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Characterization of food waste and sewage sludge mesophilic anaerobic co-digestion under different mixing ratios of primary sludge, secondary sludge and food waste

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ABSTRACT

The main aim of the present work was to characterize the mesophilic anaerobic co-digestion of food waste (FW) and sewage sludge (SS) by considering different mixing ratios of primary sludge (Pr), secondary sludge (Sc) and food waste. The experiments were carried out using batch reactors (R1-R7). The applied ratios included VS_{Pr}:VS_{Sc} (1:1 and 2:1) and VS_{FW}:VS_{SS} (0:4, 1:3, 2:2 and 4:0) with VS_x (g) as the volatile solids content of the substance x within the total feed of each reactor and VS_{SS} as the sum of VS_{Pr} and VS_{Sc}. According to the obtained results, ratio of carbon to nitrogen (C:N) was much more effective than VS_{Pr}:VS_{Sc} ratio considering VS reduction and biogas production efficiencies. A second-order polynomial curve fitted well the data of VS reduction vs. C:N, representing an optimum C:N around 16 to achieve the highest VS removal. A high R-squared linear correlation was found between data of gas production and C:N ratio according to which, with increasing the C:N within the test range (8–19.7), the biogas production increased steadily. Doubling VS_{Pr}:VS_{Sc} under lower ratios of VS_{FW}:VS_{SS} (0:4 and 1:3) positively affected both VS reduction and biogas production, while under higher VS_{FW}:VS_{SS} ratio (2:2) it influenced adversely. Doubling VS_{Pr}:VS_{Sc} also improved the digestate dewaterability under all applied ratios of VS_{FW}:VS_{SS}. A synergistic gas production was observed for the reactors with 50% share of VS_{FW} (VS_{FW}:VS_{SS} of 2:2) between which, R5 with VS_{Pr}:VS_{Sc} of 1:1 showed the highest synergistic gas production. The highest methane content (70.3%) also belonged to R5.

1. Introduction

During the last decades, along with the rapid growth of the population all over the world, sewage production has also increased sharply [1]. Treatment of such a large volume of wastewater results in the production of huge amounts of sludge [2]. Generally, sewage sludge (SS) can be categorized as primary sludge (Pr) and secondary sludge (Sc) which are generated as by-products in wastewater treatment plants (WWTP) [3,4]. The primary sludge mainly consists of settled organic matter of raw wastewater and the secondary sludge mainly consists of bacterial flocs. The organic matter in the primary sludge is more easily degradable than that in the secondary sludge [3,5]. Overally, these types of sludge are highly putrescible due to their high content of volatile solids (VS) and have unpleasant odor and various types of pathogens. For this reason, releasing them into the environment will have destructive effects on humans, animals and aquatic environments [6,7]. Therefore, safe treatment and disposal of sewage sludge is of high importance. There are several methods for treating and stabilizing sewage sludge, where aerobic digestion, anaerobic digestion, lime stabilization, incineration and composting are more common [8]. Among these methods, anaerobic digestion (AD) can be considered as a promising option due to its high performance in reducing sludge volume, reducing chemical and biological oxygen demand (COD and BOD, respectively), inactivating pathogens and converting organic matter to renewable energy [9–12].

Anaerobic digestion is a biological process in which a microbial consortium decomposes organic matter in the absence of oxygen. This ultimately results in sludge stabilization and biogas production [8]. The produced biogas through anaerobic digestion generally contains methane (60–70%) and carbon dioxide (30–40%) with a trace amount of other gases (H_2, H_2S, N_2) [5,13]. The methane content of biogas has thermal value and can be converted to different types of energy such as heating and electrical energy [5,13]. Since most of the equipment in WWTP consumes high amounts of energy, providing a fraction of that energy by anaerobic digestion is desirable [14]. The specific gas

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Nomenclature

AcoD	anaerobic co-digestion
AD	anaerobic digestion
Alk	alkalinity
BOD	biological oxygen demand
C:N	ratio of carbon to nitrogen
COD	chemical oxygen demand
EPS	extracellular polymeric substances
F:M	food to microorganism ratio
FAN	free ammonia nitrogen
FW	food waste
Ic	inoculum
Pr	primary sludge
Sc	secondary sludge
sCOD	soluble chemical oxygen demand
SRF	specific resistance to filtration
SS	sewage sludge
TAN	total ammonia nitrogen
TS	total solids
TSS	total suspended solid
TTF	Time-To-Filtration
VFA	volatile fatty acids
VS	volatile solids
VS_{Feed}	total VS added as the feed to the reactors (g)
VS_{FW}	the portion of VS_{Feed} which is related to food waste (g)
VS_{Ic}	VS content of the inoculums (g)
VS_{Pr}	the portion of VS_{Feed} which is related to primary sludge (g)
VS_{Sc}	the portion of VS_{Feed} which is related to secondary sludge (g)
VS_{SS}	sum of VS_{Pr} and VS_{Sc} (g)
WWTP	wastewater treatment plant

production from sewage sludge commonly differs from 0.75 to 1.12 m^3 kg^{-1} $VS_{destroyed}$ at standard conditions (0 °C and 1 atm) [3]. The amount and the composition of the produced biogas depends on many operational and environmental parameters such as temperature, mixing condition, type of organic content, food to microorganism ratio (F:M), pH, microbial community, etc. [15]. During the biogas production, the total solids (TS) content of the sludge including biological flocs and raw organic matters diminishes up to more than 50%. This results in production of a stabilized digestate which due to high nitrogen and phosphorus content can be used as a suitable fertilizer for agricultural purposes [8,14,16]. However, as the AD process does not eliminate all volatile solids and degradable organic matter, the digested sludge is usually not completely stable [17]. Therefore, modifying the AD process in order to produce a higher amount of biogas and to reduce the solid content as much as possible is worthwhile.

Co-digestion technology is a way that can improve the efficiency of sewage sludge anaerobic digestion. In this method different solid or liquid organic wastes are combined as co-substrates with the sewage sludge [18,19]. Anaerobic Co-digestion (AcoD) of sewage sludge with co-substrates is applied to cover the weaknesses of sewage sludge (primary and secondary sludge) including low C:N ratio, low organic content, refractory organic constituents (biological flocs in particular) and hydrolysis rate-limiting [19,20]. One of the most suitable organic wastes which can be used as co-substrate for eliminating these drawbacks and improve digester efficiency is food waste (FW) [15,20].

Food waste from residential, commercial, institutional and industrial sources is being produced at an increasing rate due to population growth and enhancing the life style standards [4,9]. The food waste characteristics in terms of carbohydrates, lipids and proteins differ considerably based on its components. According to a review study performed by Iacovidou et al. [18] carbohydrates, proteins and lipids can orderly comprise 55–78.2%, 14.4–21.3% and 13–22% of the dry weight of the food waste depending on its source and origin [18]. The most important properties of FW as a co-substrate in AcoD process are its high content of readily biodegradable organic matter and as well its high C:N ratio. According to the previous studies diluting toxic components, improving methane generation in the range of 22–127%, balancing nutrient content, enhancing C:N ratio from 6 to 9 to 14–20, increasing VS reduction from 7% to 35% and stimulating microbial activity have been reported as some advantages of using FW as co-substrate in sewage sludge AcoD process [8,15,21–27].

Several studies have been done on anaerobic co-digestion of sewage sludge and food waste in order to enhance process performance [28–31]. One of the most important challenges that almost all researchers have encountered and investigated in this regard was determining the amount of FW that should be added to the process in order to obtain the highest stability and efficiency. It has been reported in some literature that adding food waste 30–40% of the total substrate (mass ratio of VS_{FW} to VS_{Feed}) considerably improves the sewage sludge anaerobic digestion efficiency, in terms of VS reduction or gas production, under mesophilic condition [25,32–36]. However, in some other researches the optimal mixing ratio is reported to be around 50% in the same condition [24,26,28,37].

However, there is a lack of information in the literature regarding the optimal mixing ratio of primary sludge, secondary sludge and food waste in anaerobic co-digestion of sewage sludge. In other words, in most previous literature on anaerobic co-digestion of sewage sludge and food waste, no distinction has been taken into account between primary sludge and secondary sludge. For example, Cabbai et al. [30], Grosser et al. [34], Keucken et al. [31], Koch et al. [29], Kuo-Dahab et al. [26] and Liu et al. [28] used a mixture of primary sludge and secondary sludge in their co-digestion study without considering any specific ratio between them. In other studies on the same field [25,32,33,35,36], the researchers referred to the phrase of sewage sludge in general and did not report any detailed characteristics about it. In another research, Zhu et al. [24] tested three different co-digestion mixtures including FW + Pr, FW + Sc and FW + Pr + Sc at three different mixing ratios of FW to sludge, for Biohydrogen production. However, they did not investigate the effect of mixing ratio of primary sludge and secondary sludge along with the mixing ratio of FW to sludge. Accordingly, the study of the anaerobic co-digestion of sewage sludge and food waste in such a way that the effect of the mixing ratio of primary sludge and secondary sludge together with the mixing ratio of food waste to sewage sludge can be observed is worthwhile.

For this reason, the co-digestion of FW, primary sludge and secondary sludge at seven different mixing ratios was investigated in the present work. The main focus was on process performance in terms of VS reduction, gas production and methane yield at different mixing ratios. In this regard, different parameters including soluble chemical oxygen demand (sCOD), volatile fatty acids (VFA), Alkalinity (Alk), pH, total and free (unionized) ammonia nitrogen (TAN, FAN), etc. were measured.

In addition to the VS reduction and gas production, the dewaterability of the stabilized sludge is also of high importance. High digestate dewaterability reduces the costs of sludge conditioning, decreases the depreciation rate of the dewatering equipment and diminishes the disposal space constraints [38]. The digestate dewaterability is affected by various physical-chemical parameters such as particle size, EPS (extracellular polymeric substances) content, surface charge, hydrophobicity, viscosity, and bound water content of the digestate [39,40]. All these parameters depend essentially on the composition of the feed substrate and as well on the AD operational condition. Therefore, dewaterability of the digested sludge at different mixing ratios of the feed was also assessed in the present work. For this purpose, the specific resistance to filtration (SRF) of the digested sludge was measured for all mixing ratios.

The obtained results will help the designers and operators of wastewater treatment plants to more efficiently manage their sludge as a renewable energy source and convert it to a suitable fertilizer with the least possible VS content.

2. Material and method

2.1. Sludge and food waste

Thickened primary and secondary sludge samples were taken from Tehran South WWTP (Tehran, Iran). The food waste samples were collected during several days from two restaurants in the campus of the Iran University of Science and Technology. It mainly contained rice, meat, potato, breads, noodles, vegetables, and fruits. After mixing the samples and removing harsh components (e.g. bones, metals, glass, plastics and napkins) the food waste was grinded using a food waste disposer (model JD560-B0, 3200 rpm, 3/4 Hp, Hangzhou Cleesink Co., China). Since specific amounts of water, depending on the disposer type, should be added as grinding fluid [41], a water:FW ratio of 11.7 L kg^{-1} was adopted according to some previous studies [41]. The effluent of food waste disposer was allowed to settle for about 2 h (the same hydraulic retention time as the primary sedimentation tanks in Tehran South WWTP) in a 60 cm height cylinder. Then, the food waste sample was taken from the settled material at the bottom of the cylinder. All sludge and food waste samples were stored at 4 °C in a refrigerator. About 24 h before running the lab-scale anaerobic digester, the samples were allowed to gradually reach ambient temperature.

The inoculum was taken from the outlet line of the mesophilic anaerobic digesters in Tehran South WWTP. Before feeding the anaerobic reactors with the substrate, to remove the already existing degradable organic matter and degas the inoculum, the inoculum was pre-incubated at 35 °C for 3 days after since no significant gas production was observed anymore. By this way, the volume of the net biogas generated from the substrate could be precisely measured [42,43]. The characteristics of primary and secondary sludge samples, the food waste sample and the inoculum are presented in Table 1.

2.2. Lab scale anaerobic digesters

Seven batch anaerobic reactors (R1-R7) with a total volume of 2 L (1.6 L for the mixture of substrate and inoculum) were used for evaluating anaerobic co-digestion of different mixing ratios of primary sludge, secondary sludge and food waste. The reactors were made up of Plexiglas and equipped with a mechanical stirrer. An intermittent mixing mode of 5 min on/15 min off was selected for stirring of the mixture.

Three outlet ports located on the top of the reactors were used for flushing of nitrogen, sampling from the produced biogas and measurement of biogas volume (Fig. 1). Biogas production was measured by liquid displacement system with acidified water (pH = 2) as the used liquid. The liquid displacement was recorded daily for each reactor. One outlet port at the bottom of the reactors was used for sludge sampling.

Table 1
Characteristics of primary sludge, secondary sludge, food waste, and inoculum fed to the AcoD reactors.

parameter	Substrate			Ic
	Pr	Sc	FW	
TS (%)	2.40	4.96	13.90	2.30
VS (%)	2.14	4.20	12.50	1.63
VS:TS (%)	89	85	90	71
C:N	8.4	7.6	19.7	7.2

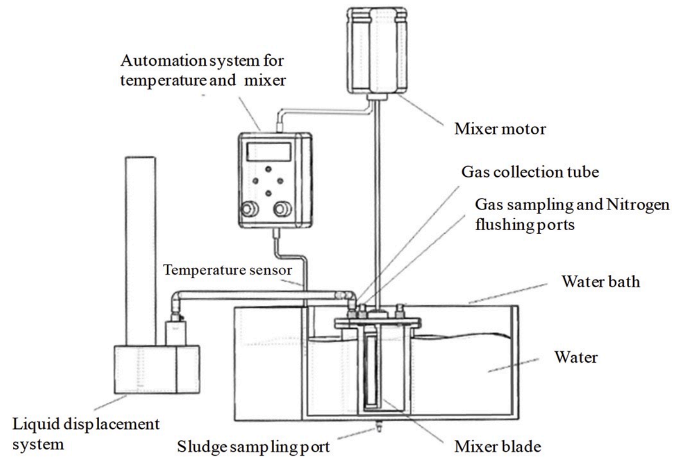

Fig. 1. Scheme of the lab scale anaerobic digesters.

The reactors were fed one time just after the pre-incubation phase and operated until the daily biogas production reached less than 1% of the total biogas production.

2.3. Operational conditions and mixing ratios

The selected substrate mixing ratios are presented in Table 2. In order to provide the same conditions for all reactors, an inoculum to substrate mixing ratio (VS_{Ic}:VS_{Feed}) of 2:1 was applied to all reactors (Table 2). This ratio was adopted according to some previous literature [28–30]. The headspace of all reactors (400 mL) was flushed with nitrogen for 3 min to create an anaerobic condition. All reactors were sealed with a rubber belt and stud bolts. The reactors were placed within a water bath equipped with a circulation system and a heater and operated under mesophilic condition (35 °C). The operating temperature was selected according to the most common optimal mesophilic temperature range, 30–38 °C, reported in literature [3,43].

2.4. Analytical methods

During the first days of the process, due to the higher changing rate of the parameter values, samples were taken at shorter time intervals (every 2 days). Afterward, the sampling time intervals became longer (every 3 or 4 days). Total solids (TS) and volatile solids (VS) were measured according to the procedures presented in standard methods [44]. VS reduction was measured according to the procedure described by Metcalf and Eddy [3]. Determination of the soluble parameters was performed by using the supernatant of digestate samples centrifuged at 5000 rpm for 10 min. Total nitrogen (TN), total ammonia nitrogen (TAN) and sCOD were measured by using HACH test kits and HACH spectrophotometer DR/4000 (USA). FAN and ammonium (NH_4^+) were calculated by using the following equations [45]:

$$FAN = \frac{TAN^*\left(K_a \div 10^{-pH}\right)}{\left(K_a \div 10^{-pH}\right) + 1} \tag{1}$$

$$NH_4^+ = TAN - FAN \tag{2}$$

where, FAN is free ammonia nitrogen (mg L^{-1}), TAN is total ammonia nitrogen (mg L^{-1}) and K_a is temperature constant (1.097×10^{-9} at 35 °C).

VFA and Alkalinity were measured according to a combination of standard titration methods (2310B & 2320B) by using 0.1 N H_2SO_4 and 0.1 N NaOH solutions [44]. A portable PC300 pH-meter (CyberScan, USA) was used for measuring pH and total dissolved solids (TDS). Viscosity was measured by using a Visco Star viscometer (POLYVISC, Switzerland). Methane content of biogas was analyzed by a Gas

Table 2
Operating conditions of the reactors.

Parameter	Reactor						
	R1	R2	R3	R4	R5	R6	R7
VS_{FW}:VS_{SS}	0:4	0:4	1:3	1:3	2:2	2:2	4:0
VS_{Pr}:VS_{Sc}	1:1	2:1	1:1	2:1	1:1	2:1	–
VS_{Feed}:VS_{Ic}	1:2	1:2	1:2	1:2	1:2	1:2	1:2
Pr (g)	236.5	308.3	186	241	128.6	176.2	0
Sc (g)	121.8	77.1	95.8	60	65.4	44.1	0
FW (g)	0	0	22.3	21	42.9	44.1	100
VS_{Feed} (g)	10.2	9.8	10.8	10.3	10.9	11.14	12.5
C:N	7.99	8.13	10.94	11.02	13.67	13.78	19.70
Ic (g)	1241.7	1214.6	1295.9	1278	1359.1	1335.6	1500
Mixture (g)	1600	1600	1600	1600	1600	1600	1600

SS: Sewage Sludge, FW: Food Waste, Pr: Primary Sludge, Sc: Secondary Sludge, Ic: Inoculum, VS_{Feed}: Total VS added as the feed to the reactors (g), VS_{Ic}: VS content of the inoculums (g), VS_{Pr}: The portion of VS_{Feed} which is related to primary sludge (g), VS_{Sc}: The portion of VS_{Feed} which is related to secondary sludge (g), VS_{FW}: The portion of VS_{Feed} which is related to food waste (g), VS_{SS}: sum of VS_{Pr} and VS_{Sc} (g).

Chromatograph (GC-2550TG, Teif Gostar Faraz Co., Iran) equipped with 1 m × 2 mm Porapak Q (80/100 mesh) packed column and TCD detector analyzer. Hydrogen was used as the carrier gas. The temperature of injector, column and detector was kept constant at 50, 50 and 40 °C, respectively. Total organic carbon was measured by the Phoenix 8000 TOC meter (Tekmar Dohrmann, USA).

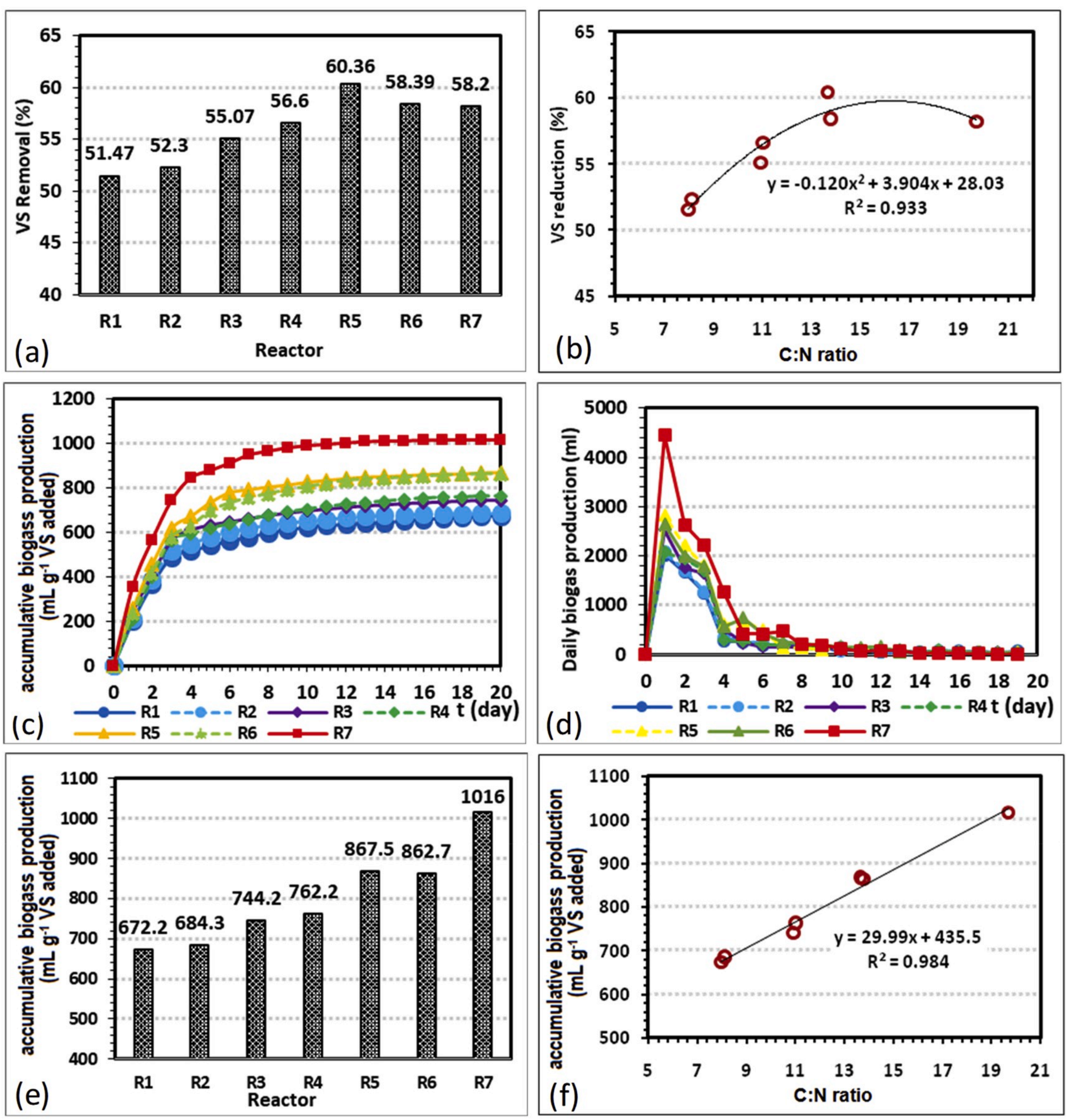

Fig. 2. VS reduction (a), correlation between VS reduction and C:N values (b), variations of accumulative biogas production (c), variations of daily biogas production (d), total biogas production (e), correlation between total biogas productions and C:N values (f).

2.5. Dewatering tests

To assess the digestate dewaterability, the Specific Resistance to Filter (SRF) was measured for all applied mixing ratios at the end of the operation period. For measuring the SRF, the modified Wisniewski and Grasmick formula [46] was used (Eq. (3)):

$$SRF\left(m\ kg^{-1}\right) = \frac{2000A^2\Delta pb}{\mu C} \tag{3}$$

where, A is the filter paper area (0.00636 m^2), ΔP is the constant vacuum pressure (kPa), μ is the filtrate viscosity (Pa.s), C is the total suspended solid (TSS) content of sludge (kg m^{-3}) and b is the Time-To-Filtration (TTF) ratio which is the slope of the curve extracted from plotting the time of filtration divided by the volume of filtrate (t V^{-1}) versus the filtrate volume (V) [46].

3. Results and discussion

3.1. VS reduction and biogas production

The microorganisms activity within the anaerobic digester results in utilization of organic matter content of the substrate and production of biogas [34]. For this reason, the amount of organic matter reduction (VS reduction) and the volume of biogas production are the two important parameters for evaluating AD performance. In the previous studies, VS reduction for the sewage sludge AcoD with FW has been reported between 33 and 85% [26,34–37,48,49]. According to the results of the present work, VS reduction for the different mixing ratios of primary sludge, secondary sludge and food waste was between 51 and 60%, indicating an almost proper performance of all reactors (Fig. 2(a)).

In Fig. 2(b), the VS reduction values for different reactors are plotted vs. the corresponding C:N ratios. As seen, a second-order polynomial curve with high regression coefficient ($R^2 = 0.9382$) fitted to the points. The curve indicates that increasing the percentage of VS_{FW} within the feed VS until reaching a C:N ratio of around 16 improves the AcoD performance in terms of VS reduction and beyond that, the VS removal will decrease. This indicates the importance of C:N ratio as a key parameter for evaluation of the nutrient balance in the AD process [30]. From another point of view, it can be stated that the reactors containing more sewage sludge as their feed, represented smaller amounts of VS removal. This can be due to the presence of some recalcitrant substances within the sewage sludge which are difficult to hydrolyze (e.g. biological flocs and their secretions). As seen in Fig. 2(a), among all reactors with the applied mixing ratios, R5 with a VS_{Pr}:VS_{Sc} ratio of 1:1, a VS_{FW}:VS_{SS} ratio of 2:2 and a C:N ratio of 13.67 showed the highest VS reduction (60.4%).

Data of biogas production are presented in Fig. 2(c-f). As seen in Fig. 2(c-d), the gas production in all reactors essentially occurred within the first 4 days of the operation period and then continued at very low rate. No significant fluctuations in gas production were observed and besides, the specific gas productions of all reactors were in the same range as the other studies which reported a normal performance for their AcoD reactors [4,31,34,36]. This demonstrates a relatively stable performance in all reactors and implies a low proportion of toxic matters and refractory organic substances within the feed.

As depicted in Fig. 2(e), with increasing the FW portion of the feed VS from 0% in R1 and R2 to 100% in R7, the specific biogas production steadily increased from 672.2 mL g^{-1} VS_{added} to 1016 mL g^{-1} VS_{added}, respectively. In other words, the reactors with more sewage sludge in their feed, produced less biogas per unit mass of the added VS. This is consistent with the results of VS reduction. Similar result has been reported by Li et al. [50] who studied the sewage sludge AcoD with different mixing ratios of FW and reported lower biogas production for the reactors with higher content of sewage sludge.

In Fig. 2(f) the biogas production values are plotted vs. C:N ratios. A linear trend with a high regression coefficient ($R^2 = 0.9883$) fitted the points. It can be found from the obtained linear trend that with increasing the C:N ratio via adding FW, the biogas production increases steadily. A similar linear trend with high regression coefficient also fitted the data of gas production with the unit of mL g^{-1} $VS_{removed}$ vs. C:N ratio (data not presented). This indicates that by increasing the percentage of VS_{FW} within the feed VS, the anaerobic fermentation and oxidation reactions change in a way that a greater part of the final product is in the form of gas.

Regarding the effect of VS_{Pr}:VS_{Sc} ratio on VS reduction and biogas production efficiencies, it can be found from Fig. 2(a) and Fig. 2(e) that under lower VS_{FW}:VS_{SS} ratios of 0:4 and 1:3, increase of VS_{Pr}:VS_{Sc} ratio from 1:1 in R1 and R3 to 2:1 in R2 and R4 slightly improved both the VS reduction and biogas production. However, such an improvement was not observed under higher VS_{FW}:VS_{SS} ratio of 2:2 so that both the VS reduction and gas production efficiencies in R6 (VS_{Pr}:VS_{Sc} ratio of 2:1) were slightly lower than those in R5 (VS_{Pr}:VS_{Sc} ratio of 1:1). Therefore it can be said that doubling of VS_{Pr}:VS_{Sc} under lower VS_{FW}:VS_{SS} ratios of 0:4 and 1:3 (R1-R4) positively affected both VS reduction and biogas production, while under higher VS_{FW}:VS_{SS} ratio of 2:2 (R5 and R6) it influenced adversely. This can be attributed to the excess amount of readily biodegradable substances in R6 which has resulted in less suitable values of VFA and pH at the beginning of the process in this reactor compared to R5. A more precise discussion in this regard is presented in sections 3.2 and 3.4.

3.2. VFA, alkalinity and pH

Volatile fatty acids (VFAs) are one of the most effective parameters on the stability of the anaerobic digestion [51]. During the hydrolysis phase of the AD process, fats and lipids are first converted into long-chain fatty acids, and then throughout the acidogenesis they change to short-chain volatile fatty acids (VFAs) [9]. The excessive concentration of VFAs acidifies the digester environment and reduces the pH to such a level that results in the process inhibition [40,52]. For this reason, regular monitoring of VFAs concentration is essential for the AD process in order to assess the performance, stability and efficiency of the system.

As seen in Fig. 3(a), the VFA concentration at the beginning of the process sharply increased due to the decomposition of lipids, proteins, polysaccharides and other macro molecule organic compounds, and then decreased rapidly. As mentioned in section 3.1, the major gas production was also occurred within this short period of time and declined afterward. For this reason, it can be said that all the hydrolysis, acidogenesis and methanogenesis stages essentially took place during the first 4 days of the process and afterward, continued at very lower rates.

It is interesting that the reactors with higher content of VS_{FW} in their feed (R5-R7) experienced severer VFA increase within the hydrolysis and acidogenesis phases. This is most probably attributed to the higher content of readily biodegradable compounds in the food waste [18]. This can be proved by higher initial sCOD concentration in these reactors in comparison with the other ones (Fig. 4(a)). In contrast, the reactors with higher content of sewage sludge in their feed (R1-R4) showed more alkalinity during the same period (Fig. 3(b)). The sudden pH drop in R6 and R7 at the 2nd day of the operation can be explained by diminution of the buffering capacity resulting from very high VFA content and VFA to alkalinity ratio (VFA:Alk) in these reactors (Fig. 3 (c-d)). Higher buffering capacity in the other reactors (R1-R4 in particular), prevented pH drop. Even a sudden increase in the pH of these reactors was occurred due to their high alkalinity content.

After the 4th day, the VFA content of the reactors did not change much due to the consumption of the most part of readily biodegradable substances. However, pH and alkalinity increased to some extent in all reactors. This indicates that the decomposition of protein substances (such as biomass) lasted all over the operation time and resulted in a

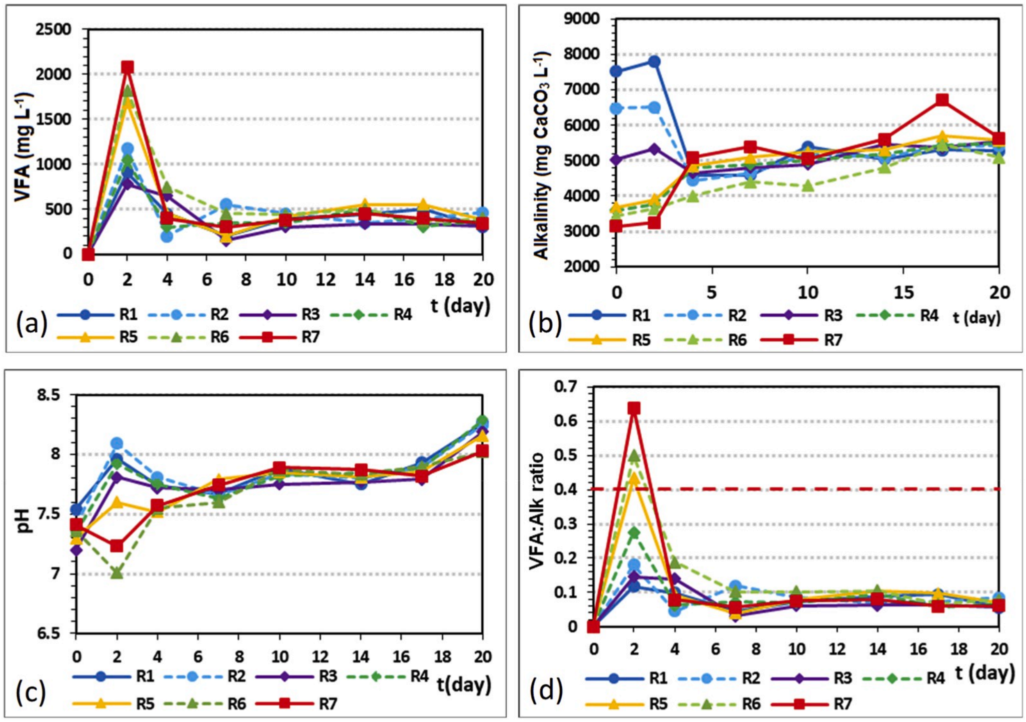

Fig. 3. Changes of VFA (a), Alkalinity (b), pH (c), and VFA:Alk (d) during the operation period.

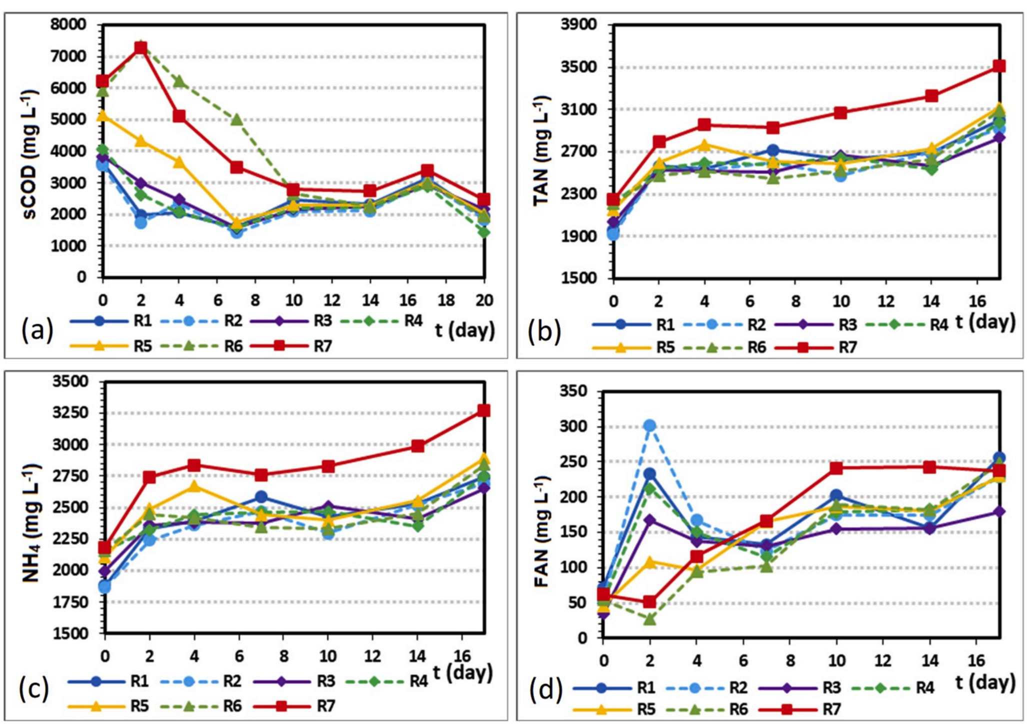

Fig. 4. Changes of sCOD (a), TAN (b), NH_4^+ (c) and FAN (d) during the operation period.

gradual release of ammonia as the main source of alkalinity [53]. The gradual increase of TAN is depicted in Fig. 4(b). Similar trends have been reported by Liu et al. [28] for changes of VFA and alkalinity during the AcoD of FW and sewage sludge.

3.3. sCOD, FAN and TAN

As seen in Fig. 4(a), the sCOD concentration in reactors R1-R5 decreased considerably during the main gas production days. In reactors R6 and R7, the sCOD increased slightly at first and then decreased. The sCOD decrease in the reactors is due to utilization of soluble organic substances that already existed within the feed or are produced from degradation of macro molecule organic matters (conversion of insoluble COD to sCOD) [35,54]. Higher content of VS_{pr} in R6 and VS_{FW} in both R6 and R7 was corresponding to higher amount of raw and readily biodegradable macro molecule organic matters in these reactors compared to the other ones. Therefore, much higher sCOD was released through hydrolysis so that an initial sCOD increase was observed [28]. Some negligible increases in the sCOD occurred further in all reactors which can be attributed to decomposition of the remained refractory macro molecule organic compounds (proteins in particular) [53].

Total ammonia nitrogen (TAN) is formed during the hydrolysis of proteins, urea and nucleic acids. This substance is generally defined as the sum of free (unionized) ammonia nitrogen (FAN, NH_3–N) and ionized ammonium nitrogen ($NH4^+$-N) [52,55,56]. Although low concentrations of ammonia are essential for bacterial growth, they can play an inhibitory role and diminish microbial activity at high concentrations. The inhibitory and toxicity effects of FAN are considerably higher than ammonium [52,55,57]. Specific ranges for toxicity and inhibitory effects of TAN and FAN are not indicated in the reports. This is because the inhibitory effect of these compounds varies depending on nature of substrates, inocula, environmental parameters (pH, temperature), acclimation periods and also the rector operating conditions [52,58].

Data of TAN, FAN and NH_4^+ are presented in Fig. 4(b–d). As seen in Fig. 4(b), TAN concentrations of all reactors were in the range of 1900–3500 mg L^{-1} which according to some previous studies bring a moderate inhibition [59]. However, as previously mentioned, any obvious malfunction or inhibitory effect during the process was not observed for the reactors. This can be due to the low increasing rate of TAN concentration in all reactors. In other words, the gradual increase of TAN concentration allowed the bacteria to adapt and not to be inhibited severely by ammonia. The FAN concentration in the reactors R1-R4 increased sharply at the early days of the operation (Fig. 4(d)). Increase of pH as a result of high initial alkalinity leaded to increase of FAN concentration in these reactors. Further reductions and increases in FAN concentration also corresponded to alkalinity and pH variations. The FAN concentration in all reactors was always less than 300 mg L^{-1} and did not exceed the toxicity range described in the literature (more than 400 mg L^{-1} [60] or 337 mg L^{-1} [61]).

To more accurately investigate the influence of FAN on the AcoD process, the specific gas productions of the AcoD reactors including R3-R6 were calculated stoichiometerically by using the actual specific gas productions of the control reactors (R1, R2 and R7). Then, the obtained values were compared by the actual gas productions of the same reactors. The brief calculations and the results are presented in Table 3. As seen in the table, for R3 and R4 the actual specific biogas productions are slightly lower than the stoichiometric values, while for R5 and R6, the actual values are slightly larger than the stoichiometric ones. In other words, addition of food waste up to 25% of the total VS (VS_{FW}:VS_{SS} of 1:3) adversely influenced the biogas production while a synergistic gas production was observed for the reactors with 50% share of VS_{FW} (VS_{FW}:VS_{SS} of 2:2). Between the reactors with 50% share of VS_{FW}, R5 with VS_{Pr}:VS_{Sc} of 1:1 represented the highest synergistic effect. The synergistic gas production in R5 and R6 can be attributed to pH values and FAN concentrations in these reactors. Higher FAN concentrations in R3 and R4 did not allow the synergistic effect to take place in these reactors. However, the lower pH values and the consequently smaller FAN concentrations in R5 and R6 were desirable for the AcoD process so that the specific biogas production synergistically increased.

3.4. Biogas analysis

The methane contents of biogas for the reactors R1-R7 are presented in Fig. 5(a). Although the highest biogas production was attributed to R7 (FW mono-digestion), R5 showed the highest methane content (70.3%) and the lowest methane content (50.4%) was obtained for R7.

Methane production is influenced by various environmental factors among them pH, temperature and VFA:Alk are accounted as the most important ones [62,63]. The commonly stated optimum pH range for the methanogenesis bacteria is 6.5–7.5 [8,64]. Even more limited ranges of 6.8–7.2 or 6.9–7.3 are reported as the best pH range for the methane formers [3,5]. Generally, pHs of more than 8 and less than 5 disrupt the activity of methanogens and stop the methane production [65]. As seen in Fig. 3(c), R1, R2, R3 and R4 had a pH of more than 7.6 during the main gas production days. This may justify the lowest methane production for R1. However, the methane production of other reactors cannot be explained in comparison to each other according to their pH values.

The VFA:Alk ratio indicates if the digester has enough buffering capacity for the volatile acids being produced. Switzenbaum et al. [63] stated that the optimum range of VFA:Alk ratio in AD process is 0.1–0.35. Some others recommended a VFA:Alk ratio of less than 0.4 to maintain the stability of the process [28,66,67]. Fig. 5(b) shows the relation between the VFA:Alk ratios of the reactors (average of the first 7 days) and their methane productions. A second-order polynomial curve fitted the points with high regression coefficient of 0.91. Such a high R-squared fitting did not obtain between methane production and pH (Fig. 5(c)). As seen in the figure, the highest methane production belongs to a VFA:Alk ratio of about 0.2 in R5 with the VS_{FW}:VS_{SS} of 2:2 and the VS_{Pr}:VS_{Sc} of 1:1. Other reactors showed lower methane production due to VFA:Alk ratio smaller or larger than 0.2.

Except for R1 with a VFA:Alk ratio of less than 0.1, which is out of the range 0.1–0.35, all other reactors had appropriate VFA:Alk ratios. Very low methane production in R1 with substrate composition of VS_{FW}:VS_{SS} of 0:4 and the VS_{Pr}:VS_{Sc} of 1:1, indicates that lack of VFA content in this reactor has resulted in low and inappropriate value of VFA:Alk ratio. Even though R2 did not contain FW in its substrate composition, but containing twofold amount of VS_{Pr} in this reactor (VS_{Pr}:VS_{Sc} of 2:1) resulted in higher VFA concentration, more appropriate VFA:Alk ratio and higher methane production in comparison with R1. Methane production in R3 and R4 can be similarly interpreted. Both reactors had a VS_{FW}:VS_{SS} of 1:3. However, R4 with the VS_{Pr}:VS_{Sc} ratio of 2:1 had a more appropriate VFA:Alk ratio (closer to 0.2) and therefore showed higher methane production than R3. Regarding methane production in R5 and R6 (VS_{FW}:VS_{SS} of 2:2), it seems that containing 50% FW within the substrate composition of R6 simultaneous with the VS_{Pr}:VS_{Sc} ratio of 2:1 in this reactor adversely effected the methane production so that the methane production in R6 was lower than that in R5. Methane production in R7 with 100% FW in its substrate also was much low in terms of percentage of the total gas production. Therefore in the case of R6 and R7, it can be stated that very high content of readily biodegradable

Table 3
Actual and stoichiometric biogas production in AcoD reactors.

Reactor	VS_{Pr}:VS_{Sc}, VS_{FW}:VS_{SS}	Gas production of the control reactors (mL g^{-1} VS_{added})	Calculations for co-digestion reactors	Gas production (mL g^{-1} VS_{added})		Difference (%)
				Stoichiometric	Actual	
R1	1:1, 0:4	672.2	–	–	672.2	–
R2	2:1, 0:4	684.3	–	–	684.3	–
R3	1:1, 1:3	–	(672.2 × 0.75)+(1016 × 0.25)	758.15	744.2	−1.9
R4	2:1, 1:3	–	(684.3 × 0.75)+(1016 × 0.25)	767.23	762.2	−0.7
R5	1:1, 2:2	–	(672.2 × 0.5)+(1016 × 0.5)	844.10	867.5	2.8
R6	2:1, 2:2	–	(684.3 × 0.5)+(1016 × 0.5)	850.15	862.7	1.5
R7	——, 4:0	1016	–	–	1016	–

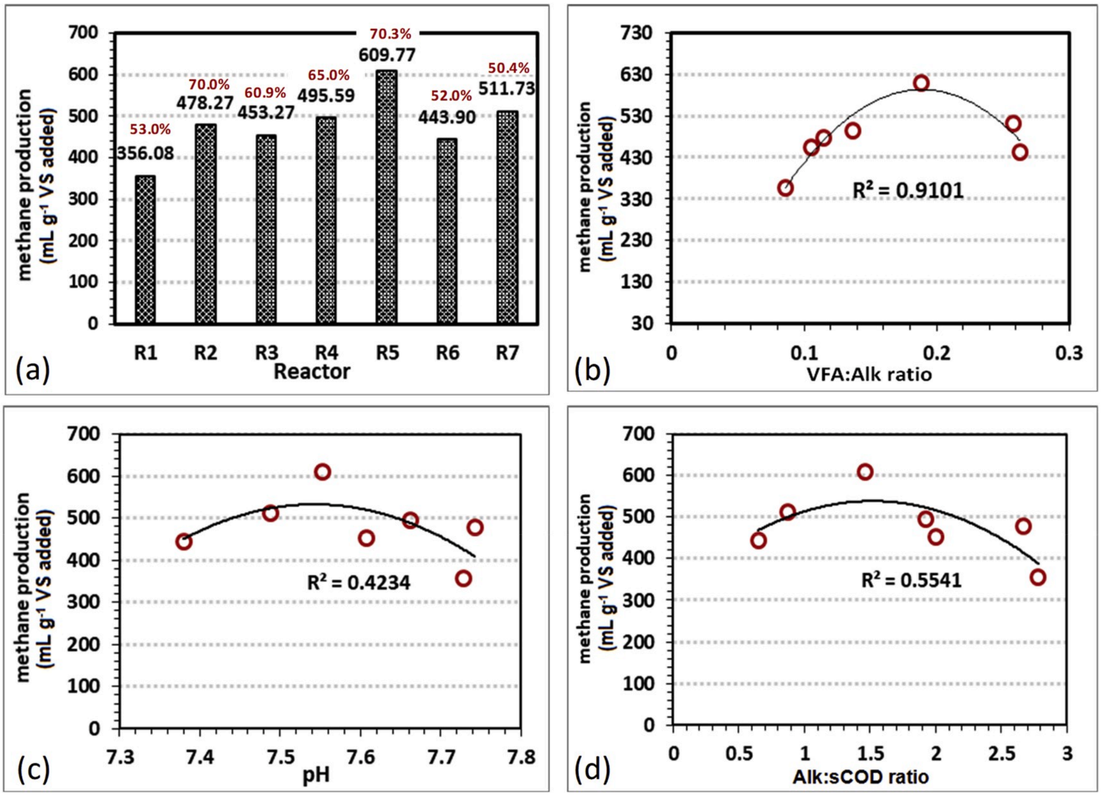

Fig. 5. Methane production value and its percentage of the total gas production (a), correlation between methane production and 7-day average VFA:Alk (b), correlation between methane production and 7-day average pH (c) and correlation between methane production and 7-day average Alk:sCOD (d).

organic matters within the substrate in these reactors has resulted in VFA:Alk ratios of higher than 0.2 so that the methane content of the produced biogas decreased considerably.

In addition to the mentioned parameters, Speece [68] stated that for an efficient methane production in AD process, the ratio of alkalinity to sCOD (Alk:sCOD) should be in the range of 1.2–1.6. Although R5 with the highest methane yield is in the indicated range (Fig. 5(d)), the overall regression coefficient of the fitted polynomial curve is very low (0.55). Accordingly, VFA:Alk ratio can be considered as the most effective factor on the methane yield of the reactors.

3.5. Sludge dewaterability

Specific Resistance to Filter (SRF) is one of the most common indicators used to evaluate sludge dewaterability [47]. This parameter evaluates the sludge resistance to the passage of its water content through a porous media under constant pressure [69]. According to the SRF formulation (Eq. (3)), the higher the SRF index, the worse sludge dewaterability and vice versa [70]. Generally, sludge with an SRF index between 10^{10}- 10^{11} (m kg^{-1}) is classified in the easy-dewatering category. More than 10^{14} (m kg^{-1}) values represent a sludge in difficult-dewatering class [70]. Data of the measured SRF values are presented in Fig. 6(a). The obtained SRF values were between 1.2×10^{14}–2.0×10^{14}. A reason for such high SRF values is that no polymeric or other types of sludge conditioner were added to sludge samples before SRF measurement. Sanin et al. [71]reported that using polymer, the SRF values decreased from 10^{13}-10^{14} to 10^{11} (m kg^{-1}). In addition, the SRF tests were carried out at the end of the operation period. As seen in Fig. 3 (c), pH of all reactors gradually increased over the operation time and reached more than 8 on the last day. As reviewed by Christensen et al. [72], at higher pH values the number of colloidal particles and the concentration of EPSs increase due to floc disintegration. EPSs provide a highly hydrated gel matrix around the bacteria and make it harder to separate water from the sludge tissue [38]. For this reason, pHs of lower than neutral pH are reported to be more desirable for sludge dewaterability [72].

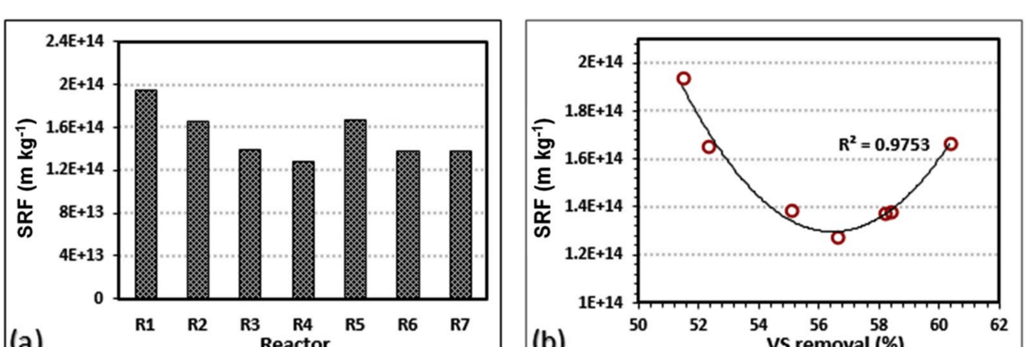

Fig. 6. SRF valuesof the reactors (a) and correlation between data of SRF and VS removal (b).

The obtained SRF values showed a good correlation with the VS removal amounts (Fig. 6(b)). According to the fitted polynomial curve, by increase of the VS removal up to a certain amount (about 57%), the SRF decreased indicating a better dewaterability of sludge. With further increase of the VS removal, larger SRF values were obtained. Taking a look at the SRF formula (Eq. (3)) it can be said that with increase of the VS removal up to a certain value, the water retention capability of biosolids (parameter b of the numerator) decreased to a degree that despite changing other variables, the SRF decreased as well. Beyond that value, the TSS concentration and the viscosity of the mixture (the parameters of the denominator) decreased to a level which resulted in increase of SRF.

Between the reactors with the same content of VS_{FW}, the reactors with two-fold VS_{pr}:VS_{sc} (R2, R4 and R6) showed lower SRF values. This can be attributed to lower content of biosolids and water retentive substances such as EPS in these reactors.

4. Conclusions

The following key points were demonstrated in the present work:

- Compared with VS_{Pr}:VS_{Sc} ratio, C:N ratio was found as a much more influencing factor on both VS reduction and biogas production efficiencies.
- A second-order polynomial curve fitted the data of VS reduction vs. C:N ratio, representing an optimum C:N ratio of around 16 for obtaining the highest VS reduction.
- A linear correlation was found between data of gas production and C:N ratio according to which, with increasing the C:N ratio from 8 to 19.7, the biogas production increased steadily.
- Among different applied conditions, R7 with a VS_{FW}:VS_{SS} of 4:0 showed the highest biogas production. However, R5 with a VS_{FW}:VS_{SS} of 2:2 and VS_{Pr}:VS_{Sc} of 1:1 represented the highest values for both VS reduction and methane production.
- Compared with pH and Alk:sCOD, VFA:Alk was distinguished as the most effective parameter on the methane yield of the reactors.
- Synergistic gas production was observed for 50% share of VS_{FW} under both VS_{pr}:VS_{sc} ratios of 1:1 and 2:1.
- Between the reactors with the same ratios of VS_{FW}:VS_{SS}, the reactors with two-fold VS_{pr}:VS_{sc} (R2, R4 and R6) showed lower SRF values which could be attributed to lower content of biosolids and water retentive substances in these reactors.

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