A novel anaerobic digestion system coupling biogas recirculation with $MgCl_2$ addition for multipurpose sewage sludge treatment

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Abstract

Sewage sludge, one of the major byproducts from wastewater treatment plants (WWTPs), contains high contents of nutritional elements that can be recycled and reutilized to make up the overuse of limited resources on Earth. Currently, the reutilization ratio of sewage sludge is relatively low, most probably due to the high operation costs for its treatment and disposal in WWTPs. This study for the first time realized biogas upgrading, resources conservation and sludge conditioning simultaneously in one anaerobic digestion (AD) reactor for sewage sludge treatment through coupling biogas recirculation with $MgCl_2$ addition. Results showed that 86% of methane content could be obtained by the intermittent biogas recirculation during AD. When $MgCl_2$ was further added, soluble orthophosphate and ammonia nitrogen were reduced by 87% and 19% during 17 days' AD. At the same time, around 120 mg/g-total solids (TS) of struvite was estimated to generate in the digested sludge, and the sludge dewaterability was enhanced by 37% when biogas recirculation was performed along with $MgCl_2$ addition. At last, not only the enhanced electricity generation from biogas with higher methane content but also the increased fertilizer potential of the digestate were analyzed in the context of a local WWTP, reflecting the great potentials and profits of this novel AD system in industrial-scale sewage sludge treatment.

Keywords: Anaerobic digestion; Biogas upgrading; Biogas recirculation; Struvite; Nutrients recovery; Sludge dewaterability

1. Introduction

Activated sludge process has been widely applied in wastewater treatment plants (WWTPs) where the disposal of one of their major byproducts, sewage sludge, accounts for about half of the total operation cost. According to the latest data released by the Ministry of the Environment (MOE) in Japan, around 167.32 million tons of sludge were generated in 2016, occupying about 43% of the total industrial wastes (MOE, 2019). In Japan, the reutilization ratio of sewage sludge reached 73% (dry solid basis) in 2017; however, only 32% was utilized as biomass (biogas, biofuel and land application) (MLIT, 2017). Landfill is still the major disposal method for sewage sludge worldwide (Zhang et al., 2017). In fact, sewage sludge contains high concentrations of chemical oxygen demands (COD), nitrogen (N) and phosphorus (P) resources that can be recovered as energy and nutrients. With the increasingly stringent regulation and large amount of sewage sludge production, both environmental and economic aspects should be considered in the real world of sewage sludge management.

Anaerobic digestion (AD) is commonly applied for sludge treatment and stabilization in the WWTPs, since the organic compounds contained can be decomposed for biogas (mainly CH_4 and CO_2) production, which can be further used for heating or electricity generation to serve industries. Moreover, the digestate can be utilized as fertilizer (Siebielec et al., 2018). In the past decades, various pretreatment methods have been attempted to enhance biogas production during AD (Cesaro et al., 2014; Li et al., 2018; Liu et al., 2019; Wang et al., 2019; Zhang et al., 2019). The produced biogas, however, still contains large amount of CO_2 (40-50%), lowering its heating value. The general processes for biogas upgrading require high energy consumption and investment cost that usually cannot be compensated by the produced CH_4 amount in small-scale industries (Miltner et al., 2017; Sahota et al., 2018). Recently, a pilot-scale in-situ methane enrichment method was examined by recirculating digestate in an AD tank after CO_2 being desorbed by air stripping, achieving 87% of CH_4 content with a CH_4 loss of 8% (Nordberg et al., 2012). However, when taking the additional equipment for CO_2 desorption and energy consumption for digestate pumping into consideration, biogas recirculation may be more economically feasible to achieve the similar targets.

The concept of biogas recirculation (or sparging or lift) has been utilized in the internal circulation (IC) reactor system for wastewater treatment, which can produce high quality biogas (80% CH_4) and generate well settleable granular sludge (Pereboom and Vereijken, 1994). Also, biogas recirculation has been used as a mixing alternative for enhanced biogas production from AD reactor (Sr et al., 1995; Sánchez-Hernández et al., 2013). At a biogas recirculation rate of 24.14 L/d, Siddique et al. (2015) obtained 29% increase in methane yield during the anaerobic co-digestion of petrochemical wastewater with activated manure. The results from Karim et al. (2005a) indicated that the biogas recirculation at 1 L/min could significantly improve the methane production from animal waste at a higher organic solid loading (3.24 g-VS/(L·d)). However, up to now, little information could be found in the literature on the changes of sludge properties and CH_4 content through biogas recirculation in the AD system of sewage sludge. As it is known,

CO_2 is 40 times more soluble in water than CH_4, thus biogas recirculation inside the AD reactor could result in the supersaturation of CO_2 in the sludge liquor, and then the increase of CH_4 content in the biogas (Sr et al., 1995; Latha et al., 2019).

In addition, the liquid phase of digested sludge contains high concentrations of orthophosphate (PO_4^{3-}) and ammonium (NH_4^+). If the liquid phase was returned to aeration tank(s) after sludge dewatering process, the wastewater treatment duration would be prolonged due to the increased N and P loadings, thus increasing the operation costs of the WWTP. Nevertheless, they are also the main essential nutrients for plant growth. As it is known, forced struvite ($MgNH_4PO_4 \cdot 6H_2O$) precipitation can be applied in industries as a side stream for simultaneous P and N recovery from the liquid digestate by adding Mg salt in an additional facility under the optimum conditions, and the recovered struvite is regarded as a slow-releasing fertilizer (Yetilmezsoy et al., 2017). Besides, Mg^{2+} addition is also helpful for the enhancement of sludge dewaterability due to the effectiveness of multivalent cations in binding with negatively charged sludge particles or biopolymers (Zhai et al., 2012). To further reduce the operation cost for sustainable sewage sludge treatment, Mg salt was directly added into the AD reactor in this study for simultaneous nutrients recovery and sludge dewaterability enhancement. Most recently, Tarragó et al. (2018) reported that suspended solids favored the aggregation of struvite crystals, who further claimed that the presence of solids was beneficial for meeting crop demands and restoring soil fertility. Thus, adding Mg salt directly into sludge might be a better choice for nutrients recovery targeting land application. Restated, little information is available on the changes of sludge properties and resources conservation when coupling biogas recirculation with Mg salt addition, which is crucial for the sustainable management of sewage sludge in WWTPs.

This study is expected to establish a novel AD system for sewage sludge treatment, aiming to increase methane content in the biogas, maintain highly bioavailable resources (especially P) in the treated sludge, and enhance sludge dewaterability to serve multiple purposes. The effects of biogas recirculation and Mg salt addition on biogas production, CH_4 content, resources conservation and sludge properties were explored using batch AD experiments. $MgCl_2$ was chosen as the Mg salt in this study due to its characteristics of neutral pH and high solubility that could avoid other phenomenon in the complicated AD system, thus less impact on the experimental results.

2. Materials and methods

2.1 *Sludge samples*

Concentrated primary sludge (PS), sampled from the Shimodate WWTP in Chikusei, Ibarak, Japan was used as the feedstock for AD in this study. Sludge samples were stored in 20 L polypropylene tanks at 4°C in laboratory. Anaerobically digested sludge (DS) was sampled from the anaerobic digestion tank in the same WWTP, which is operated under mesophilic condition and fed with the mixture of the concentrated PS and the excess sludge from the secondary sedimentation tank. Before being used as the inoculum in the AD system in this study, the sampled DS was incubated by adding the concentrated PS at a ratio of 4:1 (v/v) at 37±2°C in the laboratory for one week.

2.2 *Establishment of AD system and preparation for batch tests*

Four identical reactors as shown in Fig. 1 were fabricated, each with a working volume of 0.7 L (D×He=8.5 cm × 12.34 cm) and being connected to a 1 L graduated cylinder. The produced biogas was collected in an inverted cylinder, and the biogas volume could be read directly from the scale on the cylinder. Saturated $NaHCO_3$ solution was prepared and used in the biogas collection part to avoid the dissolution of CO_2 in water. After biogas production became stable, biogas recirculation was started at a constant flowrate controlled by a gas pump and a gas flowmeter. Then the biogas was further distributed into the AD reactor via two ceramic gas spargers with the same pore size (selected from the same batch products of the manufacturer). Before AD experiments, the reactors were tested for their gas tightness for 5 days' continuous operation of air recirculation after being filled with tap water (instead of sewage sludge). Another four reactors, with the same size but without installation of gas spargers, gas flowmeter and air pump, were also set up and used for AD without biogas recirculation.

The eight reactors were operated in parallel using batch AD experiments: 1) two control reactors (Cont.) were operated under no biogas recirculation and no $MgCl_2$ addition; 2) two reactors (+Mg) were run with $MgCl_2$ addition while no biogas recirculation; 3) two reactors (Cont.+biogasR) were under biogas recirculation while no $MgCl_2$ addition; and 4) two reactors (+Mg+biogasR) were examined under biogas recirculation and $MgCl_2$ addition, respectively.

The feedstock and the inoculum were mixed at a ratio of 2:1 (volatile solids (VS) basis). The initial total solids (TS) and VS concentrations in each reactor were about 16.04 g/L and 10.8 g/L, respectively. For the reactors with $MgCl_2$ addition, $MgCl_2$ was directly added into the mixed sludge at 1428 mg/L (15 mmol/L), according to the molar concentration of PO_4^{3-} detected in the preliminary experiments. The initial pH was adjusted to 7.00±0.02 using 4 mol/L NaOH solution. After being loaded with 0.7 L of sludge mixture, each AD reactor was purged with N_2 gas for 5 min to create anaerobic condition. All the AD reactors were placed in a temperature-controlled thermostat (37±2°C) and the AD experiments lasted for 30 days. The biogas recirculation was started from day 4 using the biogas produced by itself at a flowrate of 200 mL/min in an intermittent mode of 60 min-on/60 min-off according to our preliminary experiments. On day 17, one bottle of each condition was opened and the sludge samples were taken for parameters' determination.

2.3 *Procedures and analytical methods for general samples*

TS and VS contents, and total nitrogen (TN) and total phosphorus (TP) concentrations of the sludge samples were measured according to the standard methods (APHA, 2012). Biogas composition including CH_4 and CO_2 was analyzed by a gas chromatography (GC-8A, SHIMADZU) equipped with a thermal conductivity detector (TCD). Dissolved biogas was detected as described elsewhere (Souza et al., 2011).

To determine other parameters, 20 mL of each sample was centrifuged at 9900 rpm for 15 min. After the resultant supernatant being filtered through 0.22 μm filter membrane, the solid residue was dried at 60°C for 72 h and grounded into powder for insoluble organic carbon analysis using a total organic carbon (TOC) analyzer (TOC-V_{CSN} with SSM-5000A, SHIMADZU). Soluble organic carbon and soluble inorganic carbon in the filtrate were determined by the same TOC analyzer equipped with an ASI-V autosampler. Soluble ammonia-N was detected using the indophenol-blue method (Ivancic and Degobbis, 1984). Soluble ortho-P was measured according to the standard methods (APHA, 2012). Insoluble N, including organic N and precipitated inorganic N in the solid phase, was calculated by subtracting soluble TN from the general TN. Soluble organic N was calculated by subtracting soluble ammonia-N from the soluble TN, as very little nitrate and nitrite were detected in the filtrates.

As for P fractionation, the Standards, Measurements and Testing (SMT) harmonized procedure developed by the European Commission was used for sequential extraction of P from the solid and liquid phases (García-Albacete et al., 2012). Through this method, P can be categorized as 5 fractions, including total P (TP), organic P (OP), inorganic P (IP), non-apatite inorganic P (NAIP, associated with Al, Fe, or Mn oxides or hydroxides) and apatite P (AP, Ca-bound). NAIP and OP are deemed as bioavailable P since they can be potentially released, decomposed and utilized by plants and microorganisms.

The sample for metals content analysis was obtained by digesting the sludge with an equivalent volume of *aqua regia* solution at 150°C. After complete digestion, the volume was adjusted to its original value with deionized water. Soluble metal ions were obtained by digesting the filtrate using the mixture of HNO_3 and H_2O_2 (1:1, v/v) at 98°C. Then, the liquor containing metal ions was filtered through 0.22 μm filter membrane and quantified by inductive coupled plasma (ICP) emission spectroscopy (ICPS-8100, SHIMADZU).

The sludge samples used for surface morphology observation were taken from the Cont. and +Mg+biogasR reactors on day 30 and freeze-dried, and then characterized by scanning electron microscopy (SEM; JEOL JSM6330F, JAPAN). The existence of struvite crystals in the sludge samples were identified by X-ray diffraction (XRD, Bruker AXS D8 ADVANCE/TSM, USA).

2.4 *Determination of sludge dewaterability and settleability*

Specific resistance to filtration (SRF) and sludge volume index (SVI_{30}) were used to evaluate sludge dewaterability and settleability, respectively. SRF was determined using the vacuum filtration method as described by Wang et al. (2017). In this study, a Buchner funnel placed with a filter paper (Whatman No. 1) was used for vacuum filtration at a constant pressure of 0.07 MPa. In the determination of SVI_{30}, the sludge sample was diluted by 5 times to adjust the total suspended solids (TSS) in the range of 2000 - 4000 mg/L (Liao et al., 2001). The diluted sludge sample (after being mixed) was placed in a 100 mL graduated cylinder for 30 min settlement. Then the volume of settled sludge and the TSS of the mixed liquor were recorded and used for the calculation of SVI_{30} according to the standard methods (APHA, 2012).

2.5 *Extraction and determination of extracellular polymeric substances (EPS)*

EPS in sludge were extracted using heat extraction method (Zhen et al., 2012). In brief, the sludge sample was firstly centrifuged at 5000 rpm for 10 min, and the supernatant was collected to determine soluble EPS (S-EPS). After being washed with deionized water for three times, the sludge residue was re-suspended in 0.05% NaCl solution to its original volume. After being well shaken, it was heated at 70°C for 1 min, and then centrifuged at 5000 rpm for 10 min. The resultant supernatant was used for analysis of loosely bound EPS (LB-EPS). Later, the sludge residue was again re-suspended in 0.05% NaCl solution after being washed with deionized water for three times. After the mixture being heated at 60°C for 30 min and centrifuged at 5000 rpm for 15 min, the supernatant was collected as tightly bound EPS (TB-EPS). After these extraction procedures, all the liquid samples were filtered through 0.45 μm filter membrane. Proteins (PN) and polysaccharides (PS) contents in the EPS extracts were then quantified by using the phenol-sulfuric acid method and Lowry-Folin method with glucose and bovine serum albumin (BSA) as the standard, respectively (Dubois et al., 1956; Lowry et al., 1951).

2.6 *Statistical analysis*

All the experiments in this study were conducted in triplicate and the data were presented as their average values. One-way analysis of variance (ANOVA) was applied to analyze the statistical difference of the experimental data using Microsoft Office Excel 2016. Pearson's correlation analysis was performed by using OriginPro, Version 2018C (OriginLab Corporation, Northampton, MA, USA) to clarify the correlation between EPS and SRF. Significant difference was assumed at $p < 0.05$.

3. Results and discussion

3.1 Effect of biogas recirculation on biogas upgrading

The CH_4 contents in the reactors with biogas recirculation (+biogasR) significantly increased during AD (Fig. 2a). As shown, the CH_4 contents in all the reactors were similar before starting the biogas recirculation (day 4). After day 4, the CH_4 contents in the reactors (+biogasR) gradually increased and reached the highest (85.4-86.0%) on day 18, and then some decrease in CH_4 content was detected when the reactors were operated for a longer time. As a contrast, the CH_4 contents in the reactors without biogas recirculation kept stable, around 75% till day 30. Only addition of $MgCl_2$ showed no significant effect on CH_4 content compared with the Cont. reactor.

Fig. 2b illustrates the cumulative CH_4 production from the reactors during the 30 days' AD. Almost the same variation trend was observed on cumulative CH_4 production from all the reactors, indicating that the AD performance was not significantly affected by biogas recirculation and $MgCl_2$ addition. Similar results have been reported by Karim et al. (2005a) who claimed that biogas recirculation did not affect the methane production during AD when manure slurry was fed at low TS (5%). The cumulative CH_4 productions on day 17 amounted to 88%, 90%, 94% and 96% of the total CH_4 yield on day 30 in the reactors Cont., Cont.+biogasR, +Mg, and +Mg+biogasR, respectively. Generally, the effective biogas production duration is determined as the duration for achieving 80% of the total biogas (Huang et al., 2017). Thus, the effective biogas production duration should be shorter than 17 days for the primary sludge used in this study, which would be further shortened when biogas recirculation and/or $MgCl_2$ addition being applied. A prolonged operation duration was adopted in this study because another target of this study was to reveal the effect of biogas recirculation on CH_4 content and sludge properties.

Up to now, very few reports could be found on the obvious increase in CH_4 content through biogas recirculation during the AD process of sewage sludge. Most recently, Latha et al. (2019) reported an in-situ methane enrichment (88%) during AD with an intermittent biogas recirculation intensity of 2 L/min, which also significantly enhanced the biogas yield from co-digestion of food waste and sewage sludge at high solids content. Previous works mainly focused on the effect of biogas recirculation in a semi-continuous/continuous AD for wastewater treatment. An increase of CH_4 content from 56% to 59% was obtained by Suvajittanont and Chaiprasert (2003) during starch wastewater treatment using biogas recirculation at 14.4 L/d. The increased CH_4 content obtained in this study during the AD of sewage sludge was comparable to others. Restated, the continuous mode of AD with biogas recirculation for sludge treatment needs further investigation to confirm the feasibility of this system, since the dissolved CO_2 in sludge liquor might be again stripped out if a longer operation of biogas recirculation being applied (30 days in this study).

In order to further elucidate the carbon (C) conversion during AD, the insoluble organic C, soluble organic C, soluble inorganic C, C converted to biogas (CH_4-C, CO_2-C) and dissolved biogas C were measured and calculated. These C fractions and the pH under each condition during AD are illustrated in Fig. 3. The reduction of insoluble organic C did not show much difference among all the reactors during the experimental period. On day 17, the total organic C (TOC, soluble + insoluble) reductions were 20% (Cont.), 20% (Cont.+biogasR), 23% (+Mg) and 23% (+Mg+biogasR), in which 15-17% of TOC were detected to convert to CH_4. On day 30, the TOC reductions increased to 33%, 33%, 32% and 30% in these reactors, respectively, and the conversion ratios of TOC to CH_4 further increased to 18-19%. This phenomenon again indicated that the effective methane production duration could be shorter than 17 days, and a longer operation duration could not achieve much higher conversion of TOC in the sludge to CH_4 under the test conditions. The C as CO_2 was detected slightly lower in the reactors with biogas recirculation, decreasing by around 2% and 3% on day 17 and day 30, respectively. At the same time, the proportions of soluble inorganic C and dissolved biogas C were 12% and 11% in the reactors Cont.+biogasR and +Mg+biogasR on day 17, which increased to 14% and 13% on day 30, respectively. While little change in these two parts of C was detected in the reactors without biogas recirculation. The increased amount was comparable to the reduced amount of CO_2, implying that the reduced CO_2 in the biogas might be dissolved in the fermentation liquor or adsorbed onto the sludge particles during biogas recirculation. A higher pH value in the reactors with biogas recirculation may also be partially contributed by the increase of carbonates in the fermentation liquor. Whereas the decrease in CH_4 content after 17 days' operation (Fig. 2a) might be brought about by the escaped CO_2 which was initially dissolved or as inorganic C in the fermentation liquor, and then stripped out after reaching its saturation state in the liquid phase due to the long-term biogas recirculation.

3.2 *Nutrients conservation*

3.2.1 Phosphorus

In this study, $MgCl_2$ was added directly into the AD reactors with no additional phosphate dosage to recover P and N simultaneously. The P fractions are presented in Fig. 4 as OP, NAIP and AP in the solid phase and soluble P in the liquid phase. The sum of NAIP+OP in the solid and soluble P were defined as the total bioavailable P, and the amount of NAIP+OP in the solid was termed as solid bioavailable P. The results indicated that the total bioavailable P (87% of TP) in the sewage sludge remained almost no change before and after AD no matter whether biogas recirculation or $MgCl_2$ addition was applied or not. More specifically, the OP fraction obviously decreased on day 17 and day 30 by about 46% and 50%, respectively, probably due to the hydrolysis of OP during AD. However, the soluble P was not detected to increase correspondingly, as most of the hydrolyzed OP might be transformed into NAIP in the solid phase. Biogas recirculation was found to have slightly positive effect on the solid bioavailable P. The NAIP in the solid phase was reduced in the reactor +Mg on day 30 when compared with that on day 17, which was slightly increased in the reactor +Mg+biogasR. In addition, a slight increase of NAIP in the solid phase was also detected in the reactor Cont.+biogasR in comparison to that in the reactor Cont. on both day 17 and day 30. This observation indicates that AD process together with biogas recirculation and $MgCl_2$ addition could greatly promote P conservation in the solid phase of digested sludge. Moreover, the enhanced P conservation in the solid phase through struvite precipitation might also bring about some scaling problems in the AD tanks in practice (Ohlinger et al., 1998). Therefore, the configuration and operation conditions of the sludge treatment facilities should be further optimized to avoid scaling problems in the AD tanks and the pipelines associated with (Koga, 2019).

In short, by coupling biogas recirculation with $MgCl_2$ addition during AD process, 93% of TP could be preserved in the solid phase, in which 86% was bioavailable. Moreover, 87% and 86% of soluble P could be reduced in the reactor +Mg+biogasR on day 17 and day 30, respectively, when compared with the reactor Cont. without biogas recirculation. The removal of soluble P in this study is comparable with those obtained from the fermentation liquor through struvite precipitation. For example, Münch and Barr (2001) achieved 94% of ortho-P removal by adding $Mg(OH)_2$ at a Mg:P molar ratio of 1.3:1 to the dewatering liquid from digested sludge. Wacławek et al. (2016) reported a 81% of ortho-P removal from the liquid phase of digested sludge by adding MgO and Na_3PO_4 at a Mg:P:N molar ratio of 1:1:1.

3.2.2 Nitrogen

The changes of N species on day 0, day 17, and day 30 during AD are shown in Fig. 5. On day 17, about 19% decrease of ammonia N was detected in both reactors +Mg and +Mg+biogasR compared with the reactors Cont. and Cont.+biogasR, while the reduced amount of ammonia N in the reactor +Mg+biogasR (102.6 mg-N/L) was slightly higher than that in the reactor +Mg (97.4 mg-N/L). On day 30, the reduced amount of ammonia N was only 82.7 mg-N/L in the reactor +Mg, while it increased to 157.9 mg-N/L in the reactor +Mg+biogasR. Seen from Fig. 3, a higher pH was always detected in the reactors +biogasR, which may favor the struvite formation in these reactors. As a result, 69% and 72% of TN could be preserved in the solid phase of digestate after 17 and 30 days' AD with biogas recirculation and $MgCl_2$ addition, respectively. About 19% and 29% of ammonia N could be reduced in the reactor +Mg+biogasR, compared with those in the reactor Cont. on day 17 and day 30, respectively. This observation is comparable to the previous findings by using struvite precipitation in AD process. As pointed by Othman et al. (2009), 32% reduction in ammonia N could be achieved by adding $MgCl_2$ and Na_3PO_4 at a molar ratio of Mg:P:N =1.3:1:1 under neutral pH conditions. Uludag-Demirer et al. (2008) claimed a 23% reduction in ammonia N by adding 1750 mg/L $MgCl_2{\bullet}6H_2O$ and 3000 mg/L Na_2HPO_4 during AD at pH 7.15, with no influence on biogas production being observed.

3.2.3 Struvite formation

The formation of struvite crystals in the reactor +Mg+biogasR was confirmed by XRD and SEM (Supplementary Materials, Figs. S1 and S2). The peak positions in the XRD pattern of the sludge samples from the reactor +Mg+biogasR matched well with the reported XRD pattern of struvite crystals (Prywer et al., 2012; Chen et al., 2018). The struvite-like crystals could also be clearly noticed from the SEM image of the sludge sample from the reactor +Mg+biogasR (Fig. S1d). To estimate the quantity of the formed struvite during AD under the condition of biogas recirculation and $MgCl_2$ addition, the concentrations of soluble Mg and total Mg were measured on day 0, day 17, and day 30 (Supplementary Materials, Fig. S3a). The change in concentration of soluble Mg was consistent with the variations of PO_4^{3-} and NH_4^+. This observation again proved that biogas recirculation could accelerate struvite precipitation. The changes in molar concentrations of the increased insoluble Mg^{2+}, decreased PO_4^{3-} and NH_4^+ were calculated based on the corresponding data from the reactor Cont. on day 0, day 17 and day 30 (Supplementary Materials, Table S1). Since the Mg concentration was also noticed to decrease in the reactor Cont., the struvite concentration estimated in this study only represents the part that was increased by biogas recirculation and/or $MgCl_2$ addition during AD. In addition, the decreased PO_4^{3-} might be contributed by the precipitation with Mg^{2+} alone or the co-precipitation with other co-existing cations (e.g. Ca^{2+}), and Mg^{2+} might be precipitated with other anions (e.g. CO_3^{2-}). Thus, the decreased molar concentration of NH_4^+ in the liquid was assumed as the increased molar concentration of struvite in this study (as little NH_3 was detected in the biogas). On day 30, the increased insoluble Mg, decreased PO_4^{3-} and decreased NH_4^+ in the reactor +Mg+biogasR were 12.83, 11.43 and 11.42 mmol/L, respectively, indicating that most of the Mg added into the reactor might be precipitated as struvite. On day 17, the decreased NH_4^+ seems to be lower than that on day 30; however, the value was still higher than that in the reactor without biogas recirculation (+Mg). It could be estimated that about 7.32 mmol/L (119.56 mg/g-TS) and 11.42 mmol/L (192.96 mg/g-TS) struvite formed in the reactor

+Mg+biogasR after 17 and 30 days' AD, respectively, according to the decrease amount of NH_4^+.

3.3 *Changes in sludge properties*

3.3.1 Sludge dewaterability and settleability

The changes of SRF and SVI_{30} during AD are shown in Fig. 6. The SRF was reduced by 18% after $MgCl_2$ being added in the sludge mixture on day 0, indicating its enhanced dewaterability, most probably due to the binding capacity of divalent cation (Mg^{2+}) with negatively charged sludge particles (Sobeck and Higgins, 2002). Liu et al. (2011) compared the effect of seawater and brine on sludge conditioning, indicating that Ca^{2+} and Mg^{2+} have similar effectiveness that is greater than monovalent cations like Na^+ on sludge dewaterability and flocculation. After 17 days' AD, the SRF was further decreased in the reactors +Mg and +Mg+biogasR by 55% and 37% compared with the initial value without $MgCl_2$ addition. This observation to a greater extent agrees with Wang et al. (2018) who found that the crystallized struvite in anaerobically digested sludge could bind with EPS fractions, resulting in enhanced sludge dewaterability. However, biogas recirculation seemed to worsen the sludge dewaterability to some extent in both reactors Cont. and +Mg. In contrast, little change in SRF was observed in the reactors without biogas recirculation on day 17 and day 30. A shorter biogas recirculation (< 17 days) along with $MgCl_2$ addition can remarkably enhance the dewaterability of the digested sludge. Compared to the 16% reduction in capillary suction time (CST) by alkaline pretreatment of AD sludge at pH 8 by Shao et al. (2012), and the 14% increase in dewatered solid content after free nitrous acid (FNA) pretreatment of AD sludge by Wei et al. (2018), this novel AD system achieved higher enhancement on sludge dewaterability.

On the contrary, an opposite effect of biogas recirculation was observed on sludge settleability within 30 min. The SVI_{30} was found to decrease in all the sludge samples after AD, which was much lower in the reactors with biogas recirculation on day 30. However, after 30 min' SVI test, more small flocs were observed in the supernatant of sludge samples from the reactors with biogas recirculation on day 30. Jin et al. (2003) reported a positive correlation between floc size and SVI, indicating that a smaller floc size is beneficial for sludge settleability. While as pointed by Zhang et al. (2015), a large amount of fine colloidal particles is detrimental to sludge dewatering process. The small particles might be separated from sludge aggregates due to the shear force brought about by biogas recirculation, thus worsening the sludge dewaterability to some extent.

3.3.2 Extracellular polymeric substances (EPS)

Many previous works suggest the important role of EPS on sludge dewaterability, flocculation, and sedimentation (Liu and Fang, 2003; Tian et al., 2019). EPS are a mixture of high-molecular-weight polymers excreted by microorganisms, and its major components are proteins (PN) and polysaccharides (PS). Some EPS are negatively charged and have strong affinity for water. The results from this study show that the PS and PN contents in TB-EPS decreased, while those in LB-EPS increased after AD process under all the test conditions (Supplementary Materials, Fig. S4). Higher PS and PN contents were detected in S-EPS in the reactors with biogas recirculation compared to no biogas recirculation counterparts, while those in LB-EPS and TB-EPS were relatively lower in the reactors with biogas recirculation. An intensive mixing during AD may result in decreased excretion of extracellular polymeric substances (EPS), and breakup of sludge flocs of bacteria and archaea with negative effect on biogas production (Karim et al., 2005b). In addition, the increased S-EPS might not contribute to biogas production from AD of sludge. In this work, according to Pearson's correlation analysis, a strongly positive correlation was found between both PS and PN contents in S-EPS and SRF (R=0.8410, p=0.009 for PS; R=0.9640, $p=1.13\times10^{-4}$ for PN), while a strongly negative correlation was found between PN contents in TB-EPS and SRF (R=-0.9570, $p=1.92\times10^{-4}$). Thus, during a long-term biogas recirculation, the PN in TB-EPS might be transferred to the liquid phase, probably due to the resultant high shear force from biogas recirculation. This might destroy the structure of sludge flocs, further release small colloidal particles and thus deteriorate sludge dewaterability to some extent. Moreover, an excessive S-EPS content is detrimental to cell cohesion and stability of sludge flocs, leading to poor dewaterability (Zhang et al., 2015). A higher PN content was also detected in the TB-EPS from the sludges in the reactors with Mg addition. As expected, the added Mg^{2+} might bridge with the biopolymers in TB-EPS, partially avoiding being stripped off from the sludge particles during biogas recirculation.

3.4 *Analysis of the industrial-scale application of this novel AD system*

The primary sludge used in this study was sampled from the Shimodate WWTP in Chikusei, Ibaraki, Japan. Taken this WWTP as an example, the expected result of the industrial-scale application of this novel AD system was further discussed in this study.

From the operation data in 2017, the WWTP had an average wastewater treatment capacity of 8190 m^3/d. Averagely about 38 m^3 of concentrated sludge was daily treated by AD, and its daily biogas yield was 433 m^3/d with an average methane content of 60.2%. If all the biogas was used for electricity generation, approximately 2.48×10^5 kWh/y could be produced according to Eq. (1). The generated electricity could cover 17.3% of the total electricity consumption of the WWTP (1,431,948 kWh/y in 2017).

$$E_{biogas}(\text{kWh/y}) = (V_{CH_4} \times LHV_{CH_4} \times E_{ff})/(3.6 \times 100\%) \qquad (1)$$

where E_{biogas} (kWh/y) is the amount of electricity generated using the produced biogas per year; V_{CH4} (m^3/y) is the annual methane yield produced in the WWTP (for 365 days' operation); LHV_{CH4} (35.8 MJ/m^3) is the lower heating value of methane; E_{ff} (%) is the thermal efficiency of biogas engine (given the assumption that the same spark ignition engine was used as Porpatham et al. (2008)); 3.6 (MJ/kWh) is the coefficient for unit conversion from MJ to kWh.

In this study, the methane content in biogas was increased by 11% through biogas recirculation during AD. According to Porpatham et al. (2008), the E_{ff} of biogas engine could be improved from 26.2% to 27.1% if the CO_2 content in biogas was reduced from 41% to 30% (at a constant value of total methane amount). Thus, through applying biogas recirculation in the anaerobic digesters of the Shimodate WWTP, the E_{biogas} from the biogas would increase to 2.56×10^5 kWh/y, which could cover 17.9% of the total electricity consumption of the WWTP. The increased amount of E_{biogas} (8000 kWh) by using the novel AD system corresponds to 312,000 JPY (about 2840 USD) according to the feed-in tariff (FIT) purchase price of electricity generated from biogas (39 JPY/kWh, about 0.35 USD/kWh). According to MLIT (2018), the annual biogas production from sewage sludge in Japan is 3.3×10^8 m^3/y, in which 37% is utilized for electricity generation. If the CH_4 content could be increased from 60% to 70% in the biogas by applying the novel AD system, the E_{biogas} in Japan could be increased by 6.56×10^6 kWh/y, corresponding to 2.56×10^8 JPY (about 2.33 million USD).

In addition, the addition of $MgCl_2$ during AD could increase the P content in the solid phase of digested sludge and the sludge dewaterability. P is an essential element for crop growth, and regarding P sources Japan totally relies on imports from other countries. Nowadays, along with the increased P price which has become a serious issue, P recovery from wastewater has been strongly prompted by Japanese government (MLIT, 2018). Results from this study show that the P content in the solid phase of digested sludge was increased from 34 g-P/kg-TS to 60 g-P/kg-TS. In the case of the Shimodate WWTP, if all the dewatered sludge cake (2.3 t/d, 40%TS) was reutilized as fertilizer, the novel system tested in this study would contribute to an annual increase of 8.7 t of P, which could save at least 196,000 JPY (about 1784 USD) for importing phosphate rock from other countries (JOGMEC, 2018). When taking the huge amount of annual sludge production in Japan, 1.67×10^8 t in 2013 (MOE, 2019) into consideration, the increase amount of P resource from sludge via this novel AD system would be much more profitable and marketable. Restated, the above two value-added aspects, i.e. enhanced methane content in the biogas and P content in the solid digestate, can be greatly recognized if this novel AD system is popularized in all the WWTPs nationwide.

Furthermore, by adding $MgCl_2$ during AD with biogas recirculation in this study, the sludge dewaterability was enhanced by 37%. An enhanced dewaterability of sludge can contribute to the followings: 1) reduction of electricity consumption during dewatering process due to a shorter duration for dewatering (Chu et al., 2005); 2) reduction of energy consumption for thermal drying because of the increased solids content in the dewatered sludge (Wang et al., 2010); and 3) decrease of the addition of chemical flocculants. Eventually, all the above aspects could contribute to the reduced operation costs of the WWTP. Currently, $FeCl_3$ and lime are used as coagulants for sludge conditioning in the WWTP. More in-depth research works are demanding so as to confirm whether they are still necessary and their optimum dosages for conditioning the digested sludge from the novel AD system in the context of the whole WWTP. When this novel AD is introduced into the WWTP, the whole AD system and digested sludge conditioning can be simplified as shown in Fig. 7. Further cost reduction in operation and management is expected regarding the dewatering and disposal of the digested sludge from the WWTP.

4. Conclusions

Simultaneous biogas upgrading, resources conservation and sludge conditioning were realized in one AD system in this study. This work demonstrates the concept and possibility of simplifying the sludge treatment facilities from the viewpoint of sustainable development goals (SDGs), which might greatly reduce the costs of WWTP operation and management. During 17 days' AD with $MgCl_2$ addition and intermittent biogas recirculation, the following results could be obtained: 1) CH_4 content was enhanced to 86% in comparison to 75% in the reactors without biogas recirculation; 2) 87% orthophosphate and 19% ammonia N were conserved in the solid phase of digested sludge; 3) the sludge dewaterability was enhanced by 37%. In addition, a long-term biogas recirculation (30 days in this study) was found to worsen sludge dewaterability and decrease CH_4 content to some extent. Further optimization of operation conditions is necessary to achieve higher CH_4 content and enhanced dewaterability for practical application.

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Declarations of interest

None.

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Figures

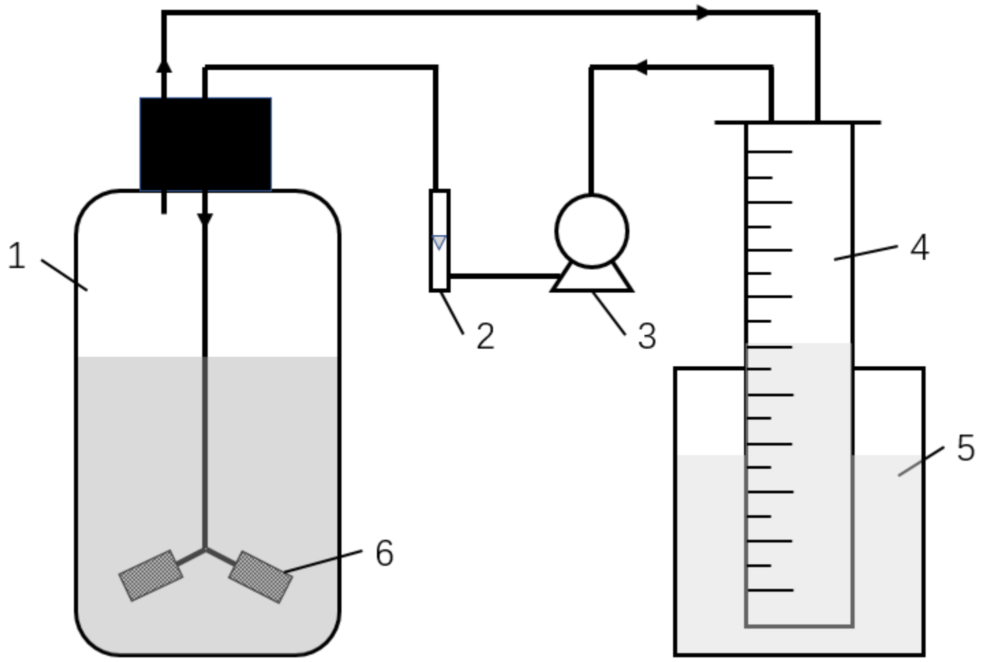

Fig. 1. Schematic of the batch anaerobic digestion system used in this study. 1. One-liter anaerobic digestion (AD) reactor; 2. Gas flowmeter; 3. Gas pump; 4. Graduated cylinder; 5. Saturated $NaHCO_3$ solution; 6. Gas sparger.

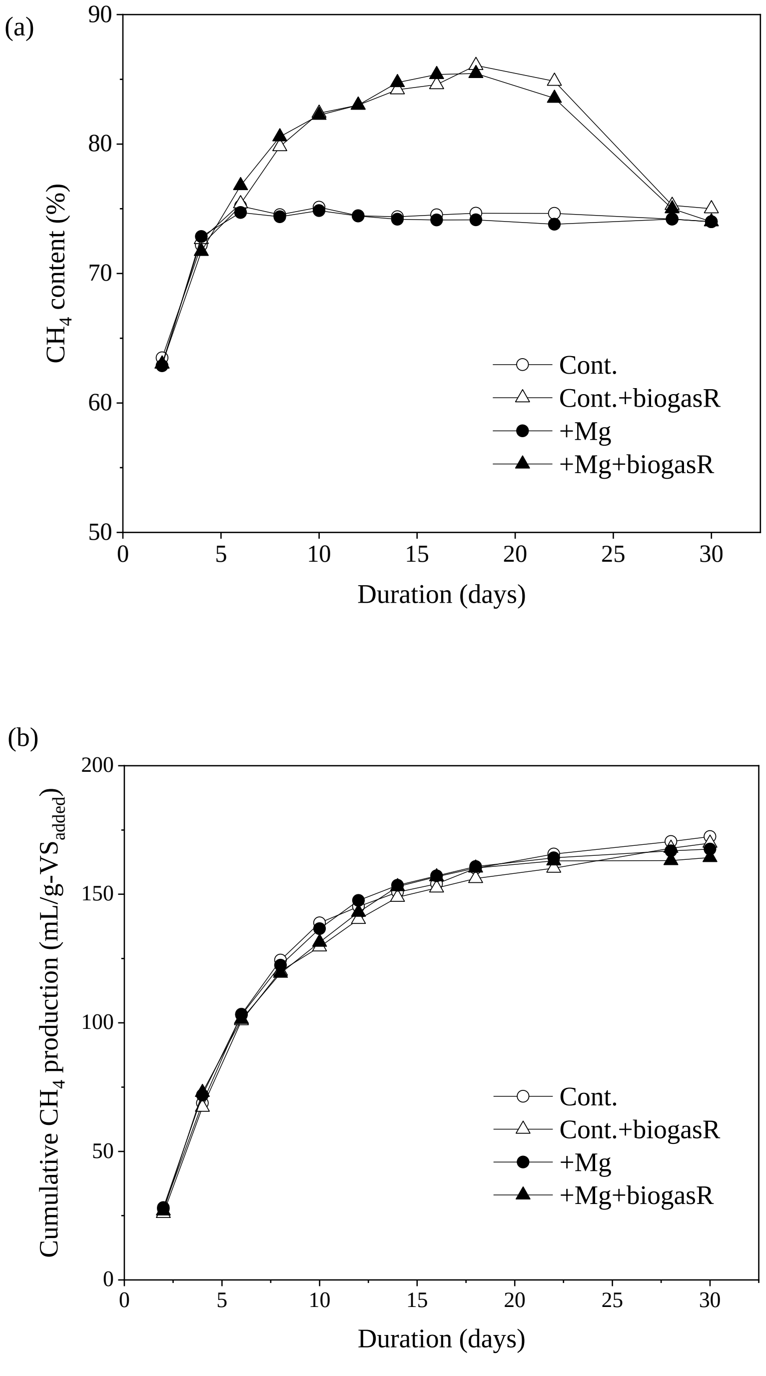

Fig. 2. Variations of CH_4 content (a) and cumulative CH_4 production (b) during 30 days' anaerobic digestion of sewage sludge with biogas recirculation (+biogasR) and $MgCl_2$ addition (+Mg). Cont.-Control reactor with no biogas recirculation and no $MgCl_2$ addition.

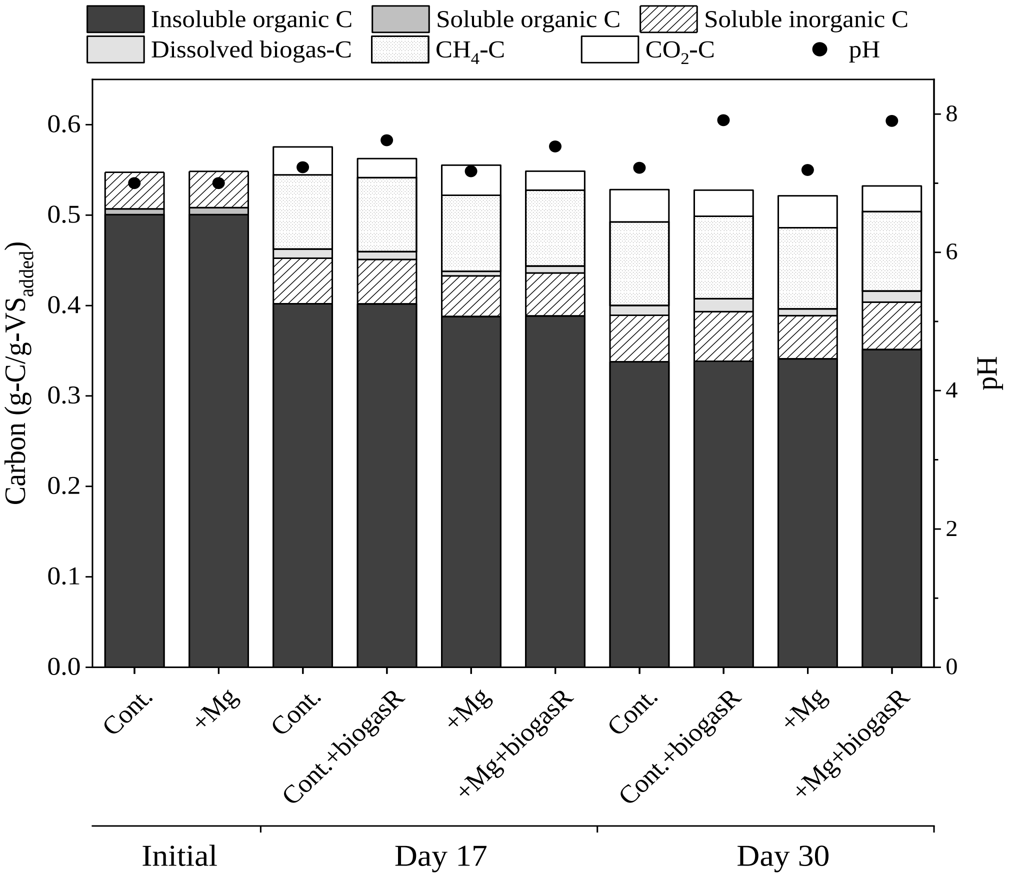

Fig. 3. Carbon (C) profiles and digestate pHs in the reactors on day 0, day 17 and day 30 during anaerobic digestion of sewage sludge with biogas recirculation (+biogasR) and $MgCl_2$ addition (+Mg). Cont.-Control reactor with no biogas recirculation and no $MgCl_2$ addition.

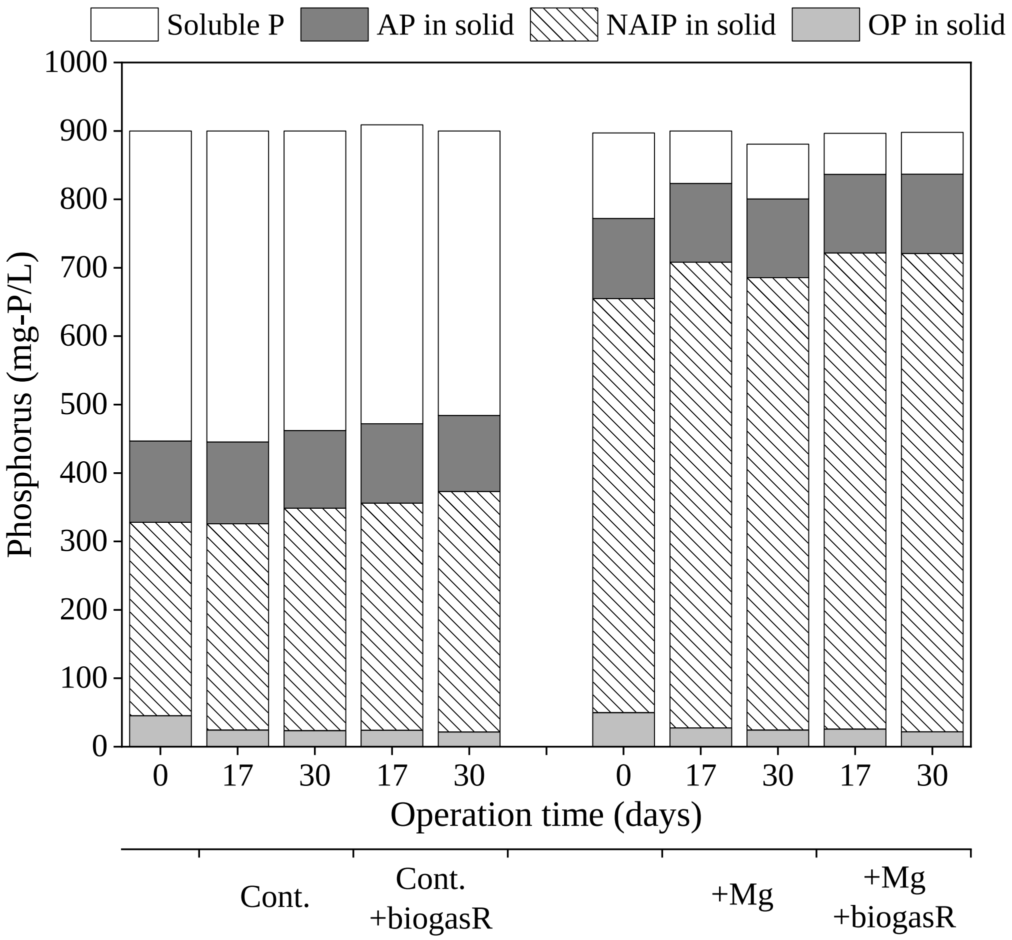

Fig. 4. Phosphorus (P) profiles in the reactors on day 0, day 17 and day 30 during anaerobic digestion of sewage sludge with biogas recirculation (+biogasR) and $MgCl_2$ addition (+Mg). The concentration of each P fraction (AP, NAIP or OP in the solid phase) was calculated based on their contents in the solid and the solids concentration in the sludge sample. Cont.-Control reactor with no biogas recirculation and no $MgCl_2$ addition.

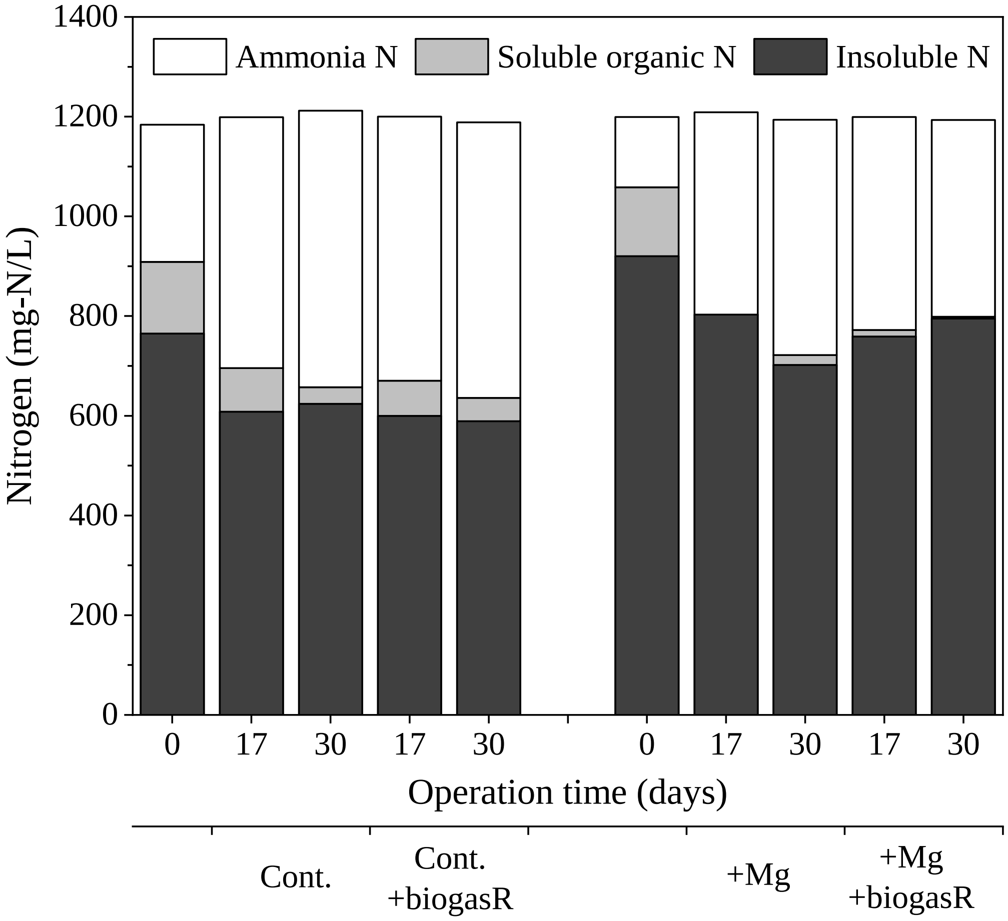

Fig. 5. Nitrogen (N) profiles in the reactors on day 0, day 17 and day 30 during anaerobic digestion of sewage sludge with biogas recirculation (+biogasR) and $MgCl_2$ addition (+Mg). The concentration of insoluble N was calculated based on its N content in the solid and the solids concentration in the sludge sample. Cont.-Control reactor with no biogas recirculation and no $MgCl_2$ addition.

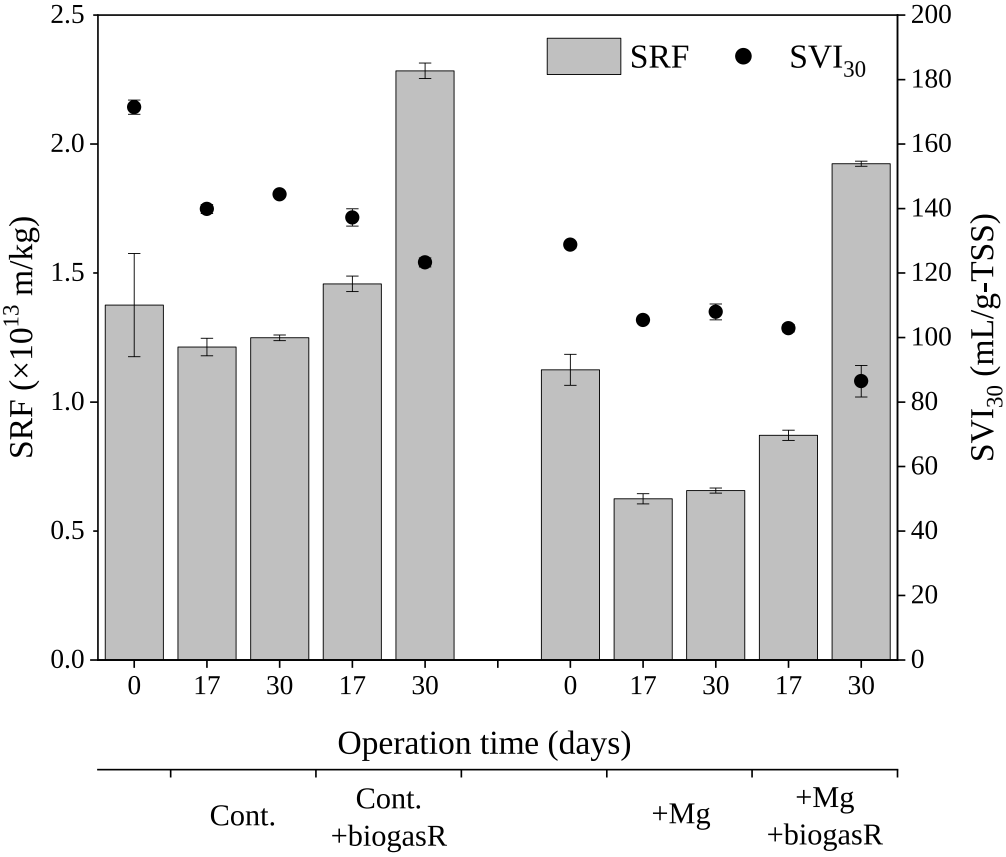

Fig. 6. Sludge dewaterability (SRF) and settleability (SVI_{30}) in the reactors on day 0, day 17 and day 30 during anaerobic digestion of sewage sludge with biogas recirculation (+biogasR) and $MgCl_2$ addition (+Mg). Cont.-Control reactor with no biogas recirculation and no $MgCl_2$ addition.

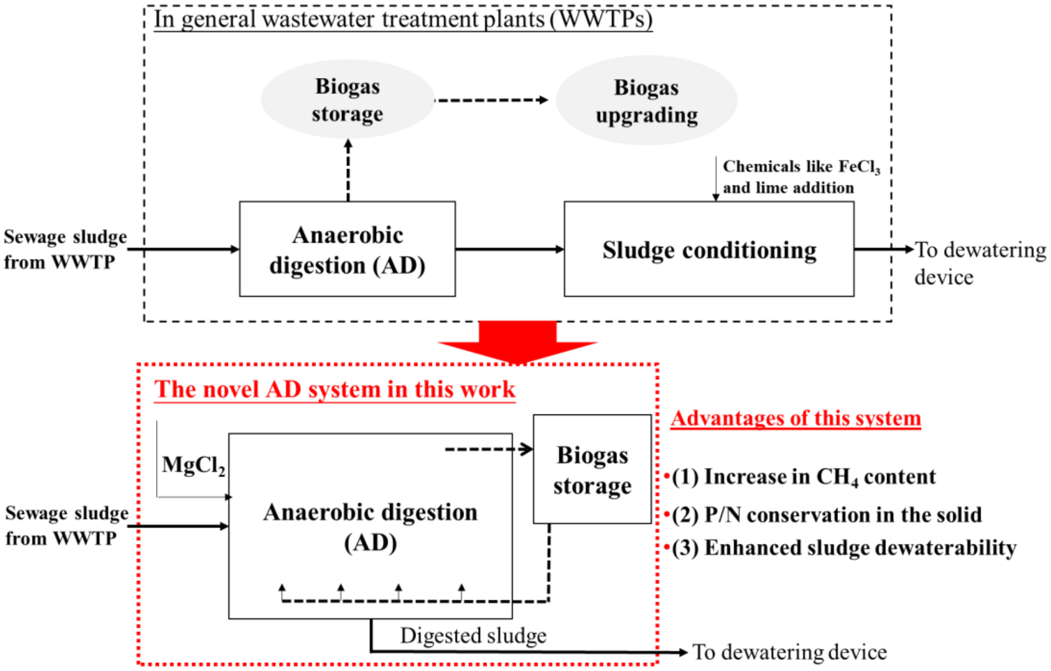

Fig. 7. Comparison between the sludge treatment processes in general wastewater treatment plants (WWTPs) and the novel anaerobic digestion (AD) system proposed in this work.