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CFD simulation and optimization of membrane scouring and nitrogen removal for an airlift external circulation membrane bioreactor

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HIGHLIGHTS

- CFD model developed for the simulation and optimization of an AEC-MBR.
- Simplified bio-kinetics and sludge rheology incorporated within the CFD framework.
- The height of gas-liquid dispersion for membrane scouring was optimized.
- Simultaneous cost-effective membrane scouring and nitrogen removal achieved.

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ABSTRACT

Cost-effective membrane fouling control and nitrogen removal performance are of great concern in airlift external circulation membrane bioreactors (AEC-MBRs). Computational fluid dynamics (CFD) model incorporating sub-models of bio-kinetics, oxygen transfer and sludge rheology was developed for the cost-effective optimization of a lab-scale AEC-MBR. The model was calibrated and validated by extensive measurements of water velocities and water quality parameters in the AEC-MBR. The validated results demonstrated that the optimized height of gas-liquid dispersion was at around 300 mm. The shear stress on membrane surface was equalized and had an average value of 1.2 Pa under an air flowrate of $1.0 \text{ m}^3 \text{ h}^{-1}$. The model further revealed that the high nitrogen removal efficiency (>90%) was achieved due to the high recirculation ratio driven by airlift force without destroying the oxygen deprivation and enrichment in the anoxic and oxic zone, respectively.

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1. Introduction

Membrane bioreactors (MBRs) with different scales, types and structures have been widely used in wastewater treatment and reclamation because of the well-recognized advantages such as good and stable quality of effluent, less excess sludge production, flexible installation and small footprint (Judd, 2011; Wang et al., 2014). However, these advantages may be overshadowed by the high energy consumption of aeration and pumping (Fenu et al., 2010). For decades, researchers and practitioners have made great

efforts to reduce the energy consumption and operating costs for MBR technology through process innovation and better operating strategy (Braak et al., 2011). Our previous work showed that the airlift external circulation MBR (AEC-MBR), which consists of an aeration tank and a membrane tank connected through holes in the clapboard separating the two tanks, was in a good performance for treating decentralized wastewater with lower energy consumption due to the lower aeration intensity and needless pumping (Fan et al., 2006). Continual innovations were made for the AEC-MBR application through membrane module design, membrane material modification and configuration optimization (Luo et al., 2015; Yuan et al., 2010; Zhang et al., 2013). Since 2003, this technology has been applied in more than 40 plants of wastewater treatment and reuse for public washrooms, hospitals, pharmaceutical factories and residential buildings in China (Zhang et al., 2013). Recently, we realized that the AEC-MBR can be converted to an

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alternative anoxic-oxic MBR system for nitrogen removal by replacing the aeration diffuser with paddle agitating in the aeration tank. A similar system called a multi-habitat MBR was proposed by (Tang et al., 2015, 2014). In their study, a high nitrogen removal rate was achieved due to the biodiversity and succession of the microbial community resulted from the formation of a multi-habitat in a single MBR. However, the comprehensive action of the stirring paddle and the aeration taken for the formation of stable internal circulation, as they mentioned, was ambiguous. In fact, the quantification and optimization of the hydrodynamics such as stirring paddle speed and aeration intensity are the prerequisites for their successful operation.

Essentially, the key issue in such systems was how to create a hypoxia environment in anoxic unit while still meets the requirement of membrane scouring in membrane unit by the adjustment of configurations and operating conditions. DO concentration distribution, related to both sides and with quick response, is the most important and direct index for such adjustment. However, the numerical quantifying and controlling of DO distribution and water quality parameters in the AEC-MBR is missing. In principle, the evolution of DO in bioreactor is a balance of oxygen transfer from gas to bulking liquid and oxygen consumption by microbial community. And it affects the removal performance of both organics and nutrients. Volumetric oxygen transfer rate (OTR) coefficient, k_{La} , is usually used for the description of oxygen source (Cockx et al., 2001). It is affected by many factors, such as configuration of bioreactor, surface tension, viscosity, temperature, and most importantly, the aeration condition including aeration intensity (Pittoors et al., 2014), aeration pattern (Capodici et al., 2015), bubble diameter and bubble shape (De Temmerman et al., 2015) and hydrodynamics (Braak et al., 2011). More than that, k_{La} is affected by the solution composition, biomass concentration and sludge rheological property in wastewater treatment systems (Pittoors et al., 2014).

Considering these factors together, CFD modeling for different bioreactor configurations, aeration devices and agitators in different types of bioreactors provided an excellent design tool for the optimization for distribution of DO concentration and other concerned components (Lei and Ni, 2014). Activated sludge model (ASM), firstly developed for conventional activated sludge processes (Henze, 2000) and then extended in MBRs (Jiang et al., 2005), contains full description of oxygen consumption and transformation of other variables. It had been incorporated within the CFD framework to have a full understanding of the spatial distribution of DO as well as other water quality parameters in oxidation ditch, anoxic tank etc. (Brannock, 2003; Lei and Ni, 2014). However, simplification of full models are preferred for target-oriented and real-time control. The most straightforward model simplifications were obtained by assuming only oxic conditions while neglecting the denitrification processes. Markus (Gresch et al., 2011) studied the impact of aeration patterns on the flow field and the ammonia removal in a full scale aeration tank via integrating CFD with a single Monod expression for nitrification in which the DO concentration was presumed to be high enough to decouple the oxygen dynamics from the rest of the process dynamics. To enhance comprehensibility, Zhao et al. (1995) used a simplified ASM1 model to describe the ammonia and nitrate dynamics in an alternating activated sludge process. Zero-order reactions were adopted to represent the denitrification rate and the exchange between the soluble $\text{NH}_4\text{-N}$ and the organic nitrogen based on-line measurements. Along this line, Julien et al. (1999) developed a reduced order model containing two sub-models respectively for anoxic and oxic condition in a single tank through which the dynamics of the oxygen, ammonia and nitrate were only considered and the slower dynamics of the processes were frozen.

The simplex method of Nelder & Mead was performed for identification of parameters of the model. Similar approach was thus adopted and the simplified model was incorporated within CFD in this study. A certain number of approximations were made from biochemical and mathematical considerations. The sensitive parameters involved in the bio-kinetic model were estimated by minimizing the sum of the squares of weighted deviations between measured data and calculations using the secant method embedded in AQUASIM (Reichert, 1995). Secant method allowed for all variables, compartments and calculations specified in the definition of parameter estimation, and over all data pairs of measured quantities for parameter estimation, which gave the user freedoms in defining the fit problem.

Besides, quantifying sludge rheological behavior to embed in the MBR process modeling for better model predictions is well worth the effort. Not only because the sludge rheological property has significant impact on oxygen transfer efficiency by retarding the fluid flow (Passos et al., 2015), but it plays an important role in membrane scouring in MBR due to the well-known power law function type relation between shear stress and sludge viscosity (Ratkovich et al., 2013).

However, to the best of our knowledge, the modeling of oxygen transfer and bio-kinetics with the consideration of sludge rheology within the CFD framework in the MBR simulation has not been reported. The objectives of this work are: (1) to develop an integrated CFD model with sub-models of oxygen transfer, simplified bio-kinetics and sludge rheology for the description of oxygen transfer and uptake, nitrogen transformation and membrane scouring, and (2) to simulate and optimize the configuration of membrane module to obtain a higher membrane scouring and appropriate DO distribution, and (3) to simulate and optimize the performance of nitrogen removal efficiency in the lab-scale AEC-MBR through the aeration adjustment. In this paper, CFD simulation and optimization of the height of gas-liquid dispersion and aeration intensity on membrane scouring and nitrogen removal was carried out with model calibrated and validated by extensive measurements of flow velocity and water quality parameters (Fig. 1).

2. Materials and methods

2.1. Experimental setup and operational conditions

Before the building up of the lab-scale AEC-MBR, the module configuration was optimized by modifying the height of gas-liquid dispersion, h_m , which showed a good sensibility on membrane scouring in our previous study (Zhang et al., 2013). Here membrane shear stresses at different h_m with a superficial aeration velocity of $50.0 \text{ m}^3 \text{ h}^{-1} \text{ per m}^2$ projected cross section area (SAR 50), which was the recommended value by the manufacturer (SINAP membrane tech Co., Ltd., China), were studied. According to the initial optimization, a lab-scale AEC-MBR with a work volume of 90.0 L was installed. It consists of an oxic tank (MBR tank) with the volume of 60.0 L and an anoxic tank with the volume of 30.0 L (Fig. 2). Five 0.07 m^2 (effective filtration area 0.05 m^2 for each membrane side) flat membrane sheets (SINAP membrane tech Co., Ltd., China) with a flux of $9.0 \text{ L m}^{-2} \text{ h}^{-1}$ were packed (Table 1). The configuration of the AEC-MBR and the sampling locations in this work were shown in Fig. 2. A clapboard with two connecting holes was inserted into a single tank, separating the tank into two units, anoxic unit and oxic unit, respectively. The membrane module was settled in the geometrical middle of the oxic unit. The two sides of each membrane sheet were named in forms of 'm_{ia}' and 'm_{ib}', where 'i' was the membrane series number along

Fig. 1. Workflow diagram for CFD simulation and optimization of membrane scouring and nitrogen removal of the AEC-MBR.

Fig. 2. Configuration and sampling locations of the lab-scale AEC-MBR.

Table 1
Geometry information and operation parameters of the lab-scale AEC-MBR.

Items	Sizes/parameters
Treatment capacity	108.0 L d ⁻¹
Oxic tank (L × W × H)	350.0 mm × 200.0 mm × 1000.0 mm
Anoxic tank (L × W × H)	175.0 mm × 200.0 mm × 1000.0 mm
Membrane module (L × W × H)	220.0 mm × 100.0 mm × 320.0 mm
Membranes' interval (mm)	10.0
HRT (h)	20.0
SRT (day)	20.0
Flat sheet membrane (L × H × D)	220.0 mm × 320.0 mm × 6.0 mm, PVDF, pore size 0.1 μm
Operation flux (L m ⁻² h ⁻¹)	9.0
Volume aeration intensity ^a (m ³ h ⁻¹)	0.5 (stage 1), 1.5 (stage 2), 1.0 (stage 3)
Orifice diameter (mm)	2.0
Connecting hole diameter (mm)	10.0
Agitator (L × W × D)	125.0 mm × 30.0 mm × 2.0 mm, rotation speed at 60 rpm

^a Volume aeration intensity of 1.0 m³ h⁻¹ is corresponded to the SAR 50.

y direction, and 'a' and 'b' indicated the front and back of each membrane sheet, respectively. The aeration diffusers were perpendicular to the membrane surfaces with a diffusers' interval s_p . The

vertical distance of gas-liquid diffusion chamber, h_m , was prescribed as 300 mm according to CFD optimization. A double-flat blade agitator with a rotation speed at 60 rpm in the anoxic tank was set for mixing. The wastewater was pumped from the feed tank into the anoxic unit with a peristaltic pump (YZ1515X-A, LONGER, China) controlled by a liquid level gauge. And treated wastewater was sucked out of the membranes by peristaltic pumps connected to pressure gauge (HSLT-PKGA23, HESHI TECH, China). The mixed liquor was driven by aeration and externally recycled between the two units through the overflow hole and return flow hole located at the top and bottom of the clapboard, respectively. At the same time, membrane sheets were scoured in the internal circulation in oxic tank. No recirculation sludge pump was needed in the entire process. The activated sludge was inoculated from a municipal wastewater treatment plant (WWTP) with a process of anoxic/anaerobic/oxic-MBR in Beijing, China. Initial concentration of mixed liquid suspended sludge (MLSS) in the AEC-MBR was around 3000 mg L⁻¹. The synthetic wastewater was composed of 400 mg L⁻¹ glucose (approximately 360 mg L⁻¹ COD), 200 mg L⁻¹ CH₃COONa (approximately 190 mg L⁻¹ COD), 191 mg L⁻¹ NH₄Cl (50 mg L⁻¹ ammonium), 35 mg L⁻¹ KH₂PO₄ (8 mg L⁻¹ total phosphorus), 100 mg L⁻¹ MgSO₄·7H₂O, 50 mg L⁻¹ CaCl₂·2H₂O, 250 mg L⁻¹ NaHCO₃ (300 mg L⁻¹ CaCO₃ alkalinity) and mg L⁻¹ of trace element solution per liter wastewater. The

trace element composed of 1500 mg L⁻¹ FeCl₃·7H₂O, 150 mg L⁻¹ H₃BO₄, 30 mg L⁻¹ CuSO₄·5H₂O, 180 mg L⁻¹ KI, 120 mg L⁻¹ MnCl₂·4H₂O, 60 mg L⁻¹ Na₂MoO₄·2H₂O, 120 mg L⁻¹ ZnSO₄·7H₂O, 150 mg L⁻¹ CoCl₂·6H₂O and 10,000 mg L⁻¹ EDTA.

Hexahedral mesh was generated for the geometry of this set up by using ICEM 15.0 (ANSYS, USA). After the grid independent test, the optimized grid used in this work was around 288,000 elements with the maximum and minimum element sizes of 30.0 mm and 3.0 mm, respectively. All the simulations were carried out in FLUENT 15.0 (ANSYS, USA) on a work station (Thinkstation D30, LENOVO, China) with a 64 bit processor (Intel® Core™ 12 Xeon CPU E5649) and 24 GB of random access memory, running at a clock speed of 2.53 GHz.

2.2. Sampling and analysis

Samples of effluent were collected once every two days to make sure the system was in a good condition and to know whether the system reached a steady state. DO concentrations were measured with a fluorescence DO meter (Multi 3410, WTW Co Ltd, German) at points of S1(25 mm, 175 mm, 25 mm), S2(175 mm, 100 mm, 750 mm), S3(325 mm, 25 mm, 500 mm), S4(385 mm, 25 mm, 750 mm), S5(510 mm, 175 mm, 500 mm), S6(510 mm, 175 mm, 25 mm), respectively (Fig. 2). Mixed liquor samples at points of S1 ~ S6 were collected once a day for 20 days during steady stages. The samples were immediately centrifuged at 4 °C, supernatant was then filtered with glass fiber filters with a pore size of 0.22 μm, and then stored at 4 °C for the measuring of COD, ammonia, nitrate, nitrite and total nitrogen. All measurements were carried out according to standard methods (APHA, 2005) within one week after the sample collecting. The measured data were evaluated by Student's *t*-test paired with the *F*-test to determine the statistical significance of the observed differences between the measured data for DO, ammonia, nitrate and nitrite at the 95% confidence level (Dochain and Vanrolleghem, 2001). After that, the average values were used for model calibration and validation.

Activated sludge samples for the determination of maximum nitrate uptake rate (NUR_{max}) was taken from the steadily operated AEC-MBR. NUR_{max} was determined as follows (Han et al., 2005): certain amounts of MLSS solution was taken out from the anoxic unit and was settled to remove the supernatant and placed on magnetic stirrer with a water bath (20 °C). The synthetic wastewater was added with the same volume of the withdrawn supernatant. First sample was taken immediately and filtered by 0.22 μm glass fiber membrane. And following samples were taken and filtered at certain time points. Nitrate and nitrite concentration was measured after all samples were collected at once. Nitrification rate was calculated through linear regression in Origin® (Origin Lab, America).

Rheology of activated sludge samples from the AEC-MBR was measured by a Haake RS6000 Rheometer® (Thermo Scientific, USA). A double gap cylinder (DG41 Ti, 0.5 mm for each gap) was used as the measure geometry with torque resolution of 0.5 μNm. The temperature of the samples was controlled at 20 °C ± 0.1 °C through use of an circulating oil bath (SC150, Thermo Scientific, USA). The measurements were performed in triplicate at shear rates ranging from 0 to 500 s⁻¹ after a 30 s strong shear with a shear stress of 500 s⁻¹. The rheological data was logged to a PC and analysed using HAAKE RheoWin™ (3.50.0012, Thermo Scientific, USA). The non-Newtonian relationships between shear stress and shear rate were fitted to the yield-pseudoplastic type Sisko rheology model (Tang and Zhang, 2014) using Matlab® (Mathworks, Inc).

3. CFD modeling

3.1. Model development

3.1.1. Governing equations

The CFD model only included the gas-liquid phases in this study since the positive buoyancy forces from rising bubbles was much greater than the negative buoyancy forces of settling sludge, and the sludge showed a solid-liquid following features even the flocculating agent was added in the lab-scale MBR (Brannock et al., 2010a; Liu et al., 2015). The Euler-Euler model was used for the multiphase flow simulation. The steady-state continuity equation for phase *q* is

$$\nabla \cdot (\alpha_q \rho_q \vec{v}_q) = \dot{m}_{pq} - \dot{m}_{qp} \quad (1)$$

where subscript *p* and *q* represent the gas phase and liquid phase, respectively. α_q is the volume fraction of phase *q*, ρ_q (kg m⁻³) is density of phase *q*, $\vec{v}_q$ (m s⁻¹) is velocity vector of phase *q*, $\dot{m}_{pq}$ (kg m⁻³ s⁻¹) is the mass transfer source from phase *p* to *q*. The steady-state momentum balance equation for the phase *q* is

$$\nabla \cdot (\alpha_q \rho_q \vec{v}_q \vec{v}_q) = -\alpha_q \nabla p + \nabla \cdot \vec{\tau}_q + \alpha_q \rho_q \times \vec{g} + \left(\vec{R}_{pq} + \dot{m}_{pq} \vec{v}_{pq} - \dot{m}_{qp} \vec{v}_{qp} \right) + \vec{F}_q \quad (2)$$

where *p* (Pa) is the pressure shared by all phases, $\vec{v}_{pq}$ (m s⁻¹) is the interphase velocity, $\vec{R}_{pq}$ (kg m⁻² s⁻²) is an interaction force between phases, $\vec{F}_q$ (kg m⁻² s⁻²) is a buoyancy force.

The interphase momentum exchange coefficient for $\vec{R}_{pq}$ was determined by Schiller–Naumann drag model:

$$C_d = \begin{cases} 24/Re(1 + 0.15Re^{0.687}) & Re \leq 1000 \\ 0.44 & Re > 1000 \end{cases} \quad (3)$$

where *C_d* is the momentum exchange coefficient, *Re* is Reynolds number.

To describe the effects of turbulent fluctuations of velocities and scalar quantities in multiphase, the RNG *k*- ϵ mixture turbulence model was applied as the closure model (ANSYS, 2014).

3.1.2. Species transport equations

The 3D steady-state species (DO, ammonia and NO_x) distribution in the AEC-MBR was predicted by solving the advection-dispersion species transport equation extended with an oxygen mass transfer rate (OTR) sub-model and Monod-like sub-models. The transport equation takes the form of

$$\nabla \cdot (\alpha^q \rho^q \vec{v}^q Y_i^q) = \nabla \cdot (\alpha^q \vec{J}_i^q) + \alpha^q S_i^q - \dot{m}_{qij} \quad (4)$$

where Y_i^q is the mass fraction of species *i* in phase *q*, $\vec{J}_i^q$ (kg m⁻² s⁻¹) is the diffusion flux of species *i* in phase *q*, S_i^q (kg m⁻³ s⁻¹) is the rate of creation by addition from the dispersed phase plus any user-defined sources, $\dot{m}_{qij}$ (kg m⁻³ s⁻¹) is the mass transfer source between species *i* and *j* from phase *q* to *p*.

3.1.2.1. Oxygen mass transfer. The oxygen transfer rate (OTR) corresponding to $\dot{m}_{qij}$ in Eq. (3) can be expressed as (Pittoors et al., 2014)

$$OTR = k_L a (\alpha S_0^* - S_0) \quad (5)$$

where $k_L a$ (s^{-1}) is the mass transfer coefficient, α is the oxygen saturation concentration ratio of wastewater to clean water (Tchobanoglous et al., 2002), S_o^* ($mg L^{-1}$) and S_o ($mg L^{-1}$) are the dissolved oxygen saturation concentration of clean water and dissolved oxygen in the liquid phase.

For bubble-water interfaces, the penetration theory developed by Higbie were widely used for volumetric $k_L a$ description in pure water and in wastewater treatment. For the sludge bulking liquid, oxygen transfer is different from that in pure water. Being transferred into the wastewater, the oxygen was then transferred into the zoogloas and finally into the cells of microbes for metabolism (Arenallos et al., 2015). MLSS concentration was found having an obvious effect on oxygen transfer by taking the vacancies of bubbles and changing their size and shape (Germain et al., 2007). The surfactants such as fatty acid and ethanol contained in the wastewater may lead a molecular film formation on the gas-liquid interface, which will hinder the oxygen transfer (Ratkovich et al., 2013). Considering these effects, $k_L a$ was modified and took the form of (Cockx et al., 2001),

$$k_L a = \beta \frac{12\alpha_b}{d_b} \sqrt{\frac{D_L \Delta U}{\pi d_b}} \quad (6)$$

where β is the coefficient considering the effect of bulk liquid property, α_b is the gas volume fraction, d_b (m) is the Sauter mean diameter of the bubbles, D_L ($m^2 s^{-1}$) is diffusivity of oxygen in the liquid phase, and ΔU ($m s^{-1}$) is the slip velocity between bubbles and bulking liquid.

Mass transfer rate was affected by bubble shape and diameter evolution, which could be captured by using of interfacial area concentration model or Lagrangian approaches (Nopens et al., 2015). However, modeling of the coalescence and breakage of bubbles would significantly increase computational consumption and was not suitable for a steady-state simulation. So, a constant mean bubble diameter with no collisions, coalescence or breakage, which was a common acceptance in many previous CFD modeling of MBRs (Liu et al., 2015) as well as other bioreactors like bubble column (McClure et al., 2014), was assumed

$$d_b = 2.9\alpha_{d_b} \left(\frac{\sigma d_o}{g\rho_L} \right)^{1/3} \quad (7)$$

where α_{d_b} is a coefficient for the form of orifice, and σ ($N m^{-1}$) is surface tension of liquid phase, d_o (m) is diameter of orifice, g ($m s^{-2}$) is gravitational acceleration, and ρ_L ($kg m^{-3}$) is the density of the liquid.

3.1.2.2. Biological kinetics of oxygen consumption and nitrogen transformation. In the ASMs framework, DO was presumed to be consumed for aerobic growth of heterotrophs and aerobic growth of autotrophs, which were affected by the concentrations of biomass and other substrates, which again involved in other kinetic processes (Henze, 2000). Since nitrogen transformation was mainly aimed in this study, only oxygen consumption, nitrification, denitrification and exchange of ammonia and organic nitrogen were described by simplified ASM1 (Zhao et al., 1995). Denitrification rate in anoxic unit, oxygen uptake rate in oxic unit and the exchange between the soluble NH_4-N and the organic nitrogen in both units were identified with the initial value calculated via the full ASM1 model. Nitrification rate was defined as Eq. (10) in which the maximum specific nitrification rate of oxic unit was measured off-line.

The oxygen consumption corresponding to S_i^q in Eq. (4) was simplified as

$$OUR_{Oxic} = OUR_{Cons} \quad (8-1)$$

$$OUR_{Anoxic} = k_1 OUR_{Cons} \left(\frac{S_o}{K_{OH} + S_o} \right) \quad (8-2)$$

where K_{OH} ($mg L^{-1}$) is the half saturation constant of oxygen, S_o ($mg L^{-1}$) is the concentration of dissolved oxygen, OUR_{Oxic} ($mg L^{-1} h^{-1}$) and OUR_{Anoxic} ($mg L^{-1} h^{-1}$) are the oxygen uptake rates in oxic unit and anoxic unit, respectively, OUR_{Cons} ($mg L^{-1} h^{-1}$) is the constant oxygen uptake rate to be identified, k_1 is the coefficient of OUR in anoxic unit.

The denitrification rate was simplified as

$$DNR_{Anoxic} = DNR_{Cons} \quad (9-1)$$

$$DNR_{Oxic} = k_2 DNR_{Cons} \left(\frac{K_{OH}}{K_{OH} + S_o} \right) \quad (9-2)$$

Where DNR_{Anoxic} ($mg L^{-1} h^{-1}$) and DNR_{Oxic} ($mg L^{-1} h^{-1}$) are the denitrification rates in anoxic unit and oxic unit, respectively, DNR_{Cons} ($mg L^{-1} h^{-1}$) is the constant denitrification rate to be identified, k_2 is the coefficient of DNR in the oxic unit.

The nitrification rate was defined by

$$NUR = NUR_{max} \left(\frac{S_{NH}}{K_{NH} + S_{NH}} \right) \left(\frac{S_o}{K_{OA} + S_o} \right) \quad (10)$$

where NUR ($mg L^{-1} h^{-1}$) is the nitrification rate, NUR_{max} ($mg L^{-1} h^{-1}$) is the maximum nitrification rate, K_{NH} ($mg L^{-1}$) is the ammonia half-saturation coefficient for heterotroph, and S_{NH} ($mg L^{-1}$) is the concentration of ammonia, and K_{OA} ($mg L^{-1}$) is the oxygen half-saturation coefficient for heterotroph.

The exchange rates between ammonia and organic nitrogen were represented by zero-order equations as follows

$$r_{A,Oxic} = R_{A,Oxic} \quad (11-1)$$

$$r_{A,Anoxic} = R_{A,Anoxic} \quad (11-2)$$

Where $r_{A,Oxic}$ ($mg L^{-1} h^{-1}$) and $r_{A,Anoxic}$ ($mg L^{-1} h^{-1}$) are the exchange rate between ammonia and organic nitrogen in oxic unit and anoxic unit, respectively. $R_{A,Oxic}$ ($mg L^{-1} h^{-1}$) and $R_{A,Anoxic}$ ($mg L^{-1} h^{-1}$) are the constant values for $r_{A,O}$ and $r_{A,A}$, respectively.

3.1.2.3. Shear stress. Shear stress was calculated by,

$$\tau_{ij} = \eta \bar{D} = \eta \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \quad (12)$$

where τ_{ij} (Pa) is shear stress, η (Pa s) is apparent laminar viscosity, $\bar{D}$ is rate-of-deformation tensor.

Apparent viscosity η is a function of all three invariants of the rate-of-deformation tensor. A shear stress-shear rate relationship with MLSS of $7.5 g L^{-1}$ was measured for the estimation of η and fitted to the Sisko model (Eshtiaghi et al., 2013) (Fig. S1).

$$\eta = 0.0012 + 0.071\gamma^{-0.567} \quad R^2 = 1.0 \quad (13)$$

where γ (s^{-1}) is the shear rate.

3.2. Boundary conditions

Membrane surfaces were treated as impermeable, stationary walls, which were free slip for air and no slip for liquid (Ndinisa et al., 2006; Yang et al., 2012). This assumption was based on the fact that the critical flux have little effect on the overall flow pattern and flat sheet membranes were non-oscillatory in lab-scale MBRs (Field and Pearce, 2011). The inlet of wastewater was positioned on the surface of anoxic tank. The outlet of wastewater was positioned on the membrane surfaces with an area of $0.0001 m^2$. Velocity inlet boundary was applied for air diffuser inlets, sewage inlet and sewage outlet, which were treated as plain

surfaces with flow rates of mass equivalent to superficial mass velocities. The influent soluble species S_i flux were prescribed accordingly. A degassing boundary condition was specified for the water-air surface (Brannock et al., 2010b). No slip boundary conditions were assigned for all other walls. The roughness constant and the roughness height at the no slip walls were default. The rotation by the submerged impeller in the anoxic unit was treated using multiple reference frame (Bannari et al., 2011). Residuals criteria of 1×10^{-7} was used for convergence of equations involving species transport, and the default residuals criteria of 1×10^{-3} was used for convergence of other variables. Flow velocity and concentration of DO, ammonia and NO_x at specific section were also monitored to make sure the simulation reaches a steady state.

4. Results and discussion

4.1. Model calibration and validation

Kinetic parameters K_{OH} , reaction rates (DNR_{Cons} , OUR_{Cons} , $R_{A,\text{Anoxic}}$, $R_{A,\text{Oxic}}$) and coefficient k_2 were found most sensitive by the relative sensitive analysis. Here, the average concentration of DO, ammonia and NO_x measured in oxic tank and anoxic tank were used for the determination of model parameters with initial values of half saturation coefficients and with reaction rates given in the full ASM1 model. With the calibrated values (Table 2), the model gave the three state variables relative errors of 1.4%, 12.0%, 1.8% in oxic tank and 2.6%, 5.7%, 13.8% in anoxic tank (stage two), respectively.

Model validation of water velocity was carried out in the AEC-MBR at an airflow rate of $1.0 \text{ m}^3 \text{ h}^{-1}$. A Particle Image Velocimetry (PIV) (EM3-03M1500, Microvec Pte. Co., LTD, China) was adopted. A certain amount of glass micro-beads (MV-H0105, Microvec Pte. Co., LTD, China) were injected into the reactor as the tracers to maintain an overall concentration of 10.0 mg L^{-1} . Planar cross-sections of $45.0 \text{ mm} \times 30.0 \text{ mm}$ were illuminated with a sheet light source (LWGL532081798, Microvec Pte. Co., LTD, China), and a high speed camera (SIGMA, Japan) taking 206 pictures per second was set up to capture the tracer particles for $15.0 \text{ s} \times 3$ times at each cross-section. To eliminate bubble reflection effects, only water velocities outside the membrane module (L1 and L2) and water velocities in anoxic unit (L3 and L4) were recorded (Fig. 2). The simulation achieved steady states after a certain of iterations (Fig. S4). The simulated local water velocity in MBR tank demonstrated that the recirculation flow between the riser and downcomer was established (Fig. S5). With a larger cross-sectional area, the observed downcomer had a much smaller water velocity than that in the riser. The even smaller water velocity near the wall and at the corner due to the wall friction and eddy-dissipation can be also observed (Fig. S5). An obvious two layers of water streamline in the anoxic unit made this unit more like a plug flow reactor (Fig. S5). A good agreement of water velocities between CFD simulations and PIV measurement results was achieved (Fig. 3). All relative errors were less than 5% suggesting that the model provided a good description of flow behavior. Model validation of water qualities was carried out during the state stages. Concentration of DO, was measured twice a day with a flu-

orescence DO meter (Multi 3410, WTW Co Ltd, German) at points of S1 ~ S6 (Fig. 2). Concentrations of ammonia, nitrite and nitrate at points of S1 ~ S6 were measured and compared to simulations. Fig. S2 shows that the simulated results provided acceptable accurate representativeness of the water quality parameters of DO, ammonia and NO_x compared to the measurements via a small modification in the parameter values (Table 2).

4.2. Optimization of membrane module configuration

High and even distribution of shear stresses on different membrane surfaces were preferred for alleviating membrane fouling (Wibisono et al., 2014). With that, MBR can be maintained with a lower superficial airflow rate, which, in this study, was helpful for energy saving and to keep the DO concentration low in anoxic unit. However, uneven distribution of shear stresses were often observed whenever the aeration diffuser was perpendicular (Ndinisa et al., 2006) or parallel (Liu et al., 2010) to the membrane surfaces. To get even shear stresses distribution on different membrane sheets, many approaches were studied including optimization of parameters in configurations (Kang et al., 2008) and in operation (Drews et al., 2010). One of the most effective approaches in our previous findings was to elevate the height of gas-liquid dispersion h_m (Zhang et al., 2013). Here h_m (from 150 mm to 600 mm) with a superficial aeration velocity of SAR 50 were studied. As shown in Fig. 4, shear stress on different membrane surfaces distributed unevenly at $h_m = 150 \text{ mm}$, e.g., higher on the middle surfaces and lower on either side membranes. With the extension of h_m , the shear stress saw growth both on the middle and the side membranes. The shear stress at the utmost membranes surfaces (m_{1a} and m_{5b}) increased faster than that of the middle ones with the increase of h_m from 150 mm to 450 mm, which finally enhanced and almost equalized the shear stresses on different membrane sheets at h_m of 300 mm and 450 mm (Fig. S3). But when h_m further extended to 600 mm, the shear stresses dropped again. The possible reasons were that water velocities didn't fully developed at low h_m whereas water velocities dropped at high h_m due to the converting of kinetic energy into potential energy and friction head loss (Douek et al., 1998). Considering the h_m 450 mm was spatial inefficiency and had a higher head loss, $h_m = 300 \text{ mm}$ was preferred in this case.

4.3. Distribution of DO concentration

For the description of the DO transportation and distribution, a 3D AEC-MBR was simulated by the integrated CFD model developed here. Fig. 5 shows the predicted distribution of DO, ammonia and NO_x at a certain hydrodynamic condition with the corresponding experimental measurements. Necessary DO concentration distribution successfully achieved in the oxic tank and anoxic tank with DO concentration of 3.5 mg L^{-1} in the oxic unit and that of 0.01 mg L^{-1} in anoxic tank, respectively (Fig. 5B). The high recirculation ($R_i = 1306\%$) didn't destroy the expected DO environment. In contrast, the nitrogen removal efficiencies turned out to be very high with the high internal recirculation ratios (Table 3). In a viewpoint of ASM1, aerobic environment is prerequisite for the growth of nitrifiers (autotrophs) in oxic unit where ammonia can be trans-

Table 2
Values of parameters for the case of SAR 50 and their variabilities for the three SARs.

Parameters	DNR_{Cons}	OUR_{Cons}	K_{OH}	K_{OA}	K_{NH}	NUR_{max}	$R_{A,\text{Anoxic}}$	$R_{A,\text{Oxic}}$	k_1	k_2
Values	6.49	33.96	0.2	0.3	0.25	23.15	−0.52	0.93	2.81	3.33
Variability	28%	21%	20%	0%	0%	0%	43%	48%	0%	28%

Fig. 3. Comparison of simulation and experimental results of water velocity in the AEC-MBR.

Fig. 4. Average shear stresses on membrane surfaces with different h_m at SAR 50.

ferred to nitrate and nitrite, which would be transformed to nitrogen gas by denitrifiers (heterotrophs) in the anoxic condition given adequate substrates (Henze, 2000). Concentrations of TN in oxic unit of CFD simulation were nearly 2.8 mg L^{-1} (Fig. 5C and D), showed the removal ratio of TN was nearly 95%, which fitted the theoretical value (Tchobanoglous et al., 2002), and thus confirmed the accuracy of the developed model (Table 3).

4.4. Coordination of nitrogen removal and membrane scouring

The nitrogen removal and membrane scouring was coordinated through the configuration modification and aeration adjustment

since high aeration intensity was preferred for membrane scouring and ammonia removal but led to a detrimental high DO concentration in anoxic tank due to the high internal circulation ratio. For configuration modification, the optimization of gas-liquid dispersion height was carried out at first. Besides, the size of connecting hole was set to be small enough to respectively form oxygen deprivation and enrichment in the anoxic and oxic zone according to the hydraulic calculation in our previous study (Zhang et al., 2013).

The aeration intensity optimization of the AEC-MBR was then studied by tuning aeration intensities from SAR 25 to SAR 75 to balance the nitrogen removal and membrane scouring. The bio-kinetic parameters, which were case based and showed different variations (Table 2), were adjusted according to the measured data. The DO concentrations increased in both the anoxic and oxic units with the increase of SAR as expected, which can explain the differences of other species distribution in these two units. For ammonia, the increase of DO concentration led to a decrease of ammonia in the oxic unit, which further led to a dilution of ammonia in the anoxic unit when the liquid with low concentration of ammonia was recirculated. So a higher aeration intensity resulted in decrease of ammonia concentration in both units. Similar situation was found for NO_x concentration in the oxic tank. The high recirculation rates led to a dilution of NO_x concentration in the oxic unit when the liquid with low concentration of NO_x from anoxic unit was recirculated. The lower NO_x concentration of SAR 25 may be contributed to the simultaneous nitrification and denitrification due to the low DO concentration in oxic unit. But the NO_x concentration in anoxic unit increased gradually due to the increase of DO and the homogenization between oxic tank and anoxic tank. As a result, further increase of aeration intensity would inhibit the denitrification process and lead to the system crash. At the same time, membrane shear stresses under different

Fig. 5. Profiles of liquid velocity (A), DO (B), ammonia (C) and NO_x (D) in the AEC-MBR at SAR 50.

Table 3

Performances of the AEC-MBR at different aeration intensities.

Parameters	SAR 25	SAR 50	SAR 75
Average DO in oxic unit (mg L ⁻¹)	0.97 ± 0.14 ^a	3.10 ± 0.24	4.98 ± 0.30
Average DO in anoxic unit (mg L ⁻¹)	0.004 ± 0.04	0.02 ± 0.13	0.08 ± 0.22
Average ammonia in oxic unit (mg L ⁻¹)	0.11 ± 0.16	0.08 ± 0.13	0.05 ± 0.10
Average ammonia in anoxic unit (mg L ⁻¹)	4.36 ± 1.13	3.41 ± 1.19	1.96 ± 1.08
Average NO _x in oxic unit (mg L ⁻¹)	2.72 ± 0.18	2.87 ± 0.17	2.36 ± 0.11
Average NO _x in anoxic unit (mg L ⁻¹)	0.10 ± 0.14	0.16 ± 0.21	0.52 ± 0.28
Removal ratio of total nitrogen (%)	94.8	94.6	95.6
Recirculation ratio (%)	1036	1306	1531
Average shear stress (Pa)	0.9 ± 0.6 ^b	1.2 ± 0.8	1.4 ± 0.8

^a The standard deviations are the deviations of the species in the oxic or anoxic tank.

^b The shear stress with a deviation obtained by averaging the shear stress at all membrane surfaces.

SARs were extracted from each membrane. The lowest shear stress occurred on the utmost surfaces of m_{1a} (m_{5b}) while the highest occurred on surfaces of m_{1b} (m_{5a}) in different SARs due to the Coanda effects (Mohd. Akbar et al., 2013). The difference of the two extreme values, however, was insignificant suggesting the success of configuration optimization (Fig. 6). Shear stresses were fitted to power-law correlation with aeration intensities due to the intrinsic relationship of shear stress and shear rate which linearly correlated to local water velocity near the membrane (Fig. 6) (Ratkovich et al., 2013). The derivative, $d(\text{Shear stress})/d(\text{SAR})$, of

the fitting functions decrease monotonically suggested that the lower SAR, the higher shear stress efficiency. However, the SAR should be high enough to obtain sufficient shear stress to avoid membrane fouling. An average shear stress of 0.5 Pa in a full scale MBR was reported while an average shear stress of 0.6–0.7 Pa in a pilot scale MBR can be achieved with the dosing of flocculent, Fe (II) (Liu et al., 2015). The average membrane shear stress can be 0.5–2.0 Pa with $\text{SAD}_m = 0.5\text{--}9.0 \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1}$ in a lab-scale MBR (Yan et al., 2016, 2015). In this study, the shear stress of 0.9 Pa at SAR 25 ($\text{SAD}_m = 1.0 \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1}$) was achieved. Operated at SAR

Fig. 6. Effects of aeration intensity on membrane surface shear stress with maximum on m_{1b} , minimum on m_{5a} and average of all membrane surfaces.

25, the aeration energy can be reduced by 50.0% and 66.7% compared to SAR 50 and SAR 75, respectively. It should be noted, though, that these conclusions were based on the modeling of a lab-scale AEC-MBR which had a lower intensity of turbulence, a homogenous biomass distribution and a neglectable membrane oscillation and filtration. Large-scale MBR modeling by models considering both evolution of biomass and soluble species, and membrane oscillation and filtration is still needed.

5. Conclusions

A three dimensional CFD model was developed to simulate the multiphase dynamics and water quality in a lab-scale AEC-MBR. The proposed model was more target-oriented and thus calculation cheaper than full models. It gave an insight of membrane scouring and nitrogen transformation in the AEC-MBR system and was used for the system optimization. The cost-effective membrane scouring and nitrogen removal was achieved by the modification of gas-liquid dispersion height and aeration intensity. The results were suitable for lab-scale AEC-MBRs and further study is needed for large-scale MBRs considering evolution both for biomass and soluble species, membrane oscillation and filtration.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.biortech.2016.07.139>.

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