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# Gas Dispersion in the Oxygen Delignification Process

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## ABSTRACT

There has been little or no knowledge about the state of gas dispersion in the oxygen delignification process. This paper presents a new on-line method for measuring oxygen bubble size distribution in the process as well as results from the studies made in the softwood and hardwood line, for example:

- In softwood line the average volumetric bubble size was about 0.1 mm and in the hardwood line it was nearly ten times higher. In both lines there was considerable variation in the measured bubble size over the long term.
- In both lines an increase in the rotation speed of the mixer had a clear decreasing effect on the bubble size, and oxygen charge had a clear increasing effect on the bubble size.
- In the softwood line no coalescence of bubbles in the reactor was observed, but in the hardwood line some coalescence of larger bubbles occurred in the reactor.
- In the test conducted in the hardwood line, the usage of defoamer had a clear increasing effect on the oxygen bubble size.
- In the hardwood line, reactor pressure had a clear effect on the delignification rate, which indicates that 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. Delignification occurs with the aid of an alkali and dissolved oxygen. Van Heiningen models the kinetics of oxygen delignification based on Equation 1 (Van Heiningen et al. 2003):

$$-(dK/dt) = Ae^{-51000/(8,314T)}([OH^-])^{0.7}(C_{O_2})^{0.7}(K)^2 \quad (1)$$

where K is the kappa number, A is the constant, T is the temperature (K) [OH<sup>-</sup>] out is the hydroxide concentration (mol/l) and C<sub>O<sub>2</sub></sub> is the concentration of dissolved oxygen.

This equation simply shows that there must be heat, a sufficient amount of hydroxide anions [OH<sup>-</sup>] and dissolved oxygen for delignification to occur. The temperature and amount of hydroxide anions are quite well known and quite easily controlled by the addition of steam and alkali. The amount of the dissolved oxygen is trickier, since it depends very much on the mass transfer rate of the oxygen from the gas phase to the water phase, which is determined by the surface area of the oxygen in the reactor. This is why the effective mixing of oxygen is an important factor for the proper functioning of the delignification process. The pulp passes in a plug-flow state through the reactor tower and mixing oxygen properly is the only way to ensure an efficient and stable delignification result. Due to the weak solubility of oxygen, there has to be small enough gas dispersion all the way inside the reactor to keep the amount of dissolved oxygen at a high level.

Up to this point, there has been little or no knowledge about the state of oxygen gas dispersion in the reactor, even though this can have a dramatic impact on the operational performance of the oxygen delignification process. In earlier papers presented at IPBC conferences (Mutikainen et al. 2014, 2017), a new imaging-based method for the characterization of gas dispersion in the oxygen delignification process was presented together with results regarding the state and effect of the different factors on dispersion bubble size in the fiber line (Kopra et al. 2017).

This paper will present more results, a summary of the earlier studies and conclusions related to the measurement, state and role of gas dispersion in the oxygen delignification process.

## 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 the imaging of pulp flow and detecting the bubbles using a machine vision system, see Figure 3. This measurement system is described more precisely in our earlier publication (Mutikainen et al. 2017). Gas dispersion measurements in the oxygen reactor were made in two fiber lines in Finland. One measurement period was performed in a line producing hardwood pulp (birch) and the other in a line producing softwood pulp.



K-patents installation valve and installation equipments



K-patents installation tube



Imaging probe

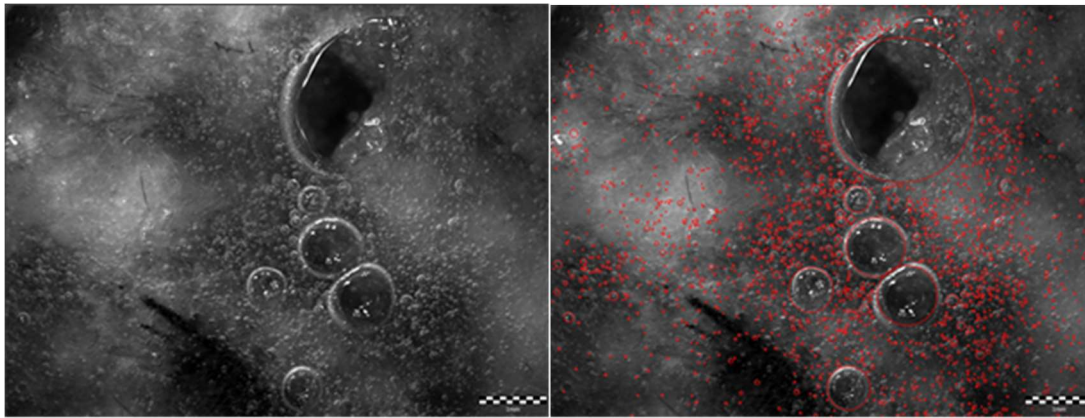


PC with on site control and monitoring

Figure 1: Essential parts of the bubble monitoring system



Figure 2: Bubble size measurement system installed in the process, including on-site control and monitoring capabilities



a) b)  
 Figure 3: a) The original image of bubble flow in the hardwood line after oxygen mixer. b) The detected bubbles are circled with red outlines on top of the image (Mutikainen et al. 2017)

## RESULTS AND DISCUSSION

### Oxygen bubble size distribution in the process

Figure 4 shows the average bubble size in the softwood line and Figure 5 in the hardwood line. In Figures 4 and 5 it can be seen that the average bubble size in the hardwood line is nearly ten times higher compared to the softwood line. In the hardwood line there is quite a large variation in bubble size from 0.3 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.

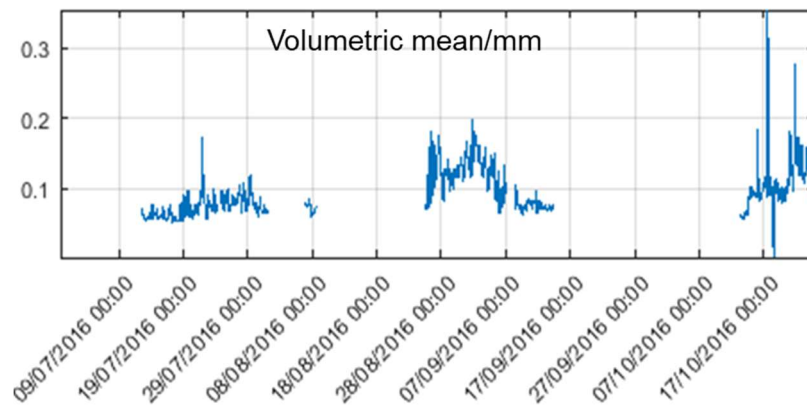


Figure 4: Volumetric mean bubble size after the oxygen mixer in a softwood fiber line

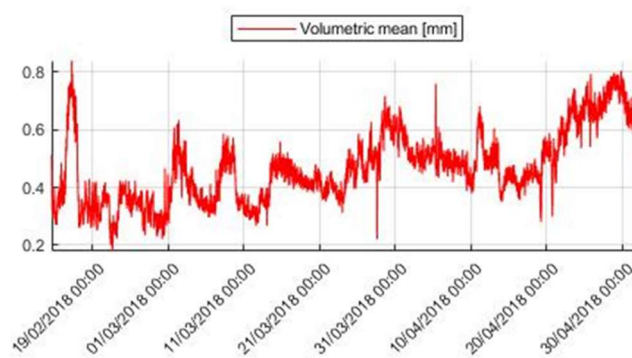


Figure 5: Volumetric average bubble size after the oxygen mixer in a hardwood fiber line during a measuring period of three months.

Figure 6 shows the volumetric bubble size distribution and an example of the image in the feed of the reactor. In the softwood line, the distribution is very narrow and there is only one peak. In the hardwood line the distribution is much wider and there are two peaks; a narrow distribution of the very small sized bubbles, the same size as seen in the softwood line, and a wide distribution of larger bubbles. The proportion and size of these larger bubbles is actually much higher, since the shape of the larger bubbles is more irregular and the machine vision system does not recognize them as bubbles so accurately.

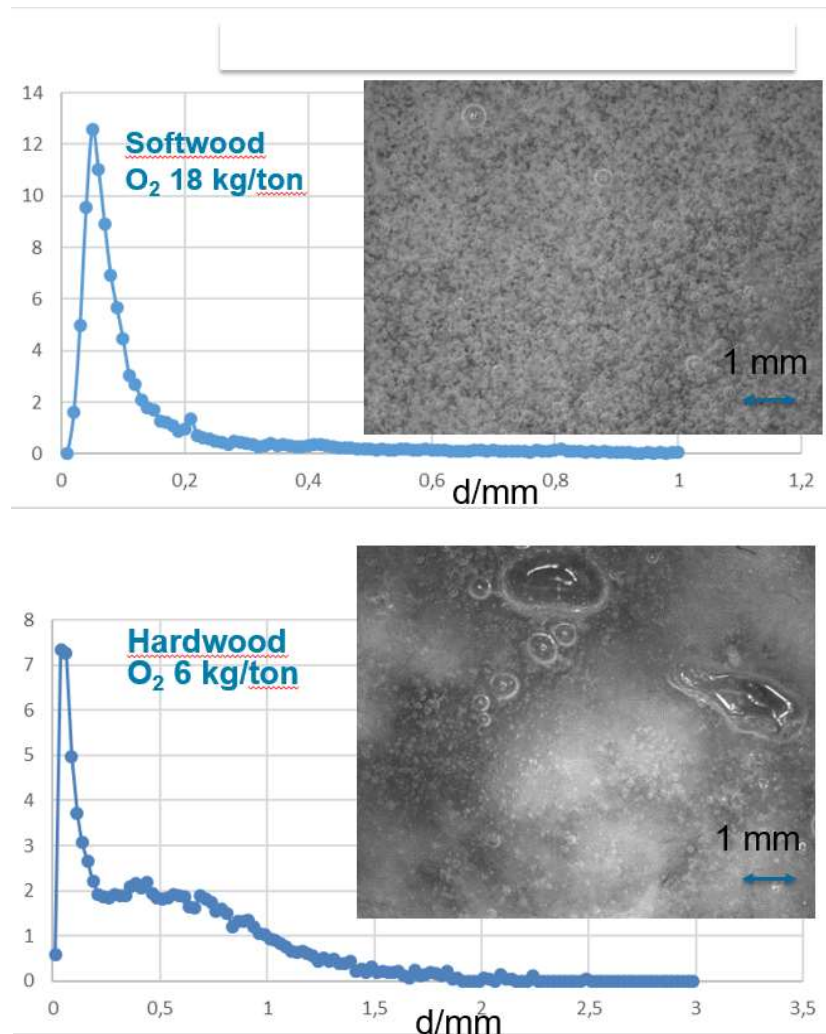


Figure 6: Typical volume weighted bubble size distribution and example of the image after the oxygen mixer in the softwood and hardwood fiber lines

Figure 7 shows the bubble size distribution and an example of the image in the top of the reactor. In the softwood line, the bubble size in the top of the reactor is slightly larger compared to the feed, which can result from the dissolution of the smaller bubbles leaving only the larger bubbles remaining. In the hardwood line there are much larger bubbles than those in the feed of the reactor, which shows that there has been some coalescence of the bubbles in the reactor. This can also be seen clearly in the images (not shown in here) taken from the top and the bottom of the reactor in the hardwood line. In the top of the reactor there are large, odd-shaped bubbles which do not exist in the feed of the reactor.

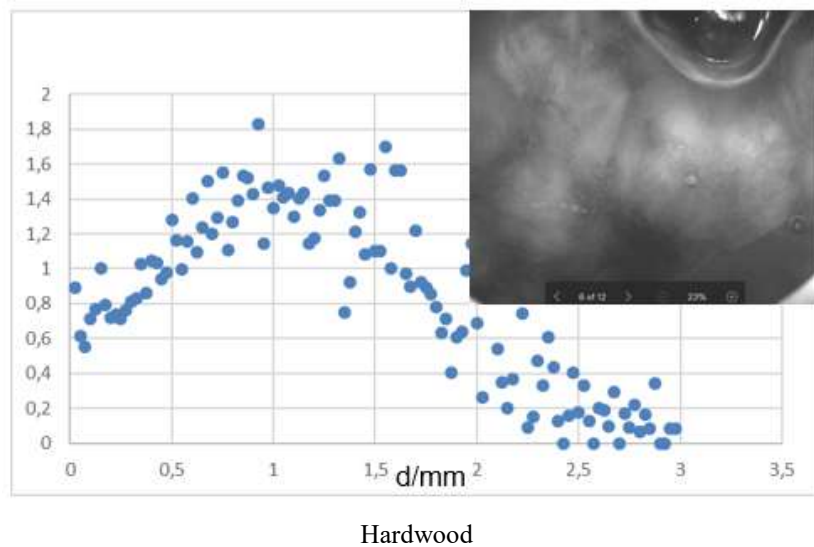
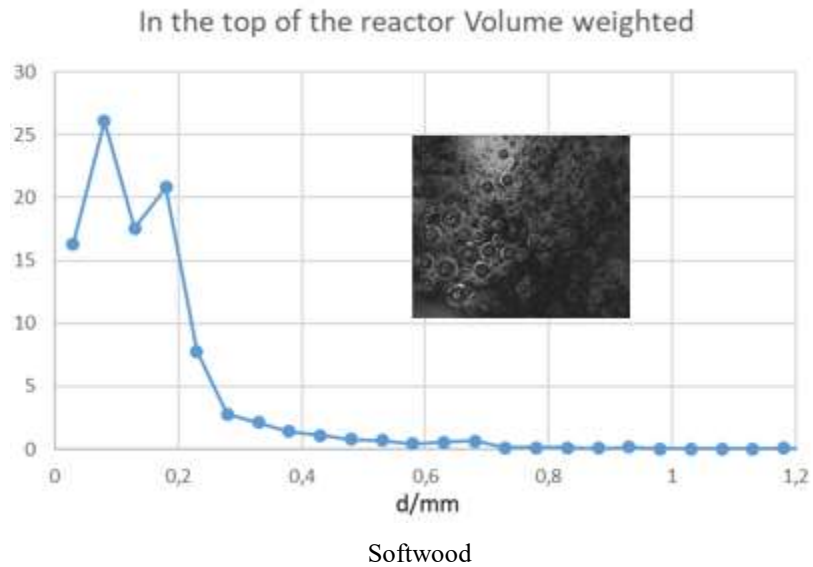


Figure 7: Volume weighted bubble size distribution and example of the image in the top of the reactor in the hardwood and softwood fiber lines

### Effect of different factors on oxygen bubble size in the process

Figure 8 shows the effect of the mixer rotation speed and oxygen charge on the bubble size in the hardwood line and Figure 9 in the softwood line. In both lines, the rotation speed of the mixer and oxygen charge had a clear effect on the bubble size. It is interesting that in the hardwood line, doubling of the oxygen charge also makes the bubble size two times larger. This means that when the oxygen charge is doubled, the surface area of the gas does not increase at all. So the oxygen charge can be quite an inefficient way to adjust the amount of dissolved oxygen and other measures, e.g. mixer rotation speed or pressure in the reactor, should be included in the kappa control loop.



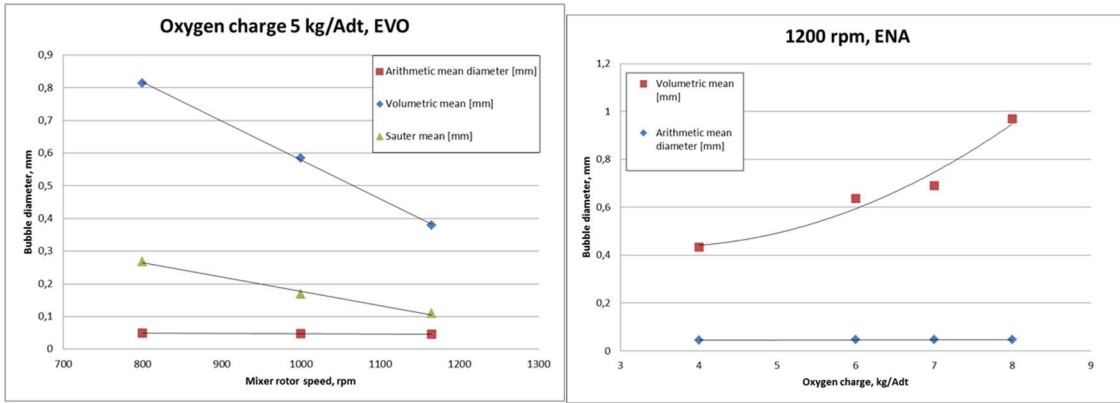


Figure 8: Effect of mixer rotation speed and oxygen charge on the average bubble size in the hardwood line

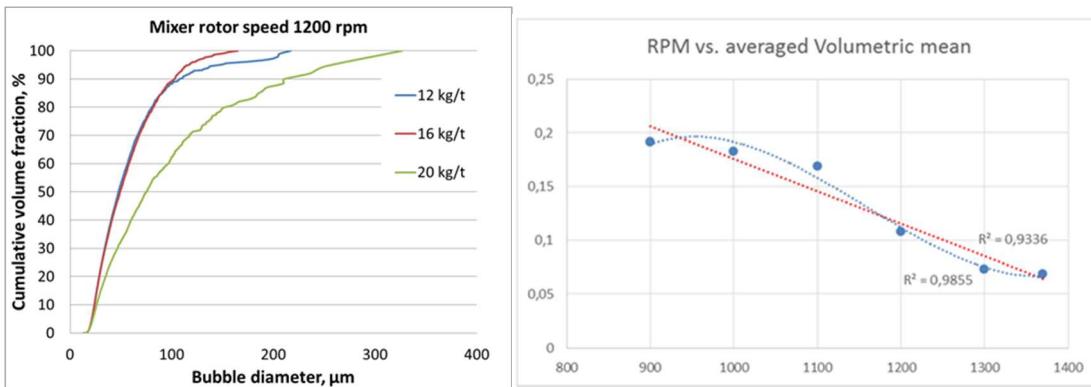


Figure 9: Effect of mixer rotation speed and oxygen charge on the bubble size in the softwood line (Mutikainen et al. 2014)

Besides the mixing process, the surface chemistry i.e. dissolved and colloidal substances in the process, has an essential role in determining the dispersion bubble size. In pure water, it is impossible to form the stable and small bubbles detected here. The differences in the bubble size in these two observed lines are very probably caused by the difference in the surface chemistry, since the mixing phenomena, i.e. the mixer model and energy consumption, are very similar in these two lines. Defoamers are used in the fiber line in order to control the gases which otherwise would disturb the 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 in the hardwood line to examine whether the usage of defoamer had an effect on the oxygen bubble size by shutting the feed of the defoamer off from the washing stage before the oxygen stage. Figure 10 shows that this had a clear decreasing effect on the bubble size in the oxygen reactor. This indicates that the defoaming chemistry and the performance of the washing stages and oxygen delignification should be optimized together.

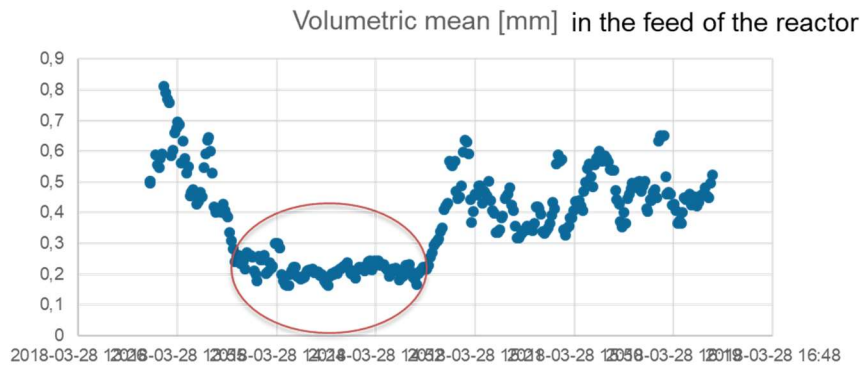


Figure 10: Volumetric average bubble diameter after the oxygen mixer, when the antifoaming agent feed to washer before oxygen reactor was shut off for the period of one hour.

### 4.3 Effect of oxygen bubble size on delignification

The essential question here is what is the quantitative effect of the oxygen bubble size distribution on the delignification. Long-term mill experiments where oxygen dispersion properties change clearly are difficult to implement in the production line and have not been done yet, but there are also other ways to obtain this information. Figure 11 shows the effect of reactor pressure on delignification. In this case, according to Figure 11 and the factorial analyses made in that study (Mutikainen et al. 2017), pressure had a significant effect on delignification. For example, increasing the pressure from 280 kPa to 380 kPa had a clear increasing effect on delignification, see the arrow in the figure 11. Pressure and bubble size together determine the amount of dissolved oxygen, and hence bubble size should also have an effect on delignification in this case.

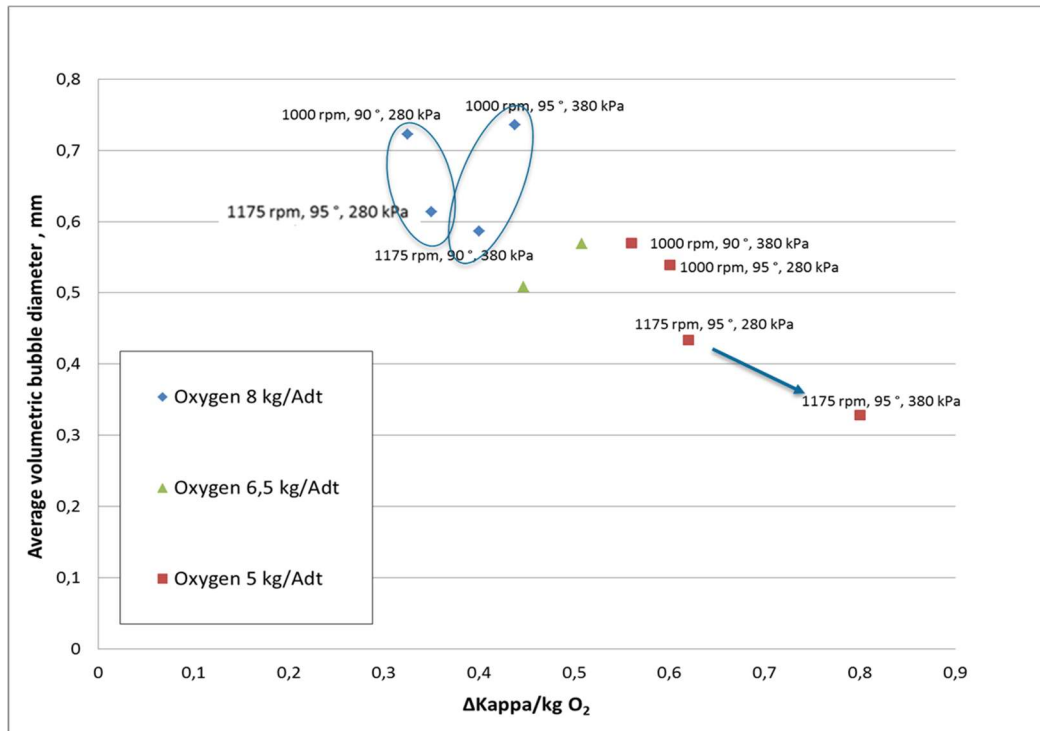


Figure 11: Effect of different factors on kappa reduction in experiments conducted in the hardwood line (Mutikainen et al. 2017)

In Figure 12 is shown the volumetric mean size and gas hold up level measured in the top of the reactor during the previously mentioned defoamer shut off test. The gas hold up level give the indication about the amount of oxygen gas leaving the reactor. In here can be seen that volumetric mean size and gas hold up level were decreased clearly. Also, during shut off there did not exist large gas bubbles in the top of the reactor, see figure 13. This all indicate that oxygen is consumed more to the delignification reactions and reactor is working better.



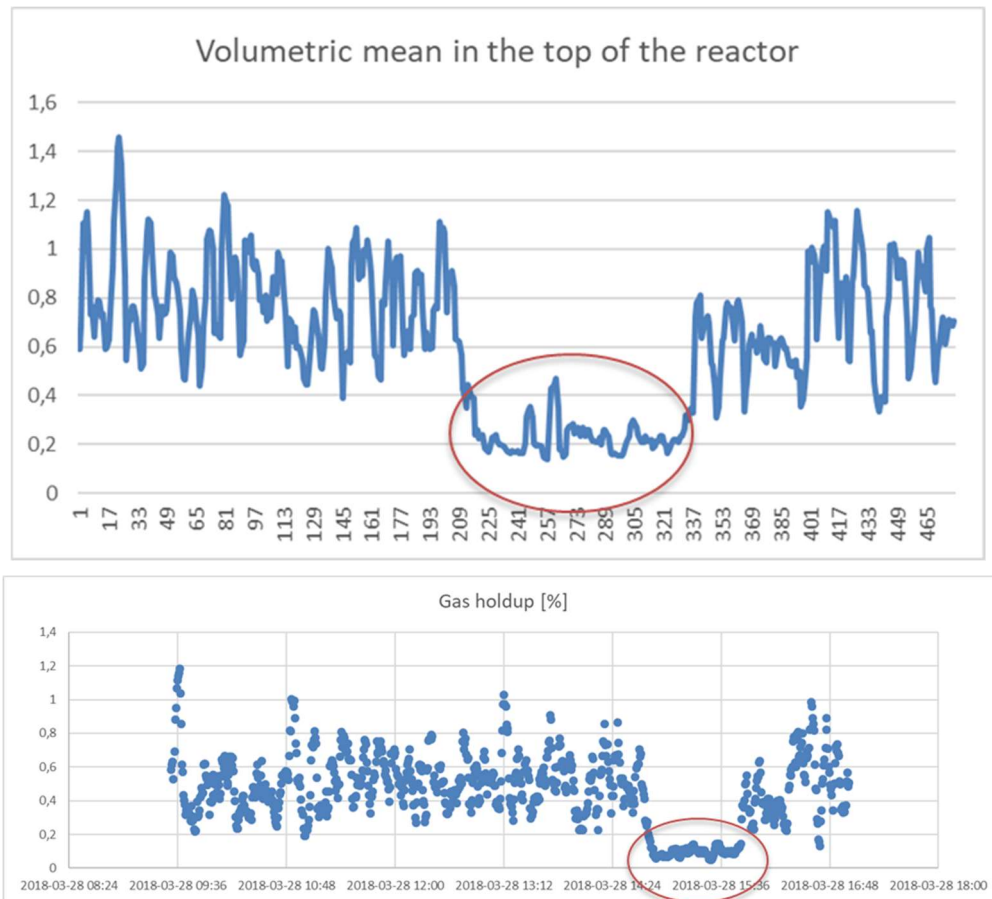
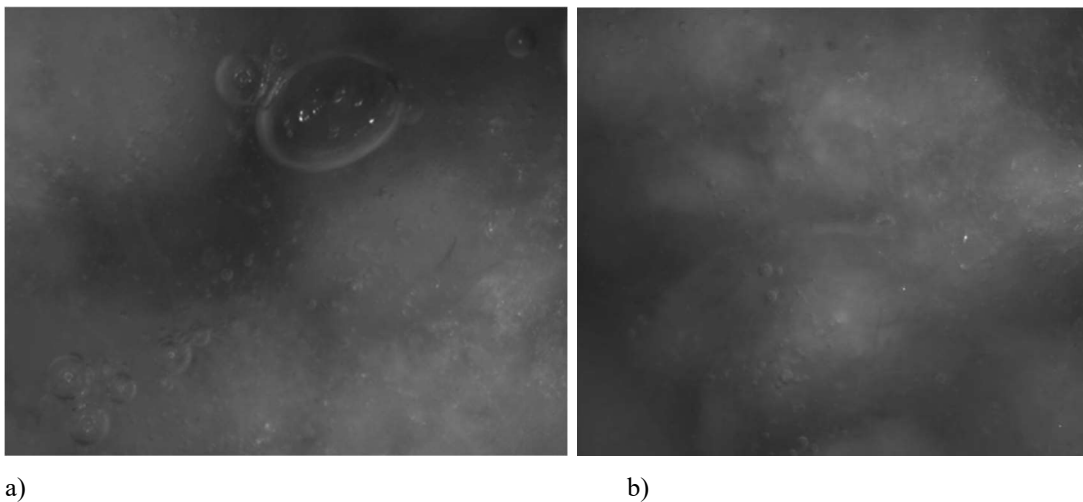


Figure 12. Volumetric mean and gash hold up level measured in the top of the reactor during defoamer shut off test.



a)

b)

Figure 13. Example of the gas dispersion in the top of the reactor a) in the normal run and b) during defoamer shut off test.

Another and more general way to quantify the effect of bubble size on kappa reduction is the physical modelling of the delignification process. In earlier studies related to the modelling (Van Heiningen et al. 2003), there was no direct physical basis to model the mass transfer of the oxygen to the liquid phase, but this is now possible to do through having information about oxygen bubble size in the reactor. In addition, with the aid of on-line bubble size measurement, the modelling can also be verified through mill measurements and experiments. This kind of study is planned to be conducted in the on-going GasOpti project in cooperation with participating companies and the University of Maine/Adrian Van Heiningen.

## CONCLUSIONS

In this presentation it is concluded that:

- New on-line bubble size measurement is working very nicely when the bubbles are small and round. When the bubbles get larger and the shape becomes irregular, the probability of detecting them gets smaller and the bubble size result obtained is smaller than the actual bubble size. This could be to some extent corrected with a function which takes this into account.
- In the softwood line the average volumetric 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.
- In both lines the increase in the rotation speed of the mixer had a clear decreasing effect on the bubble size and oxygen charge had a clear increasing effect on the bubble size.
- Coalescence of bubbles in the reactor was not observed in the softwood line, but in the hardwood line some coalescence of larger bubbles certainly occurred in the reactor.
- In the hardwood line, the reactor pressure had a clear effect on the 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 usage of defoamer had a clear increasing effect on the oxygen bubble size.

The new developed on-line bubble size measurement and mill results obtained give the possibility and base to better understand, adjust and control the oxygen delignification process. If the properties, adjustability and meaning of oxygen dispersion properties are known, this gives a totally new basis to improve the oxygen delignification process:

- It is possible to define whether there is room to improve delignification significantly 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 dispersion this will:
  - Prevent delivery of oxygen gas to the subsequent washing stage
  - Increase delignification or decrease scattering in the end kappa number
  - Give room to further develop the delignification process since one important, previously unknown parameter is now controlled.
- The oxygen mixing technology and chemistry related to the control of gases in the fiber line can be developed
- Physical modelling of the oxygen delignification stage can be improved so that this can be used in the mill as a tool for better understanding and controlling the process.

In addition 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 for quantifying the effects of defoaming agents in the washing stages.

These studies are continuing in the GasOpti project through modelling and mill studies, e.g. studying how the usage of defoamers should be optimized from the perspective of the performance of the oxygen and washing stages. Information related to bubble size in oxygen reactors in different production lines will also be collected. One particular interest is in conducting measurements and tests in a eucalyptus pulping line.

## ACKNOWLEDGMENTS

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