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## **Analysis of the Gas Holdup Evolution in a Circulating Jet-Loop Nitrifying MBR**

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# Analysis of the Gas Holdup Evolution in a Circulating Jet-Loop Nitrifying MBR\*

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## Abstract

This paper presents an analysis of the gas holdup evolution in a novel type of jet-loop membrane bioreactor (JLMBR), designed for nitrogen removal through the nitrite route application. Its configuration is inspired from airlift systems. It consists of a 60-l reactor made of an internal airlift system coupled to an external liquid recirculation loop. Hollow fiber membranes are submerged in the riser compartment. The process was intermittently fed with a synthetic ammonia solution and the gas holdup evolution was monitored for 500 to 600 days. Experiments were performed using flowrates ranging from 0.4 to 1.03 Nm<sup>3</sup>/h, and from 0 to 0.6 m<sup>3</sup>/h for air and water, respectively. This corresponded to superficial velocities from 0.004 to 0.03 m.s<sup>-1</sup> for air and 0 to 0.011 m.s<sup>-1</sup> for water. The gas holdup  $\epsilon_g$  was directly measured by the volume expansion method, using a tubular level meter located on the plant. The reported results showed that, in the absence of microorganisms,  $\epsilon_g$  ranged between 0.5 and 5.5% for the investigated range of gas liquid superficial velocities, whilst increasing from 0.5 to 4.8% only in the presence of gas (no liquid recirculation). This double influence of the air and the liquid velocities on the gas holdup was described by a multilinear correlation.

However in the presence of biosolids in the reactor, the gas holdup raised up to 6.5%, corresponding to an increase of ca. 48% (in average, with respect to data recorded on day 0). This increase in  $\epsilon_g$  was attributed to both a gas entrainment effect and an impact of the bioparticles recirculated into the reactor. Under experimental conditions investigated, the gas holdup increased linearly with the air and the liquid velocities, what corresponded to the bubbly flow regime in the system.

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This showed that, according to investigated conditions, the impact of circulated biomass was not enough to change the bubble gas flow regime.

**KEYWORDS:** aeration, gas holdup, hydrodynamics, recirculation, membrane bioreactors

## 1. Introduction

In recent years, membrane technology has increasingly been applied in conventional wastewater treatment facilities. This is in part due to improved performance of membrane processes, a large variety of membrane manufacturers throughout the world, and the decrease in membrane prices. In the context of waste water treatment and particularly for the removal of ammonia loads, the so-called nitrite route is one of the most interesting pathways to treat ammonia discharges. The main advantages of this route versus the complete oxidation of ammonia are energy savings up to 25% due to lower aeration rates, and a low consumption of organic substrate during the subsequent heterotrophic denitrification step. There are many works reported in the literature which describe a process improvement based on a reactor design modification (i.e.: Lazarova et al., 1997; Masoud et al., 2001; Nordkvist et al., 2003). However, the main problem related to these varieties of configurations resides in their transposition from an investigation to other works. As a result, many models describing the same phenomena (i.e.: mass transfer, gas holdup, etc.) are reported. Particularly, the gas holdup is an interesting process parameter that characterizes the gas flow pattern and the way the gas is distributed within the reactor. Operating two-phase systems into a circulated-liquid rectangular tank, Lazarova et al. (1997) observed that the gas holdup was affected by both the system hydrodynamics and the reactor geometry. They correlated this parameter as a power law function of the gas superficial velocity and the static liquid height. Furthermore, while  $\epsilon_g$  is intensively studied in simple bubble columns and airlift reactors (e.g., Chisti, 1989; Petersen and Margaritis, 2001), little is known about the gas holdup in these systems when membranes are integrated to keep microorganisms for activated sludge operations.

The aim of this paper is to investigate the evolution of the gas holdup in a three-phase circulated liquid jet-loop system designed to control the partial nitrification process.

## 2 Material and methods

### 2.1 Experimental apparatus

The jet-loop membrane bioreactor (JLMBR) operated in this work has been presented in Figure 1 and also in previous papers (i.e.: Kouakou et al., 2005; Kouakou, 2007). It consists of a 60-l rectangular tank inspired of airlift designs. The main difference between conventional airlift designs and the current design is a forced external liquid recirculation system which allows controlling the mixing

in the reactor, independently from the sparged gas flowrate. Built in Plexiglas, the reactor is composed of two interconnected compartments with equal volumes (the riser and the downcomer), and a total cross-section area of  $0.04 \text{ m}^2$ . The plant was intermittently fed with synthetic ammonia solution. At the top of the tank, the circulated liquid flows from the riser to the downcomer compartment through a square section area ( $\approx 64 \text{ cm}^2$ ). A submerged hollow fiber membrane (Sterapore-L, Mitsubishi;  $S = 1.5 \text{ m}^2$ , micropores:  $0.4 \text{ }\mu\text{m}$ ), located in the riser compartment, was operated at a filtration flux set to  $\sim 8.1 \text{ l.h}^{-1} \cdot \text{m}^{-2}$  corresponding to approximately  $0.4 \text{ kg.N-NH}_4^+ \cdot \text{d}^{-1} \cdot \text{m}^{-2}$  membrane. This membrane allowed maintaining nitrifying microorganisms within the reactor, avoiding their washout. At the bottom of the system, air and recirculated water were supplied through 2 separated tubular distributors built in plastic material. Both of them (distributors) are perforated by 46 circular orifices having each 1mm of diameter. Water was forced to circulate continuously while air was supplied intermittently in order to maintain the dissolved oxygen (DO) concentration at a desired level. In the riser compartment, air and water flowed up tangentially to the membrane, allowing cleaning of its surface. Air flowrate ( $0.4 - 1.03 \text{ Nm}^3/\text{h}$ ) and water flowrate ( $0 - 0.6 \text{ m}^3/\text{h}$ ), corresponding to superficial velocities of  $0.004 - 0.03 \text{ m.s}^{-1}$  and  $0 - 0.011 \text{ m.s}^{-1}$ , were controlled using two flowmeters: Brooks Instruments, R6-15B and Krohne, 164010, respectively. The dissolved oxygen (DO) concentration was monitored by a potentiometric oxygen probe (Oxypol, SON-10-17), submerged in the upper interconnection window. The value was kept constant to  $\approx 2 \text{ mg.O}_2 \cdot \text{l}^{-1}$  and the reactor temperature was set approximately constant to  $30^\circ\text{C}$ , corresponding (both DO and temperature) to near optimal conditions for the partial nitrification process (Kouakou, 2007). The gas holdup was directly measured by the volume expansion method, using a tubular level meter located on the plant.

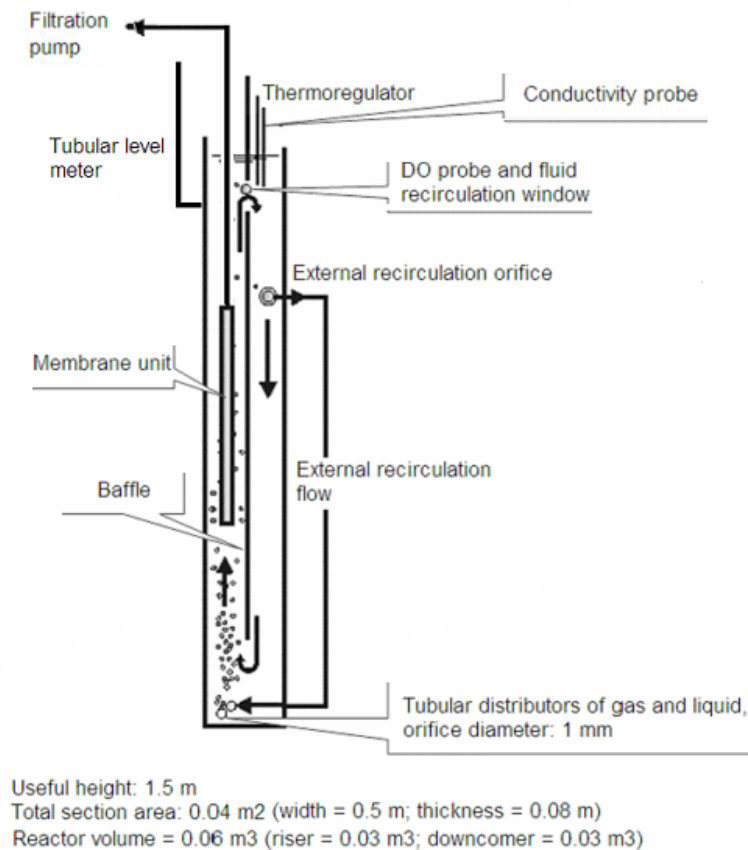


Figure 1: Reactor set-up and dimensions

## 2.2 Measurement methods

In this study, the overall gas holdup ( $\varepsilon_g$ ) was considered and measured by the volume expansion method. Estimates were obtained by comparing the ungasged ( $h$ ) and gassed ( $h_D$ ) liquid levels, as depicted in Eq. 1.

$$\varepsilon_g = \frac{(h_D - h)}{h_D} \quad (1)$$

$\varepsilon_g$  represents the average void fraction within the reactor, i.e., the ratio between the volume occupied by the gas phase and the total volume of the reactor. It can also be considered as representing the cross-sectional fraction, i.e., the ratio

between the area occupied by the gas fraction and the whole cross-sectional area of the reactor.

This method provides an averaged value of the gas holdup over the two compartments. Visual observations showed however that most of the gas holdup was confined in the riser compartment.

### 3 Results and discussion

Our investigations were performed on the gas holdup ( $\varepsilon_g$ ) evolution versus the air superficial velocity, under different external liquid superficial velocities. To better investigate the capability of the process in terms of gas retention, experiments were carried out in real operating conditions. The submerged membrane was kept in the riser compartment of the plant. Although the membrane might influence the retention measurements, this typical impact was not investigated in this study. Under the described experimental conditions (i.e., liquid,  $Q_l = 0 - 0.6 \text{ Nm}^3 \cdot \text{h}^{-1}$ ; air,  $U_g = 0.003 - 0.023 \text{ m} \cdot \text{s}^{-1}$ ),  $\varepsilon_g$  ranged between 0.5 and 4.8% of the total reactor volume, in the absence of water recirculation (bubble column conditions) (see Figure 2). However when water was recirculated,  $\varepsilon_g$  values were approximately 15% larger, corresponding to an increase of the gas holdup from 0.5 to 5.5 %. This increase of  $\varepsilon_g$  values with the liquid velocity has also been reported by Petersen and Margaritis (2001). It could be explained by a gas entrainment effect in the downcomer due to the liquid recirculation between the two interconnected compartments. The experimental data recorded in this work were fitted by a multilinear correlation expressed in (Eq. 2).

$$\varepsilon_g = \kappa U_g (1 + \chi U_l) \quad (2)$$

Estimates of model parameters and statistical uncertainties on model coefficients revealed that  $\kappa = 2.05 \pm 0.05 \text{ s} \cdot \text{m}^{-1}$  and  $\chi = 12.45 \pm 3.25 \text{ s} \cdot \text{m}^{-1}$ . The insertion of these values into the model equation (Eq. 2) led to the parity plot depicted in Figure 3. As shown, the near-perfect distribution of the recorded points around the axial bisector ( $\pm 5\%$  bounds) confirms the validity of the proposed multilinear model.

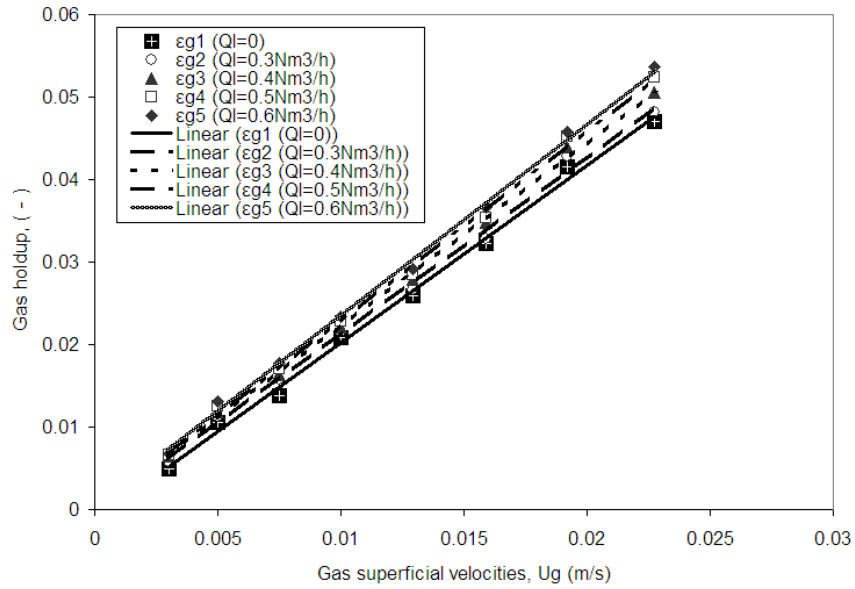


Figure 2: Impact of circulated-liquid on the gas holdup profile at different gas superficial velocities, in the absence of biological solids in the process

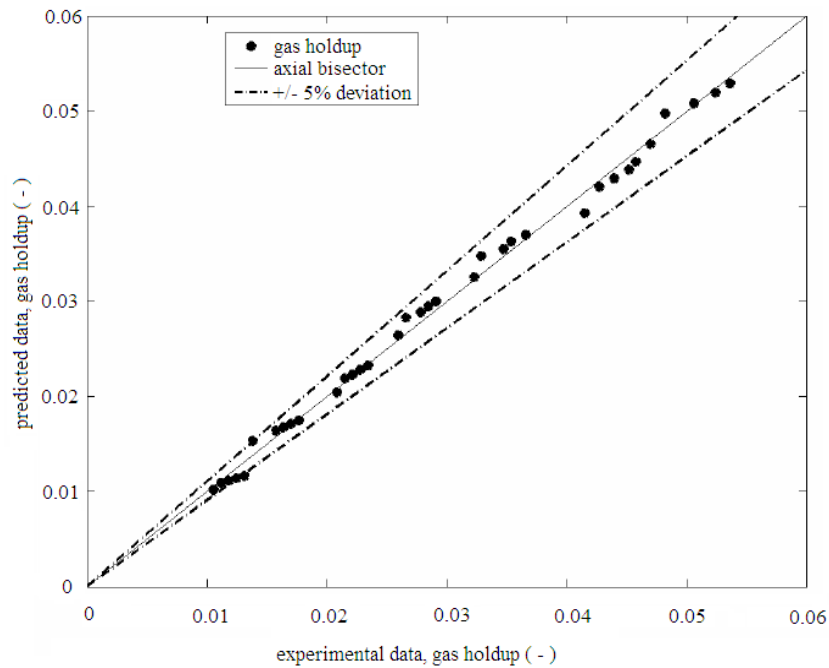


Figure 3: Parity plot of predicted  $\epsilon_g$  vs. experimental data, from the application of model Eq. 2.



The gas holdup was determined in the presence of a nitrifying flora, too. It was measured at different times of operation, under near optimal conditions of the nitrite route (pH = 8.2; DO  $\approx$  2 mg.O<sub>2</sub>.l<sup>-1</sup>; T = 30°C, HRT  $\approx$  6h, cf. Kouakou, 2007). Since low DO concentration was one of the key parameters controlling the process performances, the gas retention measurements were performed at relatively low air flowrates (0.4 – 1.03 Nm<sup>3</sup>/h). The measurement protocol was similar to the protocol adopted in the absence of microorganisms in the system. Experimental results are presented in Figure 4, both in the absence and in the presence of microorganisms.  $\varepsilon_g$  increases linearly with the gas velocity, either in the absence or in the presence of microorganisms. This proportionality between the gas holdup and the air superficial velocity is characteristic of the bubbly flow regime. In Figure 4, it is quite hard to see the evolution of the gas retention with time. Additional data are provided in Table 1. These data show that gas holdup changes are random and statistically not negligible with time. Under 520 days, estimates of gas retention increased up to 48% in average. However, this impact of biomaterials on the gas holdup increase was not enough to change the bubble gas flow regime. One explanation of the variability of  $\varepsilon_g$  could be the non linearity of solids increasing in the process combined with the fluctuation of suspended materials driven by the liquid (i.e., effects of solids settlement and cleaning operations).

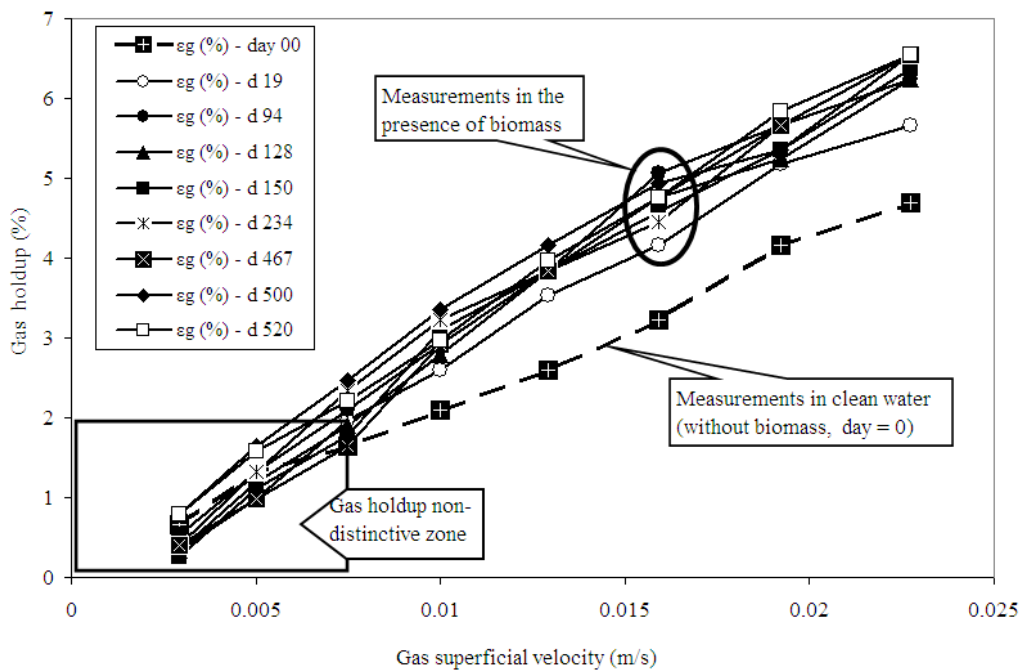


Figure 4: Comparison of gas holdup in clean water to  $\epsilon_g$  evolution with time in process media

Table 1: Changes of  $\epsilon_g$  with time (from day 0 to day 520), in process media and clean water, under circulated-liquid superficial velocities  $U_l$  ( $0 - 0.011 \text{ m.s}^{-1}$ ) and gas superficial velocities  $U_g$  ( $0.003 - 0.023 \text{ m.s}^{-1}$ )

$U_g$	$\epsilon_g$ (%)									Increase d0-520
	d0	d19	d94	d128	d150	d234	d467	d500	d520	
0.003	0.55	0.33	0.53	0.33	0.27	0.40	0.40	0.79	0.79	43 %
0.005	1.32	0.99	1.32	1.19	1.12	1.32	0.99	1.64	1.57	19 %
0.008	1.64	1.96	2.09	1.90	1.77	2.34	1.64	2.47	2.22	35 %
0.010	1.96	2.60	2.91	2.79	3.10	3.23	2.91	3.35	2.98	52 %
0.013	2.28	3.54	3.85	3.85	3.85	3.85	3.85	4.15	3.97	74 %
0.016	2.91	4.15	5.06	4.76	4.58	4.46	4.76	4.94	4.76	63 %
0.019	3.54	5.18	5.66	5.24	5.36	5.66	5.66	5.36	5.84	65 %
0.023	4.76	5.66	6.25	6.25	6.37	6.54	6.53	6.54	6.54	37 %
Average of increase percents estimated with respect to day 0.....										48±18%

Figure 4 shows also that, the presence of microorganisms definitely affects the gas holdup. At gas superficial velocities up to  $0.023 \text{ m.s}^{-1}$ ,  $\varepsilon_g$  increased from 0.5 to 4.8 % in clean water (without microorganism), whilst increasing up to 6.5% in the presence of bacteria. The magnitude of this increase (specifically 37%) is greater than the effect of liquid recirculation (15%) which was attributed to gas entrainment. This observation is somewhat general for all investigated conditions. However, even if at low gas superficial velocities the data are quite similar, the increase of gas holdup reached 43% at the end of operation time. A possible explanation of this increase in  $\varepsilon_g$  ( $\sim 48 \pm 18$  % in average) could be the effect of biomass particles disturbing the gas flow. Considering a three-phase gas-liquid-solid system, the fitting of the proposed empirical model (Eq.2) to the observed gas retention should logically lead to an increase of  $\sim 48\%$  of the model parameter ( $\kappa$ ) reported earlier. The resulting increased-coefficient, namely  $\kappa_{\text{GLS}}$  which equals to approximately  $3.03 \pm 0.06 \text{ s.m}^{-1}$ , includes the impact of biomaterials to the gas holdup visibly perceived as the increase of the slopes of the retention data in the three-phase system depicted in Figure 4.

In this study, the microorganisms continuously challenged to form as possible they could a film layer onto any surface of the reactor where harsh conditions of aeration were attenuated (i.e., especially on the walls of the downcomer compartment). Due to these specificities of the reactor, actual estimates of the overall biomass in the process were extremely tricky. However, the driven/recirculated biomass was estimated and reported at different time of the process. Results are presented in Figure 5. As shown, the data are extremely widespread. In order to better approximate the overall biomass in the reactor, a ratio of biomass concentrations was defined and estimated between the solids recirculated and the solids recovered from the film layer deposited on the membrane fibers. These measurements, performed at each period of membrane maintenance (i.e., 2 to 3 weeks) showed that in average only  $2.4 \pm 0.2$  % of microorganisms are effectively recirculated into the process. Under 600 days of operation, the concentration of biomass around the fibers was in average equal to  $210 \pm 60 \text{ g per m}^2$  of membrane, corresponding to approximately  $220 \pm 60 \mu\text{m}$  of biofilms. By assuming the nitrifying flora was uniformly laid around the membrane fibers (total area  $1.5 \text{ m}^2$ ) and specifically onto the walls of the downcomer compartment ( $1.6 \text{ m}^2$ ), the extrapolation of these data to the overall reactor revealed the biomass concentration equals  $\sim 10.9 \pm 3.1 \text{ g.l}^{-1}$ . To validate these data, the "TwoPopNitrification" kinetic model contained in the simulation tools of BioWin 2.2 software was considered. Several simulation tests were performed by varying the purge flowrate from  $0.1 \cdot 10^{-3}$  to  $3.0 \cdot 10^{-3} \text{ m}^3.\text{d}^{-1}$  and decreasing the Sludge Retention Time (SRT) from 200 to 7 days. One of the results obtained is depicted in Figure 6. This graph confirms the range of concentration of nitrifying flora ( $10.9 \pm 3.1 \text{ g.l}^{-1}$ ) accumulated by the process.

These data are quite indicative but plausible. Their accuracy is affected by the actual estimate of biomass lost during the membrane cleaning, and also, by the distribution of suspended circulated-biomass in the reactor as hydrodynamic conditions were extremely different into the two compartments.

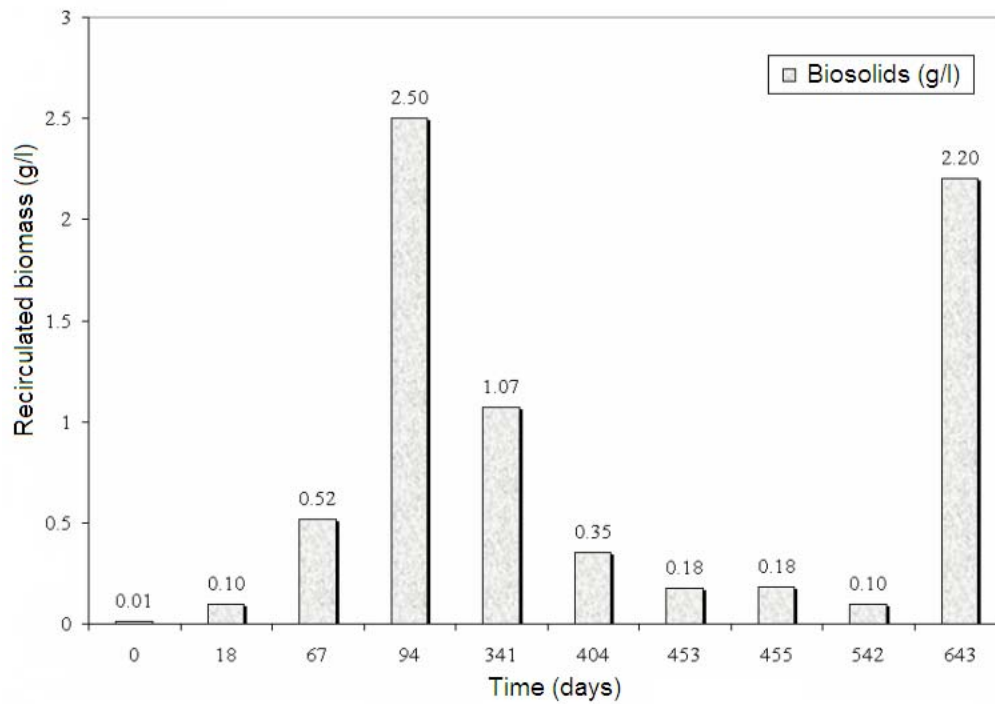


Figure 5: Recirculated/driven biomass (dry material) measured at different time of the process

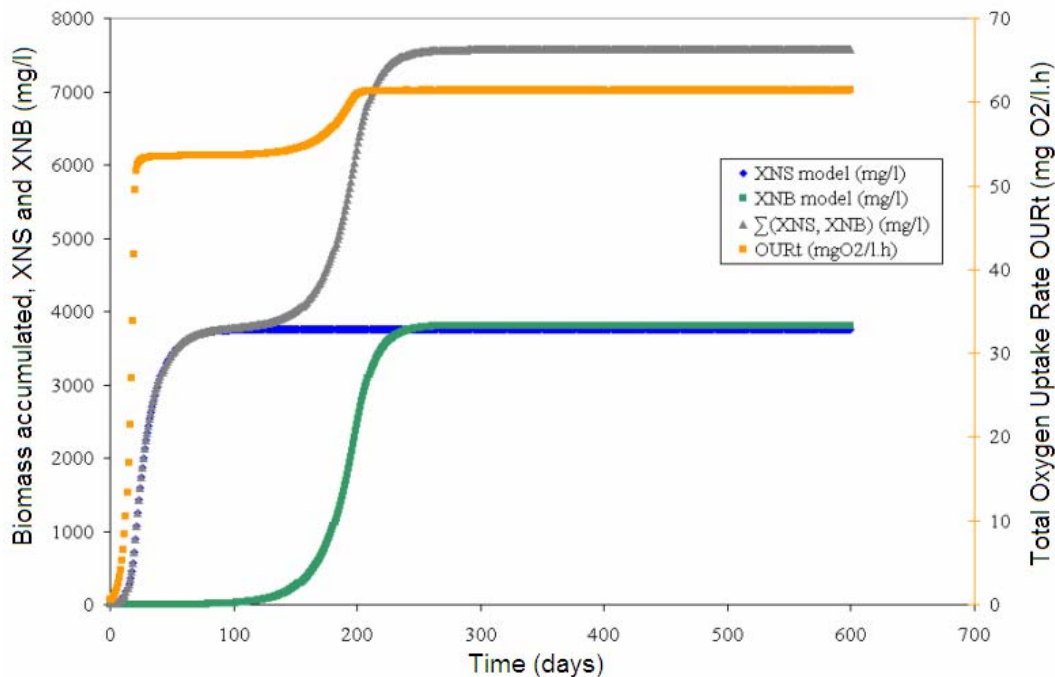


Figure 6: Simulation results of biomass accumulated by the JLMBR plant under 600 days of operation, (purge flowrate =  $1.5 \cdot 10^{-3} \text{ m}^3 \cdot \text{d}^{-1}$ , SRT = 13 days); XNS and XNB are Nitrosomonas and Nitrobacter species, respectively.

To better understand the impact of biological solids on the increase of gas holdup, the diameter of circulated particles was measured at different times of the process by the mean of a laser beam particle analyzer (Coulter LS100, Fraunhofer optical model, software n°1.53). The investigation period ranged from day 340 to 441. Although this period represents only 20% of the total investigation time (with respect to 600 days), the range (day 340–441) coincides with the steady state period of the process (Figure 6). From a biological point of view, this period is generally characterized by the stability of biomass activities e.g., biomass growth and/or biomass respiration rates indirectly represented here by the oxygen uptake rates OUR (cf. Figure 6). The investigations of the process performances and/or solids size measurements in this period of stability are representative of the process. The authors are aware that these measurements could slightly differ if the investigations were performed at the exponential phase of biomass growing. Specifically, while Figure 7 shows an example of particle size measurements at the day 358, all results are summarized in Table 2. They correspond to data recorded at different times of operation. One can see that the reported average size of bioparticles varies between 50 and 60  $\mu\text{m}$ . These values are similar to those

reported by Muller et al. (1995) and Zhang et al. (1997). These solids represent an obstacle to the gas sparged in the system. Their recirculation fragmented the gas phase into smaller bubbles, increasing the global bubble surface area flux (Finch et al., 2000). As a result, the gas holdup increased. Our results could be satisfactorily compared to the increase in the pressure drop with decreasing particle size and/or increasing particle density in a loop-seal circulating reactor operated by Sung and Kim (2002). At very low gas velocities (below  $0.01 \text{ m}\cdot\text{s}^{-1}$ , see Figure 4) corresponding to  $\varepsilon_g$  values below 2%, the gas holdup in clean water did not significantly differ from the data recorded in process media.

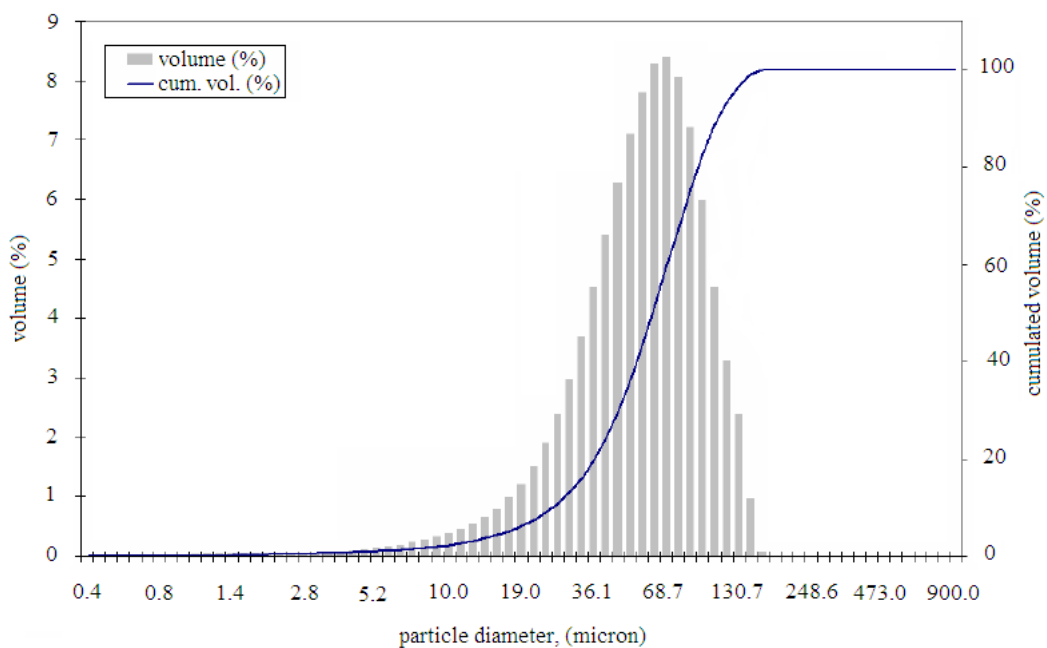


Figure 7: Result of particle size measurement at the day 358

Table 2: Diameter of recirculated aggregates and standard deviation (std)

Days	diameter ( $\mu\text{m}$ )	std ( $\mu\text{m}$ )
340	50	32
358	64	32
394	46	22
441	65	43

#### 4 Conclusion

A novel type of jet-loop membrane bioreactor has been proposed. Under 500 to 600 days of operation, the total nitrifying flora accumulated by the plant was experimentally estimated to  $\sim 10.9 \pm 3.1 \text{ g.l}^{-1}$  and confirmed by dynamic biological simulations. On the steady state period of the process, the size of circulated biomaterials was measured. The results revealed that the mean diameter of driven solids ranged between 50 and 60  $\mu\text{m}$ . These data were satisfactorily compared to those reported in the literature.

The combined action of air and circulated water flowrates on the gas holdup has been reported and modeled by a mathematical multilinear function. For the investigated ranges of air and water velocities, the gas holdup increased from 0.5 to 4.8% in clean water, whilst increased up to 6.5% in the biological media. This difference was attributed to a probable gas entrainment effect and the bubbles fragmentation due to solids recirculation into the two compartments of the system.

This impact of solid materials on the increase of gas holdup might affect the mass transfer coefficient, so the  $\alpha$ -factor in the system. A better understanding of this phenomenon and the relationship between these parameters are the next step of our investigations.

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