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Enhanced membrane-based pre-concentration improves wastewater organic matter recovery: Pilot-scale performance and membrane fouling

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A B S T R A C T

Energy neutrality in municipal wastewater (MW) treatment is an ambitious sustainability goal that is hampered by MW's low organics concentration. A potential method is to pre-concentrate MW to recover the organics and use them to produce energy via anaerobic decomposition. This study conducted a pilot-scale operation using an enhanced membrane-based pre-concentration (MPC) technique to efficiently recover organics from actual MW. Stable and highly efficient organics recovery (72–75%) was achieved during two-month operations. Concentrated organics demonstrated high COD (above 20,000 mg/L) and good anaerobic biodegradability (360–367 mL CH_4/g VS). Organics recovered by MPC exhibited the potential of net electrical energy production of 0.029–0.050 kWh/m^3, depending on the fouling situation. The energy consumption of MPC was limited by avoiding continuous intensive aeration. With effective fouling control, as much as 2.91 times more energy could be produced than that was consumed. This suggests MPC-based processes have the potential to transform MW treatment plant from energy consumers to net producers.

1. Introduction

Energy neutrality is one of most ambitious sustainability-related goals for future municipal wastewater (MW) treatment (Gong et al., 2018; Mccarty et al., 2011). Besides decreasing energy consumption by using advanced facilities and energy-efficient processes such as autotrophic deammonification, the production of energy from organic matter contained in MW has also become a subject of research. Anaerobic methane (CH_4) production has been proposed as the most practical method for recovering energy from organics (Stuckey, 2012). However, low organics content in MW, measured in terms of chemical oxygen demand (COD), generally limits its direct anaerobic CH_4 generation, making it economically unfeasible. Recently, organics pre-concentration technology, which improves organics recovery for post-anaerobic energy production, is a potential solution to this problem and has received plenty of research attentions (Behera et al., 2018; Ge et al., 2017; Gong et al., 2016; Graaff et al., 2016).

Organics pre-concentration technologies, which include both biological and physical-chemical processes, have been proposed in recent years. They include bio-flocculation (the A stage of the AB process, also mentioned as high-rate activated sludge) (Meerburg et al., 2016) and chemically-enhanced primary treatment (CEPT) (Lin et al., 2017). The "A stage" of bio-flocculation has a short sludge retention time (SRT) and high COD load. It makes use of spontaneous biological flocculation and adsorption to separate organics following a sedimentation process. The stability of the A stage process has become well known since its invention in 1970s of last century. It has been applied recently as an organics pre-concentration step at the Strauss wastewater treatment plant (WWTP) in Austria, which has achieved complete energy self-sufficiency, demonstrating the usefulness of organics recovery in WWTP energy neutrality (Meerburg et al., 2016; Wett et al., 2007). However, although the hydraulic retention time (HRT) is limited to a short period (0.5–2 h), aeration is still required in the A stage process, indicating much energy consumption and organics are lost due to the mineralisation of aerobic microorganisms (Rahman et al., 2016). Furthermore, the organics recovery efficiency of the A stage process is moderate due to limited separation in the sediment tank (~45% for the full-scale Strauss WWTP) (Wett et al., 2007). The issue of limited organics recovery efficiency is similar for coagulation

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processes such as CEPT.

In comparison, membrane filtration, particularly porous membrane including microfiltration (MF) and ultrafiltration (UF), avoids intensive aeration and has high separation efficiency. Hence, it has been proposed as an effective and compact process for recovering organics from dilute MW (Gong et al., 2015; Hey et al., 2017; Kimura et al., 2017). The main concern of the membrane process is fouling, which reduces flux and increases energy consumption (Leal et al., 2010; Nascimento et al., 2018). Moreover, organics exist in the forms of suspended, colloid and soluble materials. Thus, a certain proportion of soluble organics could not be recovered by a single membrane process. The higher the proportion of organics recovered by the pre-concentration process, the more energy self-sufficiency ratio is expected to be achieved. Highly-efficient organics recovery is essential, especially for dilute MW. To solve these issues, an enhanced membrane process that incorporates coagulation and adsorption with membrane filtration, known as enhanced membrane-based pre-concentration (MPC), has been investigated by several studies (Gong et al., 2016; Huang et al., 2017; Jin et al., 2016). Coagulation mainly removes particulate and colloidal organics, and alleviates fouling and cake layer formation. Properties (such as particle size) change during coagulation, causing variable fouling potentials (Mao et al., 2013). Adsorption promotes the pre-concentration of soluble organics and mitigates fouling by reducing the membrane pore clogging that is induced by soluble pollutants (Liu et al., 2016). Previous literature has demonstrated the feasibility of the MPC concept. However, most research has been conducted in small-scale reactors with synthesised wastewater. Long-term investigations of the performance of MPC-based organics recovery from actual MW are lacking. During organics pre-concentration, the situation that the filtration membrane faces is quite different from conventional membrane technology such as membrane bio-reactors (MBRs), which are dominated by biological processes with longer SRT and HRT. Thorough analyses of organics concentrates and their impacts on membrane fouling are also required.

In this study, the long-term performance of MPC-based organics recovery from dilute actual MW was investigated by large-scale pilot experiments. The recovery efficiency of organics and other nutrients (N and P) under various operational conditions, including SRT and chemical additions, were evaluated. The biological and physical properties of organics concentrates were analysed for their impacts on anaerobic degradability and membrane filtration. The issue of fouling during different operating conditions and control strategies was discussed. Finally, the energy balance and trade-offs of the MPC process were estimated.

2. Materials and methods

2.1. Municipal wastewater and membrane-based pre-concentration reactor set-up

Actual MW was collected after coarse filter from the Xiaojiahe municipal WWTP in Beijing, China. The pilot-scale MPC reactor contained a 24 m^2 hollow-fibre PVDF (polyvinylidene fluoride) submerged membrane module with averaging 0.02 μm pore diameter (Litree, China). The membrane module was composed of 4 curtains with about 5200 filaments in total. The membrane filament dimension was 1.00/2.00 mm for inner/outer diameter with 150 k Dalton molecular weight cut-off. The effective volume of the reactor was 280 L and its dimensions were 680 × 500 × 1720 mm (length × width × height). To enhance membrane performance, polyaluminium chloride ($[Al_2(OH)_nCl_{6-n}]_m$, PACl) and powdered active carbon (PAC, iodine value > 1000; 0.45–0.55 g/cm^3 apparent density; averaging particle size was 26.47 μm) were added as coagulants and adsorbents based on previous research (Jin et al., 2015), respectively. Every operational filtration cycle lasted for 12 min, including 10 min permeation and 2 min membrane relaxation. In order to alleviate membrane fouling, very rapid air backflushing (25 s) was applied during the 2 min relaxation period to remove cakes on the membrane surface (Jin et al., 2015). There was no additional stirring in the vessel. Flux and operational pressure were recorded automatically by electromagnetic flow meter (RHLD, China) and pressure sensor (PT124B, China, Measurement range of −0.1 MPã0.1 MPa). The hydraulic permeability *Lp* was used and defined as $J/\Delta P$ to evaluate the impacts of the extent of the concentration process on trans-membrane pressure (TMP) and flux simultaneously, where J is the flux of filtration ($L/(m^2\cdot h)$, LMH), and P is the TMP (bar). The COD, TN, NH_4^+–N and TP of the influent, permeate and concentrate were measured and recorded periodically.

The pilot experiment lasted for approximately 1600 h with two conditions of chemical dosage. As shown in Table 1, Condition I added 30 mg/L PACl and 20 mg/L PAC (Jin et al., 2015), while Condition II reduced the PAC dosage to 10 mg/L. During the first stage of Condition I (I-1), no concentrates were discharged from the MPC reactor, namely the SRT was infinite. In the second stage (I-2), about 70 L of concentrate were discharged per day with 4 d SRT. In the first stage of Condition II (II-1), the SRT was kept at 4 d and was reduced to 3 d in the second stage (II-2). After Condition I, the reactor was chemically cleaned (immersion in 350 mg/L NaClO solution for 36–48 h).

2.2. Analysis

The COD, TN, TP and NH_4^+–N of influents, permeates and concentrates were determined by colourimetric techniques using a HACH spectrophotometer (DR 5000, HACH, USA). Mixed liquid suspended solids (MLSS) were determined according to the Chinese NEPA standard methods [20]. The pH was measured using a pH meter (Sension 1, HACH, USA) and viscosity was measured using a viscosimeter (DV1, Brookfield, USA). Particle size distribution was evaluated by a grain size analyser (Mastersizer 2000, Malvern, UK). The biological methane potential (BMP) test was utilised to evaluate the methane production potential of the concentrated organics obtained from the municipal wastewater by enhanced membrane-based pre-concentration. Commercialized BMP facility was used for test (AMPTS II, Bioprocess Control, Sweden). The inoculum was digested sludge taken from sludge digester of wastewater treatment plant (Angelidaki et al., 2009). The temperature was maintained at 35 ± 1 °C. Concentrated organics in form of sludge obtained after Condition I and II was used as BMP feed respectively. The biodegradability ratio was defined as the ratio between the measured BMP and the theoretical methane potential (350 mL CH_4/g COD or 497 mL CH4/gVS with assumption of 1 g VS equal to 1.42 g COD) (Mottet et al., 2009).

3. Results and discussions

3.1. Recovery of organics and other nutrients

Enhanced COD removal and C/N separation were observed during the MPC process. As shown in Fig. 1, though influent COD fluctuated, the long-term pilot MPC reactor demonstrated high COD rejection with stable effluent. The average COD during the whole period was 231 ± 34 mg/L, while effluent COD was 32.7 ± 6.4 mg/L with removal rates as high as 85.6 ± 3.5% (Table 2). The outflow COD proportion was mainly soluble, while particulate and colloidal COD was retained in the concentrates by the membrane rejection.