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# A super high-rate sulfidogenic system for saline sewage treatment



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#### ABSTRACT

This study proposes a novel approach to resolve the challenging issue of sludge bed clogging in a granular sulfate-reducing upflow sludge bed (GSRUSB) reactor by means of introducing intermittent gas sparging to advance it into a super high-rate anaerobic bioreactor. Over a 196-day lab-scale trial, the GSRUSB system was operated from nominal hydraulic retention time of 4-hr to 40-min and achieved the highest organic loading rate of 13.31 kg COD/m<sup>3</sup>·day which is substantially greater than the typical loading of 2.0–3.5 kg COD/m<sup>3</sup>·day in a conventional upflow anaerobic sludge bed reactor treating dilute organic strength wastewater. The average organic removal efficiency and total dissolved sulfide of this system were  $90 \pm 4.2\%$  and  $158 \pm 28$  mg S/L, while organics residual in the effluent was  $34 \pm 14$  mg COD/ L. The control stage (without gas sparging) revealed that the sludge bed clogging happened concomitantly with the significant drop in extracellular polymeric substance content of granular sludge, through relevant chemical measurements and confocal laser scanning microscopy analyses. On the other hand, compared with increasing the effluent recirculation ratio (from 1.4 to 5), the three-dimensional computational fluid dynamics modeling in combination with energy dissipation analysis demonstrated that the gas sparging (at a superficial gas velocity of 0.8 m s<sup>-1</sup>) can create a 23 times higher liquid shear as well as enhanced particle attrition. Overall, this study not only developed a super high-rate anaerobic bioreactor for saline sewage treatment, but also shed light on the role of intermittent gas sparging in control of sludge bed clogging for anaerobic bioreactors.

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#### 1. Introduction

Hong Kong is home to some seven million people, but its steep mountains only leave about 263 square kilometers for people to live and work (HKSAR, 2013). Population growth, rising public aspiration for better living quality as well as economic development increase the demand for a more innovative utilization of the scarce land resources. According to the study "Enhanced Use of Underground Space in Hong Kong", the relocation of Sha Tin Sewage Treatment Works to caverns will be taken forward to release such sites for housing and other uses (DSD, 2012). In the meantime, Hong Kong has adopted seawater toilet flushing as a

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way of reducing freshwater consumption since the late 1950's (Tang et al., 2006). This project will serve as a crucial reference in demonstrating the future of sewage treatment in highly populated coastal cities despite of the extra challenges posed by the treatment of saline sewage and the requirement of a highly compact system in capture.

Several years ago, a sulfate reduction, autotrophic denitrification, nitrification integrated (SANI) process was developed to treat saline sewage, while keeping sludge production low by integrating the sulfur cycle into the carbon and nitrogen cycles in the wastewater treatment process (Wang et al., 2009). In the SANI process, the sulfate-reducing upflow sludge bed reactor acts as the first and core bioreactor for organic removal. Under a nominal hydraulic retention time (HRT) of 4 h, the reactor is capable of an average organic removal rate of 84%, and provides total dissolved sulfide of 44 mg S/L for subsequent autotrophic denitrification (Lu et al., 2012). Because of the difficulties in concentrating the soluble organic fraction, the challenge with treating dilute organic streams

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mainly lies in the short retention time, which limits the compactness of treatment system (McCarty et al., 2011). To improve the organic loading capacity and system resilience against fluctuations (e.g. changes in flow, composition, and ambient temperature over time), sludge immobilization has been adopted in methanogenic upflow anaerobic sludge bed (UASB) reactors (Lettinga et al., 1980; Rebac et al., 1995) as well as in the SANI process. Operating a GSRUSB reactor over a period of 368 days, Hao et al. (2013) achieved an average organics removal efficiency of 80% and the highest organic loading rate (OLR) of 6.4 kg COD/m³·day (with an HRT of 1 h).

However, the issue of greater concern with upflow anaerobic granular sludge bed reactors is that when granules come into contact with wastewater of low organic strength, problems including preferential flows and dead zones emerge (van Haandel et al., 2006). This universal phenomenon of sludge bed clogging can lead to a reduction in the volume of active biomass and disappointing reactor performance (van Haandel and Van Der Lubbe, 2007). Even though some mitigation measures have been proposed, such as effluent recirculation, a pre-acidification step, or controlling the amount of sludge (van der Meer, 1979; Seyfried, 1988; Pereboom and Vereijken, 1994), sludge bed clogging is still not completely eliminated and often leads to higher costs in both setup and operation (Henze, 2008). Unlike in the methanogenic UASB, dissolved sulfide is produced as the major energy yield in the GSRUSB near neutral pH. With respect to the degree of mixing without biogas production, the GSRUSB reactor has a higher chance of encountering sludge bed clogging than the methanogenic reactor.

Hence, the overarching goal of this study was to develop a novel approach to resolve the problem of sludge bed clogging in a GSRUSB reactor by applying intermittent gas sparging for disaggregation of clogged sludge so as to develop a high-rate GSRUSB reactor in the SANI process or other sulfate reduction-based biotechnology for wastewater treatment. On the other side, it could serve as an important study for examining the possible limiting factors in conventional upflow anaerobic granular sludge bed reactors having a low biogas production rate.

#### 2. Materials and methods

#### 2.1. Laboratory-scale GSRUSB reactor

A lab-scale GSRUSB reactor, coupled with a recirculation unit, was fabricated from a plexiglass column having an internal diameter of 55 mm, giving a total working volume of 2.0 L (Fig. S1) (Supplementary material). The anaerobic biomass was originally obtained from a local anaerobic sludge digester. Using the same granulation approach adopted by Hao et al. (2013), mature granular sludge with an average particle diameter of 360 um was first cultivated, before being seeded into the reactor for operation in this study. An effluent recirculation ratio (recirculating effluent to feeding influent) of 1.4 was adopted in this study while a ratio of 5 was previously applied by Hao et al., 2013. Inside the GSRUSB reactor, intermittent inert gas (i.e. nitrogen gas) sparging at intervals of 10 s per hour was applied (at a gas flow rate of 40 mL/s), and a plate separator was installed to prevent excessive biomass washout during the gas sparging. Besides, the recirculation unit served as the function of external settler for the retention of washout granular sludge during the gas-sparging. The ambient temperature in the laboratory was  $22 \pm 1$  °C.

The GSRUSB reactor was fed with synthetic saline sewage according to previous research (Wang et al., 2009; Hao et al., 2013). Seawater (with an average sulfate concentration of 2.7 g/L) acted as the major sulfur source. Acetate, glucose and yeast were used as the

carbon source. The mixture of seawater, stock solution (organic carbon and trace nutrients) and tap water yielded average influent concentrations of 330 mg COD/L, 540 mg  $SO_4^{2-}/L$  (or 180 mg S/L), and 30 mg NH<sub>4</sub>-N/L, and a pH of 7.2. The GSRUSB reactor was operated continuously for 196 days without discharge of sludge. Four operational stages with different organic loading rates are summarized in Table 1. Stage 5 acted as the control stage without applying intermittent gas sparging.

#### 2.2. Physical and chemical analysis

The mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) were determined according to Standard Methods (APHA, 2005). Total organic carbon (TOC) was analyzed instead of chemical oxygen demand (COD) to prevent chloride and sulfide from interfering with the saline samples. A conversion ratio between TOC and COD was determined to be 2.78 in this study. Dissolved sulfide was measured by the methylene blue method (APHA, 2005), while pH and temperature were determined using a multimeter electrode (WTW Multi3420). Sulfate ions were analyzed using an ion chromatograph (HIC-20A Super), which was equipped with a conductivity detector and an IC-SA2 analytical column.

The physical properties of the sludge were characterized by the sludge volume index (SVI) and particle size distribution (PSD). Sludge volume indices at 5 min and 30 min (SVI<sub>5</sub> & SVI<sub>30</sub>) were measured in a 1-L graduated cylinder. The size distribution of the sludge was determined using a laser diffraction particle size analyzer (LSI3 320, Beckman Coulter). Extracellular polymeric substances (EPS) were extracted from the granular sludge using a formaldehyde-NaOH method (Liu and Fang, 2002). The content of polysaccharides (PS) in EPS was then quantified using the phenolsulfuric acid method with glucose as the standard (Dubois et al., 1956). The content of protein (PN) in EPS was determined by a modified Lowry colorimetric method (DC Protein Assay, BioRad) with bovine albumin serum as the standard (Frølund et al., 1995). The total quantity of the extracted EPS was determined from the weight of the lyophilized solids. The spatial distribution of EPS content in the granules was visualized microscopically according to the method described by Chen et al. (2007).

#### 2.3. Three-dimensional computational fluid dynamics modeling

A three-dimensional (3-D) computational fluid dynamics (CFD) model was applied to characterize the hydrodynamic conditions inside the GSRUSB reactor (Table S1) (Supplementary material). In the simulation, the wastewater flowed into the GSRUSB reactor uniformly through the 25 inlets at its bottom, with a liquid velocity of 0.58 cm/s for each inlet (Table S2). The liquid in this gas-liquid-solid three-phase model was assumed to be a Newtonian fluid (Le Moullec et al., 2008). The set of momentum and continuity equations for each phase was subsequently solved using the Eulerian—Eulerian multiphase model, which is commonly applied in

Summary of the reactor performance under various stage.

Stage	1	2	3	4	5
HRT	4-hr	2-hr	1-hr	40-min	1-hr
OLR (kg COD/m³day)	1.95	4.02	8.91	13.31	8.64
COD removal (%)	86.3%	90.8%	92.4%	90.9%	55.3%
Sulfide (mg-S/L)	122.2	164.7	177.6	171.4	62.3
Volumetric activity (gSO4 <sup>2-</sup> L <sup>-1</sup> d <sup>-1</sup> )	4.4	11.86	25.57	37.02	8.97
$\Delta$ COD/ $\Delta$ S ratio	2.42	1.85	1.94	1.98	3.12
PS/PN ratio	0.88	0.81	0.98	0.82	0.27

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