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 inline 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 10 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 use 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 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.

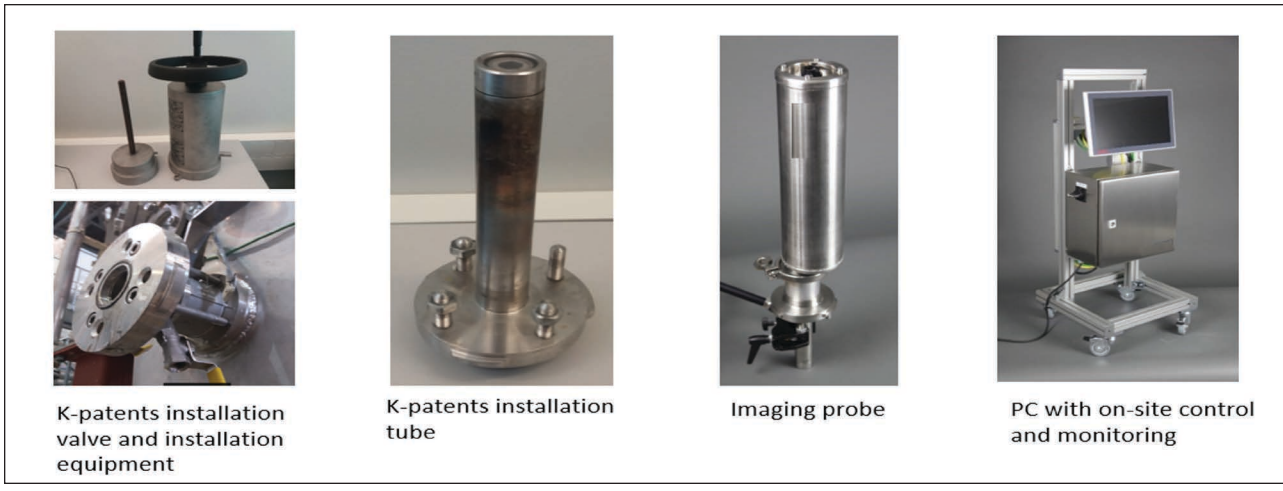
Application: Information related to gas dispersion offers the possibility to develop and improve the performance of the oxygen delignification process.

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 [1]. The amount of the dissolved oxygen is less defined, because 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 [1,2]; 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 [3].

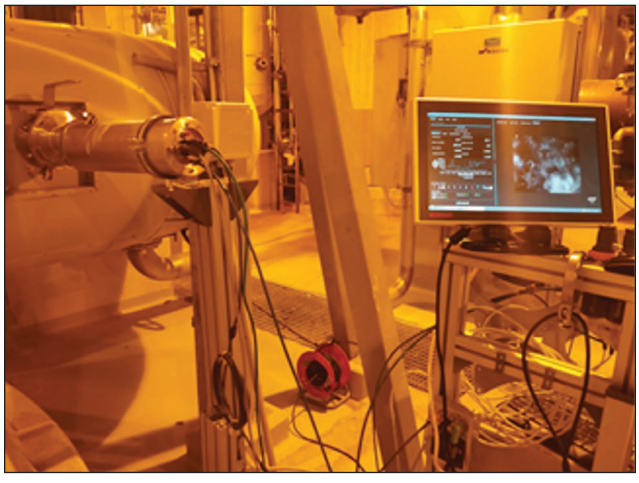
Up to this point, there has been little-to-no knowledge about the state of oxygen gas dispersion in the reactor, even though this factor can have a major impact on the operational performance of the oxygen delignification reactor. In our earlier papers [4-11], a new image-based method to characterize 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 paper 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

Online bubble size measurement was conducted using a continuous Pixact Bubble Monitoring (PBM) system (Pixact Ltd.; Tampere, Finland), as shown in **Figs. 1-2**. The measurement is based on imaging of the pulp flow and detecting the bubbles using a machine imaging system (**Fig. 3**). This measurement system has been described in detail in an earlier publication [8]. 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.



1. Essential parts of the bubble monitoring system [3].



2. Bubble size measurement system installed in the process that includes onsite control and monitoring capabilities [4].

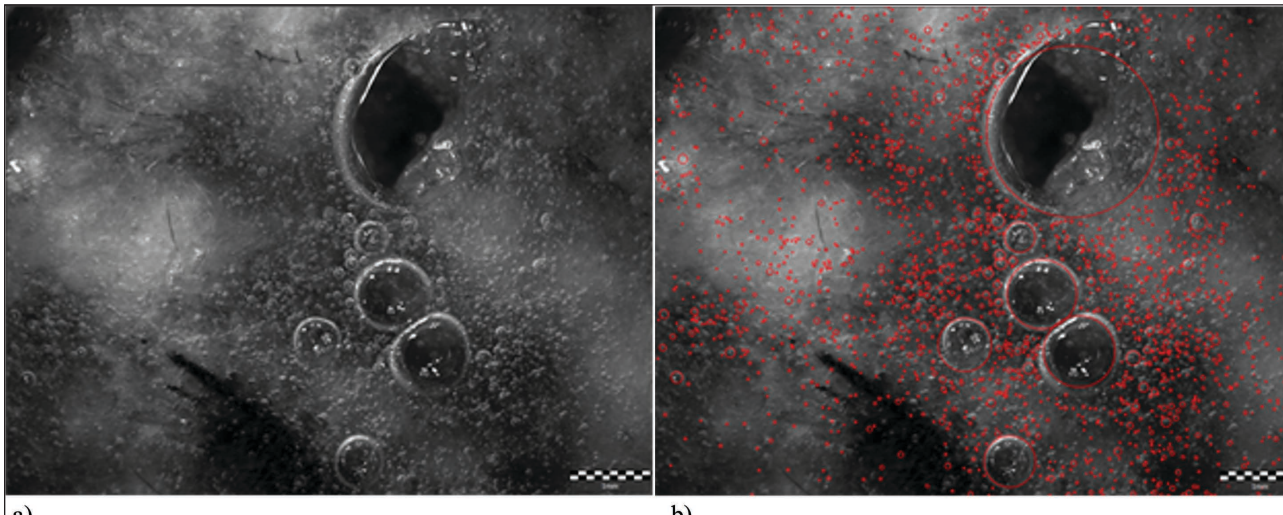
RESULTS AND DISCUSSION

Oxygen bubble size distribution in the process

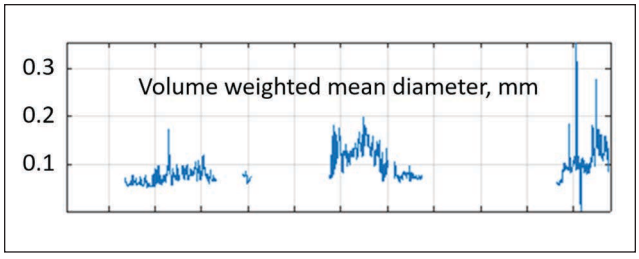
Figure 4 shows the volume-weighted mean bubble diameter in the softwood line, and **Fig. 5** shows the volume-weighted mean bubble diameter in the the hardwood line. The volume-weighted mean bubble diameter (d_v) is calculated as:

$$d_v = \frac{\sum_{i=1}^{N} d_i V_i}{\sum_{i=1}^{N} V_i} \quad (1)$$

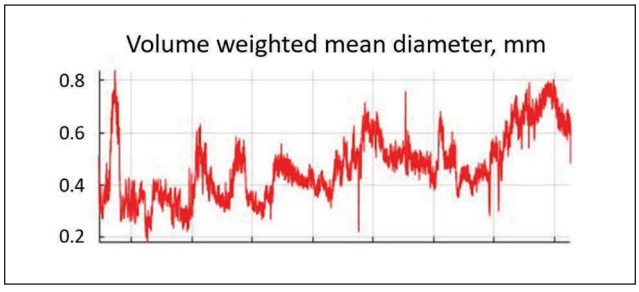
where:
d_i = diameter of the individual bubble
V_i = volume of the individual bubble
N = number of bubbles



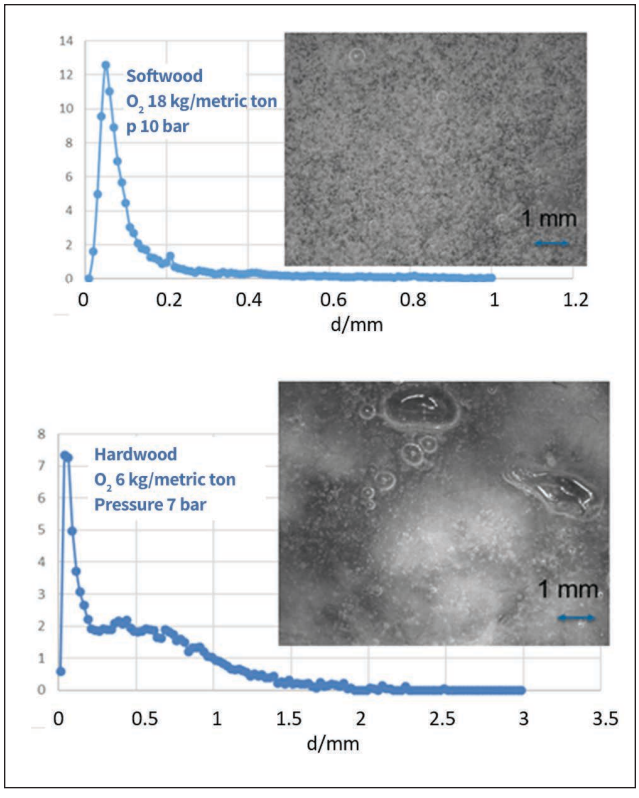
3. a) Original image of bubble flow in the hardwood line after oxygen mixer; and b) detected bubbles by the machine imaging system are outlined in red [8].



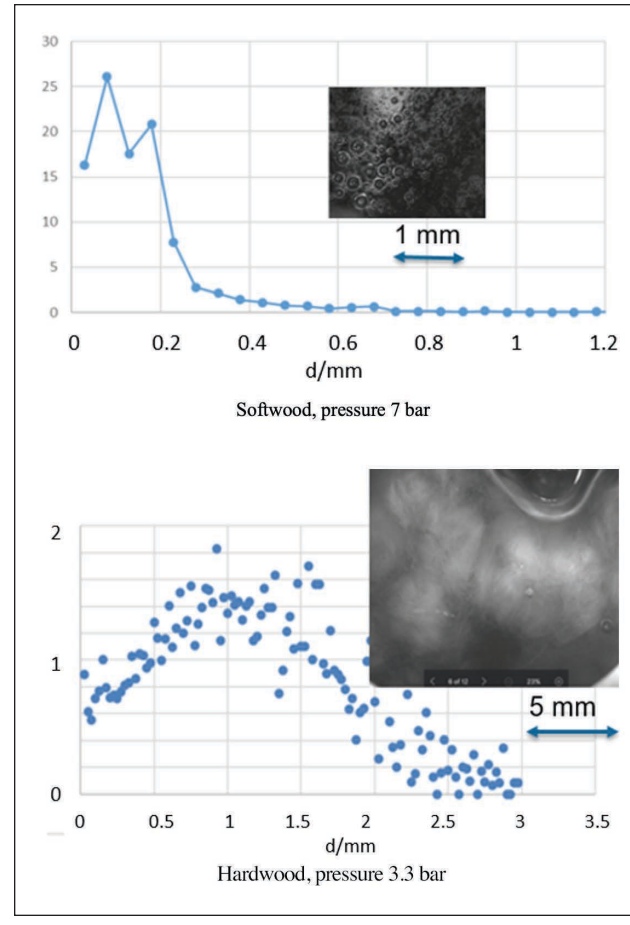
4. Volume-weighted mean bubble diameter after the oxygen mixer in a softwood fiber line during a measuring period of three months [5].



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



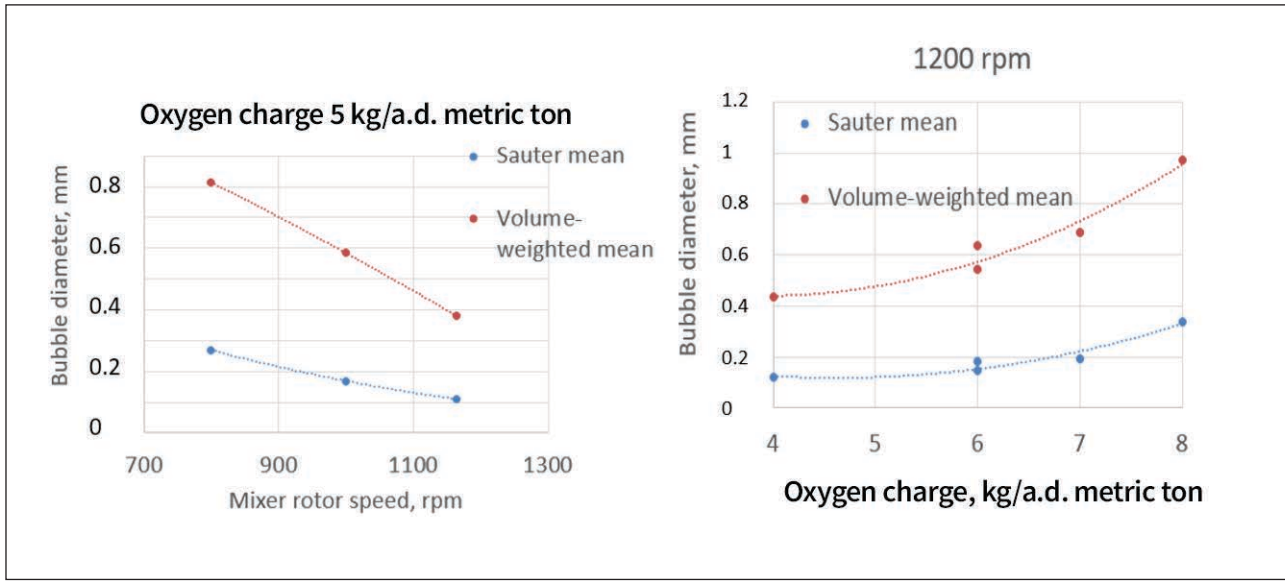
6. Typical volume-weighted bubble size distribution, and image examples after the oxygen mixer in the softwood (top) and hardwood (bottom) fiber lines. The length step of bubble diameter used in calculation was 0.01 mm, so the y-axis indicates the percent of the bubbles existing in that category (e.g., with hardwood, over 7% of the bubbles exist in the category from 0.02 to 0.03 mm) [5].



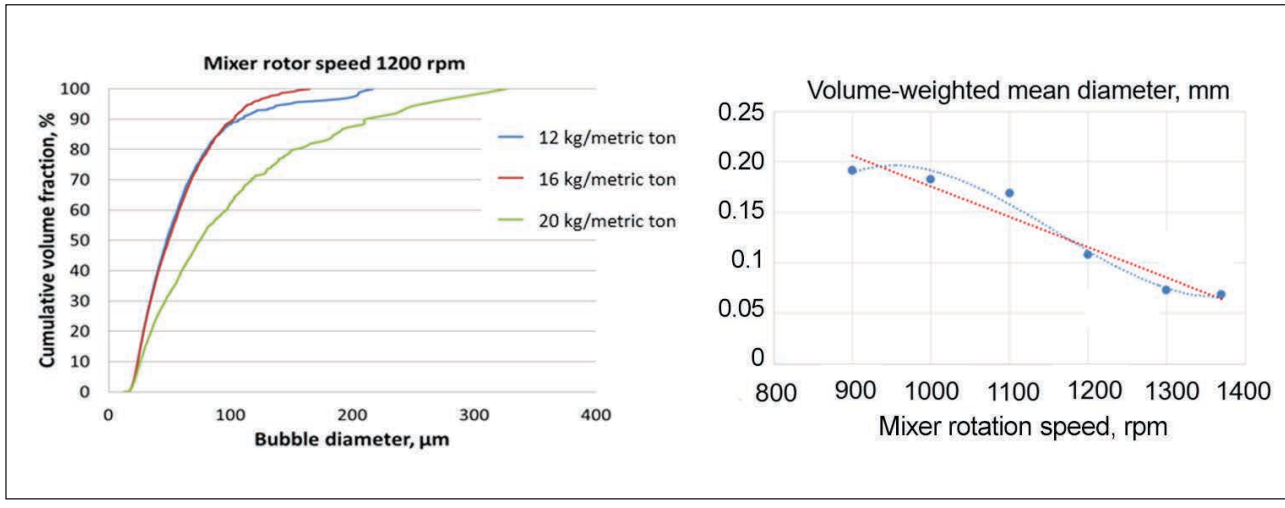
7. Volume-weighted bubble size distribution, and image examples from the top of the reactor in the hardwood (top) and softwood (bottom) fiber lines [5].

In Figs. 4-5, it can be seen that the volume-weighted mean bubble size in the hardwood line is about five times higher than that of the softwood line. The actual difference beween the two is likely greater due to limitations with the machine imaging system. There are a lot of small bubbles that are not detected with the softwood, whereas there are big and oddly shaped bubbles that are not recognized as bubbles with the hardwood. 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 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, because the shape of the larger bubbles is more irregular; the machine imaging system does not recognize the irregularly shaped bubbles as accurately.



8. Effect of oxygen charge and mixer rotation speed on the mean bubble size in the hardwood line [5].

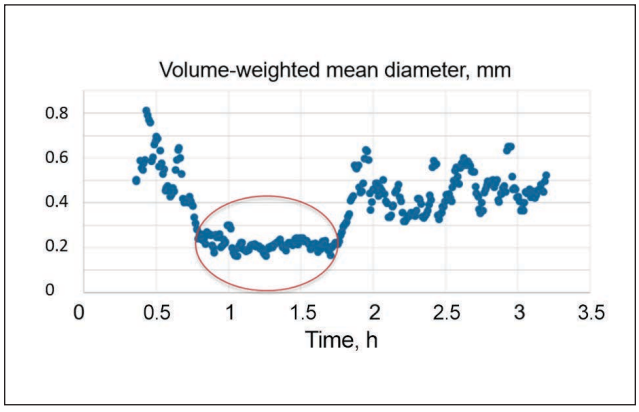


9. Effect of oxygen charge and mixer rotation speed on on the bubble size in the softwood line [11].

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 than 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 number of pictures taken from the top of the reactor (not shown here) also show that, with hardwoods, there exist large, odd-shaped bubbles that do not exist in the feed into the reactor. According to these observations, it was concluded that if the bubbles were sufficiently small (e.g., smaller than 0.5 mm), then there was 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.

Effect of different factors on oxygen bubble size in the process

Figures 8-9 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 improves or does not 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



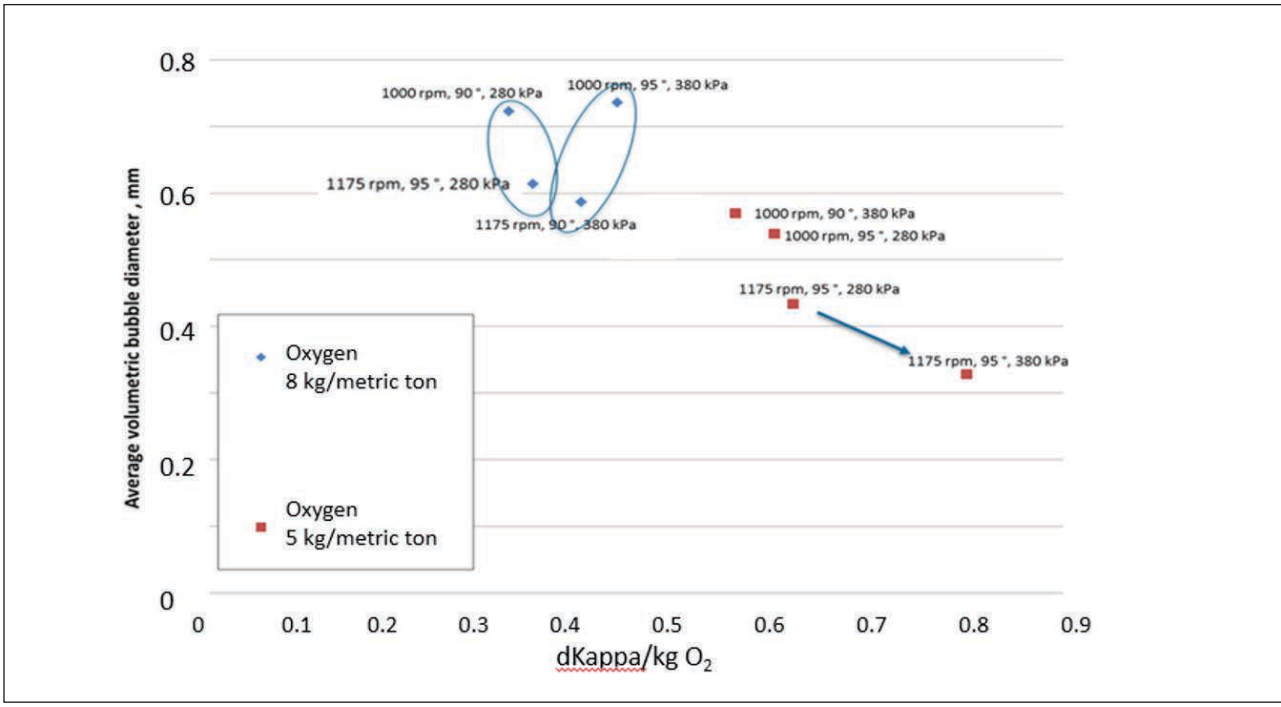
10. Volume-weighted mean bubble diameter after the oxygen mixer when the defoamer feed to the brownstock washer prior to the oxygen reactor was shut off for a period of 1 h [5].

amount of dissolved oxygen; other measures, such as mixer rotation speed or pressure in the reactor, should be included in the kappa number control loop.

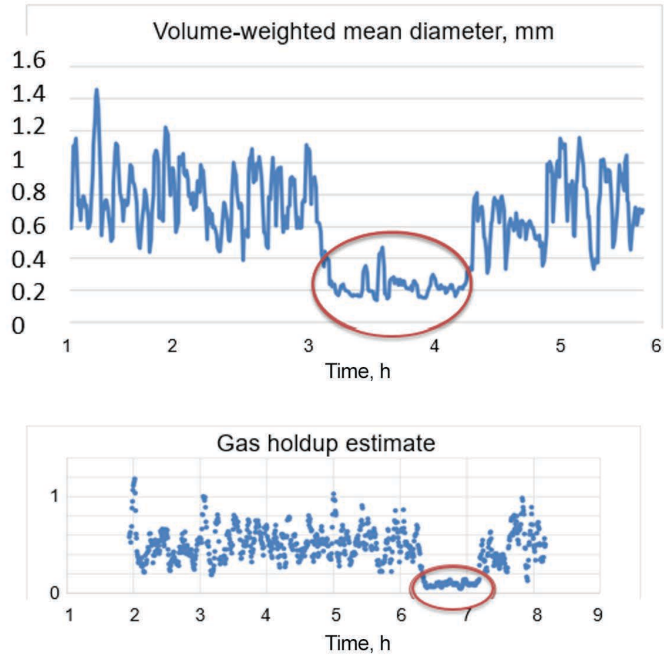
Besides the mixer control varibles, the surface chemistry of the pulp slurry (i.e., dissolved and colloidal substances) has an essential role in determining the bubble size of the dispersed gas. In pure water, it is impossible to form stable and small bubbles as detected by the machine imaging system. 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 consumption) 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.

Effect of oxygen bubble size on delignification

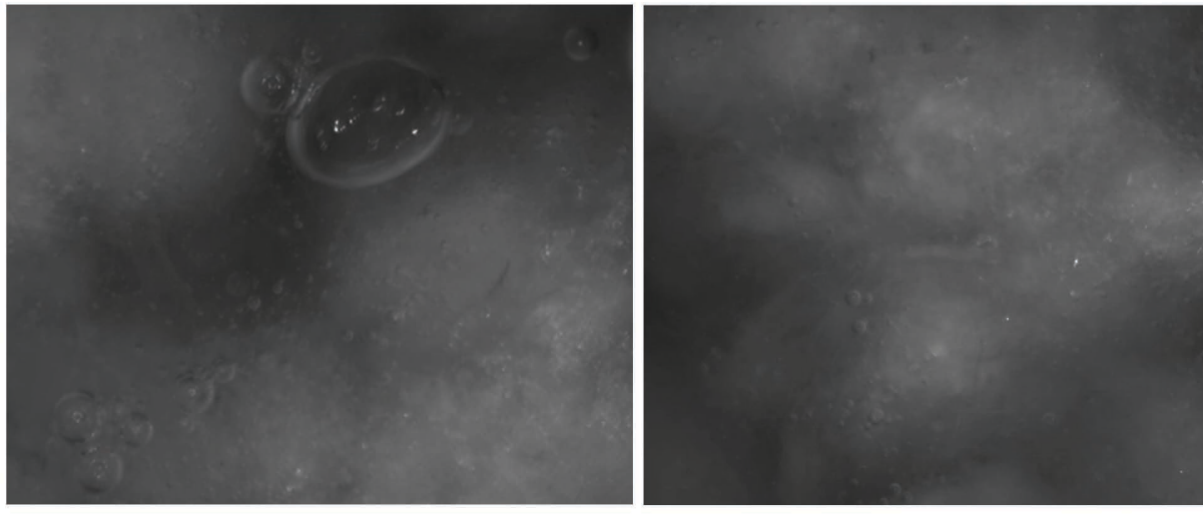
The essential question here is what is the quantitative effect of the oxygen bubble size distribution on delignification. Clearly, long-term mill experiments where oxygen dispersion properties change are difficult to implement in the production line and have not been done yet; however, there are other ways to obtain this information. **Figure 11** shows the effect of reactor pressure on delignification. In a previous study [8] 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 Fig. 11). Both pressure and bubble size determine the amount of dissolved oxygen; hence, bubble size should also affect delignification.



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 refers to the pressure at the top of the reactor. The pressure at the bottom of the reactor was about 700 kPa [8].



12. Volume-weighted mean diameter and gas holdup estimate measured at the top of the reactor during the defoamer shut-off test [5].



13. Example of gas dispersion at the top of the reactor: a) during a normal run, and b) during the defoamer shut-off test [5].

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 simply obtained by calculating the total volume of the bubbles that has been regognized by the imaging system. This estimate is not same as gas void fraction (X_g), but it gives a possible indication of how X_g changes. 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 (**Fig. 13**). These observations indicates that more oxygen is consumed with the delignification reactions, which suggests that the reactor is working better.

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 [1], 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 online 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 the University of Maine.

CONCLUSIONS

The new online 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 online 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

ABOUT THE AUTHORS

In the Fiber Laboratory at South-Eastern Finland University of Applied Sciences, we have conducted research work related to the mixing phenomena in pulp and paper processes for over 20 years. Mixing of gases has been one subject studied, and there have been indications that there is the potential to improve such processes by better understanding the state and behavior of oxygen gas dispersion, especially for the oxygen delignification process.

When we began these studies, there was no suitable measurement for quantifying the oxygen bubble size, and it took many years to develop this measurement. Over time, we discovered that, in some cases, gas dispersion is not good enough to produce the best performance for the oxygen stage and that use of defoamer may have a large negative effect on the formation of gas dispersion.

Now, there is the possibility to study gas dispersion and its effects, so there is much potential for improving industrial oxygen delignification stages around the world. In the short term, mills that feel their process is not performing with the best possible efficiency can inspect to determine if this issue is related to the poor dispersion of oxygen gas. Longer term, this practice can yield improvements related to structure, control, and chemistry of the process.

Our next step is to obtain more information about the state of gas dispersion in different processes and to more precisely define the effects of gas bubble size on delignification.

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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, online 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. **TJ**

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