doi: 10.1111/1471-0307.12252

IJDT684.

REVIEW

Anaerobic granular reactors for the treatment of dairy wastewater: A review

DOGAN KARADAG,[1,2]* OGUZ EMRE KOROGLU,[1] BESTAMI OZKAYA,[1] MEHMET CAKMAKCI,[1] SONIA HEAVEN,[2] CHARLES BANKS[2] and ALBA SERNA-MAZA[2]

[1]*Department of Environmental Engineering, Faculty of Civil Engineering, Yildiz Technical University, Istanbul, Turkey, and* [2]*Faculty of Engineering and Environment, University of Southampton, Southampton, SO17 1BJ, UK*

Considerable research has been conducted on the treatment of dairy wastewater by anaerobic granular reactors. Upflow anaerobic sludge blanket (UASB) reactors, anaerobic sequencing batch reactors (ASBR) and static granular bed reactors (SGBR) are the conventional granular reactor types most commonly applied in dairy wastewater treatment. Hybrid systems have also been developed to increase treatment efficiency and overcome the operational problems associated with the treatment of this substrate. Effects of parameters including temperature, organic loading and operating protocols on the performance of granular reactors are summarised. Individual and hybrid granular reactors are evaluated based on organic matter removal and methane production capacity.

Keywords Dairy wastewater, Granular reactor, Methane, Organic loading.

*Author for correspondence. E-mail: dogankaradag@gmail.com

INTRODUCTION

A billion tons of dairy products is consumed worldwide each year, and global demand has been increasing significantly with rising population and living standards (Faye and Konuspayeva 2012). Dairy production facilities are one of the most important industrial wastewater sources, as a large volume of water is used in all production steps and in equipment cleansing. Effluents from dairy production systems are rich in carbohydrates, proteins and fats which are major sources of wastewater pollution (Prazeres *et al.* 2012; Traversi *et al.* 2013). Dairy process wastewaters are typically high-strength effluents with high concentrations of organic matter, suspended solids and oil–grease along with varying amounts of other pollutants (Table 1).

Anaerobic treatment has the dual benefits of reducing pollution and producing renewable energy, and dairy process wastewater with its high organic content is thus a valuable potential resource for energy production. In anaerobic conditions, organic matter can be directed towards the production of hydrogen (H_2) or methane (CH_4). A previous review (Karadag *et al.* 2014) showed how conventional anaerobic reactors and hybrid systems have been successfully employed to convert dairy industry wastewaters to H_2. In comparison with H_2 generation, CH_4 producing systems convert nearly all organics to final products and are less sensitive to changes in operational conditions (Shuizhou *et al.* 2005).

Anaerobic reactors can be operated using suspended, granular or biofilm micro-organisms. The superior settling characteristics of granules allow higher biomass concentrations to be maintained in the reactor and prevent the easy washout of micro-organisms: compared to other reactor types, granular reactors may also have the advantages of withstanding shock loads and of stable operation under high organic loadings (Leitão *et al.* 2006; Couras *et al.* 2014; Lim and Kim 2014). Upflow anaerobic sludge blanket (UASB) reactors, expanded granular sludge bed (EGSB) reactors and anaerobic sequencing batch reactors (ASBR) are well-known high-rate granular reactors and have been successfully employed for the treatment of high-strength wastewaters from different sources (McHugh *et al.* 2003; Turkdogan *et al.* 2013;).

Table 1 Dairy industry wastewater characteristics

Wastewater Source	*pH*	*COD (g/L)*	*Solids (g/L)*	*Nitrogen (mg/L)*	*Phosphorus (mg/L)*	*FOG (g/L)*	*References*
Cheese production	6.7	2.93	2.75 (TS)	36 (TN)	21	0.294	Gutierrez *et al.* (1991)
Ice cream	6.96	4.94	1.1 (TSS)	NA	NA	NA	Hawkes *et al.* (1995)
Cheese production	5.5–9.5	1.0–7.5	0.5–2.5	NA	NA	NA	Monroy *et al.* (1995)
Whey	4.3–8.7	5.4–77.3	3.9–58.9 (TS)	500–5600 (TN)	NA	0.4–5.7	Kalyuzhnyi *et al.* (1997)
Butter production	5.8	1.91	1.72	NA	NA	NA	Strydom *et al.* (1997)
Cheddar cheese production	6.2	7.62	6.34	106 (TKN)	20	NA	Danalewich *et al.* (1998)
Milk and cream bottling plant	8–11	2–6	0.3–1.0 (TSS)	50–60	20–50	0.3–0.5	Ince (1998)
Whey	3.92	74.5	9.38 (SS)	145.6 (TN)	124	NA	Erguder *et al.* (2001)
Dairy wastewater	NA	18	7.18 (TSS)	329 (TKN)	637	4.89	Arbeli *et al.* (2006)
Whey	5.5–6.6	50–70	55–65 TS	NA	NA	NA	Najafpour *et al.* (2008)
Simulated milk	6.5–7.0	0.15–11	NA	75–550 (TN)	12–88	NA	Vlyssides *et al.* (2008)

NA, not available; SS, suspended solid; TS, total solid; TN, total Nitrogen; TKN, total Kjeldahl nitrogen; FOG, fats, oil and grease.

Although many anaerobic systems have been documented for the treatment of dairy industry wastewaters (Demirel *et al.* 2005; Arbeli *et al.* 2006; Lim and Kim 2014), there is no report focusing specifically on the treatment of dairy wastewater by anaerobic granular reactors. In the present work, publications on anaerobic granular reactors treating dairy wastewater have been reviewed from the viewpoint of operational strategies and performance has been compared on the basis of organic matter removal and methane production.

ANAEROBIC GRANULAR REACTORS

Anaerobic granular reactors have proven successful for treatment of high-strength food-processing wastewaters (Ibrahim *et al.* 2013; Kushwaha 2015; Rajagopal *et al.* 2013). Reactor studies on dairy wastewaters have mostly been conducted in a pH range of 6.6–7.2 (Hawkes *et al.* 1995; Buntner *et al.* 2013), while some researchers have operated at higher pH (7.1–7.6) (Banu *et al.* 2008). In some cases, the desired pH in the reactor has been maintained by adding acid or basic chemicals (Gavala *et al.* 1999; Banu *et al.* 2008), while in other studies, untreated wastewater was fed directly to the reactor without pH adjustment (Passeggi *et al.* 2012). A decrease in pH is usually associated with accumulation of volatile fatty acids (VFAs) within anaerobic reactors and this is especially common in the start-up period when the reactor performance in terms of removal of organics and production of CH_4 is lower. In order to prevent the rapid increase in VFAs, the reactor needs to have a sufficient quantity of alkalinity. The required alkalinity can be supplied by the addition of buffer chemicals such as $NaHCO_3$ (Gutierrez *et al.* 1991; McHugh *et al.* 2006), $NaCO_3$ or NaOH (Kalyuzhnyi *et al.* 1997; Bandara *et al.* 2011) into the wastewater feed or directly to the reactor. On the other hand, anaerobic degradation of proteinaceous compounds in the dairy wastewater releases amino groups and ammonia which contribute to the generation of alkalinity (Rajeshwari *et al.* 2000). Banu *et al.* (2008) reported an improvement in the alkalinity of an anaerobic reactor treating dairy wastewater due to the increase in protein content when the wastewater concentration was increased.

Studies on dairy wastewater treatment in anaerobic granular reactors have mainly been performed at mesophilic temperatures such as 30 °C (Mockaitis *et al.* 2006; Ganesh *et al.* 2007) or 35–37 °C (Kim *et al.* 2004; Leal *et al.* 2006; Najafpour *et al.* 2008). Only a few studies have been conducted at psychrophilic (10–20 °C) (McHugh *et al.* 2006; Tawfik *et al.* 2008; Bialek *et al.* 2013) and thermophilic temperatures (55 °C) (Zielinska *et al.* 2013). The main drawbacks of anaerobic treatment at psychrophilic temperatures are the decrease in CH_4 in the gas phase due to increased solubility at lower temperatures and the low growth rate of the methanogenic community. Researchers were able to recover dissolved methane and increase COD removal efficiency by installing a degassing membrane unit at the outlet of a UASB reactor (Bandara *et al.* 2011).

Heating of granular reactors has been accomplished by water jacket (Gutierrez *et al.* 1991; Najafpour *et al.* 2008), electrically (Hawkes *et al.* 1995), using microwave (Zielinska *et al.* 2013) or by maintaining the reactor in a temperature-controlled room (Ramasamy *et al.* 2004; Kundu *et al.* 2013). Zielinska *et al.* (2013) compared the effect of microwave and water jacket heating on the performance of anaerobic hybrid reactors. It was reported that microwave heating significantly improved biogas production by enhancing methanogenic diversity. Similarly, other researchers reported that improvement in CH_4 production with microwave-heated anaerobic reactors is associated with improvements in microbial diversity and growth (Banik *et al.* 2003; Kwiatkowsk *et al.* 2012).

Granular reactors have been fed on real dairy wastewater from whey production (Gavala *et al.* 1999; Erguder *et al.* 2001), ice-cream production (Goodwin *et al.* 1990; Borja and Banks 1994; Hawkes *et al.* 1995), cheese production (Gutierrez *et al.* 1991) or simulated wastewater that was prepared by mixing whole milk or milk powder with water (Haridas *et al.* 2005; Belancon *et al.* 2010). In their study, Ganesh *et al.* (2007) used washing wastewater from a dairy plant as substrate in a UASB. Some researchers initially fed reactors with simulated substrate or diluted dairy wastewater to adapt the microbial consortium to operational conditions (Belancon *et al.* 2010). The other strategy applied for easy adaptation was operation in batch mode before continuous feeding (Kalyuzhnyi *et al.* 1997; Ganesh *et al.* 2007). In continuous reactor systems, the amount of substrate is commonly optimised based on organic loading rate (OLR), which can be adjusted by changing influent COD concentration or hydraulic retention time (HRT). It was reported that mass transfer and anaerobic degradation rates are more favourable at high OLR, while slow-growing methanogens are easily washed out at lower HRTs (Kundu *et al.* 2013).

UPFLOW ANAEROBIC SLUDGE BLANKET REACTORS

Compared to other anaerobic reactors, UASB reactors have some advantages, such as simple construction, low operational cost and high organic removal efficiencies (Latif *et al.* 2011). At relatively short HRTs, UASB can develop a high concentration of granular sludge with good settling properties and a rich methanogenic population (Ozgun *et al.* 2013). UASB operation on dairy wastewater has been initiated by transferring granules from other UASB reactors (Borja and Banks 1994) or by self-granulation from various inoculum sources (Goodwin *et al.* 1990; Hawkes *et al.* 1995). Ramasamy *et al.* (2004) investigated the treatment of dairy wastewater by two UASBs with self-granulated and transferred granules. It was reported that the self-granulated UASB gave lower organics removal at the initial operational stage but there was no difference in the performance of both reactors in long-term operation.

In self-granulation, the start-up procedure is crucial for the development of granules with high methanogenic activity and good settling properties. The long start-up time of up to several months is a bottleneck in UASB operation; however, Najafpur *et al.* (2008) were able to shorten the start-up duration by developing tubular flow in a UASB reactor. McHugh *et al.* (2006) reported that inoculation with a high concentration of well-settling granular sludge contributes to successful and rapid start-up of reactors. Granules developed from sludge with low-volatile solids and poor settling characteristics lead to easy washout of micro-organisms, low organics removal and poor biogas production. The carbon content of the wastewater also significantly affects the physical structure and microbial diversity in granules, while the presence of carbohydrates and additives such as natural and cationic polymers can improve granulation (Yang and Anderson 1993; Ramasamy *et al.* 2004). Vlyssides *et al.* (2008) reported that Fe^{2+} supplementation provided a 56% increase in granule diameter in a UASB. The positive effect of iron was associated with the generation of inert nuclei from ferrous sulphide precipitates with the attachment of biomass around the nuclei. Iron accumulation within granules also interacts with exopolysaccharide polymers along with sulphide ions, and the granule colour can become darker (McHugh *et al.* 2006). The structure and functions of granular layers have also been well documented by researchers. Satoh *et al.* (2007a, b) performed microsensor measurements in granule layers from a UASB treating dairy wastewater and revealed that granules have considerable microbial diversity and different anaerobic processes are carried out in different distinct layers of a granule. Complex organics are hydrolysed at the surface of the granule, while simple organics are fermented to fatty acids, alcohols and H_2 in the middle layers, and then, CH_4 is produced in the inner layers.

Organic loading rate has a considerable effect on granulation; however, conflicting results have been reported on the relationship between these parameters. Although rapidly increasing OLR was reported as helping to enhance granule formation (Gutierrez *et al.* 1991; Hawkes *et al.* 1995), some researchers have indicated that a sudden increase in OLR can cause loss of specific sludge activity, washout of granules and reduction in COD removal efficiency (Yan *et al.* 1988; Ramasamy *et al.* 2004). Ramasamy *et al.* (2004) recommended start-up of UASB with a low organic loading of 2.4 kg COD/m^3/day to allow easy acclimation to dairy wastewater. It has been reported that the addition of more readily biodegradable carbon sources, such as methanol, in the start-up period can improve granule formation, settling velocity and reactor stability (Cayless *et al.* 1990). On the other hand, high nitrogenous content in the wastewater may increase sludge loss by up to 60% due to the agitation of the granular bed by nitrogen bubbling (Yan *et al.* 1989; Goodwin *et al.* 1990). Design faults in the phase separator and high lipid content of wastewater may negatively affect granulation, while a low acetate concentration inside the UASB reactor enhances granulation (Hawkes *et al.* 1995). Additionally, anaerobic bacteria treating whey tend to produce large amounts of sticky exopolymers which adversely affect granulation (Janczukowicz *et al.* 2008).

Effect of organic loading on UASB

Comprehensive studies have been performed to optimise OLR values and incrementing strategies for efficient UASB operation, but contradictory results have been reported for dairy wastewater treatment. Variations in optimum OLR values have been related to differences in wastewater characteristics, operational conditions and design parameters for UASB. Applied OLR values in UASB studies and performance comparisons with other reactors are given in Table 2. For successful operation of UASB on whey, stepwise increment of influent COD is recommended (Yan *et al.* 1989) and VFA concentrations should be closely monitored before changing OLR as the highest CH_4 production could be obtained at the lowest acetic and propionic acid levels (Yan *et al.* 1988). Yan *et al.* (1989) obtained 97% COD removal with 57% CH_4 content at OLR of 2 kg COD/m^3/day for whey treatment, while Gavala *et al.* (1999) reported that OLR up to 7.5 kg COD/m^3/day has no negative effect on UASB stability. Yan *et al.* (1993) suggested the optimal influent concentration for system stability at an HRT of 5 days was 25–30 g COD per L, corresponding to an OLR of 5 kg COD/m^3/day. Yu *et al.* (2002) reported that biogas production from simulated dairy wastewater increased with increasing OLR up to 12 kg COD/m^3/day, whereas further increases in OLR negatively affected the performance of UASB at both mesophilic and thermophilic temperatures. Kalyuzhnyi *et al.* (1997) obtained stable COD removal of over 90% for the treatment of whey without sludge washout at 35 °C when a UASB was operated at OLR of 28.5 kg COD/m^3/day, but further increases in OLR caused washout and deterioration in the reactor performance. When the temperature was reduced to 25 °C, the microbial community rapidly adapted to the new conditions and gave a COD removal efficiency of greater than 90% at OLR of 7.0–9.5 kg COD/m^3/day. The authors concluded that the OLR limit to avoid washout in UASB at submesophilic temperatures is 9.5 kg COD/m^3/day and the optimal OLR for whey treatment at 35 °C is 28.5 kg COD/m^3/day.

Yan *et al.* (1988) indicated that increasing the OLR by decreasing HRT is more effective than increasing COD concentration in terms of maintaining the microbial community structure and methane production in UASB treating dairy wastewater. However, different optimum HRT values have been reported for stable UASB operation on dairy wastewater. Hwang *et al.* (1992) recommended keeping the HRT above 0.8 day for successful operation of UASB, while Gutirrez *et al.* (1991) obtained stable COD removal of around 97% at HRT of 0.17 day. Erguder *et al.* (2001) stated that 95–97% COD removal from whey is possible at HRT values of 2–5 day. On the other hand, UASB operation at very short HRT promotes VFAs generation, while accumulation of propionate deteriorates reactor performance by inhibiting methanogenic archaea. Similarly, Borja and

Table 2 Comparison of anaerobic granular reactors treating dairy wastewater

Reactor	*Wastewater*	*Temp. (°C)*	*pH*	*HRT (day)*	*OLR (kg COD m^3/day)*	*Inf. COD (g/L)*	*COD removal (%)*	*CH_4 yield (m^3 CH_4 per kg COD)*	*References*
UASB	Whey		7–8	2–5	10.4–24.6	42.7–55.65	91.9–97.0	0.424	Erguder *et al.* (2001)
UASB	Whey	35	6.6–7.3	6–40	1.5–7.3	12–60	79–99	NA	Gavala *et al.* (1999)
UASB	Cheese production	35	7.3	0.07–0.5	4.6–31	1.7–2.34	88–97	0.29–0.33	Gutierrez *et al.* (1991)
UASB	Whey	20–35	7.0–7.5	2.3–12.8	3–28.5	5–77	90–99	NA	Kalyuzhnyi *et al.* (1997)
UASB	Simulated dairy	30	NA	0.13–0.5	2.4–13.5	1.44	37–96.3	NA	Ramasamy *et al.* (2004)
UASB	Washing wastewater	30	6.8–7.4	0.25–0.75	0.80–9.60	0.6–2.0	75–85	NA	Ganesh *et al.* (2007)
Hybrid	Simulated milk	37	NA	0.75–5	2.22–31	10–77.5	78	0.27	Kundu *et al.* (2013)
Hybrid	Dairy	12–20	7–8	0.75–2	5–13.3	5–10	52.3–91.9	NA	McHugh *et al.* (2006)
Hybrid	Whey	36	6.5	1.5–2	7.9–45.42	50–70	77–97.5	NA	Najafpour *et al.* (2008)
Hybrid	Simulated dairy	35, 55	NA	1	1,2	1,2	64–76	0.037–0.245	Zielinska *et al.* (2013)
Hybrid	Butter production	35	7.8		0.97	1.84	91	0.287	Strydom *et al.* (1997)

Banks (1994) observed instability in UASB performance when HRT was reduced from 5 day to 0.4 day during the treatment of ice-cream processing wastewater.

Effect of temperature on UASB performance

Treatment studies of dairy wastewater in UASB have mainly been conducted in mesophilic temperatures (Goodwin *et al.* 1990; Borja and Banks 1994; Vlyssides *et al.* 2009), while a few UASB reactors have been operated in the thermophilic range. Thermophilic UASB treatment of dairy wastewater has mainly been conducted as a preliminary acidification step (Yu and Fang 2000). Operation of UASB fed on simulated milk wastewater at 37 and 55 °C indicated that temperature has no effect on COD reduction and acidification level; however, biogas production was higher at thermophilic temperature (Yu *et al.* 2002). In recent years, operation of anaerobic reactors at psychrophilic temperatures has gained great attention as there is no need for an energy input to maintain higher temperatures (Colins *et al.* 2013). During the operation of anaerobic reactor at lower temperatures, microbial growth rate and methanogenic activity decrease and biomass washout occurs (Akila and Chandra 2007; Janczukowicz *et al.* 2008). In order to overcome these disadvantages, psychrophilic reactors are recommended to operate at higher HRT (Chu *et al.* 2005) and to start up with a large amount of biomass (Rebac *et al.* 1999).

Operation of two-phase UASB reactors has been recommended for efficient treatment of dairy parlour wastewater at ambitious temperatures (Luostarinen and Rintala 2005). Start-up of a UASB reactor with mesophilic digester sludge and stepwise decreasing of the reactor temperature prevented the washout of biomass and provided over 80% COD removal at 10, 15 and 20 °C. At 30 °C, Ramasamy *et al.* (2004) obtained over 96% COD removal by UASB at HRT of 3 h, but this decreased slightly when the HRT was increased to 12 h. The highest COD reduction was at OLR of 10.5 kg COD/m^3/day and reactor performance dropped when OLR was further increased. The researchers indicated that optimal reactor performance for the treatment of dairy wastewater could be obtained with HRT of 3 h and OLR of 13.5 kg COD/m^3/day. Buntner *et al.* (2011) operated a UASB reactor for the treatment of low-strength dairy wastewater (600 mg COD per L) at an ambient temperature range of 23–17.5 °C. They reported that the UASB had high tolerance to changes in temperature and organic loading, while COD removal efficiency and methane content were around 80%.

Tawfik *et al.* (2008) employed different operational schemes to overcome the difficulties in psychrophilic operation of UASB. Domestic wastewater was mixed with dairy influent at 30% as it has easily biodegradable organics, and a high solid retention time (76 days) was applied. Achievement of constant organic removal efficiencies with the average values of 69% COD along with 72% total suspended solids (TSS) removal was associated with effective hydrolysis and degradation at long sludge retention times. The highest reactor performance was obtained at OLR of 3.4 kg COD/m^3/day, and regular intentional sludge discharge from the UASB prevented washout of granules. Application of activated sludge treatment after the psychrophilic UASB provided an excellent overall COD removal of 98.9%.

Effect of supplementation with trace elements

It has been reported that supplementation of nutrients and trace metals significantly improves COD removal and biogas production in UASB (Hawkes *et al.* 1992). Murray and van den Berg (1981) reported that supplementation of Ni^{2+}, Co^{2+} and Mo^{2+} enhanced the treatment performance of food industry wastewater in UASB by increasing the amount of methane-producing archaea. During the treatment of undiluted whey, Erguder *et al.* (2001) achieved the operation of UASB at lower HRT (2–5 day) by supplementation with nutrients and trace elements. It has been reported that Zn^{2+} up to 10 mg/L slightly enhances acidogenesis, while Cu^{2+} is toxic even in trace amounts during the treatment of dairy wastewater. The inhibitory effects of Zn^{2+} and Cu^{2+} increase at higher concentrations, while Cu^{2+} is 1.4–4.3 times more toxic than Zn^{2+} in degradation of carbohydrates and protein (Yu and Fang 2001). Vlyssides *et al.* (2012) obtained over 98% COD removal during the treatment of simulated milk wastewater with ferrous iron addition, which was 24% higher than for a control reactor not receiving ferrous iron. The researchers indicated that iron addition increases anaerobic treatment performance by promoting microbial growth, granule diameter, absorption of carbon materials and precipitation of sulphate (Vlyssides *et al.* 2007).

Operational problems in UASB

Dairy wastewater contains a significant quantity of lipids, and accumulation of these in a UASB causes several operational problems including sludge flotation, biomass washout, mass transfer reduction, impairment of sludge settling capacity and lower sludge activity (Passeggi *et al.* 2012). The research indicated that the lipid concentration should be less than 100 mg/L for successful treatment of dairy wastewater in mesophilic conditions. On the other hand, Leal *et al.* (2006) reported that dairy wastewater with a high lipid content of up to 1000 mg/L could be successfully treated in UASB. Degradation of lipids could be improved by the addition of extracellular enzyme or application of pretreatment. The researchers applied various pretreatment methods to prevent lipid-related problems. It has been reported that Fenton oxidation before a UASB reactor could be a solution for this problem (Yu and Fang 2001). Fenton oxidation removed 80% of lipids by converting them into soluble organics or fully mineralising them. Fenton treatment plus UASB provided satisfactory COD removal of over 90%, along with 85% methane content at 27 °C. Ferrous iron addition through Fenton oxidation also enhanced the sludge activity and granule formation, with excellent carbon removal and also sulphide precipitation. Cammarota *et al.* (2001) obtained COD removals below 50% during the operation of UASB fed on dairy wastewater with high lipid content. Leal *et al.* (2006) hydrolysed dairy wastewater with the enzyme of *Penicillium restrictum* and obtained COD removal efficiencies of 90% at low fats, oil and grease (FOG) concentrations. However, COD removal efficiency decreased from 91% to 82% when the FOG concentration in the raw wastewater increased from 200 to 1000 mg/L. Accumulation of lipids under the biogas hood in UASB also prevents effective escape of biogas and causes break-up of the UASB bed, which results in poorer contact between micro-organisms and substrate with limited biofilm formation and lower methane yields (Gutierrez *et al.* 1991; Hawkes *et al.* 1995). Blonskaja and Vaalu (2006) recommended sludge recirculation from a sedimentation tank to the UASB to prevent biomass wash-out. Passeggi *et al.* (2012) recirculated biogas bubbles into a UASB to enhance the gas escape and obtained enhanced treatment efficiency.

Long-chain fatty acid (LCFA) is a hydrolysed product of lipids, and LCFA accumulation causes operational problems, including limited substrate removal and decrease in reactor performance (Alves *et al.* 2001; Pereira *et al.* 2003). Proper design of the UASB is important; otherwise, the small reactor diameter may cause a piston effect resulting in biomass flotation even at very low LCFA concentrations. Inhibition by high concentrations of LCFA could be eliminated by dilution, addition of adsorbents and changing feeding patterns. Palatsi *et al.* (2009) indicated that adsorbent addition is a more reliable strategy for industrial treatment plants, while bentonite binds LCFA and improves the recovery time of the anaerobic reactor.

Intermittent feeding has been proven effective as a method to improve the performance of various anaerobic reactors during the treatment of complex wastewaters with high lipid content (Del-Pozo *et al.* 2000; Nadais *et al.* 2006; Neves *et al.* 2009). In intermittent operation, feeding of the reactor is stopped for a certain time to allow complete degradation of substrates accumulated within the reactor. Nadais *et al.* (2011) compared the performance of continuous-fed and intermittent-fed UASB for dairy wastewater treatment and obtained enhanced COD removal and 25% higher methane production with intermittent feeding. The improvement in reactor performance was related to higher organic matter degradation and better adaptation of the biomass to the complex substrates. Furthermore, Nadais *et al.* (2005) achieved an increase in OLR of up to four times with intermittent operation when treating dairy wastewater in UASB.

Methanogenic archaea in UASB are sensitive to inhibition by dissolved oxygen, toxic chemicals, ammonia, accumulated VFAs and heavy metals (Yenigun and Demirel 2013). Buntner *et al.* (2011) reported that inhibition of methanogens due to a high concentration of methanol reduced COD removal below 20% with CH_4 content less than 50%. Accumulation of VFAs is a common problem with UASB operation, where it causes a decrease in pH and inhibits methanogenic activity (Omil *et al.* 2003). Among the VFAs, the amount of propionate should be monitored closely as it is toxic to methanogens at concentrations higher than 2000 mg/L (Yan *et al.* 1988; Kundu *et al.* 2013). Problems related to VFA accumulation could be eliminated by the addition of alkalinity or by decreasing the OLR. Ghally *et al.* (2000) recommended reseeding the UASB only if alkaline addition did not recover the reactor. Mixing of dairy wastewater with other waste streams may also prevent inhibition of the methanogenic community. Demirel *et al.* (2013) reported that ice-cream production residues contain a large amount of sulphate and mixing them with wastewater from an ice-cream-producing plant at appropriate ratios eliminated sulphur inhibition. Chemicals in wastewater from the control laboratory in a dairy industry plant could be highly toxic for methanogens, and mixing with other effluents is beneficial for efficient anaerobic degradation (Lopez-Fiuza *et al.* 2002).

Different operational schemes for UASB

When an anaerobic reactor fails due to low pH, a two-stage process becomes essential for improving the biogas production rate and methane yield (Rajeshwari *et al.* 2000). Higher performance and stability were reported when dairy wastewater was treated in a two-stage UASB system. In two-stage operation, wastewater is acidified in the first reactor and CH_4 is produced in the second reactor. Generally, a continuous stirred tank reactor (CSTR) is installed prior to the UASB to acidify the dairy wastewater. Diamantis *et al.* (2014) compared the performances of varying configurations of CSTR and UASB. The experimental results indicated that the use of a CSTR followed by biomass separation and recirculation prior to the UASB provided complete acidification of carbohydrate and higher COD removal of up to 90%, along with superior methane yield. Recirculation of separated biomass to the CSTR also eliminated alkali addition and the UASB achieved a stable treatment performance at OLR values of up to 20 kg COD/m^3/day.

Kim *et al.* (2004) compared the effect of LCFA on the performance of single-stage UASB and a two-stage system of CSTR plus UASB. They reported that LCFA did not cause significant problems in two-stage systems up to 3.5–4.0 g LCFA-COD per L, and COD removal was over 95%. Two-stage treatment gave stable organic removals in all conditions, whereas the performance of a single-phase system deteriorated when the influent wastewater had 4.0 g LCFA-COD per L. Degradation of lipids in a CSTR enhanced UASB stability and treatment efficiency. The two-stage treatment provided 1.19- and 1.42-fold higher COD removal and CH_4 production, respectively (Kim *et al.* 2006).

In some cases, a UASB is inadequate to meet discharge limits and requires a posttreatment step for dairy wastewater. The application of a membrane bioreactor (MBR) after UASB was reported with excellent COD and suspended solids removal in stable operational conditions, while no or very little nitrogen and phosphorus reduction was achieved (Buntner *et al.* 2011). Buntner *et al.* (2013) combined a UASB with a two-compartment aerobic MBR for the treatment of dairy wastewater at ambient temperatures (17–25 °C). The UASB and the combined system were operated at HRTs of 10, 15 and 11 h for the UASB and 14, 20 and 15 h for the combined system. COD removal was 66–85% after the UASB while it increased to 99% in the combined system along with 73% methane content. Erguder *et al.* (2001) operated two UASB reactors in series for the treatment of whey and stated that the second UASB gave a slight improvement in COD removal; however, this enhancement could be achieved by increasing the HRT of a single-stage UASB.

OTHER GRANULAR REACTORS

Anaerobic sequencing batch reactor

Anaerobic sequencing batch reactor (ASBR) can be operated with granular or immobilised biomass (Fuzzato *et al.* 2009) and has been successfully employed for the treatment of dairy and other wastewaters (Dugba and Zhang 1999; Bodik *et al.* 2002; Mockaitis *et al.* 2006). Ndon and Dague (1997a,b) studied the treatment of low-strength nonfat dry milk wastewater in an ASBR at various temperatures, substrate concentrations and HRTs. Granule development was observed at a HRT of 12 h, while no granulation was present when the HRT was increased. Granule formation at this short HRT was due to the selection of good settling particles at higher hydraulic loadings, while lighter particles were washed out of the reactor. Operation of the ASBR at higher organic loadings is recommended for the treatment of low-strength wastewater as the OLR provides the necessary nutrients for granules. The granular ASBR achieved over 85% COD removal at all substrate concentrations when operated at temperatures over 15 °C; however, treatment performance at 15 °C declined at elevated substrate concentrations. The deterioration in CH_4 production at lower temperature was associated with biomass washout due to the low biomass settling velocity. Similarly, Banik *et al.* (1997) investigated the effect of lower temperatures (5, 15 and 25 °C) on granule activities in an ASBR treating nonfat dry milk. The ASBR was inoculated with mesophilic biomass, and no significant variation was observed in the microbial community when the temperature was decreased stepwise. At lower temperatures, mesophilic bacteria were active and had the capability for rapid degradation of organics to acetate and propionate even after prolonged operation. Mockaitis *et al.* (2006) employed an ASBR containing granular biomass for the treatment of whey at 30 °C. The mechanically stirred reactor gave stable treatment performance at all organic loadings studied and achieved organic matter removal efficiencies close to 90%. Addition of $NaHCO_3$ as a buffering chemical increased the organic loading capacity of reactor. Biomass flotation occurred due to release of carbonic gas, however, and the amount of floating biomass increased with increasing alkalinity supplementation. Pretti *et al.* (2011) compared the performance of an ASBR and an anaerobic sequencing biofilm batch reactor (ASBBR) for the treatment of wastewater from a dairy plant after fat separation and found the ASBR gave superior performance especially at a 24-h cycle. Matsumoto *et al.* (2012) used an ASBR followed by an aerobic sequencing batch reactor to maximise the removal of organics and nitrogen: the ASBR was able to achieve 91% COD removal over a 24-h cycle at an OLR of 4.5 kg COD/m^3/day.

The static granular bed reactor

The static granular bed reactor (SGBR) is a novel anaerobic reactor which has no mixing process and can offer significant energy savings. The reactor operates in downflow mode, while the upper part contains a headspace for gas collection (Debik and Coskun 2009). During the treatment of simulated wastewater composed of nonfat dry milk and sucrose, the SGBR demonstrated significantly higher performance at a HRT of 8 h with an average COD removal of 90.7% compared to 77.5% in a UASB (Evans and Ellis 2005, 2010). A pilot-scale (42.5 m^3 working volume) SGBR treating dairy processing wastewater at ambient temperature (10–29 °C) in Tulare, California, was operated for 7 months at HRT from 9 to 48 h and OLR from 0.63 to 9.72 kg COD/m^3/day. The system consistently achieved over 90% COD removal, and average TSS removal was over 80% (Park *et al.* 2012). Trials at the same site using a 5.7 m^3 pilot-scale SGBR showed significant accumulation of nondegraded particulate organics at HRT of less than 18 h and OLR greater than 3.5 kg COD/m^3/day, especially at lower temperatures; these were removed by backwashing through valves in the side of the reactor (Oh *et al.* 2015).

Hybrid reactors

Hybrid reactors have been developed by combining the properties of granular and biofilm systems to increase the treatment efficiency of conventional granular reactors (Najafpaur *et al.* 2008; Passeggi *et al.* 2012). Hybrid systems are constructed by modifying conventional anaerobic reactors. In two-compartment reactors, granule and biofilm communities are grown in different compartments and work simultaneously. Wastewater is firstly fed to the granule-containing compartment at the bottom of the reactor, and a biofilm is developed on various support media in the upper part of the reactor. These reactors have been reported to provide a buffering effect against shock loading (Strydom *et al.* 1997; McHugh *et al.*2006).

Córdoba *et al.* (1995) converted the flow mixing chamber of an anaerobic filter, comprising 30% of the total reactor volume, into a UASB. Formation of granules was observed after 4 weeks of operation on a dairy industry wastewater at 30 °C. Performance was compared to that of an unmodified anaerobic filter. It was found that the hybrid filter achieved 40% higher COD removal efficiencies and 65% higher volumetric gas production. OLR could also be increased more rapidly on the hybrid system. Strydom *et al.* (1995) examined the performance of a hybrid UASB reactor fed on synthetic dairy wastewater in response to stepwise changes in influent concentration (3.7–10.3 g COD per L) and in HRT (1.7–4.1 days). The optimum methane yield was achieved at 1.9-day HRT. It was concluded, however, that a two-phase system would be necessary for full-scale treatment.

Belancon *et al.* (2010) established a hybrid UASB with polyurethane support material above the gas–liquid separation zone to improve solids retention. The reactor was fed with dairy wastewater and operated at 30 °C at a HRT of 1 day. Inserting the supporting media increased the biomass amount and the reactor remained in stable condition despite the loss of 18% of biomass due to flotation of granules. Organic matter removal was 86% in the sludge bed, while biofilm bed improved COD removal up to 93% with a methane production rate of 1.8 L/L/day.

Haridas *et al.* (2005) operated a Buoyant filter Bioreactor (BFBR) for the treatment of simulated dairy wastewater. Reactor liquor content was mixed by biogas recirculation, and a scum recirculation facility was installed into the reactor to improve mixing. The BFBR was operated at a HRT range of 7.5–11.3 h and the start-up period lasted several months. In the reactor, granulation occurred by an unknown mechanism and the sludge was in irregular granule shape with a size of 2 mm. Organic matter removal gradually improved with ageing of the sludge, and COD removal efficiency was above 85% at all organic loadings. The highest COD removal was 90% at maximum OLR of 10 kg COD/m^3/day with the methane yield of 0.37 m^3/kg COD. Scum formation inside the BFBR was observed during the increment of OLR; however, the accumulated scum was degraded when wastewater feeding stopped.

Ozturk *et al.* (1993) operated a laboratory-scale hybrid UASB (HUASB) for the treatment of dairy wastewater at mesophilic conditions. The bottom 40% of the HUASB was designed as a UASB, and the upper 60% was filled with cylindrical plastic rings. The HUASB was operated at a HRT of 0.21–0.96 day, and the OLR was gradually increased from 2.54 to 17 kg COD/m^3/day. The highest COD removal was 87% at 10 kg COD/m^3/day and a HRT of 18 h. The researchers also reported that rapid OLR increment caused some problems in the gas–liquid separator. However, the stability of the reactor was not negatively affected by increasing OLR up to 17 kg COD/m^3/day with an average COD removal efficiency of 75%.

McHugh *et al.* (2006) constructed a hybrid EGSB by separating it into three chambers through installing a circular baffle with a ball to retain more biomass within the system. The researchers investigated the effect of OLR and temperature on the treatment of high-strength whey (10 g COD per L). The temperature was changed from 20 to 12 °C by decreasing 2 °C at every step. COD removal was between 90 and 95%, and the reactor gave a stable performance with low VFAs concentrations when temperature was stepped down. The highest COD removal was at 14 °C and an OLR of 13.3 kg COD/m^3/day. When the temperature was reduced to 12 °C, the reactor performance deteriorated due to VFAs accumulation and disintegration of the granular sludge. Biomass concentration in the hybrid reactor increased with the rapid development of biofilm on the matrix. The biofilm also prevented the washout of biomass from the reactor and enhanced COD removal by approximately 3%. Colins *et al.* (2013) compared the performances of a hybrid EGSB with a conventional EGSB and an anaerobic filter (AF) for the treatment of simulated whey in psychrophilic conditions. The experimental results indicated that diluted and high-strength whey could be treated with all reactors at satisfactory organic removal efficiencies. At 12 °C, the hybrid EGSB provided higher COD removal (95%) when operated at higher OLR (5–15 kg COD/m^3/day) compared to lower OLR (0.5–1.5 kg COD/m^3/day). On the other hand, the hybrid EGSB reactor had higher COD removal of 70% than the AF with 61% when treating high-strength whey (10 g COD per L) at 15 °C. The researchers indicated that the upper fixed-film section of the hybrid EGSB offered a polishing step for the degradation of acidified wastewater from the initial upflow-bed stages.

Zielinska *et al.* (2013) constructed a pilot-scale hybrid reactor having the properties of a UASB and an AF. The lower chamber of the hybrid reactor was full of suspended sludge, while the biofilm was immobilised on polyethylene particles in the upper chamber and the reactor was operated in upflow mode. The study was carried out at two temperatures of 35 and 55 °C and two OLRs of 1 and 2 kg COD/m^3/day. The researchers also compared the reactor performance with convection and microwave heating. Microwave heating stimulated the growth of highly diverse methanogenic community, while thermophilic conditions caused changes in microbial community. It was reported that the presence of methanogens within the reactor was sensitive to changes in OLR, while the highest biogas production was at lower OLRs. In mesophilic conditions, biogas production was almost sevenfold higher compared to thermophilic temperatures, while microwave heating provided higher biogas production and methane content in all conditions studied. At mesophilic temperature, increases in OLR resulted in reduction in COD removal and the highest COD removal was 76% at 2 kg COD/m^3/day at 55 °C. Banu *et al.* (2008) operated a similar hybrid reactor for the treatment of dairy wastewater using plastic cut rings for biofilm attachment media. Organic loading was gradually increased from 8 to 20 kg COD/m^3/day, and the highest reactor performance was at an OLR of 19.2 kg COD/m^3/day. When the OLR was increased to 20 kg COD/m^3/day, COD removal efficiency was less than 65% along with a decrease in methane production. The authors explained that the decrease in reactor performance at higher OLR was related to the reduction in methanogenic activity at low pH caused by VFA accumulation. They recommended installing a posttreatment step to meet discharge limits and obtained 95% COD removal and 96% BOD with a solar photocatalytic reactor.

Conventional granule reactors are also modified to overcome operational problems such as biogas liberation and scum formation. Passeggi *et al.* (2012) modified a UASB to avoid the problems caused by the FOG content of dairy wastewater. The researchers installed a scum extraction device to remove accumulated oily scum under the hood and also provided out a lamella settler for the UASB effluent, while settled solids were reintroduced periodically into the reactor. Thus, biomass washout was prevented and the biomass content in the reactor was preserved. The modified reactor was operated without pH adjustment even though the pH of raw wastewater varied between 5.0 and 11.5, which led to a reduced use of chemicals during operation. Furthermore, the HRT in the modified reactor was reduced by 22% and the required total treatment volume was reduced by 40% compared to a conventional treatment system. The modified reactor also required less investment and operational costs, but 13% less biogas was obtained due to the greater gas losses.

CONCLUSIONS AND RECOMMENDATIONS

In this review, dairy wastewater-treating anaerobic granular reactors were evaluated. Several reactor systems have been constructed for dairy wastewater treatment, and many different operational strategies have been developed to maximise methane production. Reactors were compared based on COD removal efficiencies and methane production performance. Among the individual reactor systems, the UASB reactor has been widely preferred for dairy wastewater treatment due to its simple construction, ease of operation and high performance. Research has mainly been conducted in mesophilic conditions, and few reports are available at elevated temperatures. Studies at low temperatures provide promising results for organic removal and methane production; however, more work should be carried out to increase the performance of reactors. In recent work, most studies have been performed with hybrid systems combining the properties of granular and biofilm systems. Reports reveal that optimisation of organic loading has the primary effect on successful performance of granular reactors. Finally, it can be concluded that further research should be conducted on current reactor technologies to enhance energy production and organics removal from dairy wastewater.

ACKNOWLEDGEMENTS

The authors thank the ERA-NET BBSRC (AmbiGAS, Project Number: BEN6-12-19, BB/L000024/1) for financial support. Dr. Karadag gratefully acknowledges the postdoctoral research scholarship by The Scientific and Technological Research Council of Turkey (Tubitak).

REFERENCES

Akila G and Chandra T S (2007) Performance of an UASB reactor treating synthetic wastewater at low-temperature using cold-adapted seed slurry. *Process Biochemistry* **42** 466–471.

Alves M M, Vieira J A M, Pereira R M A, Pereira M A and Mota M (2001) Effect of lipids and oleic acid on biomass development in anaerobic fixed-bed reactors. Part I: Biofilm growth and activity. *Water Research* **35** 255–263.

Arbeli Z, Brenner A and Abeliovich A (2006) Treatment of high-strength dairy wastewater in an anaerobic deep reservoir: analysis of the methanogenic fermentation pathway and the rate-limiting step. *Water Research* **40** 3653–3659.

Bandara W M K R T W, Satoh H, Sasakawa M, Nakahara Y, Takahashi M and Okabe S (2011) Removal of residual dissolved methane gas in an upflow anaerobic sludge blanket reactor treating low-strength wastewater at low temperature with degassing membrane. *Water Research* **45** 3533–3540.

Banik G C, Ellis T G and Dague R R (1997) Structure and methanogenic activity of granules from an ASBR treating dilute wastewater at low temperatures. *Water Science Technology* **36** 149–156.

Banik S, Bandyopadhyay S and Ganguly S (2003) Bioeffects of microwave—a brief review. *Bioresource Technology* **87** 155–159.

Banu J R, Anandan S, Kaliappan S and Yeom I T (2008) Treatment of dairy wastewater using anaerobic and solar photocatalytic methods. *Solar Energy* **82** 812–819.

Belancon D, Fuzzato M, Gomes D R S, Cichello G C V, Pinho S, Ribeiro R and Tommaso G (2010) A comparison of two bench-scale anaerobic systems used for the treatment of dairy effluents. *International Journal of Dairy Techology* **63** 290–296.

Bialek K, Cysneiros D and O'Flaherty V (2013) Low-Temperature (10 C) anaerobic digestion of dilute dairy wastewater in an EGSB bioreactor: microbial community structure, population dynamics, and kinetics of methanogenic populations. *Archaea* **346171** 1–10.

Blonskaja V and Vaalu T (2006) Investigation of different schemes for anaerobic treatment of food industry wastes in Estonia. *Proceedings of the Estonian Academy of Sciences* **55** 14–28.

Bodik I, Herdova B and Drtil M (2002) The use of upflow anaerobic filter and AnSBR for wastewater treatment at ambient temperature. *Water Research* **36** 1084–1088.