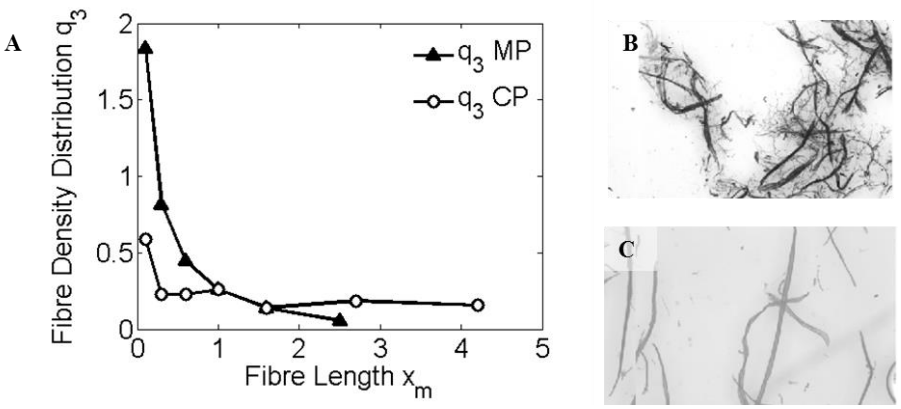

Figure 2: A: Fibre length distribution of the used mechanical pulp (MP) and chemical pulp (CP). B: Microscope image of MP fibres. Fibre show a high degree of fibrilation and interaction. C: less fibrilated CP fibres.

The pulp suspension was prepared according to standard EN ISO 5263-1 and EN ISO 5263-2. After addition of the frother, the pulp suspension was diluted to a concentration of 0.10%.

3. Experimental Procedure and Evaluation

3.1. Flotation Procedure

Air flow had to be assured before filling the pulp suspension into the flotation cell to prevent the sparger from blocking. The experiments were performed for ca. 20 min to 30 min, depending on the froth stability and the resulting amount of product. Being a batch experiment, the suspension height decreases during the experiments by ca. 30 to 50 mm. This results in an increase of the froth height. Thus, the froth retention time was not constant over the experiment.

Detailed experimental settings are listed in Table 2. Experiment MPNS-4 (involving mechanical pulp and a needle sparger) could not be evaluated since the froth was not stable enough to produce any product.

Repeatability of the experiment was assured by repeating experiment MPFS-4 and CPFS-2 on two different days. We compared the flotation rate and the fines content of the flotation product. Results did not differ significantly.

Table 2: List of experiments and their settings. MP and CP denotes mechanical pulp and chemical pulp respectively. FS and NS donates the frit sparger and needle sparger system. Froth height denotes the initial froth height after filling of the flotation cell. The horizontal line devides the first set of experiments from the second set of experiments.

Exp.No.	Pulp	Sparger	Utility	Air Flux [L/min]	Time [min]	Froth height [mm]
MPFS-0	MP	FS	-	4	30	10
MPFS-4	MP	FS	-	4	30	107
MPNS-4	MP	NS	-	4	30	110
CPFS-2	CP	FS	-	2	17	124
CPNS-2	CP	NS	-	2	30	122
CPFS-4	CP	FS	-	4	20	30

CPFS-4-S	CP	FS	stirrer	4	20	30
CPFS-4-W	CP	FS	spray nozzle	4	20	30

3.2. Measurement Techniques

Samples were taken from the initial pulp suspension and from the resulting froth. The flotation rate, froth consistency, and amount of fines were determined gravimetrically. The flotation rate is the total mass of recovered froth, including water and fibres divided by the time of flotation. For the consistency, fibres had been removed from the suspension by filtration, and then oven dried at 110°C before weighting. Froth consistency was expressed relative to pulp suspension consistency. Thus the reported results state the increase in consistency. The amount of fines was determined according to SCAN-CM 66:05 from the Scandinavian Pulp, Paper and Board Testing Committee using a Britt Jar dynamic drainage device [45]. Fines are defined as the fraction passing a sieve of 76 µm mesh size. The reduction in fines content in the froth compared to the fines content in the pulp was reported.

The volume-based fibre length distribution q_3, and the volume weighted average fibre length L_3 were measured with a L&W Fiber Tester + (Lorentzen+Wettre, Sweden) according to ISO 16065-2:2014 standard. The classes for MP and CP were defined as [0.0, 0.2, 0.4, 0.8, 1.2, 2.0, 3.0] mm, and [0.0, 0.2, 0.4, 0.8, 1.2, 2.0, 3.4, 5.0] mm respectively. The froth fibre distribution $q_{case,froth}$ was compared to the feed fibre distribution $q_{case,feed}$ using a representation similar as the one proposed by Zhu and Tan [22]. Specifically, the so-called "feed deviation" (FD) was reported in this previous work. FD-q_1 (Eqn. 1) states the deviation based on the length-based fibre distribution, and FD (Eqn. 2) states the deviation based on the volume-based fibre distribution. Whilst we use FD to compare our experiments, we use FD-q_1 to compare our results to those of Zhu and Tan [22].

$$FD-q_1\left(x_m\right)=\frac{q_{1,case,froth}}{q_{1,case,feed}}-1 \qquad \textbf{Eqn. 1}$$

$$FD\left(x_m\right)=\frac{q_{3,case,froth}}{q_{3,case,feed}}-1 \qquad \textbf{Eqn. 2}$$

Similarly, the deviation of flotation results was compared to the standard case (CPFS-4), and reported as the base case deviation (BCD, Eqn. 3). Furthermore, the critical fibre length *cs* was calculated. This length represents the fibre length below which a reduction of the particle content is observed. Thus, *cs* can be viewed as a typical cut size for the fractionation.

$$BCD(x_m) = \frac{q_{3,case,\,froth}}{q_{3,case,\,feed}} - \frac{q_{3,base,\,froth}}{q_{3,base,\,feed}} \qquad \textbf{Eqn. 3}$$

The separation efficiency g (Eqn. 4) is a measure to compare the recovery of fibres. It states the total mass of solid material recovered in the froth compared to the initial mass of pulp suspension. It is defined as:

$$g = \frac{m_{froth}}{m_{pulp}}. \qquad \textbf{Eqn. 4}$$

4. Results and Discussion

4.1. Effect of Froth Height, Pulp Type, and Bubble Size

Flotation rate, froth consistency and fines content of the froth compared to the initial pulp suspension were recorded and results are listed in Table 3. Results highlight the importance of a controlled bubble size in flotation experiments. Table 3 and Table 4 summarize operational results and key changes in the pulp composition (fibre length). Figure 3, compares the relative deviation in fibre length FD for each type of pulp and sparger system.

Table 3: Flotation rate, froth consistency and fines content in dependence on the froth height, bubble size, and pulp type. Froth consistency and fines content are reported as relative values comparing froth to pulp suspension.

Exp.No.	Flotation Rate [g/min]	Relative Froth Consistency	Relative Fines Content	Separation Efficiency *g*
MPFS-0	63.36	1.10	1.00	0.210
MPFS-4	27.89	2.01	0.81	0.167
MPNS-4		No stable froth		
CPFS-2	76.37	3.16	0.25	0.303
CPNS-2	0.39	51.59	-	0.061

Froth height

MPFS-0 compared to MPFS-4 shows the importance of the froth height to allow a separation of fines. Even the consistency increased slightly, the amount of fines remained on the same level and the ratio is 1 (MPFS-0). Increased froth height yielded a fibre consistency increased by factor of 2 (MPFS-4). Liquid preferred drained from the froth leading to thickening of the pulp suspension. Furthermore, in experiment MPFS-4 the fines content was reduced by 19%, and consequently the volume averaged fibre length $L_{3,MP}$ increased by 35%, from 0.844 mm to 1.143 mm (see Table 4).

Table 4: Cut size *cs* and volume-averaged fibre length for the first set of experiments. An increase in fibre length reflects a reduction of fines content.

Exp.No.	*cs* [mm]	$L_{3,feed}$ [mm]	$L_{3,froth}$ [mm]
MPFS-4	0.57	0.844	1.143
CPFS-2	1.48	2.129	2.544
CPNS-2	1.72	2.129	2.815

Type of Pulp

Results of the flotation experiments showed a clear difference between MP and CP. Flotation of MP was not possible with large bubbles, even when using a higher amount of frother. Comparing results involving froth with small bubbles, i.e., MPFS-4 and CPFS-2, a higher reduction of fines was found for CP (Figure 3). Fines content could be reduced by 35% for MPFS-4 and by 75% for CPFS-2. For CPFS-2, amount of fibres smaller than 1.48 mm decreased whilst for MPFS-4 only a decrease for fibres smaller 0.57 mm was found. This clearly states a difference in the behavior of the fibres in the froth.

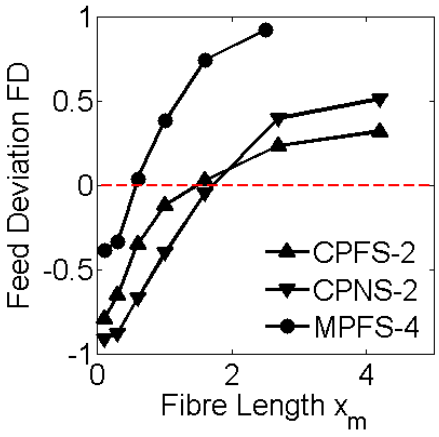

Figure 3: Relative feed deviation for the first set of experiments.

At this point, we can only hypothesize, that fibre and fibre-fines fibrils interact with bubbles and hence stretch over a larger area. Thus, froth stability is reduced in case of MP, and fines movement in the plateau border, important for the drainage of particles, is hindered. Thus, a pronounced effect of bubbles size on the separation of long fibres can only be shown for CP.

Bubble Size

Comparing reported values in Table 3 and Table 4 we find that, (i) a larger amount of CP fibres (g = 0.303) could be recovered for small bubbles of ca. 1 mm (CPFS-2), and (ii) that for larger bubbles of ca. 3 mm (CPNS-2) a better removal of fines from the froth was achieved (increase of fibre length L_3 by ca. 30%). Figure 3 shows that for CPNS-2 only 5% of initial fines and fibres < 0.1 mm are found in the froth. However for both bubble sizes, the intersection with the 0 deviation line, and thus the cut size cs_3 based on the volume based fibre length distribution, was similar (CPFS-2: cs_3 = 2.544 mm, CPNS-2: cs_3 = 2.815 mm). Above, the deviation flattens compared to the steep increase of FD for smaller fibres. From this data we conclude that there is a threshold in fibre length for which the removal rate (due to drainage) decreases rapidly with the fibre length.

Comparison to Literature

In Figure 4 we compare the deviation of the length based fibre length distribution of our findings to the data reported by Zhu and Tan [22]. They performed flotation experiment of MP (TMP) and CP (Kraft) in a column flotation with bubbles of size 1 mm. However as a difference to our experiment, the reported data are time dependent results for steady froth. This means that the froth was not growing and no new feed was introduced into the froth. Reported change in distribution for CP (Kraft) and for MP (TMP) is after 15 s and 29 s respectively. This means that MP needed longer drainage time to yield similar results. Literature data and results for CPFS-2, which was performed with bubbles around the same size show similar trend of the results. The cut size cs_1 based on fibre length based fibre length distribution is similar and around 0.8 mm. cs_1 for CPNS-2 is higher. Interesting is, that cs_1 is similar despite the differences including the type of frother and the flotation cell. This strengthens the conclusion that the bubble size has a comparable strong impact on the fibre mobility in the froth and hence the length based fractionation. The results for MP (MPFS-4 compared to TMP) differ strongly. As stated above, in the experiments by Zhu and Tan, a longer drainage time was given to MP compared to CP. This is not the case in our experiments where the drainage height was given by the experimental setting. This highlights the differences in fines mobility for MP and CP. Better fractionation of MP in our experiment could be achieved by (i) increasing the froth height, or (ii) by letting the froth rest afterwards.

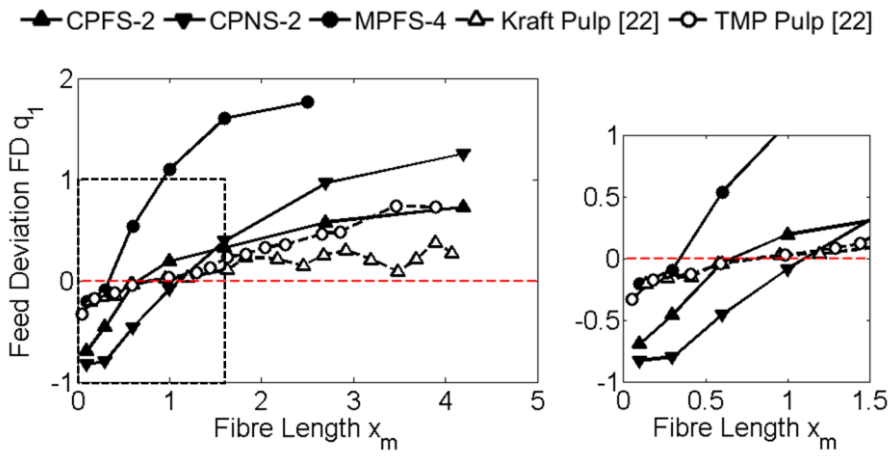

Figure 4: Comparison of the feed deviation for fibre length based fibre length distribution FD-q_1. Our results (filled markings) are compared to literature results (open markings) by Zhu and Tan [22]**.**

Bubble and Particle Interaction

The fractionation process of entrained fibres and fines differs from conventional separation by flotation process as (i) the fractionation takes place in the froth, and (ii) particle do not interact with the bubbles [11,23,24]. Thus, phenomena interesting for true flotation, i.e., the interaction of particles with the bubble's boundary layer are not relevant and not looked at in this study.

4.2. Effect of Stirring and Washing on the Separation of Fibres from Pulp Suspension

After successful separation of fibres based on literature on froth flotation, we aimed to improve the process. We modified the flotation cell (i) adding internals for controlled bubble rise, and applied (ii) stirring at the vicinity of the suspension forth interface, and (iii) washing of the froth. Flotation experiments had been performed for 20 min at an air flux of 4 L/min using frit sparger (FS). The resulting froth fibre distribution was compared to the feed fibre distribution (FD) and to a defined standard experiment (BCD). First the performance of the air loop flotation cell (CPFS-4) was compared to the standard flotation cell (CPFS-2) used in the first set of experiments (Figure 5). The impact of utilities was then compared to the basic air loop flotation cell CPFS-4 (Figure 6 and Figure 7). Results are presented in Table 5 and discussed below.

Table 5: Results table for second set of experiments.

Exp.No.	Utility	Separation Efficiency g	cs [mm]	$L_{3,feed}$ [mm]	$L_{3,froth}$ [mm]
CPFS-4	-	0.161	1.381	2.200	2.604
CPFS-4-S250	Stirrer, 250 rpm	0.257	1.328	2.124	2.690
CPFS-4-S400	Stirrer, 400 rpm	0.238	1.435	2.158	2.573
CPFS-4-WOP	Spray Nozzle, 1 L/h	0.161	1.374	2.173	2.649
CPFS-4-WG	"Gloria", 5.5 L/h	0.044	1.829	2.073	2.847

Air Loop Flotation Cell – Modification of the Flotation Cell

In the original flotation cell we found strong recirculation zones depleted of air bubbles. To maintain a regulated up-flow of air bubbles and fibre suspension we added baffles to the flotation cell (see the Appendix for details). Similar as in an air-loop reactor, bubbles and fibres suspension will rise in the center and settle on the side promoting a more steady flow. Consequently, only in this (smaller) central region froth growth and recovery of fibres from the suspension can take place. However, since the froth layer is not confined to the center region, the froth flows sideways, and accessible area for drainage increases with respect of the area of fibre transfer into the froth. Compared to previous CP flotation trials with the standard flotation cell, the air flow rate was increased, and froth height was reduced yielding in summary a lower froth retention time. Additionally, collection of froth differs between the settings. See here Figure a in the Appendix. For the first set of experiments froth was collected from the shorter side of the box (sampling position A), and for the second set of experiments froth had been collected from the longer side of the box (sampling position B). Changes had been necessary due to the addition of a stirrer.

Despite these major changes to the flotation settings, the separation of fibres was found to be largely similar. Only a slight increase in fines reduction was found for the air loop flotation cell (Figure 5, right panel). However, the separation efficiency between both settings differs and dropped nearly by a factor of 2 for the modified air loop flotation cell. The result is

attributed to the reduced cross sectional area of bubble rise, and hence a lower amount of suspension was transferred into the froth.

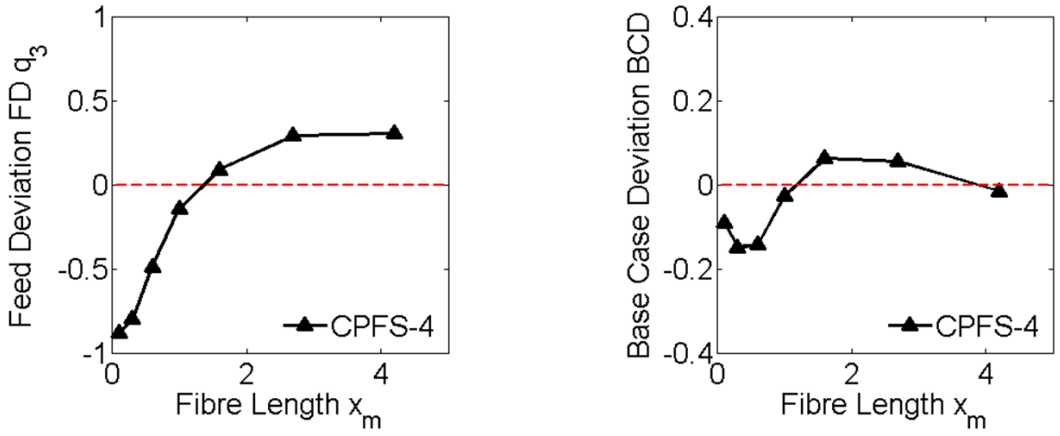

Figure 5: Left panel: Feed deviation q_3 for the standard air loop reactor experiment CPFS-4. Right panel: Corresponding base case deviation BCD, comparing the air loop flotaiton cell to the quasi-2D flotation cell.

Stirring in the Vicinity of the Pulp Suspension Interface

The separation efficiency *g* nearly doubled in the trials with stirring compared to CPFS-4 (see Table 5, and Figure 6). Literature suggests an increase in water recovery, and hence particle transfer from the suspension into the froth due to stirring [13,33,46]. Additionally, for the air lift flotation cell, stirring also leads to a distribution of bubbles, fibres, and fines throughout the cross section, increasing the area available for froth layer formation. This leads to an increase in the solid transfer to the froth. Separation efficiency was comparable to the first set of experiments, CPFS-2.

Increasing the stirrer speed from 250 rpm to 400 rpm did not affect the separation efficiency *g* but had an impact on the fibre length distribution of recovered fibres (Figure 6). For moderate stirrer speed of 250 rpm, only little deviation from the base case was found. Differently, the amount of fines reduced for higher stirrer speed of 400 rpm. Following above argument, we can assume larger transfer of water into the froth. Consequently the drainage increases and more of the fines are washed from the froth. L_3 could be increased by ca. 26%, from 2.124 mm to 2.690 mm. Higher stirrer speed would affect the bubble size and thus altering further flotation parameters.

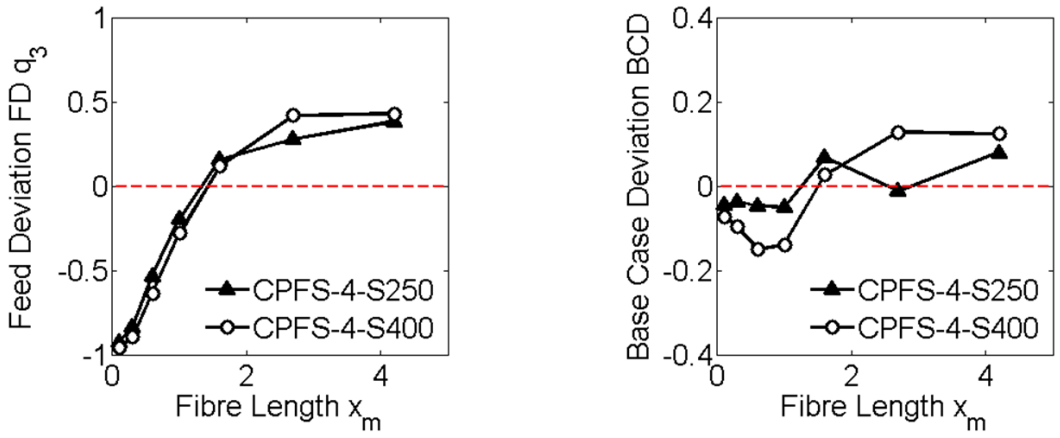

Figure 6: Left panel: Feed deviation q_3 for stirring experiments. Right panel: Corresponding base case deviations (i.e., a comparison with the standard experiment CPFS-4).

Note that stirring in the vincinity of the froth interface is different to stirring in a conventional flotation cell as used by Eckert et.al. in their experiments [24]. In their set-up, the stirrer was installed in the feed pipe at the bottom of the column to produce air bubbles. We employed gentle stirring in the vicinity of the interface to promote mixing and eventually bubble/fibre interaction.

Effect of Washing by Top Spraying

At a rate of recovered water, which was around 3 L/h, no effect was achieved for washing at a spray rate at 1 L/h, whilst a larger reduction of recovered fibres and fines, i.e. separation efficiency g droped by 70%, was found for a spray rate of 5.5 L/h.

Results for lower washing rate go in hand with the findings of Robertson et.al. [40] who stated, that the net flux of water has to be from the froth to the pulp suspension to reduce fibre loss in flotation de-inking. For increased water drainage, plateau borders collapse and solid material is immobilized and consequently not washed from the froth [37,38].

Results for higher washing rate are in accordance with literature [40,42,47] where a reduction of fibre loss from 50% to 96% was reported when applying top spraying to promote washing of fibres from the froth. A larger reduction of fines, which was anticipated, was found and L_3 increased by ca. 37% from 2.073 mm to 2.847 mm (Figure 7). With those settings, separation of fibres was similar to CPNS-2, i.e., a high selectivity of the fractionation, but at a low total

recovery. Also, froth growth reduced compared to CPFS-4. Still, froth was more stable (and froth handling easier) when using a large washing rate, comparted to a froth consisting of large bubbles.

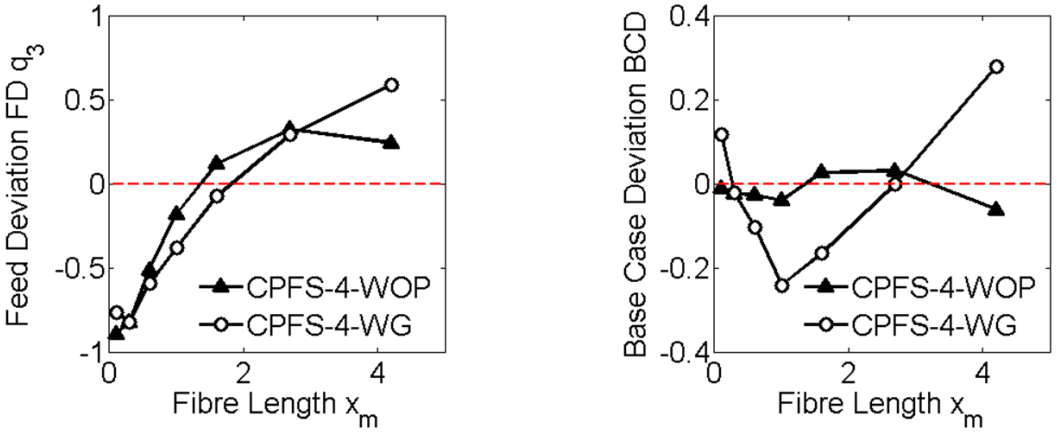

Figure 7: Left panel: Feed deviation q_3 for stirring experiments. Right panel: Corresponding base case deviations (i.e., a comparison with the standard experiment CPFS-4).

4.2.1. Grade Efficiency

The grade efficiency *T* combines the changes in fibre length distribution expressed by FD with the separation efficiency *g*, and hence states the recovery per fibre length class:

$$T\left(x_m\right) = g\,\frac{q_{3,case,\,froth}}{q_{3,case,\,feed}} = g\left(FD+1\right). \qquad \textbf{Eqn. 5}$$

Following we compare the results for the standard air-loop flotation cell, and the best results for the stirred and washed cases to highlight the effect of the modification on the fibre separation (Figure 8). The slope of the grade efficiency curve is a measure for the size selective separation. For comparison we use the ratio of the grade efficiency of the largest fibre length class to the smallest fibre length class: $T\left(x_{\max}\right)/T\left(x_{\min}\right)$. Results are listed below in Table 6.

Table 6: Grade efficiency *T* for the largest and the smallest fibre length class for cases CPFS-4, CPFS-4-S400, and CPFS-4-WG. The ration of both is a measure of the process selectivity.

	CPFS-4	CPFS-4-S400	CPFS-4-WG

$T(x_{max})$	0.211	0.341	0.070
$T(x_{min})$	0.019	0.010	0.010
$T(x_{max})/T(x_{min})$	11.11	34.10	7.00

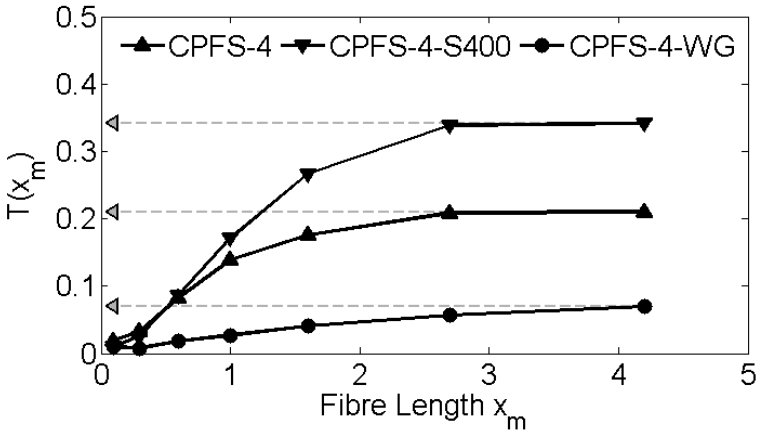

Figure 8: Grade efficiency curve T for cases CPFS-4, CPFS-4-S400, and CPFS-4-WG. The grade efficiency, thus the recovery, of the largest fibre length class is indicated on the y-axis.

None of the cases achieved values larger than 0.4, stating that the recovery per length class in one unit is always below 40%. To improve the separation of long fibres we suggest increasing the flotation time and/or reducing the pulp suspension volume. Recovery of the smaller fibres is low, i.e. < 0.05 for all cases.

For the stirred case at 400 rpm (CPFS-4-S400) ca. 34% of the two largest fibre classes could be recovered. Compared to the standard case this is an increase by ca. 60% due to stirring. For smaller fibre length classes the grade efficiency *T* reduces sharply. Fibres could be recovered from the pulp suspension at high selectivity, i.e. the ratio $T(x_{max})/T(x_{min})$ is 34.1.

Washing the froth led to a depletion of fibres from the froth and only 7% of the largest fibres could be recovered from the suspension. This findings support literature findings suggesting top spraying to prevent the removal of fibres in de-inking flotation [40,41]. The ratio $T(x_{max})/T(x_{min})$ is 7, which is comparably low. However as a difference to the other cases, the slope of the grade efficiency curve is nearly linear. In total only a small amount of fibres < 1 mm was recovered when utilizing top spraying.

5. Conclusion

Literature from de-inking flotation, fibre fractionation by froth flotation, and mineral flotation was reviewed. Experiments were planned following the suggestions from literature, and focused on maximizing the selective separation of large fibres from pulp suspension. Our flotation trials were conducted using two different pulps in a bubble column setup. Our experimental setup allowed us to control (i) the bubble size, (ii) the intensity of turbulence (i.e., stirring), and (iii) the froth washing rate. Differences in the performance due to the internal flow pattern are highlighted, and narrowed down to differences in the bubble rise cross section that clearly affects froth growth.

In summary, our results support findings from literature that the selective separation of long fibres from pulp suspension via flotation is mainly affected by phenomena in the froth. Specifically, we confirm that entrainment causes the collection of pulp fibres. Subsequent drainage of water from the froth back into the pulp suspension leads to a considerable increase of product consistency (i.e., the froth layer). Smaller fibres are removed from the froth by the draining water, and we have measured a reduction of fibres < 76 µm content of -19% for MP and -75% for CP when using appropriate process conditions (i.e., small bubbles). Clearly, the size dependent fractionation takes place in the froth layer, and is effective for non-fibrillated fibers, i.e., CP. The exact mechanism that causes the less effective fractionation behavior of mechanical pulp is unclear. A possible explanation is that the (strongly fibrillated) mechanical pulped fibres destabilize the froth due to an increased bubble bursting rate. We speculate that this phenomenon leads to a rapid collapse of the froth layer, such that the washing effect (of draining water) is not observed.

Finally, we have identified the optimal settings for the successfully removal of small fibres from the flotation froth under laboratory conditions. Those are (i) a long drainage time, and (ii) comparably big bubbles in the froth. However, the bubble size is limited by froth stability. Artificially increasing the drainage rate by top spraying of wash water led to a significant reduction of separation efficiency and to a relative increase of the longer fibre fraction. Thus, washing has a similar effect as the generation of a froth consisting of large bubbles.

Stirring had a key effect on the separation efficiency. A uniform distribution of bubbles, and fibres lead to an increase in transfer rate from the suspension into the froth. Consequently

water drainage rate increases benefiting the selective separation of long fibres from the pulp suspension at considerable high separation efficiency. 34% of the largest fibres, those have a length of 3.4 mm to 5 mm, could be recovered in the laboratory experiment.

In summary, a size selective separation of long fibres, a valuable material for many industries, is possible by froth flotation involving gentle stirring. The success of a possible industrial application will depend on finding a tradeoff between high flotation rates (at the cost of lower separation efficiency) and a clean product (i.e., a low content of small particles in the product). Furthermore, the efficient separation of added chemicals (in our case soap from tall oil) will affect the future exploitation of flotation as a fractionation technology. This is due to the need of a well-controlled froth layer and the required intense washing that necessitate the addition of chemicals.

6. Outlook and Application

The applicability of recovered fibres from suspension naturally depends on the coverage of fibres with frother. Using tall oil, the hydrophobicity of the fibres might decrease. Such fibres are known to be of use in construction materials [48], and might be used in biocomposites [6,8].

Future studies should include the type of frother and frother concentration. Depending on the desired product, optima can be found for already coated fibres, or clean (i.e., washed) fibres. Additionally, the influence of inorganic fillers on the fractionation performance should be looked at in future studies. This is since many process streams in the pulp and paper production contain inorganic material, which is often undesired.

Changing the column set-up to a stack system would allow a fast change of column height. Consequently, the froth height can be easily adjusted, allowing the study of froth evolution, i.e., wetness or solids concentration in the froth, as a function of the froth height.

Scaling the process for industrial purposes, the cross section of column needs to be increased, and consequently the effect on the overall hydrodynamics in the column needs to be investigated. Thus, studies using a pilot scale process might be advisable to ensure a proper scale up to a full industrial scale.

Naturally, our findings can also benefit flotation operations where the loss of fibres shall be prevented, i.e. deinking flotation or the removal of resins from effluent streams [29,49]. Increased froth height, a homogenous bubble distribution, along with spray washing, and increased bubble size, will reduce the loss of organic material. However, bubble size will likewise influence the adherence of smaller hydrophobic particles in the suspension [50].

7. Acknowledgement

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9. Appendix – Flotation Cell

The flotation (Figure a) cell had the dimensions of 900 x 250 x 80 mm and is a 1:2 model of a bubble column used by Becker et.al. [43]. Two spargers were mounted at the bottom for aeration. Froth was removed at a height of 610 mm for the first set of experiments, and at a height of 805 mm for the second set of experiments. Consequently, the maximum filling of the bubble column was 12 L and 14.7 L, respectively.

For the second set of experiments internals were mounted dividing the flotation cell in riser and downer zone similar to an air lift reactor [44]. Fibres rise with the bubble swarm in the center and settle in the downer zone. Furthermore, a 4 blade radial stirrer with diameter of 50 mm (Lactan, Austria) promoting fluid motion beneath the fluid/froth interface and spray nozzles introducing a flow of wash water were added. The stirrer, was mounted in the center of the flotation cell 40 mm above the internals which left a clearance to the suspension/froth interface of 80 mm. Spray nozzles used are a one-component spray nozzle (Danfoss A30-256.10, Austria) yielding a wash rate of 1 L/h, and the pressurized hand sprayer ("Gloria Drucksprüher" Hobby 100, Germany) yielding a wash rate of 5.5 L/h.

A needle sparger (NS) and a frit sparger (FS) system were used for aeration yielding bubbles of different size. The NS system consisted of 18 high precision tubes (Injecta, Germany) per sparger with an inner tube diameter (Di) of 0.8 mm and an outer tube diameter (Da) of 1.6 mm. The bubble size was measured to be 3.57 mm and 3.32 mm for air flow rate of 1 L/min and 0.5 L/min per sparger. The FS system consisted of a porous disc (Type P4, Duran Group, Germany) with median pore size of 10 µm to 16 µm. The diameter of the porous discs was 26 mm, resulting in a free surface of the porous disc of $0.53 \cdot 10^{-3}$ m². The bubble size was measured to be 1.11 mm and 0.47 mm for air flow rates of 2 L/min and 0.5 L/min per sparger, respectively. Evaluation of the bubbles size for the porous disc sparger was performed by Sopat (Germany) according to the procedure described by Maaß et.al. [51]. Note that the bubbles size was measured with the addition of frother, however in the absence of fibres. Unfortunately, it was impossible to include fibres during the bubble size measurements, since they would have prevented optical access.

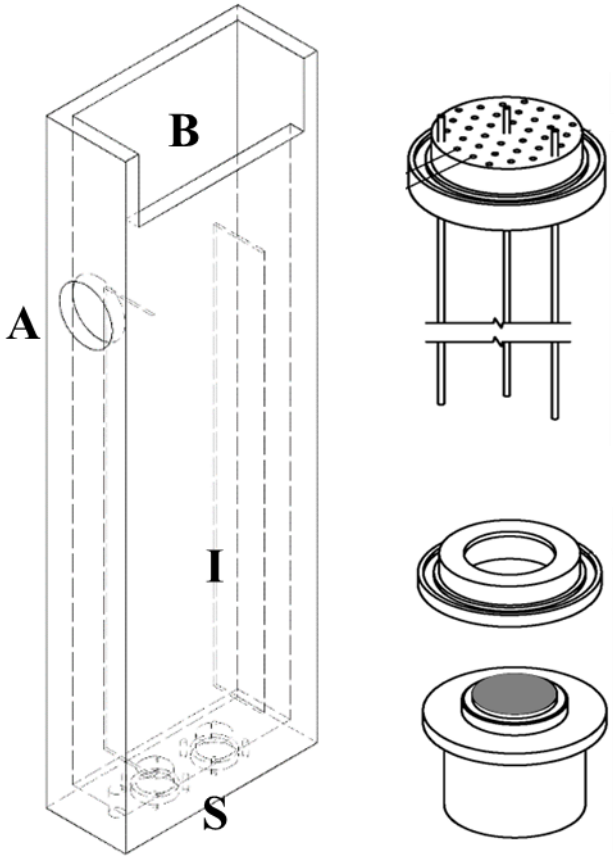

Figure a: Sketch of the 2D flotation cell and sparger system. A and B denote the froth collection point for the first and second set of experiments respectively. I denotes installed internals. S marks the mounting of the sparger. Needle sparger (NS) system and frit sparger (FS) system are presented next to the flotation cell. For the FS, the frit is placed between to PTFE rings.