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Gas Dispersion in the Oxygen Delignification Process

JARI KÄYHKÖ, HEIKKI MUTIKAINEN, KARI PELTONEN, RIKU KOPRA AND MARKUS HONKANEN

ABSTRACT

There has been very little knowledge about the state of gas dispersion in the oxygen delignification process even though this has a major impact on the performance of the reactor. This paper presents a new continuous in-line method for measuring oxygen bubble size distribution in the reactor, as well as results from studies conducted in softwood and hardwood lines. This new measurement worked well and new information about oxygen bubble size, as well as how different reactor conditions affected the distribution, was obtained. For example:

- In the softwood line, the mean volume weighted bubble size was about 0.1 mm, whereas in the hardwood line, this size was almost ten times higher. For both lines, there was considerable variation in the measured bubble size over the long term.
- For both lines, an increase in mixer rotation speed caused a discernible decrease in the bubble size, and an increase in oxygen charge caused a discernible increase in the bubble size.
- In the softwood line, no coalescence of the bubbles in the reactor was observed, but in the hardwood line, some coalescence of the larger bubbles occurred.
- In the test conducted in the hardwood line, the usage of brownstock washer defoamer caused a discernible increase in oxygen bubble size.
- In the hardwood line, reactor pressure had a noticeable effect on the amount of delignification, which indicated that that improving mass transfer of oxygen e.g. by decreasing the oxygen bubble size in this case should also have an increasing effect on the delignification.

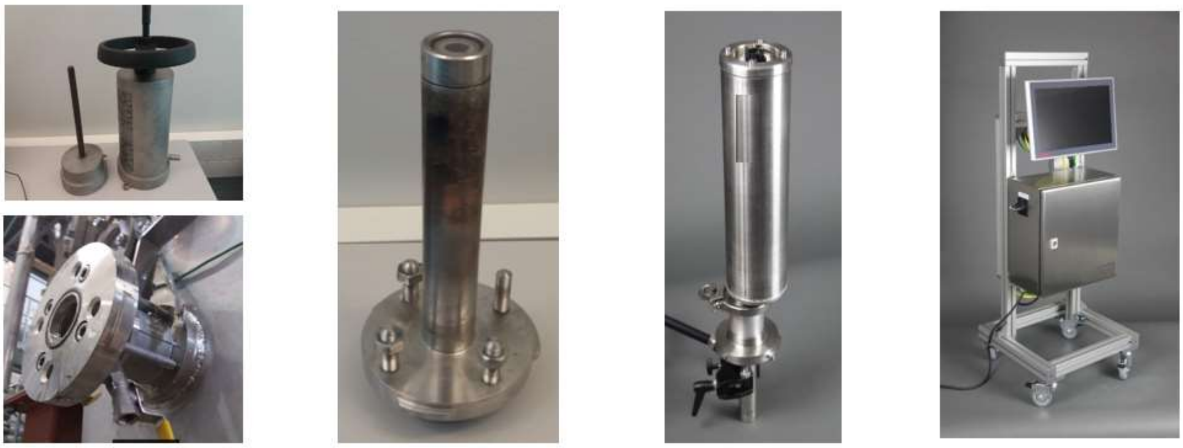
INTRODUCTION

Oxygen delignification is an essential part of the pulp production process. Oxygen delignification occurs at high temperature with the aid of alkali and dissolved oxygen. The temperature and amount of hydroxide anions are known and easily controlled parameters [11]. The amount of the dissolved oxygen is less defined since it depends very much on the mass transfer rate of the oxygen from the gas phase to the fiber, which is determined by the amount of oxygen applied and its bubble size in the reactor. Due to the weak solubility of oxygen, there has to be proper and complete gas dispersion through the reactor to keep the amount of dissolved oxygen at a high level. This is why effective mixing of oxygen gas is an important factor for the proper functioning of the oxygen delignification process. It has also been shown that industrial oxygen delignification reactors on average work at 80% of that observed in a laboratory reactor [1,11]; it has been assumed that the reason for this observation is the lower oxygen mass transfer with the industrial-scale reactors when compared to laboratory reactors [2].

Up to this point, there has been little to no knowledge about the state of oxygen gas dispersion in the reactor, even though this factor can have a major impact on the operational performance of the oxygen delignification reactor. In our earlier papers [3-10], a new image-based method to characterize gas dispersion in the oxygen delignification reactor has been presented. These studies have characterized the state of and the effect of different factors on the dispersed bubble size in the fiber line. This article will present a summary of the earlier studies and conclusions related to the measurement, state, and role of gas dispersion in an industrial oxygen delignification reactor.

MATERIALS AND METHODS

On-line bubble size measurement was conducted using a continuous Pixact Bubble Monitoring (PBM) system (Figures 1 and 2). The measurement is based on imaging of the pulp flow and detecting the bubbles using a machine imaging system (Figure 3). This measurement system has been described in detail in an earlier publication [7]. Gas dispersion measurements in the oxygen reactor were conducted on two fiber lines at a mill in Finland. One measurement period was performed with a fiber line producing hardwood pulp (birch), whereas the other was performed with a fiber line producing softwood pulp.


K-patents installation valve and installation equipments

K-patents installation tube

Imaging probe

PC with on-site control and monitoring

Figure 1: Essential parts of the bubble monitoring system [2].

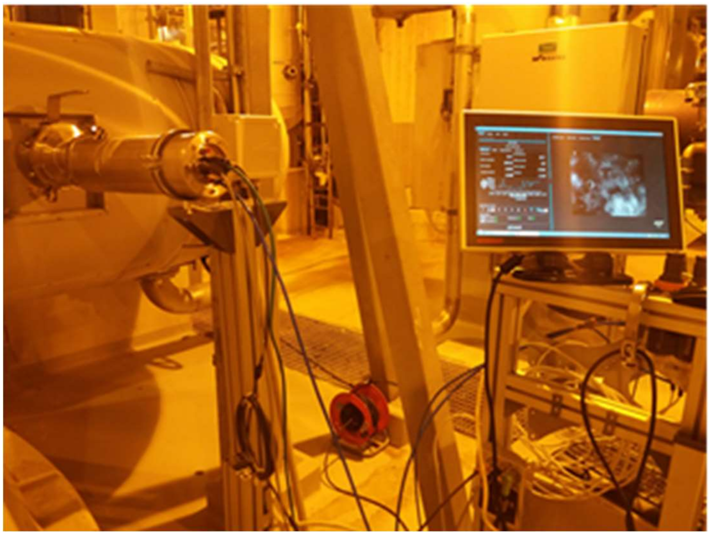
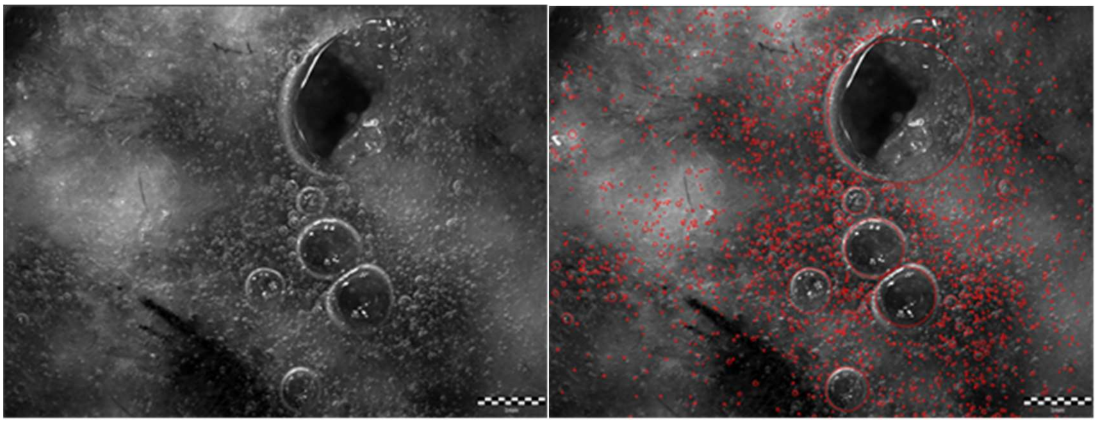


Figure 2: Bubble size measurement system installed in the process that includes on-site control and monitoring capabilities [3].



a) b)

Figure 3: a) Original image of bubble flow in the hardwood line after oxygen mixer; and b) detected bubbles by the machine imaging system are outlined in red [7].

RESULTS AND DISCUSSION

Oxygen bubble size distribution in the process

Figure 4 shows the volume weighte mean bubble diameter in the softwood line and Figure 5 shows the mean bubble diameter in the the hardwood line. The volume weighte mean bubble diameter is calculated as;

$dv = \Sigma (di*Vi)/ \Sigma (Vi)$, i goes from 1 to N Eq. 1

Where;
di; diameter of the individual bubble
Vi; volume of the individual bubble
N; number of bubbles

In these figures it can be seen that the mean bubble size in the hardwood line is about five times higher than that of the softwood line. The actual difference is much higher since with softwood there is lot of small bubbles wich are not detected properly and with hardwood there exist big odd shape bubbles which machine vision system does not regognice as bubbles. For the hardwood line, there is a large variation in bubble size from 0.2 to 0.8 mm. In the softwood line, there is also a clear long-term variation in bubble size from 0.07 to 0.18 mm.

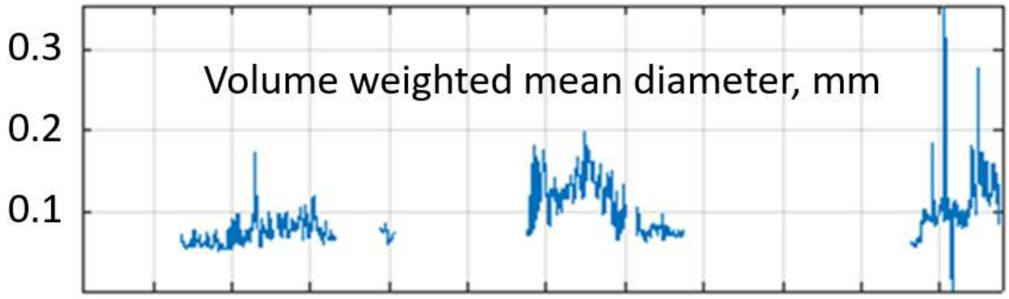


Figure 4: Volume weighted mean bubble diameter after the oxygen mixer in a softwood fiber line during a measuring period of three months [4].

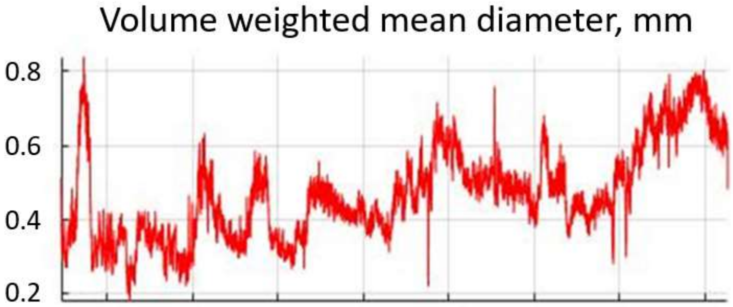


Figure 5: Volume weighted mean bubble size after the oxygen mixer in a hardwood fiber line during a measuring period of three months [4].

Figure 6 shows the volume weighted bubble size distribution along with an example of the image at the feed of the reactor. In the softwood line, the distribution is very narrow with a singular peak. In the hardwood line, the distribution is much wider and there are two peaks. There is a narrow distribution of very small-sized bubbles, which is similar to that seen in the softwood line, along with a wide distribution of larger bubbles. The proportions and sizes of these larger bubbles are actually much higher since the shape of the larger bubbles is more irregular; the machine imaging system does not recognize the irregularly shaped bubbles as accurately.

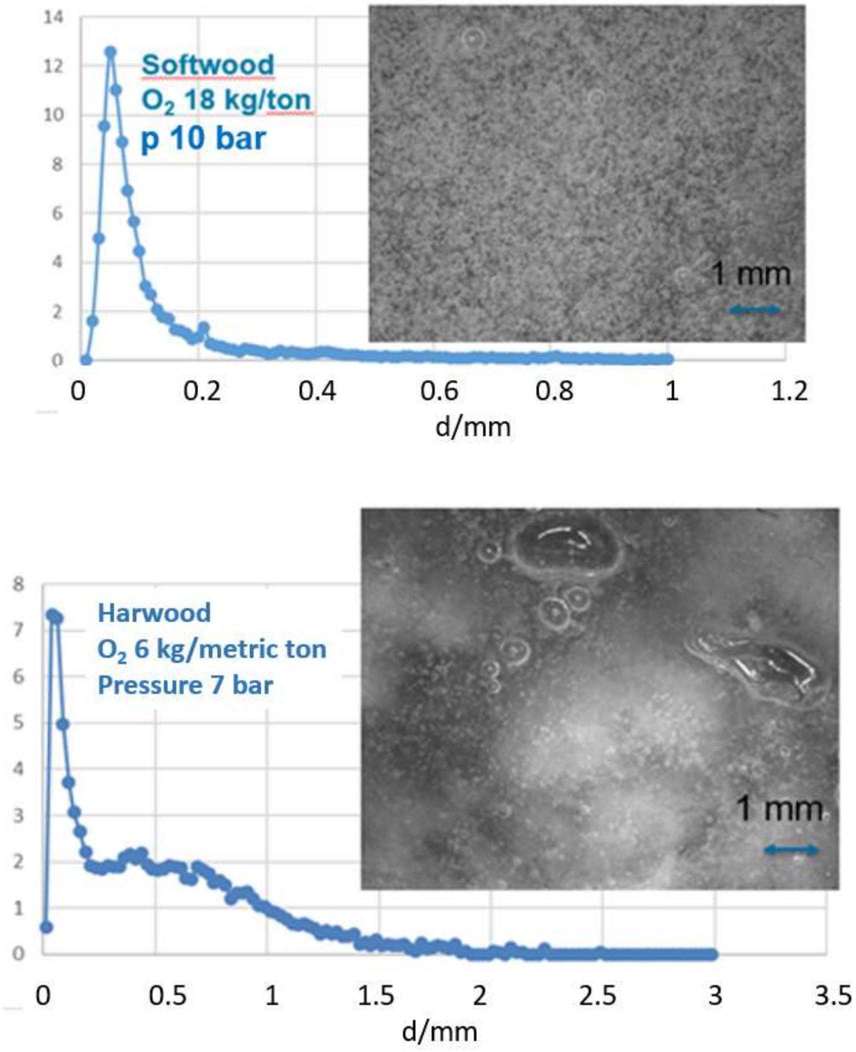
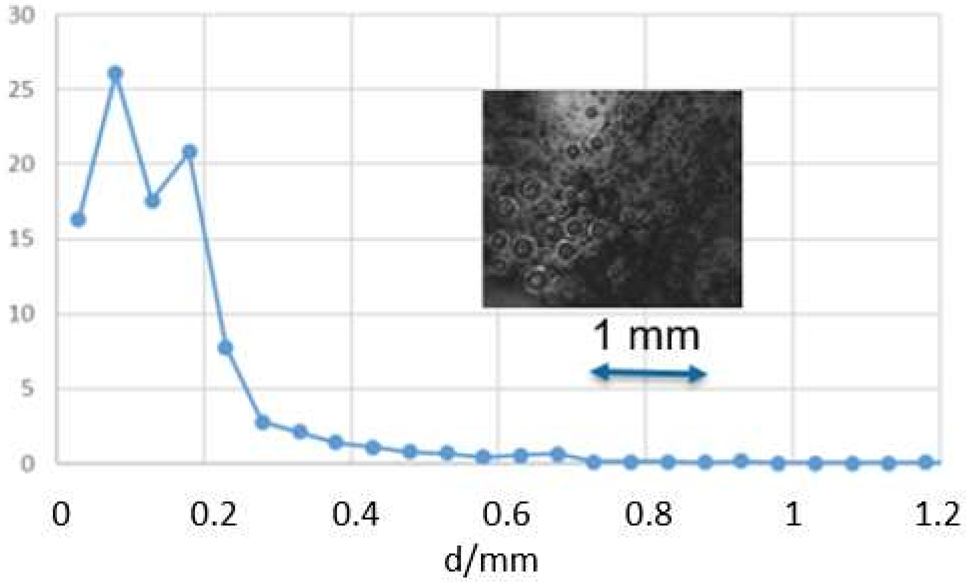
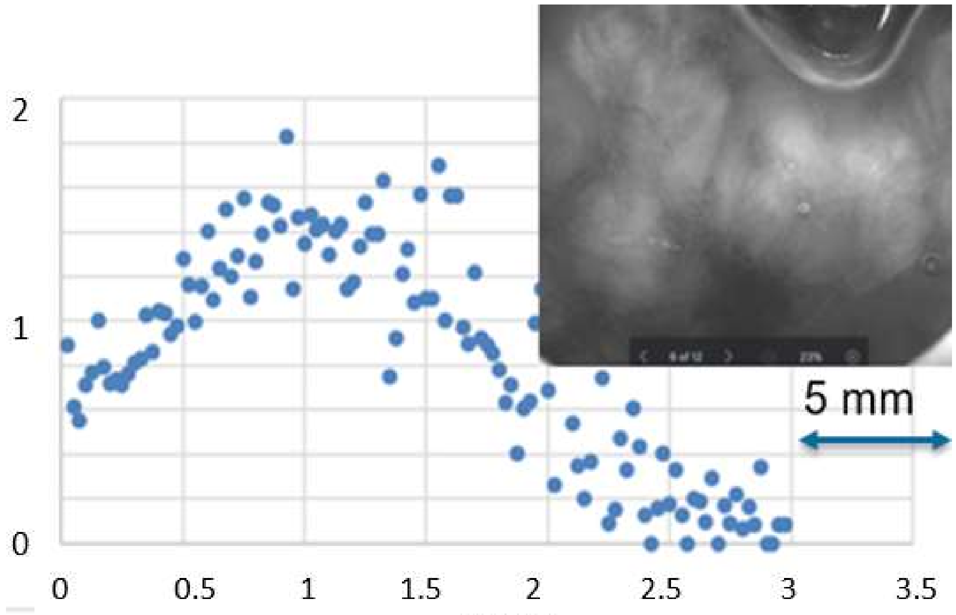


Figure 6: Typical volume weighted bubble size distribution and example of the image after the oxygen mixer in the softwood (top) and hardwood (bottom) fiber lines. The length step of bubble diameter used in calculation was 0.01 mm, so the Y-axis tells how many prosent of the bubbles exist in that category. E.g. with hardwood over seven prosent of bubbles exist in the category from 0.02 to 0.03 mm [4].

Figure 7 shows the volume weighted mean bubble size distribution along with an example of the image at the top of the reactor. In the softwood line, the bubble size at the top of the reactor is slightly larger to that of the feed, which can result from the dissolution of the smaller bubbles while leaving the larger bubbles unaffected. In the hardwood line, the bubbles were much larger than those of the feed into the reactor, which shows that there has been some coalescence of the bubbles within the reactor. The large amount of pictures taken from the top of the reactor (not shown here) also show that with hardwood there exist large, odd-shaped bubbles that do not exist in the feed into the reactor. According to these observation, it was concluded that if the bubbles were sufficiently small, e.g. smaller than 0.5 mm, then there were no appreciable bubble coalescence that occurred within the reactor. All of the bubble size measurements shown here, especially those in the case of softwood, but also for hardwood, indicated that the oxygen gas traveled through the reactor at the same speed as the pulp slurry.



Softwood, pressure 7 bar



Hardwood, pressure 3,3 bar

Figure 7: Volume weighted bubble size distribution and example of the image in the top of the reactor in the hardwood (top) and softwood (bottom) fiber lines [4].

Effect of different factors on oxygen bubble size in the process

Figures 8 and 9 show the effect of the mixer rotation speed and the oxygen charge on the bubble size in the hardwood line and the softwood line, respectively. For both fiber lines, the mixer rotation speed and the oxygen charge had a clear impact on the bubble size. It is interesting with regards to the hardwood line that doubling the oxygen charge caused the bubble size to double. This means that when the oxygen charge is increased, it is not clear how this imporove or does it improve the mass transfer of oxygen in the reactor. Hence, in this case adjusting the oxygen charge could be be an inefficient way to adjust the amount of dissolved oxygen; other measures, e.g. mixer rotation speed or pressure in the reactor, should be included in the kappa number control loop.

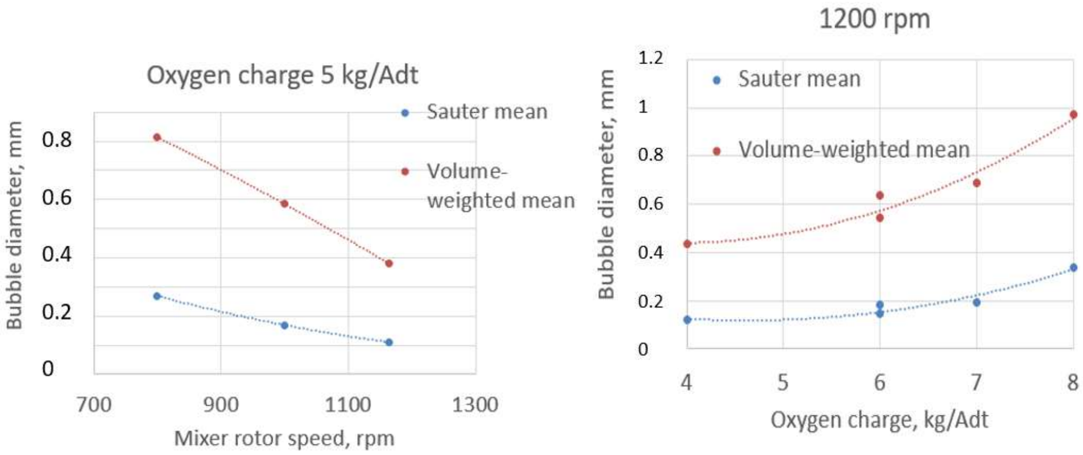


Figure 8: Effect of oxygen charge and mixer rotation speed on the mean bubble size in the hardwood line [4].

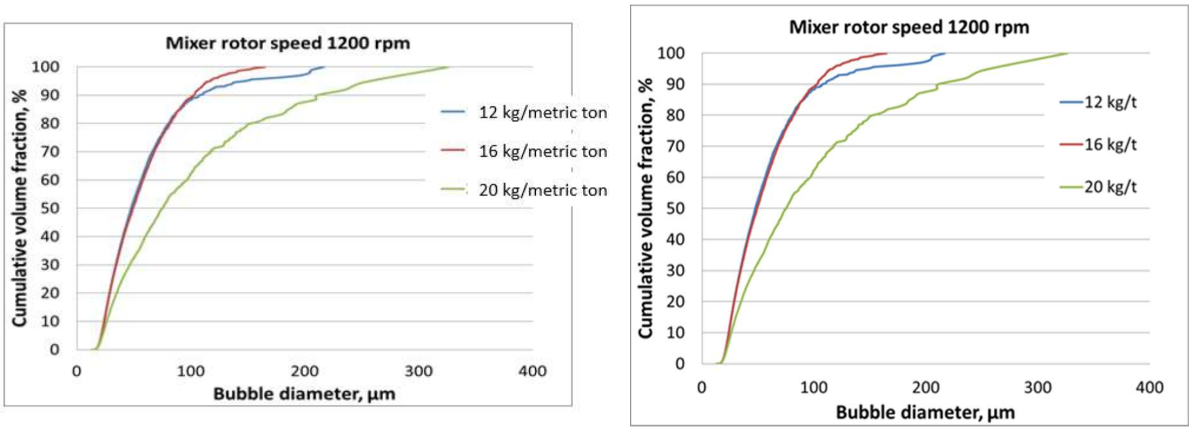


Figure 9: Effect of oxygen charge and mixer rotation speed on on the bubble size in the softwood line [10].

Besides the mixer control varibles, the surface chemistry of the pulp slurry, i.e., dissolved and colloidal substances, has an essential role in determining the bubble size of the dispersed gas. In pure water, it is impossible to form stable and small bubbles as detected by the machine imaging system. The differences in the bubble size in these two observed fiber lines are probably caused by differences in their surface chemistries since the mixing phenomena, i.e. the mixers and their energy consumptions, were nearly identical. Brownstock washer defoamers are used in the fiber line to control foam generation that would otherwise disturb pulp washing. The defoamers promote the coalescence of the gas bubbles, and when the bubbles are larger, they can be separated from the pulp and filtrates more easily. A test was conducted with the hardwood line to examine if the usage of the defoamer affected the oxygen bubble size by turning off the defoamer feed to the washing stage before the oxygen stage. Figure 10 shows that this had a clear effect by decreasing the bubble size in the oxygen reactor.

This indicates that the defoaming chemistry and the performance of the prior washing stages should be optimized simultaneously along with oxygen delignification performance.

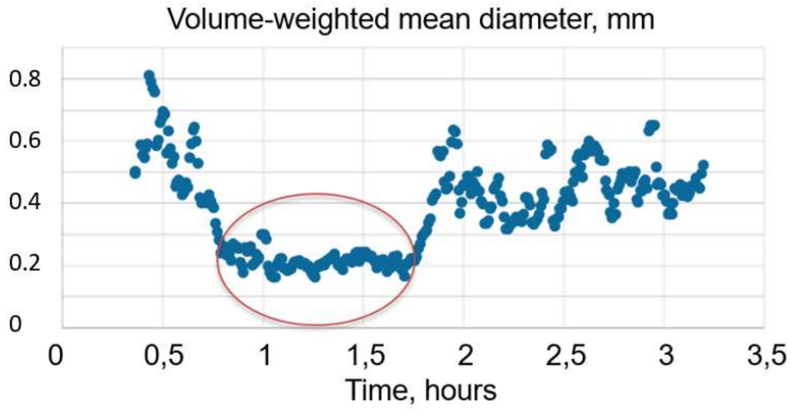


Figure 10: Volume weighted mean bubble diameter after the oxygen mixer when the defoamer feed to the brownstock washer prior to the oxygen reactor was shut off for a period of one hour [4].

Effect of oxygen bubble size on delignification

The essential question here is what is the quantitative effect of the oxygen bubble size distribution on delignification. Clearly, long-term mill experiments where oxygen dispersion properties change are difficult to implement in the production line and have not been done yet; however, there are other ways to obtain this information. Figure 11 shows the effect of reactor pressure on delignification. In this previous study [7] where factorial analyses were done, oxygen pressure had a significant effect on reactor delignification. For example, increasing the pressure in the top of the reactor from 280 kPa to 380 kPa clearly increased reactor delignification (see arrow in Figure 11). Both pressure and bubble size determine the amount of dissolved oxygen; hence, bubble size should also affect delignification.

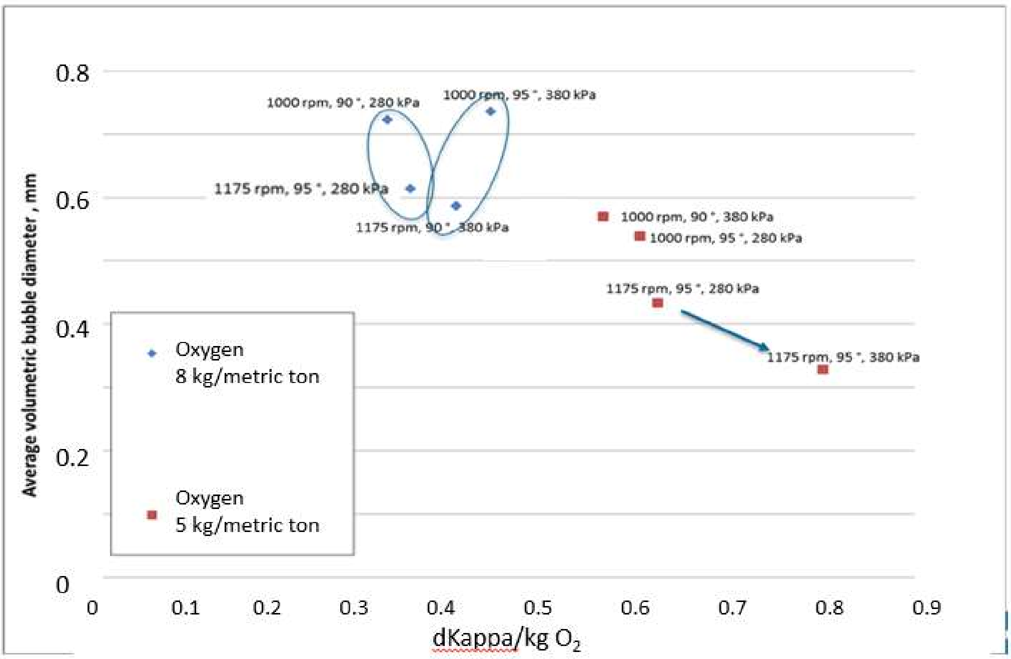


Figure 11: Effect of different oxygen reactor factors on kappa number reduction in experiments conducted with the hardwood fiber line. The inlet kappa was 17 and pressure value referres to the pressure in the top of the reactor. The pressure in the bottom of the reactor was about 700 kpa [7].

Figure 12 shows the volume weighted mean diameter and gas holdup estimate measured at the top of the reactor during the previously mentioned defoamer shut-off test. The gas holdup estimate indicates the amount of oxygen gas leaving the reactor. The gas hold up estimate is obtained simply by calculating together the total volume of bubbles which has been regognised so this is not same as gas void fraction (Xg) but it gives indication how Xg is changing. It can be clearly seen that the volume weighted mean diameter size and gas holdup decreased. Also, during the defoamer shut-off, there were no large gas bubbles observed at the top of the reactor (Figure 13). These observations indicates that more oxygen is consumed with the delignification reactions, which suggests that the reactor is working better.

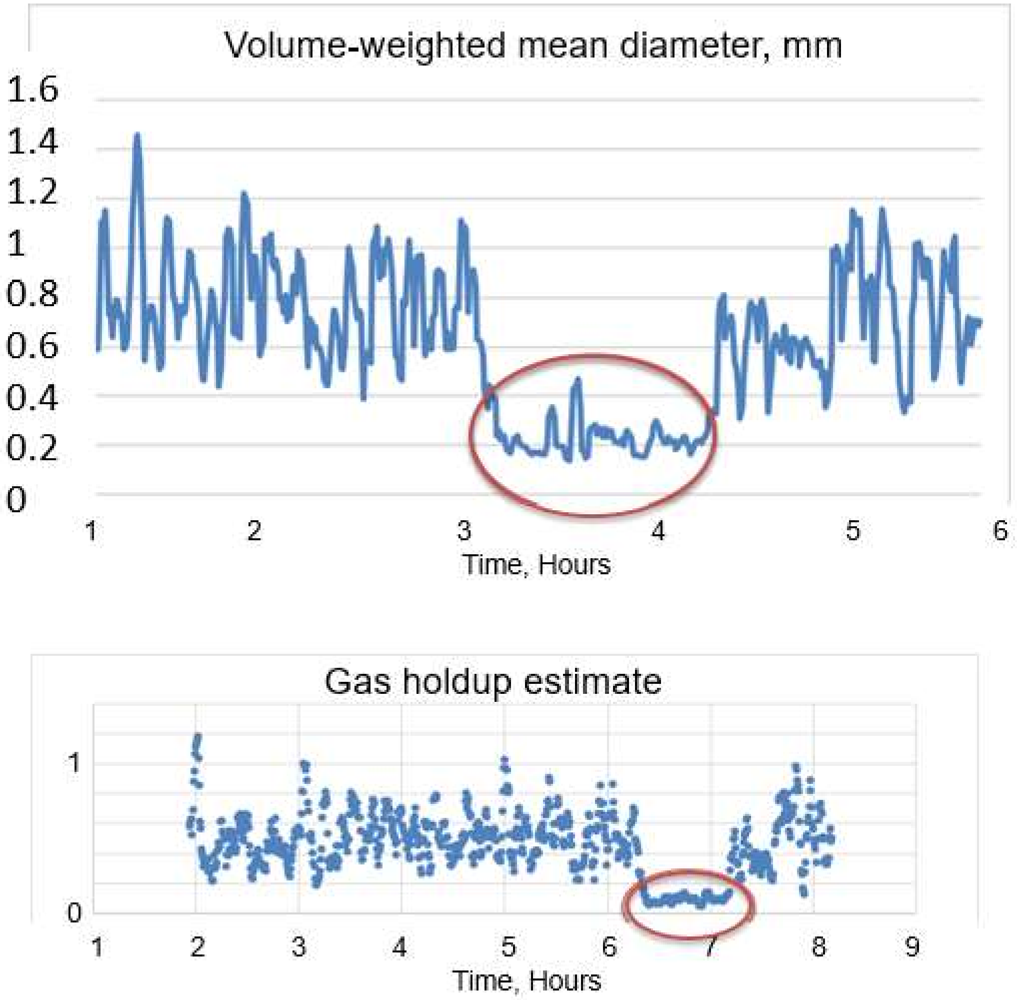
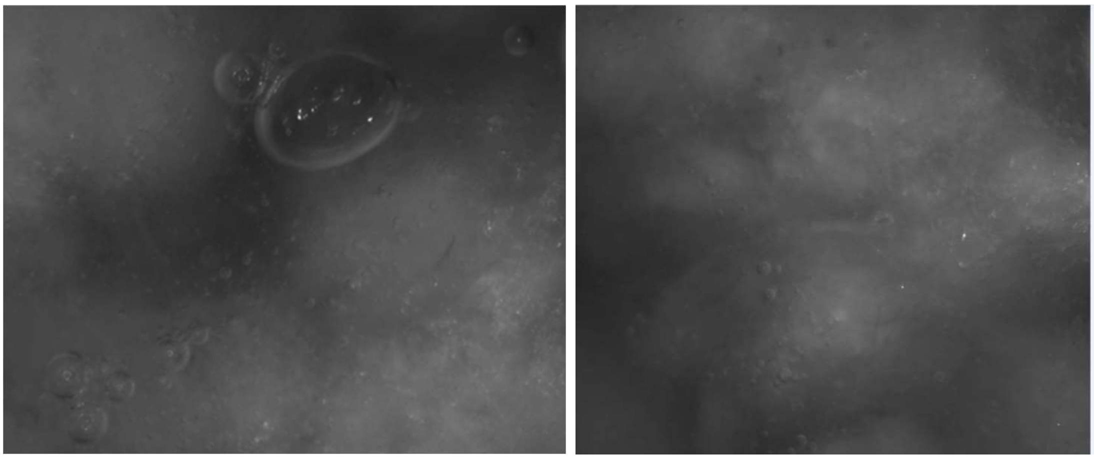


Figure 12. Volume weighted mean diameter and gas holdup estimate measured in the top of the reactor during the defoamer shut-off test [4].



a) *b)*

Figure 13. Example of gas dispersion at the top of the reactor: a) during a normal run, and b) during the defoamer shut-off test [4].

Another and more general way to quantify the effect of bubble size on kappa reduction is the numerical modeling of the delignification process. In an earlier modeling study [11], there was no direct basis to model oxygen mass transfer of the gas to the liquid phase. However, this physical phenomenon can be accurately modeled by having information about oxygen bubble size in the reactor. Also, with the aid of on-line bubble size measurement, the modeling can be verified through mill measurements and experiments. Currently, this kind of study is being conducted with our partners, South-Eastern Finland University of Applied Sciences and University of Maine.

CONCLUSION

The new on-line bubble size measurement works well when the bubbles are small and round. When the bubbles become larger and the shape becomes irregular, the probability of detecting them becomes smaller and the bubble size result obtained is smaller than the actual bubble size. In the softwood line, the mean volume weighted bubble size was about 0.1 mm and in the hardwood line, it was nearly ten times larger. The mixing of oxygen in these two lines was very similar, so the difference must come from the difference in the surface chemistry between these two lines. In both lines, there was considerable long-term variation in the measured bubble size.

The increase in the rotation speed of the mixer had a clear decreasing effect on the bubble size and the oxygen charge had a clear increasing effect. Coalescence of bubbles in the reactor was not observed in the softwood line, but in the hardwood line, some coalescence of larger bubbles occurred. In the hardwood line, the reactor pressure had a clear effect on delignification, which indicates that decreasing the oxygen bubble size, in this case, should also have a similar effect. In the test in the hardwood line, the use of brownstock washer defoamer caused an increase in bubble size.

The newly developed on-line bubble size measurement and the obtained mill results provide the basis for better understanding, adjusting, and controlling the oxygen delignification process. If the properties, adjustabilities, and meaning of oxygen dispersion properties are known, this provides a new basis for improving the oxygen delignification process:

- It is possible to define whether there is room to improve delignification appreciably by decreasing the bubble size in the oxygen reactor.
- If the amount of dissolved oxygen in the reactor can be kept at the optimal level through the control of the oxygen feed and its dispersion, this will:
    - Prevent gas entrainment in the stock slurry entering the subsequent washing stage.
    - Increase delignification or decrease variablility in the post-reactor kappa number.
    - Improve the delignification process, since one important, previously unknown parameter can now be controlled.
- The oxygen mixing technology and chemistry related to the control of gases in the fiber line can be developed.
- Physical modeling of the oxygen delignification stage can be improved so that this can be used by the mill as a tool for better understanding and controlling the process.

Also, on-line bubble size measurement can be used to study and control issues related to the gases in the fiber line more widely, for example to quantify the effects of defoaming agents in the washing stages.

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