Rheological behaviors of anaerobic granular sludge in a spiral symmetry stream anaerobic bioreactor
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ABSTRACT

The rheological behaviors of the anaerobic granular sludge (AGS) in a spiral symmetry stream anaerobic bioreactor (SSSAB) were investigated. Shear-thinning behavior, thixotropic behavior, concentration–viscosity behavior and temperature–viscosity behavior were evaluated based on the constitutive equation of the AGS. The results indicated that the Herschel-Bulkley model was able to adequately describe the constitutive relation of AGS in the SSSAB. The AGS also showed shear-thinning behavior as well as thixotropic behavior. The critical shear rate and network strength of the AGS were 61.8 s⁻¹ and 497.0 W m⁻³, respectively. The relationship between the apparent viscosity and the sludge concentration was illustrated and explained by the Woodcock formula. The relationship between apparent viscosity of the AGS and temperature could be modeled using the Arrhenius equation. The AGS was significantly thermo-sensitive and its mean energy of activation was 14.640 kJ mol⁻¹. Notably, it was necessary to consider such behaviors in the hydrodynamic modeling of SSSAB in which shear condition, sludge concentration and temperature were in non-uniform distribution.

INTRODUCTION

Anaerobic bioreactor (AB) technology achieved rapid development in recent years. In the late 1970s, as the representative of the second generation ABs, the upflow anaerobic sludge blanket (UASB) process (Lettinga et al. 1997), developed by Dr Gatz Lettinga et al., has been widely used in practical wastewater treatment. Since then the emergence of the third generation ABs including expanded granular sludge bed (EGSB) and internal circulation (IC) (Pereboom & Vereijken 1997) anaerobic reactors, have been receiving broad attention, due to their advantage of relatively improved sludge retention ability, high volume load, and so on (Lettinga et al. 1997). It is noteworthy that the technological innovation of high-rate ABs contributes to the enhancement of anaerobic granular sludge (AGS) keeping capacity. Therefore, AGS keeping capacity is one of the key factors of the high-efficiency of ABs.

A patent bioreactor, Spiral Symmetry Stream Anaerobic Bioreactor (SSSAB) (Patent number: ZL201,210,054,218.6, China), was invented by our research group in recent years. The reactor’s sludge bed is spirally and symmetrically divided into three chambers by three elliptic plates, which helps to maintain the stability of the AGS (functional bacteria) and to strengthen mass transfer between AGS and the substrate. In addition, each chamber is provided with a gas collection pipeline to collect up-moving biogas in time and then reduce the effect of the bubble wake stream entrainment, enhancing SSSAB’s retention capacity of AGS.

AGS is a kind of non-Newtonian fluid (Pevere et al. 2000), and its rheological behaviors might also be greatly influenced by many elements, such as substrate composition, substrate concentration, hydraulic retention time (HRT), pH, temperature, etc. (Hulshoff Pol et al. 2004) It impacts not only the design and operation of a bioreactor, but also mass transfer between AGS and the substrate and the release of generated biogas from aqueous phase to gaseous phase (Yang et al. 2007). The rheological behavior of AGS is therefore a significant factor influencing anaerobic process.

As a powerful tool for characterizing the non-Newtonian properties of sludge suspensions, rheology can quantify flow behaviors on a scientific basis (Dentel 1997). Since the
1960s (Dick & Ewing 1967), rheological parameters have been gradually applied to the research of sludge in the field of wastewater treatment: the area of the hysteresis loop was used as an important parameter for indicating the overgrowth of filamentous bacteria species in activated sludge (Guibaud et al. 2005). Limiting viscosity and yield stress were regarded as two parameters for guiding the sludge transport and sludge dewatering process (Tixier et al. 2005). Additionally, sludge network strength was considered as a measure of sludge cohesion (Niraula et al. 2004).

However, as we all know, most research has focused on the rheology of activated sludge and aerobic granular sludge in the literature (Ratkovich et al. 2013; Sanin 2004; Yang et al. 2009; Yuan & Wang 2013; Ruiz-Hernando et al. 2014). Little systematic research has been carried out in the field of rheological behaviors of AGS, especially AGS in a high-rate AB. Therefore, the main objective of this study was to explore the rheological behaviors of AGS in a SSSAB including shear-thinning behavior, thixotropic behavior, concentration–viscosity behavior and temperature–viscosity behavior based on the constitutive equation of the AGS, providing useful information to enrich rheological parameters of the AGS and further optimize the operation of SSSAB.

MATERIALS AND METHODS

Experimental set-up and operation

The schematic diagram of the experimental set-up is presented in Figure 1. The SSSAB was made of plexiglass, the configuration parameters of SSSAB were: bed (column) diameter 100 mm, bed height 800 mm, separation unit diameter 200 mm and height 220 mm, respectively. The upper and lower diameters of the three-phase separator were 50 mm and 180 mm, respectively. The available volume and bed volume of bioreactor were 4,000 mL and 2,500 mL, respectively. Wastewater was pumped into the distribution zone of the reactor (bottom) by a peristaltic pump (Figure 1(2)), then was mainly treated in reaction zone of the reactor (middle), finally leaving the reactor in the separation zone (top) via the three-phase separator (Figure 1(5)). In the SSSAB three elliptic plates were 120° spirally and symmetrically set (Figure 1(8)). So the sludge bed was divided into three chambers including top, middle and bottom part. Each chamber was provided with a gas collection pipe (Figure 1(9)) to achieve respective biogas collection.

The SSSAB was fed with synthetic wastewater containing sucrose (6,000 mg L⁻¹), NH₄Cl (530.0 mg L⁻¹), CaCl₂·2H₂O (100.0 mg L⁻¹), trace element solution according to Tang et al. (2011) (1.0 mL L⁻¹), and nutrition solution (32 mL L⁻¹), which contained beef extract 0.6 g L⁻¹, yeast extract, tryptone 1.8 g L⁻¹, KH₂PO₄ 7.54 g L⁻¹, and MgSO₄ 0.22 g L⁻¹. The NaHCO₃ was used to adjust pH value (7.0–8.0) and to satisfy alkalinity requirements according to the index of effluent. The chemical oxygen demand (COD) of influent was constantly 5,000 mg L⁻¹, and hydraulic retention time was 24 hours. The SSSAB was not equipped with insulation measures, and the environmental temperature was 10 ± 5°C, which was lower than the optimum temperature of 35°C for the middle temperature anaerobic fermentation process.

Seed sludge and testing sludge

The seed sludge was obtained from an EGSB reactor used for treating pulping and papermaking wastewater in a plant. The average diameter of the seed sludge was 1.606 mm, and its density was 1.052 g cm⁻³. Moreover, the inoculation amount of the seed sludge was approximately 2.5 L.
To ensure that the rheological characteristics of the testing sludge could well represent the rheological characteristics of AGS in the SSSAB, the testing sludge was made up of an equal quantity of samples from different sampling places (see Figure 1). The average diameter of the testing sludge was 1.426 mm, and its density was 1.060 g cm$^{-3}$. The total suspended solids (TSS) of the testing sludge was 25.2 g L$^{-1}$.

**Analysis of the granular sludge**

The granular sludge size was analyzed by a dynamic image analyzer for granule size and shape with measuring range between 0.002 cm and 0.5 cm (QICPIC, Germany). The density of the granular sludge was determined according to Xiaoguang et al. (2013). The TSS was measured according to *Standard Methods* (APHA 2012).

**Rheological measurement**

The constitutive equation, the shear-thinning behavior and the thixotropic behavior were tested using a rotational viscometer (DV-III, Brookfield Engineering Laboratories, Inc., USA). And No. 0 rotor and 500 mL beakers were selected for the tests of the above rheological behaviors. The temperature was kept constant by using a water bath. Moreover, those behaviors were tested in the steady shear mode. In the constitutive equation and the shear-thinning behavior tests, shear rate was increased linearly from 0 to 1000 s$^{-1}$ in 4 minutes, and the rheogram of shear stress as a function of shear rate was recorded for the AGS with the viscometer. In the thixotropic behavior tests shear rate was increased linearly from 0 to 1000 s$^{-1}$ in 4 minutes (ascending path was drawn meanwhile), then maintained constant at 1000 s$^{-1}$ for 1 minute, then finally decreased linearly from 1000 s$^{-1}$ to 0 in 4 minutes (descending path was drawn meanwhile).

In the concentration–viscosity behavior and the temperature–viscosity behavior tests, the viscosity was directly determined by a viscometer NDJ-5S (China) in five shear conditions, corresponding to shear rate from 0.7–8.8 s$^{-1}$.

**RESULTS AND DISCUSSION**

**The constitutive equation**

The constitutive equation of the AGS is a mathematical description to reflect the deformation of a body under the influence of stress (Ma et al. 2014). The most commonly used constitutive equations (Eshtiaghi et al. 2013) with an additional yield stress to represent the non-Newtonian behavior of sludge suspensions are the Bingham model, the Herschel–Bulkley model and the Casson model. The experimental values of shear rates and shear stress are shown Figure 2. The fitting results of three models indicate that the Herschel–Bulkley model shows the highest regression coefficient (0.998), higher than 0.996 of the Casson model and 0.964 of the Bingham model. Hence, the Herschel–Bulkley model is adopted as the constitutive equation of the AGS, that is, the constitutive equation of AGS is $\tau = 4950 + 287\gamma^{0.592}$.

As is illustrated in the constitutive equation, the value of yield stress $\tau_y$ is 4,950, which physically means beyond such yield stress the AGS exhibit fluid-like characteristics, and below such value the AGS exhibit solid-like characteristics. This feature is significant in process design and quality assessment of the sludge (Eshtiaghi et al. 2013). Moreover, the consistency index $k$ value of AGS is 287, less than 531 of a certain anaerobic digestion sludge at TSS of 25.5 g L$^{-1}$ and temperature of 283 K (Baudet al. 2013). The power-law index $n$ value of the AGS is 0.592, which means that the AGS is a kind of pseudoplastic fluid (its $n$ value is less than 1). It is notable that $n$ can represent the deviation degree from the AGS to Newtonian fluid, that is, the smaller than 1 the $n$ value becomes, the more flow character deviates from Newtonian fluid (Sanin 2004). It was studied in the literature (Baudet al. 2013) that a certain anaerobic digestion sludge displayed $n$ value of 0.308 under the same condition, indicating that anaerobic digestion sludge deviates further from Newtonian fluid than the AGS.

**The shear-thinning behavior**

As a kind of pseudoplastic fluid, the AGS behavior is shear-thinning. The apparent viscosity of such fluid is then a function of the shear rate. According to Equation (1), the apparent viscosity is directly proportional to the shear stress, inversely proportional to the shear rate. Then the shear rate-viscosity curve can be drawn (line with blocks in Figure 2) to characterize the shear-thinning behavior of the AGS.

$$\eta_{app} = \frac{\tau}{\gamma} \quad (1)$$

A rapid decrease in viscosity of liquid–solid suspensions at lower shear rates could be observed in Figure 2. In order
to determine the strongest viscosity decrease point accurately, the intersection of the two regression lines for low and high ranges of shear rates was calculated and a critical shear rate of $61.8 \text{s}^{-1}$ ($\gamma_{\text{crit}}$) was obtained. If the shear rates increased below this value, the viscosity would dramatically decrease. Conversely, the viscosity would decrease slowly if the shear rates increased beyond this value.

It can also be seen from Figure 2 that apparent viscosity became constant at a higher shear rate, which was called limiting viscosity ($\eta_\infty$) at the infinite shear rate (Ratkovich et al. 2015). The $\eta_\infty$ of the AGS at TSS of 25.2 g L$^{-1}$ and temperature of 283 K was 22.1 mPa·s, which was higher than the $\eta_\infty$ of the sludge in a EGSB reactor treating brewery wastewater and the sludge in a UASB reactor treating paper-making wastewater under the same conditions (Pevere et al. 2000).

### The thixotropic behavior

The thixotropic behavior refers to the time-dependent disintegration of internal structure as a result of the application of shear stress (Ratkovich et al. 2015). It was reported by the research of Mu & Yu (2006) that the sludge in a UASB reactor showed thixotropic behavior. As is demonstrated in Figure 2, the shear stress is higher on the ascending path than descending path, attributing to the thixotropic properties of the AGS.

In order to quantify the thixotropic behavior of the AGS, the network strength has been adopted. There exists intertwined network structure in AGS, and the network strength is a kind of energy required to break down such structure. With increasing energy, network strength gets larger. Moreover, the area of the hysteresis loop, or the area between the ascending path and the descending path (Figure 2), could be taken as a measure of the degree of network strength of the AGS (Yen et al. 2002), which could be evaluated as follows:

$$A = \int_0^t \tau d\gamma(t)$$

where $t$ (s) is the time that shear force exert on AGS.

The area of the hysteresis loop in Figure 2 was 497.0 W m$^{-3}$ on the basis of Equation (2). It suggests the energy required to break down network structure of 1 m$^3$ AGS in the SSSAB was 497.0 W. This value is obviously larger than 100.0 W of activated sludge reported by others (Guibaud et al. 2005; Tixier et al. 2003), indicating that the AGS owns higher network strength than activated sludge. The high network strength of the AGS is probably due to the formation of dense granules depending on both hydraulic and pneumatic scour.
The concentration–viscosity behavior

The viscosity, including apparent viscosity and limiting viscosity, has been widely used as a parameter for characterizing sludge rheology (Eshtiaghi et al. 2015). It was found that the limiting viscosity of sludge increases with solids content, or TSS (Mu & Yu 2006). However, in order to acquire the limiting viscosity of the AGS, it required extremely high shear rate, which commonly could not be achieved in daily operation of anaerobic reactors. The shear rate in the anaerobic reactor was about 8 s⁻¹ (Pereboom & Vereijken 1994; Wu et al. 2009), therefore the concentration–viscosity behavior for a range of shear rates of 0.7–8.8 s⁻¹ was investigated. Figure 3(a) represents variations of apparent viscosities of the AGS at different TSS and T of 283 K. When TSS was 50.6 g L⁻¹ and shear rate was 2.0 s⁻¹, the highest apparent viscosity of the AGS could reach 38.4 mPa s. Figure 3(a) also clearly showed a linear increase (R² greater than 0.97) in apparent viscosity at a TSS concentration of 25.2–50.6 g L⁻¹ when shear rates were 0.7–8.8 s⁻¹. Considering the AGS has high fraction of suspension and interact with each other, the tendency can be explained based on the Woodcock formula (Zuo et al. 2011), which gives the average distance h (mm) between AGS in terms of the particle size d (mm) and the volume fraction φ (%) as follows:

\[
h = \frac{1}{3\pi\phi} + \frac{5}{6} - 1\]

in which

\[
\phi = \frac{TSS}{\rho \times 1000} \times 100\%
\]

where \( d = 1.426 \) mm; the unit of TSS is g L⁻¹; \( \rho \) is the density of AGS, 1.06 g cm⁻³.

From Equations (3) and (4), it can be calculated that the average distance between the AGS decreases from 1.854 to 1.068 mm when TSS increases from 25.2 to 50.6 g L⁻¹. This attributes to an increase of particle–particle interactions. Hence the flow resistance between the AGS also increased as well, suggesting the increase of the apparent viscosity of the AGS.

The temperature–viscosity behavior

Temperature in an anaerobic reactor, another important factor affecting the rheological properties of the AGS, usually varies with seasonal change if insulation measures are not effective enough. Therefore, the influence of temperature on the apparent viscosity of the AGS was studied. As we can see from Figure 3(b), an increase in temperature results in a decrease of apparent viscosity in different shear rates (0.7–8.8 s⁻¹), because the thermal motion of the AGS is more violent at a higher temperature, and then the network strength between the AGS is weakened, leading to a decrease in viscosity (Mu & Yu 2006). The influence of temperature on the apparent viscosity of the AGS could be described by an
Arrhenius type equation

\[ \eta_{\text{app}}(T) = Ke^{E_a/RT} \]  

where \( \eta_{\text{app}} \) (mPa-s) is the apparent viscosity of AGS at temperature \( T \), \( K \) is the pre-exponential factor, \( E_a \) (J·mol\(^{-1}\)) is the energy of activation for viscosity, and \( R \) (J·mol\(^{-1}\)·K\(^{-1}\)) is the universal gas constant. Taking the logarithm (base 10):

\[ \lg \eta_{\text{app}}(T) = \lg K + \frac{E_a}{2.303RT} \]  

Equation (6) is a linear equation, in which the slope is \( E_a/2.303R \) and the intercept is \( \lg K \). Hence the energy of activation of the AGS could be computed in different shear conditions. Modeled by Equation (6), the mean pre exponential factor and the mean energy of activation are calculated for 0.212 mPa s and 14.640 kJ mol\(^{-1}\), respectively. The high correlation coefficient value (all greater than 0.95) indicates that the temperature affected the rheology of the AGS according to the Arrhenius equation. Furthermore, Liu et al. (2009) found that the mean energy of activation of duck manure, cattle manure and chicken manure were 8.018 kJ/mol, 11.337 kJ/mol and 8.285 kJ/mol, respectively. It was suggested (Liu et al. 2009) that the bigger \( E_a \) of fluid represents the greater thermo-sensitivity for such fluid, so we can deduce that the AGS is more sensitive to thermo-change than livestock manure.

CONCLUSIONS

The Spiral Symmetry Stream Anaerobic Bioreactor is a kind of novel anaerobic reactor. The Herschel–Bulkley model was able to adequately describe the constitutive relation of AGS in a SSSAB. The AGS showed shear-thinning behavior as well as thixotropic behavior. The average distance between the AGS decreases from 1.854 to 1.068 mm while TSS increases from 25.2 to 50.6 g L\(^{-1}\), leading to an increase of the apparent viscosity of the AGS. Furthermore, the apparent viscosity of the AGS decreased with increasing temperature. The mean energy of activation of the AGS was 14.640 kJ mol\(^{-1}\).

Due to the non-uniform distribution of shear condition, sludge concentration and temperature in SSSAB, it is necessary to consider such behaviors in its hydrodynamic modeling, which functions as an indicator on the design of larger scale SSSAB.

ACKNOWLEDGEMENTS

The authors would like to thank the National Natural Science Foundation of China (Grant no. 12ZR1208087), the Shanghai Natural Science Foundation of China (Grant no. 12ZR1400800), Doctoral Fund of Ministry of Education of China (New Teachers) (Project no. 20120075120001), and the Fundamental Research Funds for the Central Universities.

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First received 28 February 2015; accepted in revised form 7 May 2015. Available online 25 May 2015