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The impact of solid/floc holdup on oxygen transfer in a rotating hollow fiber membrane bioreactor under endogenous conditions

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ABSTRACT

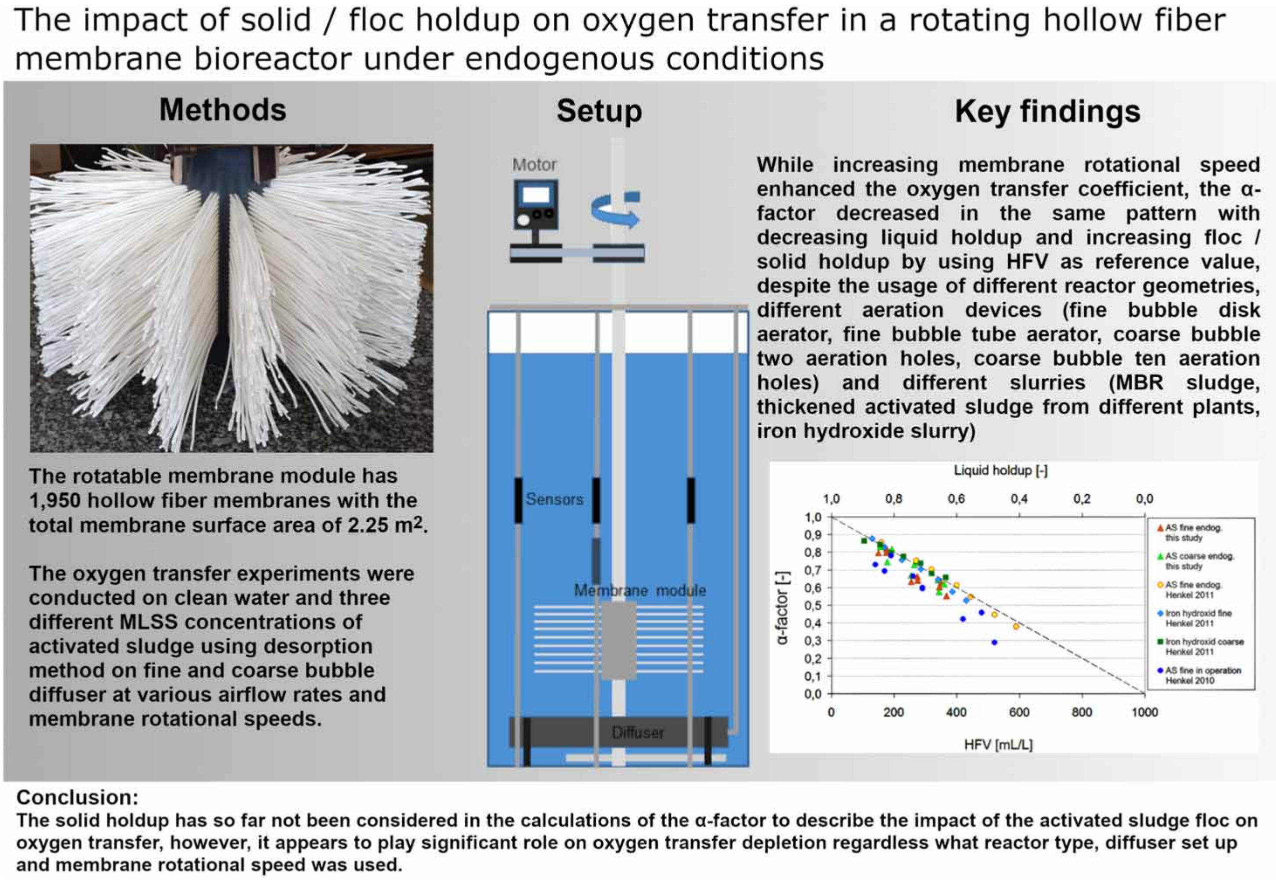
A set of oxygen transfer experiments in clean water and three different activated sludge concentrations were conducted with fine and coarse bubble aeration in a rotating hollow fiber membrane bioreactor to observe the impact of different rotational speeds on the oxygen transfer rate. The results showed that with increasing membrane rotational speed, the oxygen transfer coefficient enhanced while the α-factor showed similar values at comparable sludge concentrations and solid/floc holdups. The highest improvement rates occurred during the experiments with coarse bubble aeration at 50 rpm and the lowest specific airflow rate. The solid/floc holdup appears to universally impact oxygen transfer depletion regardless of what reactor type, diffuser setup and membrane rotational speed were used in the wastewater experiments.

Key words: membrane bioreactor, oxygen transfer, solid holdup, α-factor

HIGHLIGHTS

- The rotating hollow fiber membrane improved the oxygen transfer coefficient.
- Increasing membrane rotational speed did not improve the α-factor.
- The solid/floc holdup concept is introduced to explain oxygen transfer depletion.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Gas transfer measurements are routine in the field of multi-phase flow studies. If the impact of particles is investigated the solid holdup is introduced, which describes the fraction of solids within the total volume of the suspension (Sun & Furusaki 1989). The aeration of activated sludge belongs to the biggest applications in the field of multi-phase flow studies and it consumes 50–60% of the energy consumption in a wastewater treatment plant (WWTP) (Chen *et al.* 2022). One fundamental problem for slurries that consist of activated sludge flocs or hydroxide flocs is the determination of the solid/floc holdup and its impact on oxygen transfer. Surrogate parameters used to express the impact of the sludge concentration on oxygen transfer are the mixed liquor (volatile) suspended solids concentration (MLSS, MLVSS) (Kayser 1967; Wolfbauer 1977; Günder 1999; Krause 2005; Germain *et al.* 2007; Henkel 2010; Capodici *et al.* 2019; Kim *et al.* 2020) or the apparent viscosity of the slurry (Krampe 2001; Rosenberger 2003; Krause 2005; Durán *et al.* 2016; Campbell *et al.* 2019; Ali *et al.* 2022). However, these parameters lack describing that activated sludge is a three-phase mixture, consisting of flocs (gel/solid phase), free available water (liquid phase) and air bubbles (gaseous phase). The sensors that measure the oxygen concentration in activated sludge measure the oxygen concentration in the liquid phase whose viscosity does not change. It is the number of flocs that increases with increasing sludge concentration and it is the floc that interacts with the bubble and the impurities of the water.

Henkel (2010) developed a method called hydrostatic floc volume (HFV) to approximate the free water content/liquid holdup and the floc volume/solid holdup in activated sludge and iron hydroxide slurries (see Materials and Methods). By comparing oxygen transfer experiments using different concentrations of activated sludge flocs and iron hydroxide flocs, it was shown that both slurries follow the same pattern of oxygen transfer depletion if the results were compared against the HFV, independently of whether coarse bubble or fine bubble aeration systems were used. These results could not be explained by the common theory that the apparent viscosity is triggering oxygen transfer depletion.

This study tested the effect of a novel rotating membrane bioreactor (MBR) device and the impact of different rotational speeds on oxygen transfer rate at various sludge concentrations. It is known that an additional agitation device typically improves oxygen transfer in slurry systems compared to systems without agitation (Kubsad *et al.* 2004; Barrera-Cortés *et al.* 2006; Di Palma & Verdone 2009; Mahdariza *et al.* 2022). Fine and coarse bubble aeration devices were tested because both are also used in practice for crossflow aeration in traditional MBR systems.

The experiment series also allowed us to recheck the impact of the solid holdup/floc volume and apparent viscosity on oxygen transfer in activated sludge. Following the current theory of the impact of apparent viscosity, it was expected that with increasing rotational speed the α-factor would improve and that coarse bubble aeration would show higher α-factors compared to fine bubble aeration.

2. MATERIAL AND METHODS

2.1. Oxygen mass transfer kinetics in clean water and activated sludge

If air is released to the water it will turn into air bubbles, and then all gases present in the air bubble will start moving from the gas to the liquid phase, driven by the Brownian motion, until they reach a state of equilibrium and become dissolved in water. The basic theory used for the calculation of oxygen transfer into water is the Two-film Theory from Lewis & Whitman (1924), which states that the transfer rate can be expressed in terms of an overall transfer coefficient and resistances on either side of the interface. Only if the liquid phase is completely mixed and any concentration gradient in the liquid phase is eliminated by turbulence, the basic equation is as follows:

$$\frac{dc(t)}{dt} = k_{\mathrm{L}}a \cdot (c_{\mathrm{L}}^{*} - c(t)) \tag{1}$$

with $dc(t)/dt$ representing the increase of oxygen concentration in respect of time (g/(m^3·h)); $k_{\mathrm{L}}a$ representing the oxygen transfer coefficient (h^{-1}); $c_{\mathrm{L}}*$ representing the oxygen saturation concentration (g/m^3); $c(t)$ representing the oxygen concentration at time t (g/m^3).

However, when reactive particles/flocs are present, the standard diffusion equation needs to be adapted. The overall volume of the solids reduces the volume of the liquid phase and thus cannot be neglected. According to Sun & Furusaki (1989), the standard equation changes into:

$$\frac{dc(t)}{dt} = \frac{k_{\mathrm{L}}a}{\varepsilon_{\mathrm{L}}} \cdot (c_{\mathrm{L}}^{*} - c(t)) - r \tag{2}$$

and

$$\varepsilon_{\mathrm{L}} + \varepsilon_{\mathrm{P}} = 1 \tag{3}$$

with ε_{L} representing the liquid holdup (–); ε_{P} representing the solid/floc holdup (–); r representing the reaction term (mol/(L·s)).

A consequence of this equation is that at a theoretical solid/floc holdup of 1, the $k_{\mathrm{L}}a$ is 0.

To describe the practically observed differences in clean water and wastewater oxygen transfer studies the α-factor was introduced (Kayser 1967):

$$\alpha\text{ - }factor = \frac{k_{\mathrm{L}}a_{\mathrm{wastewater}}}{k_{\mathrm{L}}a_{\mathrm{clean\ water}}} \tag{4}$$

The α-factor is used to estimate the required standard oxygen transfer rate (SOTR), which is one of the key parameters in wastewater engineering.

2.2. Experimental setup

The experiments used the same membrane module from previous experiments (Mahdariza *et al.* 2022), which consists of 1,950 hollow fiber (HF) membranes with one side potted horizontally into the permeate tube and sealed at the other side

(Figure 1). The material of the HF fiber is polyvinylidene difluoride (PVDF) with a pore size of 0.3 μm. The total surface area of the membrane module is 2.25 m^2, which leads to a packing density of 81 m^2/m^3. Due to the horizontal membrane alignment and the applied rotation, all membranes are in contact with the air bubbles even though only one segment of the membrane prototype is aerated.

The measurements were carried out in a transparent tank filled with clean water (softened tap water) and activated sludge with a volume of 1 m^3. Softened drinking water was chosen to prevent any hardness precipitation during the experiments and water storage. The 3–5 g/L MLSS concentration activated sludge (AS I) was obtained from the WWTP in Königsbach, Germany (55,000 PE). Meanwhile, the activated sludge with 7–9 g/L MLSS (AS II) and 11–13 g/L MLSS (AS III) was obtained from the WWTP in Kassel, Germany (340,000 PE). The sludge thickening process was completed at the treatment facility before being transported to the pilot plant for the oxygen transfer experiments. The experiments were performed in batch mode, which means no feed or filtrate was added nor taken during oxygen transfer measurements. A Flexnorm 500 diffuser (OTT System GmbH & Co. KG, Langenhagen, Germany) was used as a fine bubble air diffuser, while a self-made coarse bubble air diffuser using a PVC pipe was used as a coarse bubble aerator with two 3-mm holes. The airflow rates and dissolved oxygen (DO) concentrations were measured with a thermal flow sensor TA Di 21.6 GE (Hoentzsch GmbH, Waiblingen-Hegnach, Germany) and three oxygen electrodes with 10–15 s response time (PRONOVA Analysentechnik GmbH & Co., Bad Klosterlausnitz Germany), respectively. In addition, the ambient parameters, i.e., air temperature, pressure and partial humidity, were documented through a weather station. Figure 2 shows the setup of the pilot plant.

2.3. Sludge thickening

Since the MLSS concentration of the raw activated sludge from WWTP Kassel varied from 2 to 4 g/L, sludge thickening processes were conducted in advance for AS II and AS III experiments. The raw activated sludge was filled into two containers and sedimented for 1 h. Afterwards, the 60–80% supernatant was taken out and the sedimented sludge from both tanks was collected into one tank. The process was repeated until the targeted amount of 1 m^3 thickened sludge was obtained. However, due to the variation of raw sludge for different collecting times in addition to the presence of some floating sludge as a result

Figure 1 | Rotatable HF membrane module.

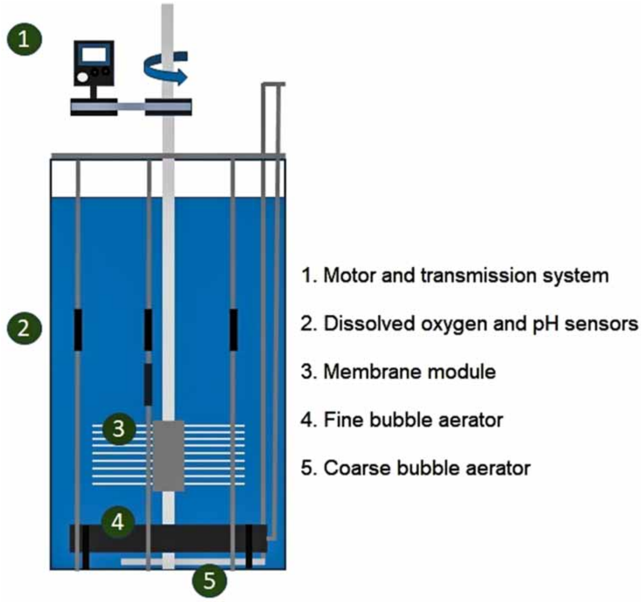


Figure 2 | Setup of the pilot plant of a rotatable membrane prototype system (Mahdariza *et al.* 2022).

of the respiration process during sedimentation, the obtained thickened sludge MLSS concentration had some small variation over all experimental weeks.

2.4. Sludge characteristics

HFV measurements were conducted before and after the experiment. A 1 L sample of the activated sludge was transferred into a 1 L measuring cylinder. Different to the sludge volume index (SVI) measurement developed by Dick & Vesilind (1969) which measures the settled flocs after 30 min, the sample was left until the settled floc volume remained constant.

In addition to HFV, the MLSS, MLVSS, soluble chemical oxygen demand (sCOD), temperature and conductivity were also measured. Table 1 shows an example of measured sludge characteristics and HFV values during 1 week of coarse bubble experiments on 11–13 g/L MLSS sludge for 30 and 50 rpm. As shown, the values were relatively stable during the week of experiments. Still, a trend in decreasing MLSS, MLVSS and HFV is recognizable with increasing days.

It is worth noting that each experimental week did not have an identical initial MLSS value, since the MLSS concentration of raw wastewater which was thickened did not have the same value as well. During all activated sludge experiments, the prepared activated sludge was aerated for at least 12 h in order to maintain constant respiration rates and to nullify the potential impact of impurities on oxygen transfer. Therefore, the results mainly reflect the impact of activated sludge on oxygen transfer depletion. Furthermore, Campbell *et al.* (2020) highlighted the effect of filamentous organisms on oxygen transfer

Table 1 | Sludge characteristics during one experimental week with 11–15 g/L MLSS concentration

Experiment day	Sampling time	MLSS (g/L)	MLVSS (g/L)	HFV (mL/L)	Conductivity (μS/cm)	sCOD (mg/L)	Temp. (°C)
1	Morning	12.00	7.98	380	1,040	77.0	9.0
	Afternoon	12.10	8.01	360	1,068		11.3
2	Morning	11.80	7.73	370	1,112	86.8	9.3
	Afternoon	11.90	7.88	370	1,135		10.2
3	Morning	11.40	7.39	340	1,204	88.5	9.4
	Afternoon	11.40	7.49	340	1,263		10.9
4	Morning	11.20	7.27	350	1,340	90.7	9.6
	Afternoon	11.30	7.37	340	1,383		10.5

efficiency. However, the SVI values for sludge experiments in this study were below 150 mL/g, which means that the presence of filamentous organisms is limited, hence, it can be assumed that the distortion in oxygen transfer due to filamentous organisms is neglectable. Some key properties of AS I, AS II and AS III are listed in Table 2.

2.5. Oxygen transfer measurement and calculation

In this study, the desorption method (Wagner *et al.* 1998; DWA-M 209 2007) using pure oxygen was selected for calculating the k_La value to guarantee comparable results to previously conducted fine bubble experiments on clean water and 3–5 g/L MLSS concentration of activated sludge (Mahdariza *et al.* 2022). Three different rotational speeds (0, 30, 50 rpm) were tested at three different airflow rates (1, 2, 4 m^3/h). However, for coarse bubble experiments at 7–9 g/L (AS II) and 11–13 g/L (AS III) MLSS concentration, additional experiments with an airflow rate of 5 m^3/h were conducted, because at an airflow rate of 1 m^3/h, not sufficient oxygen could be transferred to satisfy oxygen consumption caused by endogenous respiration.

The oxygen concentration was recorded by three oxygen sensors during the experiment at a constant airflow rate and constant membrane module rotational speed. In order to fulfill the requirement according to the guideline from DWA-M 209 (2007), during all experiments, the airflow rate was maintained to have fluctuation less than ± 10% and the temperature difference between the beginning and the end of the experiment did not exceed 2 °C. The decrease in recorded oxygen concentration was then determined by non-linear regression to produce an average k_La value from all three sensors. The obtained k_La value was normalized to the standard conditions (k_La_{20}) at a water temperature of 20 °C and an atmospheric pressure of 1,013 hPa, and with the correction factor for a salt concentration of 1 g/L, due to significant differences between the salt content of clean water and wastewater (DWA-M 209 2007).

This procedure was performed in tap water and activated sludge with three different MLSS concentrations. Afterwards, the polynomial trend lines of calculated k_La_{20} values from different specific airflow rates for each membrane module rotational

Table 2 | Sludge characteristics during all experimental weeks

		Tap water	AS I	AS II	AS III
Fine bubble					
MLSS	g/L	–	3.9 (± 0.3)	8.9 (± 0.9)	12.4 (± 0.8)
MLVSS	g/L	–	2.5 (± 0.1)	5.7 (± 0.7)	8.2 (± 0.5)
sCOD	mg/L	–	–	44 (± 2)	44 (± 5)
Conductivity	µS/cm	811 (±0)	1,481 (±126)	1,170 (±80)	852 (± 213)
pH	–	6.9 (±0.0)	6.9 (± 0.0)	7.4 (± 0.2)	7.3 (± 0.2)
Temperature	°C	17.7 (±0.4)	15.6 (± 0.9)	14.3 (± 1.6)	10.2 (± 0.6)
Loss on ignition	%	–	35 (± 2)	36 (± 1)	34 (± 0)
SVI	mL/g	–	–	78 (± 3)	92 (± 4)
HFV	mL/L	–	167 (± 13)	268 (± 18)	354 (± 13)
Endogenous respiration	mgO_2/ (gMLVSS·h)	–	–	1.6 (± 0.1)	2.6 (± 0.4)
Coarse bubble					
MLSS	g/L	–	3.7 (± 0.1)	8.0 (± 0.3)	11.5 (± 0.4)
MLVSS	g/L	–	2.6 (± 0.1)	5.0 (± 0.2)	7.6 (± 0.3)
sCOD	mg/L	–	–-	–	74 (± 19)
Conductivity	µS/cm	811 (±0)	1,392 (±293)	1,003 (±142)	1,277 (±79)
pH	-	6.9 (±0.0)	6.9 (± 0.0)	6.3 (± 0.5)	6.6 (± 0.3)
Temperature	°C	18.0 (±0.5)	14.4 (± 2.5)	19.2 (± 1.2)	9.6 (± 0.9)
Loss on ignition	%	–	32 (± 1)	37 (± 1)	34 (± 1)
SVI	mL/g	–	–	82 (± 2)	83 (± 7)
HFV	mL/L	–	179 (± 15)	265 (± 10)	347 (± 10)
Endogenous respiration	mgO_2/(gMLVSS·h)	–	–	2.2 (± 0.5)	2.0 (± 0.2)

speed were generated. Finally, the comparison between the k_La_{20} value of activated sludge and clean water at a specific airflow rate defined as the α-factor was calculated by dividing the trendline equation at a certain sludge concentration by the equation obtained during the clean water experiment.

3. RESULTS AND DISCUSSION

Four experiment series (clean water, AS I, AS II and AS III) were performed with coarse and fine bubble diffusers at different sludge concentrations to investigate the impact of rotation and solid/floc holdup on oxygen transfer. Sampling was executed after the sludge was aerated overnight, before and after the experiments, which also ensured that all sludges had the same conditions of endogenous respiration. In Tables 3 and 4 the results of oxygen transfer experiments are summarized. The table also incorporates the results of Mahdariza *et al.* (2022).

3.1. Impact of rotation and airflow rate on oxygen transfer

Generally, coarse bubble experiments show lower oxygen transfer rates at the same specific airflow rate and sludge concentration compared to fine bubble experiments. The bigger bubble size of coarse bubble aeration (16–20 mm) compared to fine bubble aeration (2–3 mm) leads to a lower interfacial area and therefore lower oxygen transfer rates at the same specific airflow rate.

All results have in common that with increasing airflow rate and increasing rotational speed the k_La value increases, except for the experiment with coarse bubble aeration at 30 rpm and 12 g/L MLSS concentration at a specific airflow rate of 5 $m_N^3/(m^3{\cdot}h)$. The exception for coarse bubble aeration can be explained by the higher solid holdup/floc volume during the transfer experiments with rotation compared to no rotation, which has an additional negative effect on oxygen transfer.

The biggest improvement was observed for coarse bubble aeration at a rotational speed of 50 rpm and a specific airflow rate of 2 $m_N^3/(m^3{\cdot}h)$. The achieved oxygen transfer coefficients are nearly as high as for the fine bubble experiments at the same airflow rate and rotational speed. This can be explained by the impact of the membrane module. For fine bubble aeration, the rotation at 50 rpm caused such a high circular fluid flow force to the bubbles that they were bypassing the membrane fibers (Mahdariza *et al.* 2022). However, coarse bubble formation and rising behavior are governed by the liquid inertia and gas momentum forces and are only little impacted by the fluid flow forces. Consequently, the coarse bubbles were still rising straight up to the membrane module. Once the bubbles hit the rotating fibers they disintegrated, forming fine bubbles and now the fluid force evenly distributed these bubbles in the reactor. This effect decreased with increasing airflow rate for

Table 3 | Coarse bubble experiment results

				Specific airflow 2 ($m_N^3/(m^3{\cdot}h)$)			Specific airflow 4 ($m_N^3/(m^3{\cdot}h)$)			Specific airflow 5 ($m_N^3/(m^3{\cdot}h)$)		
Experiment	**MLSS (g/L)**	**MLVSS (g/L)**	**HFV (mL/L)**	**k_La (1/h)**	**Imp.[a] (%)**	**α (–)**	**k_La (1/h)**	**Imp. (%)**	**α (–)**	**k_La (1/h)**	**Imp. (%)**	**α (–)**
0 rpm Tap				2.64			5.23			6.52		
30 rpm Tap				3.39	+28		5.96	+14		6.94	+06	
50 rpm Tap				6.10	+131		9.16	+75		9.56	+47	
0 rpm AS I	3.70	2.50	155	2.28		0.87	4.31		0.82	5.23		0.80
30 rpm AS I	3.87	2.60	193	2.71	+19	0.80	4.88	+13	0.82	5.78	+10	0.83
50 rpm AS I	3.63	2.53	178	4.44	+94	0.73	6.83	+58	0.75	7.27	+39	0.76
0 rpm AS II	8.14	5.19	275	1.87		0.71	3.97		0.76	5.10		0.78
30 rpm AS II	8.07	5.07	265	2.33	+24	0.69	4.38	+10	0.74	5.30	+04	0.76
50 rpm AS II	7.71	4.85	254	3.95	+111	0.65	6.14	+55	0.67	6.57	+29	0.69
0 rpm AS III	11.30	7.36	340	1.41		0.54	3.48		0.67	4.76		0.73
30 rpm AS III	12.10	8.01	360	1.88	+33	0.55	3.76	+08	0.63	4.70	−1	0.68
50 rpm AS III	11.33	7.42	343	3.11	+120	0.51	5.32	+53	0.58	6.10	+28	0.64

[a]Improvement compared to 0 rpm.

Table 4 | Fine bubble experiment results

Experiment	MLSS (g/L)	MLVSS (g/L)	HFV (mL/L)	Specific airflow 1 ($m^3_N/(m^3 \cdot h)$) k_La (1/h)	Imp.[a](%)	α (–)	Specific airflow 2 ($m^3_N/(m^3 \cdot h)$) k_La (1/h)	Imp. (%)	α (–)	Specific airflow 4 ($m^3_N/(m^3 \cdot h)$) k_La (1/h)	Imp. (%)	α (–)
0 rpm Tap				4.12			7.36			11.17		
30 rpm Tap				5.25	+27		9.40	+28		14.39	+29	
50 rpm Tap				6.52	+58		11.61	+58		17.50	+57	
0 rpm AS I	3.60	2.40	175	3.20		0.78	5.92		0.80	9.92		0.89
30 rpm AS I	4.30	2.70	175	4.23	+32	0.80	7.53	+27	0.80	11.38	+15	0.79
50 rpm AS I	3.90	2.50	150	4.92	+54	0.76	9.05	+53	0.78	14.93	+50	0.85
0 rpm AS II	9.20	6.23	274	2.71		0.66	4.85		0.66	7.48		0.67
30 rpm AS II	8.47	5.38	275	3.43	+27	0.65	6.07	+25	0.65	9.00	+20	0.63
50 rpm AS II	8.61	5.43	255	4.14	+53	0.64	7.37	+52	0.63	11.06	+48	0.63
0 rpm AS III	13.27	8.82	367	2.35		0.57	4.12		0.56	5.95		0.53
30 rpm AS III	11.87	7.84	350	3.40	+45	0.65	5.99	+45	0.64	8.73	+47	0.61
50 rpm AS III	12.10	8.05	345	3.88	+66	0.60	6.97	+69	0.60	10.76	+81	0.61

[a]Improvement compared to 0 rpm.

coarse bubble aeration because at the higher gas holdup and heterogeneous flow regime, a portion of the fine bubbles again coalesced and formed larger bubbles.

The even increase in oxygen transfer rates for fine bubble experiments with increased rotational speed at a specific airflow rate can mainly be explained by smaller bubble formation at the orifice due to increased liquid flow forces and the change in flow pattern from straight upwards to more circular caused by the rotation of the module. Both effects increased the gas holdup and consequently lead to a steady increase in oxygen transfer at the chosen airflow rates.

3.2. Impact of rotation and solid/floc hold up on oxygen transfer

The effect of increased airflow rate and rotational speed had only little effect on the α-factor at comparable floc volumes and sludge concentrations (see Tables 3 and 4).

The results indicate that the α-factor from all applied airflow rates for each MLSS concentration and membrane rotational speed presented in Figures 3 and 4 follows the same pattern no matter which rotational speed or which aeration system is used. Neither for fine bubble aeration nor for coarse bubble aeration, it could be observed that an increase in rotational speed improved the α-factor. All measurements show quite similar values. Only for coarse bubble aeration at a rotational speed of 50 rpm, a clear decrease in α-factor is measured.

This result is contrary to most of the current literature where oxygen transfer depression with increasing sludge concentration is mainly explained by the effect of apparent viscosity on activated sludge (Krampe & Krauth 2003; Durán *et al.* 2016; Campbell *et al.* 2019). Accordingly, the non-Newtonian pseudoplastic fluid properties of activated sludge should have caused an increase in α-factor with increasing rotational speed and higher α-factors should have been observed for coarse bubble aeration. The results of Figures 3 and 4 contradict this theory.

A similar conclusion was drawn by Henkel *et al.* (2011) when comparing fine bubble and coarse bubble aeration systems using iron hydroxide flocs and activated sludge flocs. Again, the non-Newtonian pseudoplastic fluid properties of the activated sludge should have theoretically caused higher α-factors for coarse bubble aeration due to the shear-thinning effect. No significant difference in the α-factor could be determined by Henkel *et al.* (2011) if the solid holdup/floc volume (HFV) was used to correlate the results.

Based on the results of this study and Henkel *et al.* (2011), activated sludge flocs are behaving similarly to solid particles on oxygen transfer. Studying activated sludge under the microscope shows that the sludge floc creates its own cluster and clearly separates from the free water content (Mesquita *et al.* 2013; Campbell 2020). This is corroborated by the structure of granular activated sludge flocs with spherical solids/particles.

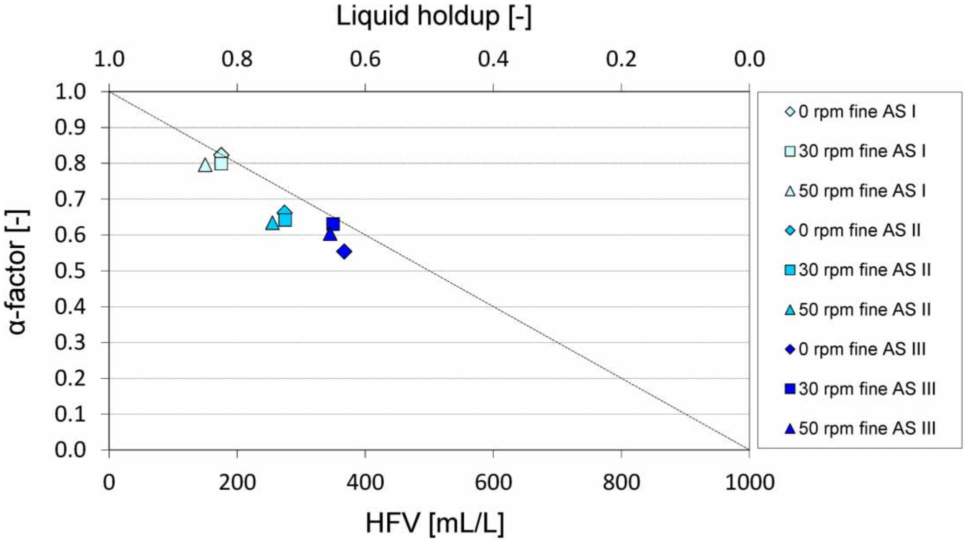


Figure 3 | Average α-factors for fine bubble aeration at various rotational speeds under endogenous conditions.

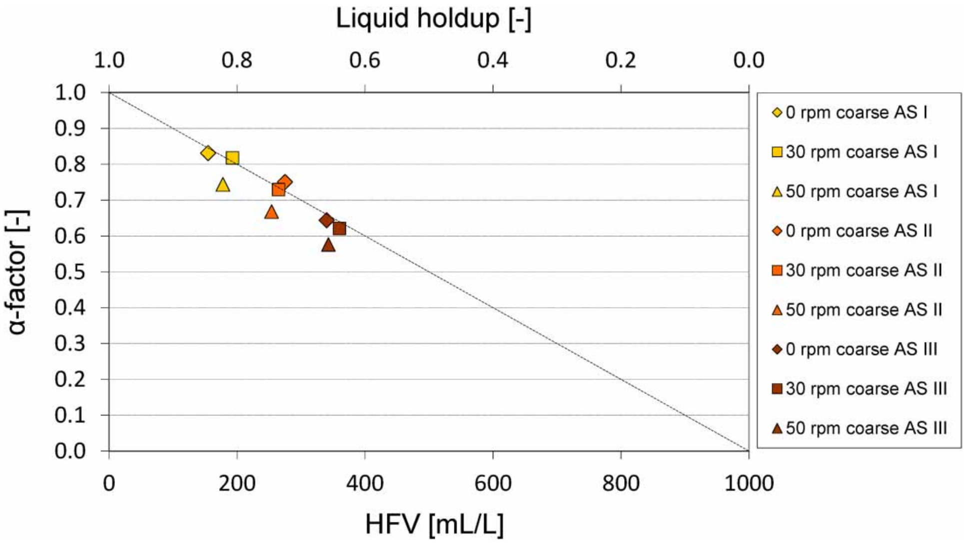


Figure 4 | Average α-factors for coarse bubble aeration at various rotational speeds under endogenous conditions.

By using Equation (2) and correcting the oxygen transfer results in these experiments by the reduced liquid holdup, which can be estimated by using the HFV, the corrected α-factors are in the range of 0.9–1.0. The results are compared to obtained α-factors from Equation (4), as shown in Table 5.

Consequently, oxygen transfer coefficients into the liquid phase achieved during stabilized activated sludge experiments are comparable to the clean water experiments. This is supported by experiments from Henkel (2010), Kayser (1967) and Steinmetz (1996), who measured and investigated the impact of the pure liquid phase on the α-factor of activated sludge plants without deriving a significant impact of the wastewater effluent or MBR filtrate.

In summary, despite the usage of different reactor geometries, different aeration devices (fine bubble disk aerator, fine bubble tube aerator, coarse bubble two aeration holes, coarse bubble 10 aeration holes) and different slurries (MBR sludge, thickened activated sludge from different plants, iron hydroxide slurry), the α-factor decreases in the same pattern with decreasing liquid holdup and increasing floc/solid holdup by using HFV as reference value (Figure 5).

Table 5 | The comparison of obtained α-factors from two different equations

	AS I			AS II			AS III		
***α*-factor fine bubble**									
rpm	0	30	50	0	30	50	0	30	50
$(k_La_{waste}/k_La_{clean})$	0.82	0.80	0.80	0.66	0.64	0.63	0.55	0.63	0.60
solid/floc holdup ε_P	0.18	0.18	0.15	0.27	0.28	0.26	0.37	0.35	0.35
$(k_La_{waste}/((1-\varepsilon_P)\ k_La_{clean}))$	1.00	0.97	0.94	0.91	0.88	0.85	0.88	0.97	0.92
***α*-factor coarse bubble**									
rpm	0	30	50	0	30	50	0	30	50
$(k_La_{waste}/k_La_{clean})$	0.83	0.82	0.74	0.75	0.73	0.67	0.64	0.62	0.58
Solid/floc holdup ε_P	0.16	0.19	0.18	0.28	0.27	0.25	0.34	0.36	0.34
$(k_La_{waste}/((1-\varepsilon_P)\ k_La_{clean}))$	0.98	1.01	0.91	1.04	0.99	0.90	0.98	0.97	0.88

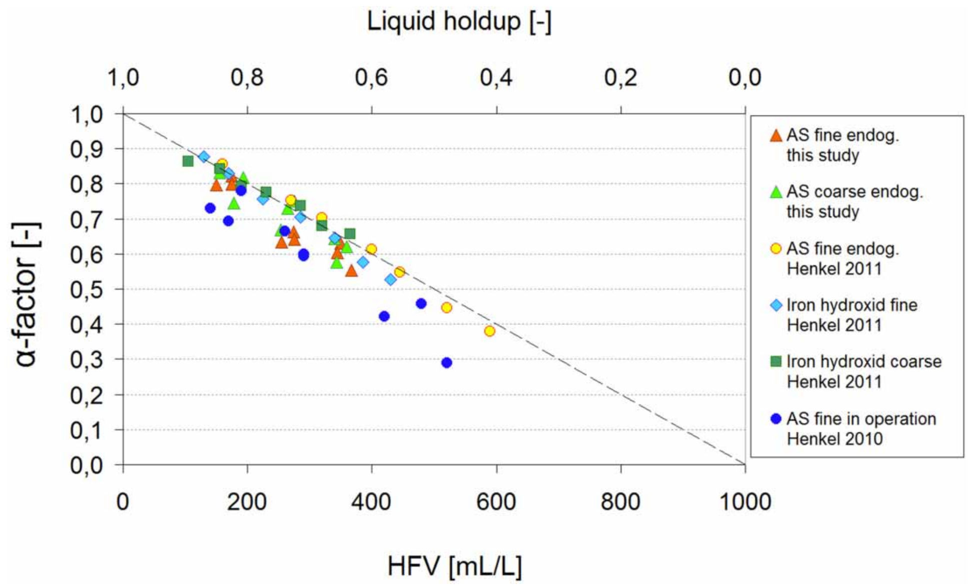


Figure 5 | Summary of HFV experiments with fine and coarse bubble aeration under endogenous conditions and in operation.

To the best of the authors' knowledge, a consistent survey of these influencing factors on oxygen transfer has not been published yet. Due to the fact that the suspended solids concentration (MLSS) is still used as the main parameter to compare oxygen transfer results, the impact of activated sludge flocs on oxygen transfer is until now not included in the majority of the studies. Several authors aimed to disclose the impact of activated sludge flocs by running experiments with different materials, e.g., aluminum hydroxide, activated carbon, peat and bentonite clay, using similar suspended solid concentrations and comparing the oxygen transfer results (van der Kroon 1968; Steinmetz 1996; Henkel 2010; Blanco Zúñiga *et al.* 2021). However, the suspended solids concentration does not reflect the volumetric fraction of the different materials. Iron hydroxide flocs occupy for example a different volume at the same suspended solids concentration compared to activated sludge flocs, as shown in Figure 6.

But also activated sludge taken from a wastewater plant that operates without primary sedimentation shows a different floc volume to suspended solids ratio compared to activated sludge from a wastewater plant with primary sedimentation due to higher content of silt, clay, and sand (Henkel *et al.* 2011). In addition, the sludge retention time (SRT) impacts the floc volume as it influences the organic content of the activated sludge floc. Plants running at higher SRTs typically show a lower loss on ignition of the sludge compared to plants that operate at low SRT (Foladori *et al.* 2010). Finally, a finding from Wu *et al.*

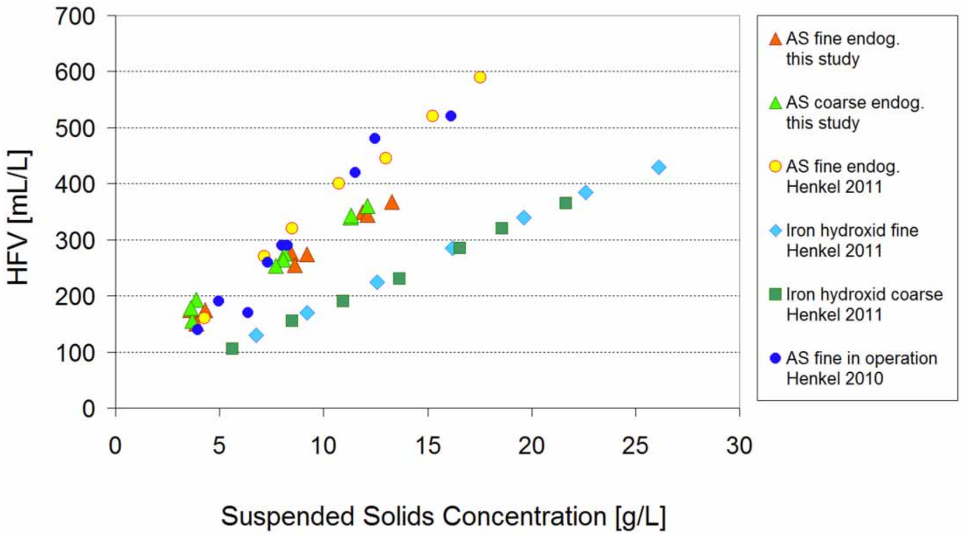


Figure 6 | Suspended solids concentration vs. HFV under endogenous conditions and in operation.

(2021) presented the influence of floc size and circularity on oxygen uptake rate, which is supported by a study by Burger *et al.* (2017) who showed that filamentous bacteria influence floc morphology, impacting oxygen transfer but also the free available water content and solid/floc holdup. These diverse impact factors on floc volume are not reflected by the MLSS concentration and thus apparently different α-factors were obtained in the past by using only MLSS concentration as the reference value.

3.3. Practical implications

The still-existing lack of a common understanding of which parameters rule oxygen transfer in activated sludge is mainly caused by the fact that important parameters like the impact of the flocs (MLSS, MLVSS, HFV) and the impact of impurities (surfactants, polymers, adsorbed organics) are discussed independently although they are interconnected, e.g., by Maximilian Schwarz *et al.* (2021).

The floc volume (solid holdup) is directly linked to the mass of sludge in the system and because of this, it also governs parameters like the food-to-mass ratio (F/M) or the SRT and it is responsible for the amount of adsorbed organic matter to the floc and the dissolved impurities in the sludge, which can additionally impact oxygen transfer (Gillot & Héduit 2008; Rosso *et al.* 2008; Schwarz *et al.* 2021; Bencsik *et al.* 2022).

Acknowledging these interdependencies, the worst oxygen transfer conditions occur where a high load of impurities that influence oxygen transfer (high F/M ratio, low SRT) and high floc volumes (solid holdup, total suspended solids (TSS) concentration) jointly appear (Schwarz *et al.* 2023). This is for example the case for sequencing batch reactors that do not use additional agitation. Just after the sedimentation phase when aeration is used to expand the settled sludge bed (high floc concentration) and still the amount of adsorbed organic to the floc is high (high F/M ratio), the lowest α-factors are observed. This has been confirmed by Cecconi *et al.* (2020) and Strubbe *et al.* (2023), who reported α-factor values as low as 0.2 in such applications. This is even lower than the typically measured α-factor of 0.3–0.4 in the raw wastewater influent (Kayser 1967; Henkel 2010) or 0.40–0.45 in the activated sludge plants running at an SRT of 2.0 (Kroiss & Klager 2018; Schwarz *et al.* 2021). On the contrary, aerobic stabilization plants, which use low TSS concentrations (low floc volume) and low F/M ratio (high SRT) specifically at the end of the aeration basin, show α-factors as high as 0.85 (Gillot & Héduit 2008; Schwarz *et al.* 2021), as long as no filamentous bacteria occur (Campbell & Wang 2020).

4. CONCLUSIONS

Oxygen transfer experiments were conducted with a new rotating type of HF membrane module using fine and coarse bubble aeration with different airflow rates and membrane rotational speeds.

- For both fine and coarse bubble experiments oxygen transfer coefficients rise with increasing rotational speed of the membrane at the same solid/floc holdup and sludge concentration.
- The improvement of oxygen transfer rate at 30 rpm is on average higher for fine bubble aeration (25%) compared to coarse bubble aeration (10%). At 50 rpm, the highest improvement rate could be observed for coarse bubble aeration at the lowest airflow rate tested (100%). However, with increasing airflow rate, this improvement rate decreases again significantly for coarse bubble aeration while for fine bubble aeration, it stays nearly constant.
- Despite the very distinct impact of rotation and airflow rate on oxygen transfer in activated sludge, the α-factors showed quite similar values for both fine and coarse bubble aeration at comparable sludge concentrations and solid/floc holdups.
- The solid holdup or liquid holdup has so far not been considered in the calculations of the α-factor to describe the impact of the activated sludge floc on oxygen transfer in wastewater engineering. However, the results in this study and previous studies indicate the need to do so, as it appears to universally impact oxygen transfer no matter what reactor type (bubble column, airlift reactor), diffuser setup (disk aerator, tube aerator, fine bubble, coarse bubble) and rotational speed (30 rpm, 50 rpm) was used in the wastewater experiments. Practically, the individual solid/floc holdup can be correlated to the MLSS concentration of each wastewater treatment plant (WWTP).
- The study could not confirm that coarse bubble aeration compared to fine bubble aeration systems generally create higher α-factors and that the α-factor generally increases with increasing turbulence (Stenstrom & Gilbert 1981). Consequently, these statements cannot be generalized for the impact of the solid holdup and the liquid holdup in floc suspensions.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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