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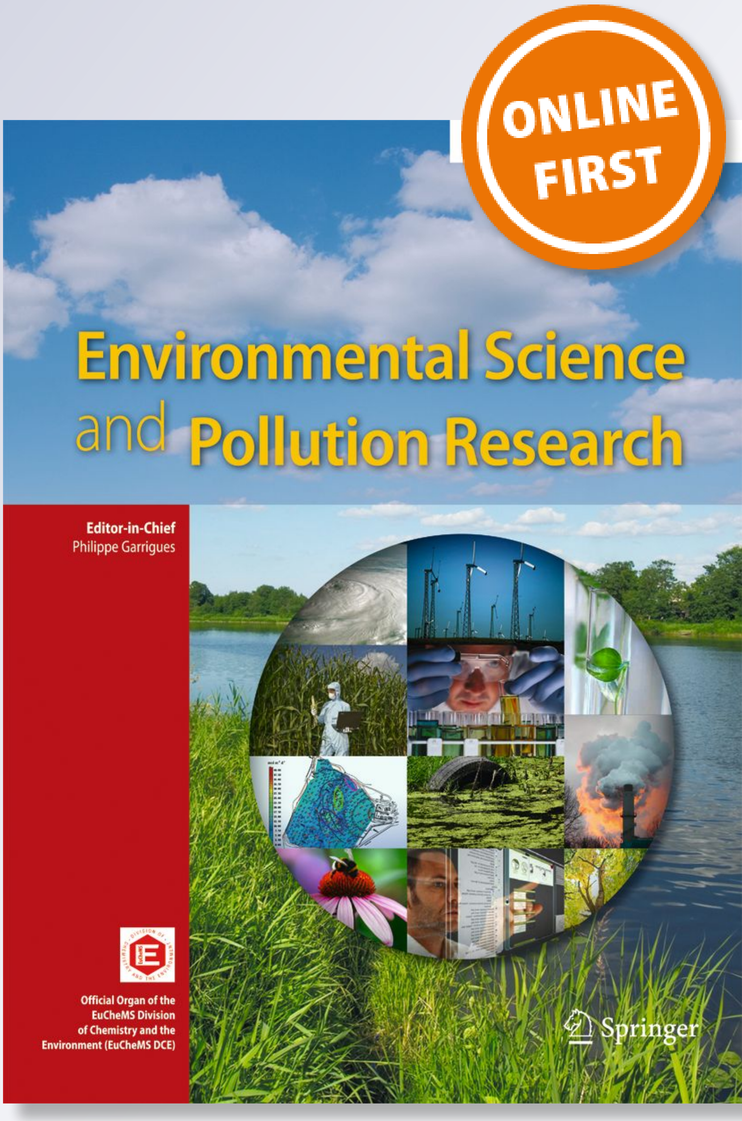
Environmental Science and Pollution Research

ISSN 0944-1344

Environ Sci Pollut Res
DOI 10.1007/s11356-018-4018-3



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https://doi.org/10.1007/s11356-018-4018-3

APPROPRIATE TECHNOLOGIES TO COMBAT WATER POLLUTION

A full-scale study of external circulation sludge bed (ECSB) system for anaerobic wastewater treatment in a whiskey distillery

Yu-Chung Lin [1] • Chen-Hua Ni [2] • Chin-Yi Wu [1,3] • Justin Chun-Te Lin [3]

Received: 4 June 2018 / Accepted: 17 December 2018

Abstract

Waste liquid streams from distillery were a hurdle in conventional wastewater treatment due to extreme high chemical oxygen demand (COD) and fluctuating feed conditions. A recently commissioned full-scale external circulation sludge bed (ECSB) was applied at a malt whiskey distillery in northeast Taiwan. Start-up of the new ECSB system, which has a total volume of 490 m^3 with diameter of 6.55 m (*ø*) and height of 15.9 m (*H*), was performed by gradual increasing influent flow rates from zero to the design value of 300 $m^3\ day^{-1}$ in the first 90 days. In the subsequent 204 days, both influent flow rates (0–389 $m^3\ day^{-1}$) and COD concentrations (2.8–18.1 kg L^{-1}) were highly fluctuated due to diverse batches from the distillery. However, effective bioremediation (COD removal 95.1 ± 2.4%) and biogas production (1195 ± 724 L day^{-1}) were achieved in this system. Intensively, the Imhoff tests were carried out and shown the settled solids concentration by 0.5 ± 0.4 mL L^{-1}, while size distributions of granular sludge were analyzed and observed by SEM-EDS. In addition, developments of the anaerobic systems (including lab, pilot, and full scale from the simplest reactor to the latest ECSB) applied in whiskey wastewater treatment were reviewed with their operational parameters for comparing performances of various anaerobic systems. In general, real-time monitoring and feasible operation strategies were critical to successfully run the system by producing clean energy simultaneously. It provides more economically attractive and sustainable-to-adopt ECSB not only an end-of-pipe process but also a bioresource technology.

Keywords Whiskey distillery wastewater · Anaerobic treatment · External circulation sludge bed · Biogas production · Operation strategy · Process monitoring

Introduction

Traditionally, malt whiskey was produced by only three raw materials, i.e., cereals (malted barley and wheat), water, and yeasts (Bathgate 2016), through a series of fermentation process, including malting, milling, mashing, fermentation, distillation, maturating, blending, and bottling (Russell and Stewart 2014). However, wastewater of the whiskey-making process was very complex and generally regarded as a treatment hurdle, not only in its high strength but also in significant volume. Most of the liquid residues in the wash still left after the first distillation (a.k.s. pot ale) and waste streams generated in the spirit still after the distillation (a.k.s. spent lee) with 6% and < 4% dry matter (Graham et al. 2012) and resulted in high suspended solids (SS) in the wastewater. Typically, chemical oxygen demand (COD) concentrations in the pot ale, which contains yeast, inorganic salts, and organic compounds (e.g., a wide variety of unfermented sugars), were ranging 60–70 g L^{-1}, while biochemical oxygen demand (BOD) values were in 15–45 g L^{-1}. Less COD (3–5 g L^{-1}) and BOD (1.5–2.0 g L^{-1}) concentrations were in the spent lees, but also contain a number of volatile organics (Goodwin and Stuart 1994; Goodwin et al. 2001). The total COD values in the spent wash from a malt whiskey distillery and in pot ale from a grain distillery in Scotland, UK, were characterized as 46.3 and 61.5 g·L^{-1}, respectively; the total solids, respectively, were 23 and 17 g L^{-1} (Mallick et al. 2010). Even the whiskey raw material ingredients and production processes remained constant, and measured COD and BOD in pot ale were varied in the ranges of 38.5–62.9 and 13.0–35.3 g·L^{-1}, respectively

Responsible editor: Philippe Garrigues

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Published online: 11 January 2019

(Graham et al. 2012). In addition, high sulfate, phosphate, and nitrogen content as well as colorants (e.g., polyphenols, caramels, melanoidins) in the distillery wastewater (DWW) make it difficult to treat. The melanoidins in DWW (Agarwal et al. 2010) possessed antimicrobial activities (Arimi et al. 2014), which are recalcitrant coloring pollutants that cause serious environmental problems and health threats in human and animals as well as decreasing biodegradation in the treatment. Moreover, the high volume of liquid wastes generated from whiskey distillation (Zero Waste Scotland 2015). It is estimated that 10 L of pot ale is discharged per 1 L of pure alcohol (LPA) in the case of malt whiskey and 18 L of spent wash per LPA of grain whiskey (Tokuda et al. 1999). Due to the high concentration of organic compositions as well as significant amounts, it is normally difficult to degrade by the conventional aerobic biological treatment process.

Comparing with century distillers in Europe, America, and Asia (Russell and Stewart 2014), the whiskey industry in Taiwan is just in the infant stage. Although the first bottler released about 10 years ago, there are already a number of whiskey bottles awarded (Stout 2015). The liquid residues from pot ale and spent lee after whiskey making were mixed as distillate wastewater in our study with the mixed COD concentration was as high as 20–35 g L^{-1}. Inadequate treatment of DWW resulted in eutrophication and pollutions in the received water bodies, while inappropriate land discharge/disposal of stillage induced soil pollution, soil pore clogging, salinization, acidification, inhibition of microbial activity, and fertigation problems (Fuess and Garcia 2014, 2015). Ecotoxicological and health hazards of distillery wastewater were recently reviewed by Chowdhary et al. (2018), and environmental hazards of distillery spent wash were highlighted by Mohana et al. (2009). A total of 43 organic pollutants were identified in DWW and some were categorized as endocrine disrupting chemicals. Besides, new discharge regulation on fermentation industries (including alcohol distilleries) was limited the COD, BOD, and SS in the final effluents as low as 150, 50, and 50 mg L^{-1}, respectively. Therefore, there is an emergent need to treat such high-strength DWW more efficiently and sustainably. A full-scale anaerobic bioreactor using the latest external circulation sludge bed (ECSB) reactor to treat such high-strength DWW was present as this is the first whiskey distillery in Taiwan. In addition, practical operational problems, such as system start-up and suffering extremely high fluctuations in the feed, were demonstrated the robust of the ECSB system. To maintain stability in the anaerobic bioreactor and to provide vulnerable microorganism, a sustainable environment to grow is critical in the operation of a full-scale plant. Therefore, the monitoring process parameters and timely response were valuable. Lastly, simultaneously biogas production along with DWW treatment in facilitating renewable energy (methane) utilized in the distillery was demonstrated.

Overall process flow diagram and system design of ECSB

External circulation sludge bed (ECSB) system, which is the third-generation high-rate anaerobic bioreactors, was applied to treat distillery wastewater from an award-winning whiskey distillery in Ilan County, Taiwan. The high-strength wastewater (COD of 30,000 mg L^{-1}) firstly flows into an equalization tank, as EQ tank shown in Fig. 1a, for buffering fluctuations of flow rates and concentrations from various distillery waste liquids and other streams of wastewaters. The feed flow rate of the ECSB system was designed as 300 m^3 day^{-1} and pH in the distillery WW was generally less than 6.5 and the temperature was varied between 30 and 40 °C. In order to meet strict local discharge surcharges, fulfill increasing capacity of the distillery, and minimize the operation costs, the engineering project was carried out in 2016–2017 for a more efficient treating of the high organic wastewaters and recovering biogas generated from the anaerobic bioreactor as a green energy source. Therefore, the modified external circulation sludge bed system was build up to take the mission. One of innovation part is the ECSB system using an additional tank (neutralization (NT) tank) as a gate of input (WW influent) and output (WW effluent) of the anaerobic digester. It not only buffer nutrient fluctuations in the influents, but also mixing chemicals efficiently prior to entering the main anaerobic bioreactor and served as external circulation. Thus, a highly loaded section in the lower ECSB tank and a polishing section in the upper ECSB tank (*D* 6.55 m, *H* 15.9 m, *V* 490 m^3) can be functionalized as designed. The calculated height-to-diameter ratio (*H*/*D*) was 2.43. A three-dimension diagram of the NT tank, ECSB reactor, H_2S scrubber, waste sludge pump, and feed/recirculation pumps area were shown in Fig. 1b as well as the picture. The NT tank provided efficient external circulation to facilitate mixing of wastewater and granular sludge, as well as survived as a buffer to eliminate shock loading and to recycling alkalinity. The closing venting not only avoided odor emissions but also prevented the corrosion of H_2S gas which resulted from biochemical reactions. The feed flow rate from the NT tank to the ECSB was designed as 170 m^3 h^{-1} and the maximum COD organic loading was designed as 9000 kg COD day^{-1}; thus, the calculated VLR of the ECSB tank was 18.4 kg COD·m^{-3} day^{-1}. With the integration of two three-phase (gas-liquid-solid, GLS) separators inside the ECSB reactor, upflow velocity can increase up to 5.05 m^3 h^{-1}. Therefore, anaerobic granular sludge can be efficiently maintained in the reactor without loss of biomass and effluents from the system contained much lower organics, which were acceptable for subsequent aerobic treatment. Solid outputs from the ECSB tank flow into the waste sludge storage tank, while liquid

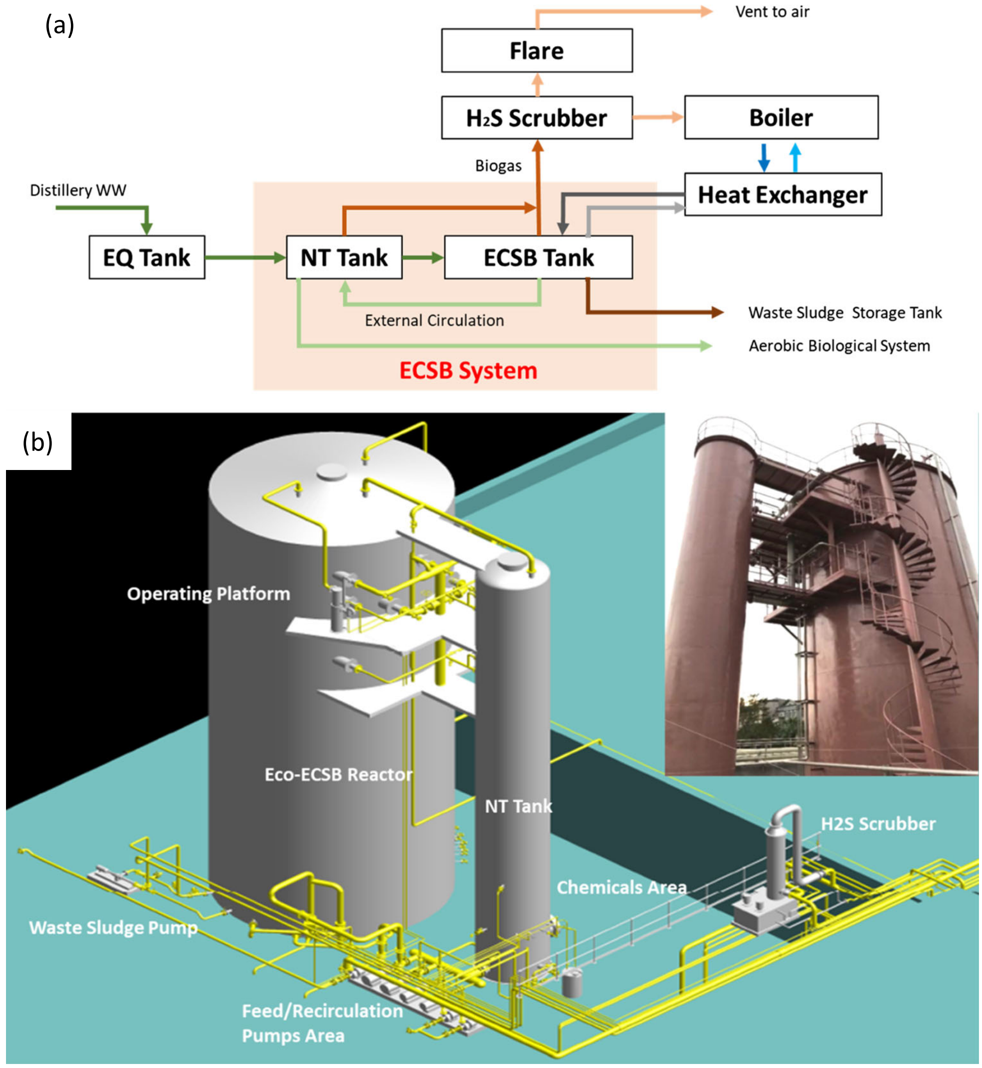

Fig. 1 **a** Process flow diagram of ECSB system and biogas recovery system; **b** 3D diagram of the high-rate anaerobic ECSB system and a picture of the full-scale reactor

supernatant from the anaerobic system flowed into the aerobic biological system. Patent design of gas collection piping system in the ECO-ECSB™ can check fouling under the G-L-S separator during operation. In case fouling occurred, it is also capable to clean the separator directly either by the biogas or from the external water pipes. Therefore, the system can continuously run without intensively scheduled checks or maintenance.

In order to keep the stability of anaerobic treatment system, it is critical to maintain system operational temperatures higher than 25 °C, especially winters of the distillery in northeast Taiwan were generally cold. Therefore, to utilize renewable energy from biogas was attractive in this case. The biogas produced by anaerobically biochemical reactions, either from a previous anaerobic system or the updated ECSB system, was collected to a scrubber for removal H_2S firstly. Then, the biogas was then burn in flare in a heat water boiler, wherein heats were recycled via two heat exchangers and provided the two anaerobic systems under the required temperatures.

Results and discussions

Progress of DWW treatment technologies and developments of anaerobic reactors

Various treatment processes to treat distillery wastewater (DWW) have been reviewed, either biological approaches (Hutnan et al. 2003; Pant and Adholeya 2007; Kharayat 2012) or physicochemical treatment (Mohana et al. 2009; Prajapati and Chaudhari 2015), while some emphasized on specific reactor (Melamane et al. 2007) or pollutants (Arimi et al. 2014). Krishnamoorthy et al. (2017) recently characterized distillery wastewater and suggested a green phycoremediation by using microalgae. Pant and Adholeya (2007) compared different reaction types, organic loading rates (OLRs), retention time, and COD and BOD removal for DWW treatment; state-of-the-art developments on the applying various anaerobic biological systems on this field were updated in Table 1 and comprehensively reviewed below.

Table 1 Chemical reactions in the four stage of anaerobic digestion

Hydrolysis	$C_6H_{10}O_4 + 2H_2O \rightarrow C_6H_{12}O_6 + H_2$	Eq. (1)
Acidogenesis	$C_6H_{12}O_6 \longleftrightarrow 2CH_3CH_2OH + 2CO_2$	Eq. (2)
	$C_6H_{12}O_6 + 2H_2 \longleftrightarrow 2\ CH_3CH_2COOH + 2H_2O$	Eq. (3)
	$C_6H_{12}O_6 \rightarrow 3\ CH_3COOH$	Eq. (4)
Acetogenesis	$CH_3CH_2COO^- + 3H_2O \longleftrightarrow CH_3COO^- + H^+ + HCO_3^- + 3H_2$	Eq. (5)
	$C_6H_{12}O_6 + 2H_2O \longleftrightarrow 2CH_3COOH + 2CO_2 + 4H_2$	Eq. (6)
	$CH_3CH_2OH + 2H_2O \rightarrow CH_3COO^- + 3H_2 + H^+$	Eq. (7)
Methanogenesis	$CH_3COOH \rightarrow CH_4 + CO_2$	Eq. (8)
	$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$	Eq. (9)
	$2CH_3CH_2OH + CO_2 \rightarrow CH_4 + 2CH_3COOH$	Eq. (10)

pKa values of individual VFA compounds (e.g., acetic, propionic, butyric, citric acid) in anaerobic digestion can be found in Sun et al. 2016)

The Gibbs free energy of biochemical reactions during acetogenesis and methanogenesis can be found in Fatih Demirbas and Balat (2009)

Bacterial involved in acidogenesis (stage II), acetogenesis (stage III), and methanogenesis (stage IV) can be found in Abbasi et al. (2012)

In the early work of Goodwin and Stuart (1994), two lab-scale upflow anaerobic sludge blanket (UASB) reactors were used to anaerobic treat pot ale from a commercial malt whiskey distillery. Effects of dilution ratios of pot ale and trace elements supplementation on the OLR, hydraulic retention time (HRT), biogas production, and volatile fatty acid (VFA) degradation were investigated. The UASB reactor operated at 35 °C was further tested to treat pot ale from an experimental malt whiskey distillery (Goodwin et al. 2001), and the team found the performance was limited by conversion VFA to methane and granulation. In contrast to the previous operational temperatures within mesophilic range (25–45 °C), a thermophilic (45–65 °C) pilot-scale UASB reactor, which was operated at 55 °C to facilitate methanogenic activity, was used to treat cane-molasses vinasse from an alcohol distillery wastewater in Thailand for a period of 430 days by Harada et al. (1996). Although COD removals were only 39–67%, nearly 83% COD conversion to methane was yielded. Furthermore, COD concentrations were distinguished into four fractions, i.e., soluble (Sol-COD), suspended solids (SS-COD), which were recovered as methane gas (CH_4-COD) and used in sulfate reduction (SO_4-COD) and identified different trends with adjusting OLRs. Biodegradability of the cane- and malt-vinasse (another distillation plant in Japan) was also compared. Another pilot-scale anaerobic treatment for whiskey pot ale (Tokuda et al. 1999) using an upflow anaerobic filter (UAF) removed 76% COD and yield biogas at a rate of 0.75 m^3/kg COD_{Cr}, with methane content in 65–75% and minor H_2S in 2000 ppm. A granular bed anaerobic bed reactor (GRABBR), which used an anaerobic baffled reactor (ABR) coupled with the UASB concept, was proposed for treating whiskey distillery wastewater (Akunna and Clark 2000) and demonstrated better reduction in COD (90–96%), BOD (80–82%), and biomass retention under four investigated HRTs. The system, which was supposed to run at a near plug-flow hydraulic regime, was found in encouraged phase separation of granular methanogens and non-granular acidogens. In South Africa, wine wastewaters (grape wine as a raw material in column distillation) and unsettled and settled grain wastewaters (wort as a raw material in pot distillation) were compared by Laubscher et al. (2001), who set-up two lab-scale UASB reactors to identify operational problems occurred in a full-scale UASB system. The full-scale UASB system was also studied by Wolmarans and Villiers (2002), who focused on another practical issue, i.e., system start-up associated with various grape seasonal operations. As biological systems often suffer a hurdle in each initial stage and take about 1- to 2-month period to recovery, this study applied a new approach to shorten the start-up period to 1 week by controlling the volumetric loading rate (VLR) between 4 and 8 kg COD m^{-3} day^{-1}.

Another high-strength malt whiskey distillery wastewater (COD 30.1–50.7 g L^{-1} and BOD 15.6–22.1 g L^{-1}) in Turkey was tested by a lab-scale UASB, in which two-stage anaerobic columns were connected in series with one aerobic flask (Uzal et al. 2003). Even operated at OLRs as high as 39 kg COD m^{-3} day^{-1}, the reductions in BOD and COD of the system were as high as 99.5% and 98.2%, respectively. Despite being with or without nutrient supplemented, the net total gas production of the system was between 0.019 and 0.020 m^3 kg^{-1} COD-removed, whereas CH_4 content in the biogas was 77 ± 5%. A combination of UASB and an internal filter was built up by Kumar et al. (2007) as an anaerobic hybrid reactor to treat distiller spent wash. They examined precisions of kinetic models for prediction of bacterial growth, effluent substrate concentration, and biogas yield in the case of treating such high-strength wastewater. A lab-scale UASB operated at mesophilic (37 °C) was tested for treating distiller's acidic grains wastewater (pH 3.8) with original high COD (44.6 g L^{-1}) and obtained reductions of 80–97.3% (Gao et al. 2007). Similar to the aforementioned two-stage UASB system, an anaerobic biphasic fixed film bioreactor (AB-FFB) comprised by an acidogenic reactor (AR) and a methanogenic reactor (MR) was developed to treat a very acidic (pH 3.0–4.5) distillery spent wash (Acharya et al. 2011). The AB-FFB was efficient to remove 50–80% COD at different OLRs and a kinetic model can be used to interpret. Bacterial community structures of the AR and MR were further identified. Anaerobic digestion of grain stillage has been compared within three reactors (Schmidt et al. 2013), including continuous stirred tank reactor (CSTR), fixed bed reactor (FBR), and anaerobic sequencing batch reactor (ASBR). Although no significant differences have been observed within the three reactors, the CSTR system without biomass immobilization can achieve a stable process at HRTs below 10–14 days. An anaerobic upflow staged sludge bed (USSB) with multiple internal solid-gas separators was developed to treat molasses-based wastewater by Onodera et al. (2013). The high-concentration (43–120 g COD L^{-1}) synthetic wastewater was treated by three anaerobic units controlled at 35 °C, (including an acidification reactor, an USSB, and an UASB) and then by an aerobic unit, i.e., a two-stage trickling filter (TF) reactor. The USSB showed good efficiency of both COD reduction percentage (80–87%) and methane recovery (70–80%) at various OLRs (11–43 kg COD m^{-3} day^{-1}). continuously up to 749 days. A high-rate hybrid UASB reactor for treating a combination of domestic waste activated sludge and distillery's spent wash was reported (Prajapati and Chaudhari 2015) that COD, BOD, and SS reduction percentages achieved 65%, 40.4%, and 87%, respectively, at HRT of 5 days. Saner et al. (2016) also investigated effects of OLRs on reduction percentages of COD, BOD, and biogas production in a bench-scale UASB, which controlled at 37 °C, under a constant HRT of 47.1 h and fed by hot spent wash from a distillery industry for up to 2 years. A multi-stage pilot-scale system, including an UASB, anoxic-aerobic membrane bioreactor (An/Ae-MBR), and a post-treatment via chemical precipitation or adsorption, was used to treat high-strength winery wastewater (Petta et al. 2017) with influent COD of 44.6 g L^{-1} and pH at 3.8.

Various types of anaerobic reactors (Wilkie et al. 2000; Metcalf and Eddy Inc. et al. 2014) have been developed to treat high-strength wastewater (Fang 2010; Khanal et al. 2016) and simultaneously for biogas processing (Fatih Demirbas and Balat 2009). External circulation sludge bed (ECSB) is regarded as the third generation of anaerobic wastewater treatment process (Tauseef et al. 2013; Guo et al. 2018), while expanded granule sludge bed (EGSB) was the second generation with the modification of the first-generation UASB. A review has been addressed for the development of UASB, EGSB, and another type of anaerobic granular treatment processes (Lim and Kim 2014). Main features of the three processes were introduced, and advantages, limitations, and applications were all listed. Commercially available advanced EGSB-type reactors, including IC and ECSB, in the market, have been shortly introduced (Meyer and Edwards 2014). We illustrated the development of AD reactors and updated ECSB in Fig. 2. Various integrated biorefinery wastewater designs, which applied AD and membrane processes, were proposed to achieve circular economic effluent treatment (Bilad et al. 2011). A two-stage EGSB, which consists of a pre-acidification (PA) tank, was designed as the first stage for fermentation and acidification. Then, an EGSB digester as the second stage for acetogenesis and methanogenesis was designed to treat distillery wastewater and to recover biogas simultaneously (Ghorbanian et al. 2014a, b). Impact of supplemental hydrogen on biogas quality enhancement and substrate reduction percentage efficiency were investigated (Ghorbanian et al. 2014a). They also used four feed COD concentrations of 30, 20, 10, and 5 g $L^{-1}$to present high, medium-high, medium, and low strength, respectively, and investigated the impact of HRT at constant OLRs (3, 5, 7, and 9 g COD·L·day^{-1}) on mesophilic digestion of 35 °C (Ghorbanian et al. 2014b) in the two-stage lab-scale reactors. Two EGSB reactors connecting anaerobic filter (AF) have compared treatment performances of brewery effluent under psychrophilic (15 °C) and mesophilic (37 °C) anaerobic digestions for 194 days (Connaughton et al. 2006). The maximum obtained OLR and HLR were 4.47 kg COD m^{-3} day^{-1} and 1.33 m^3 m^{-3} day^{-1}, respectively. Two lab-scale EGSB studies were conducted recently to evaluate a full-scale high-rate anaerobic digester bioreactor (ECSB) at a Scottish whiskey distillery from the aspects of treatment performances and microbial ecology (Connelly et al. 2017). Superficial velocity (V_{up}) of EGSB was usually significant higher (6–15 m h^{-1}) than UASB (V_{up} 0.5–2 m h^{-1}) and taller. Intensive contact between incoming organic matter and sludge of EGSB was provided by the higher kinetic energy of influent.

Nowadays, high-rate anaerobic bioreactors were not only as efficient end-of-pipe wastewater treatment approaches but also can be regarded as resource conversion technologies (van Lier 2008). Regenerated resources, including nutrients, water, and energy (Tauseef et al. 2013), can be obtained from various anaerobic processes for energy conversion, e.g., combined heat and energy (Hosseini and Wahid 2014). Recovered biogas from anaerobic bioreactors can be either directly used as clean

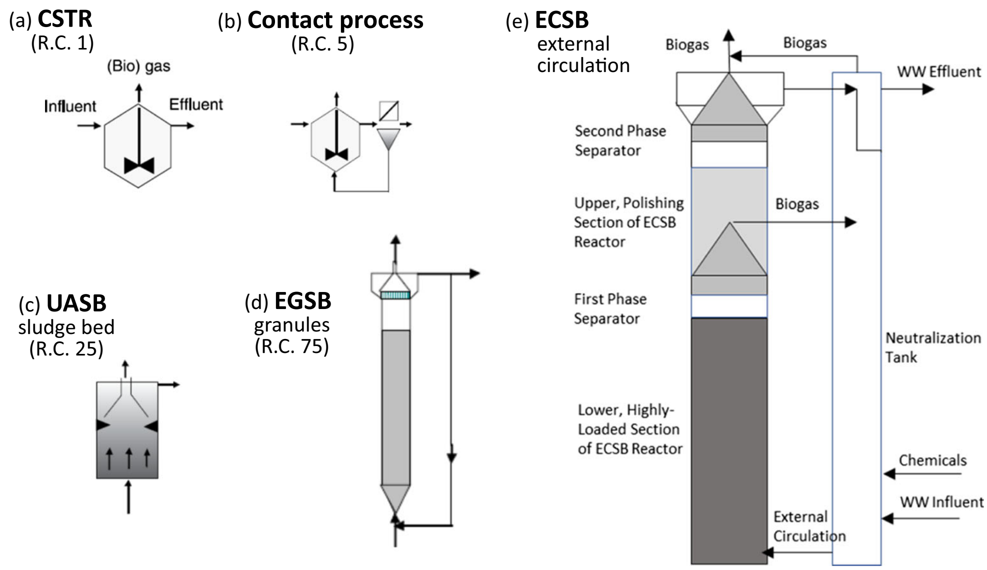

Fig. 2 Schematics of anaerobic digester evolutions from **a** CSTR, **b** contact process, **c** UASB, **d** EGSB, modified from (van Lier 2008) (R.C relative capacity) to the latest **e** ECSB

and renewable energy for cooking, lightening via burning or indirectly used for producing electricity. Benefits of utilizing the biogas produced can reduce costs of treating organic wastewater, as well as reducing GHG emission. An overview of reducing global warming and generating energy by biogas was provided by (Abbasi et al. 2012). Factors affecting biogas production efficiency from anaerobic biochemical process, including loading rate, pH, temperature, retention time (HRT and SRT), specific (contact) surface of the substrate (wastewater), carbon to nitrogen (C/N) ratio, toxicity, dilution, mixing/agitation, light, solid residue/slurry, and pathogens, were extensively reviewed (Abbasi et al. 2012; Mao et al. 2015). Successful circular economic business models by using malt distillery by-products, such as pot ale, spent wash, and whiskey draft (contains 23% dry matter), were reported for extraction of biofuel, polyphenols, algae, and proteins production (Zero Waste Scotland 2015). A pot ale deproteination process (Barrena et al. 2018) was not only capable to recovery protein as valuable materials for fish feeding, but also beneficial to wastewater treatment process (reducing total COD up to 35% and soluble copper of 30–35%). A three-stage pot ale wastewater treatment process proposed by Uzukwu et al. (2017), which can be used to recycle solid contents as animal feed in the first-stage of solid-liquid separation and precipitated sludge as fertilizer in the second-stage of alkaline precipitation (pH 8–11). They estimated volume of annular wastewater which can be reduced one fifth by the third-stage evaporation in a medium-size whiskey distillery; meanwhile, COD for feeding subsequent anaerobic process can be concentrated (from 46 to 224 g L^{-1}) as organic-rich influent with adjustable nutrient and mineral contents (i.e., ammonia, phosphorus, calcium, magnesium, copper) by preliminary pH balancing. Sustainable paths for managing solid and liquid waste from distilleries were performed by (Weber and Stadlbauer 2017).

COD reduction percentage and biogas production

The third-generation UASB reactor, i.e., external circulation sludge bed (ECSB), was startup to treat whiskey distillery wastewater from December 22, 2016, and continuously monitored nearly 300 days. As shown in Fig. 3a, feed flow rates were highly fluctuations because distillation batches were varied from time to time. Until Oct. 11, 2017, the whole ECSB operation duration was distinguished as three stages: (1) system start-up and sludge conditioning phase (initial 90 days), (2) system stabilization phase (from the 91th to 109th day, subsequent 20 days), (3) the third phase (from the 110th to 294th day, a total of 183 days), as shown in Fig 3a. In the first phase, daily influent flow rates of the ECSB system were gradually increased to the design value (i.e., 300 m^3 day^{-1}) by carefully adjusted flow rates from an anaerobic buffer tank in the distillery wastewater treatment plant. The corresponding OLR at the end of phase 1 (the 90th day) was eventually reached to 4000 kg COD·day^{-1}. As shown in Fig 3b, even though influent COD concentrations were varied between 2825 and 18,140 mg L^{-1} in phase 1, COD concentrations in the ECSB effluent were constantly maintained (average of 590 mg L^{-1} from the 12th to 90th day) and COD reduction percentage was good (average of 93.8% in the same duration). In the second phase, influent feed flow rates were kept to 300 m^3 day^{-1} except for the few days fall to 168 m^3 day^{-1}. Biogas production in phase 2 was significantly decreased from 1720 to 639 m^3 day^{-1}, in

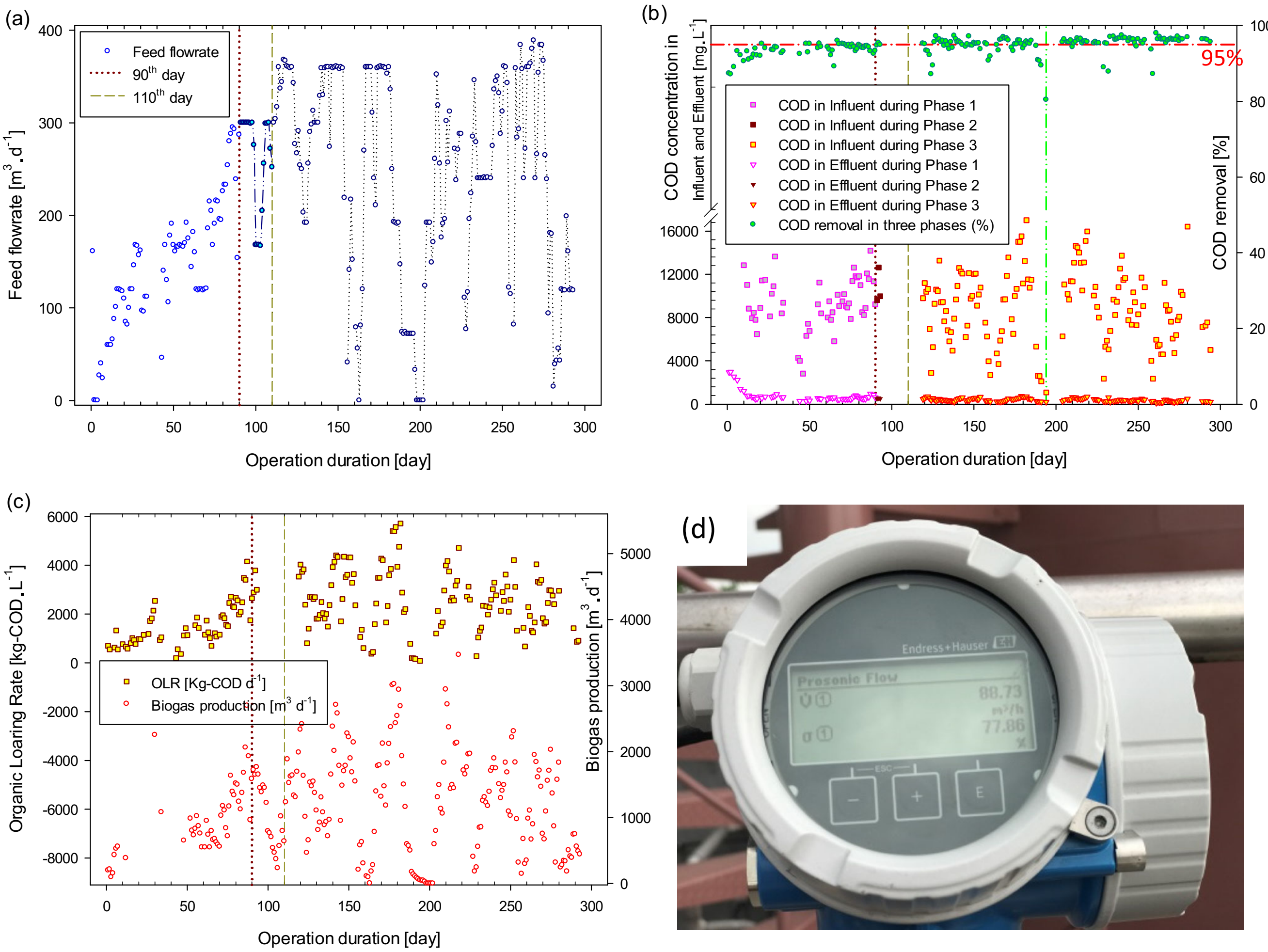

Fig. 3 **a** Feed flow rates in three phases (separated by dotted and dashed lines on 90th and 110th day, respectively); **b** COD in the influent, effluent, and removal; **c** OLRs and biogas production rates; **d** biogas monitoring transmitter

contrast to the increasing trends which appeared in phase 1 (up to 2694 m^3 day^{-1} in the 87th day), as shown in Fig 3c.

In phase 3, influent flow rates were highly fluctuated (up to 389 m^3 day^{-1} at the 269th day, and down to zero at the 269th and 198th–202nd day) and quite often over the system designed treated value (300 m^3 day^{-1}), as shown in Fig 3a. Even though influent flow rates and COD concentrations varied dramatically (the lowest of 1090 at the 194th day and the highest of 16,417 mg L^{-1} at the 280th day) in phase 3, the average effluent COD concentration was kept in an average of 401.8 ± 124.6 mg L^{-1} (from the 110th to 294th day) and average COD reduction percentage of 95.1 ± 2.4% was achieved. This is one of the main challenges in treating alcohol distillery wastewater as waste liquid flow rates from each batch of pot ale and spent wash which were varied sharply. However, a constant feed flow rate and COD concentrations were essential to a stable anaerobic biological system. Taking an example at the 194th day, influent COD concentration was only 1090 mg L^{-1} and the effluent COD was 213.8 mg L^{-1}, resulting to the lowest COD removal of 80.4% in the whole three phases. As shown in Table 1, our results on COD reduction percentage (~95%) were better than reported COD reduction percentage of 70.7 ± 11.7% in another full-scale ECSB for treating whiskey wastewater (Connelly et al. 2017). Therefore, the robustness of the ECSB system was demonstrated that it is capable to suffer the extreme fluctuation ranges in feed and provide excellent performance in treating such high-strength wastewater. Specific gas production has been identified close to OLR in the case of treatment distillery spent wash via an anaerobic process (Kumar et al. 2007), where the biogas yield was predicted by a mathematical model. Although it is difficult to investigate kinetic modeled in a full-scale AD system, the lab-scale study can provide as a reference in practical operation.

Typically, biogas is comprised 55–70 vol.% methane, 30–45 vol.% CO_2, < 2 vol.% N_2, and around 500 ppm

H_2S (Fatih Demirbas and Balat 2009). Anaerobic digestion (AD) process occurs in the following four steps: (1) hydrolysis, (2) acidogenesis, (3) acetogenesis, and (4) methanogenesis. Table 1 listed chemical reactions in the four stages of AD, which can be seen that the methane was generated in the last phase (methanogenesis). Methane was produced by methanogenic organisms via Eqs. (8) and (9) which can be seen elsewhere (Metcalf and Eddy Inc. et al. 2014). The CO_2 was generated from Eqs. (2), (6), and (8), while hydrogen was generated from Eqs. (1) and (5). The calculated theoretical gas production used the stoichiometric CH_4 production (1 g COD is equivalent to 395 mL of methane) (Uzal et al. 2003). Schmidt et al. (2013) compared three lab-scale anaerobic digestion processes, i.e., CSTR, FBR, and ASBR, from the production of dried distiller grains with soluble (DDGS). Although no significant differences of methane production (57.3–62.3%) within the three reactors, H_2 concentrations varied significantly between the reactors. A modified methane generation model (MMGM) has been developed to predict the biogas production of a full-scale UASB reactor for brewery wastewater treatment (Enitan et al. 2015). Similar benefits by using full-scale anaerobic biological treatment process to treat plot ale and wash waters on cost reduction in fossil fuel demand as well as saving carbon footprints were also reported (Anonymous 2016). A distillery in Scotland successfully turns 1000 m^3 of malt whiskey distillery co-products per day into 16 MW h of renewable heat during full operational for 1 year.

Monitoring of VFAs, pH, temperature, and alkalinity recovery

Volatile fatty acids (VFAs) and alkalinity were two key process parameters in performance monitoring of anaerobic digestion (Sun et al. 2016). VFAs represent short-chain fatty acids consisting of six or fewer carbon atoms (C_2−C_6 including iso-forms of C_4–C_6) that can be distilled at atmospheric pressure. In anaerobic digestion, VFAs were converted from hydrolyzed simple organics (e.g., monosaccharides, amino acids, and long-chain fatty acids (LCFA)) after acidogenesis. Since VFA is regarded as a critical intermediate factor and most methane is produced via metabolic routes involving them, it is important to monitor its variations in the feed as well as in the reactor. Goodwin et al. (2001) also indicated that UASB performance was limited by conversion VFA to methane and granulation. Contents of VFAs in the influent and effluent of the ECSB system were monitored throughout the whole operational period, as shown in Fig. 4a. Average VFAs in the influent and effluent were 43.0 ± 18.3 and 1.5 ± 1.7% meq L^{-1}, respectively. Although fluctuations of VFAs in the influent were quite big (a max. of 150.1 meq L^{-1} in the initial (the 6th day) and a min. of 6.0 meq L^{-1} in the end (the 194th day)), the average removal of VFAs (97.4 ± 1.7%) in the whole operation duration appeared excellent.

The pH values in the influent and effluent as well as in the ECSB system were also monitored with respect to time, as shown in Fig. 4b. As there is a buffer bank in front of the ECSB system, the pH values were generally ranged in neutral (pH 5.5 and 7.5). However, average in the influent was more acidic (pH 6.2 ± 0.4) than that in the ECSB tank (pH 7.1 ± 0.2) and the effluent (pH 7.7 ± 0.6).

Alkalinity levels in the influent and effluent of the ECSB system were depicted in the bottom of Fig. 4c. The increment between the influent and effluent was plotted in the top of the same figure. The alkalinity in the influent was regarded as low (average of 6.0 ± 4.7 meq L^{-1}) while alkalinity in the effluent was much higher (average of 52.8 ± 13.3 meq L^{-1}). The raising alkalinity in the ECSB effluent was resulting from dissolved carbon dioxide, which generated from anaerobic microorganisms in the reactor during methanogenesis. In addition, alkalinity increment with respect to time showed a slowly decreasing trend (slope of − 0.0473) which resulted from a relative significant decreasing trend of the alkalinity in the effluent (slope of − 0.0576) to the end of our monitoring date. Addition of alkalinity generally needs at least a concentration of 2–3 kg L^{-1} as $CaCO_3$ (40–60 meq L^{-1}) to maintain pH with the characteristic high concentration of $CO_{2(g)}$ and was one of the main expenditures in anaerobic wastewater treatment (Metcalf and Eddy Inc. et al. 2014). With an external circulation in this system, alkalinity in the effluent can be recycled with the blending of the influent. This can reduce required chemicals (liquid caustic soda) for pH adjustment and subsequent operational costs (Yamada et al. 2013), as well as can contribute to reducing related carbon footprints, as the very close relationship between VFA and various forms of alkalinity has been highlighted (Kumar et al. 2007). The monitoring of both can be a helpful index on modification of operational strategies.

As summarized in Table 2, anaerobic systems (most of them are UASB reactors or modified system) for distillery wastewater treatment generally applied mesophilic operation (generally in 25–45 °C, more specifically either in 35 or 37 °C), while few operated thermophilically (45–65 °C, or defined even in a smaller range, 50–57 °C, by Metcalf and Eddy Inc. et al. 2014). For example, Harada et al. (1996) operated a pilot-scale UASB reactor at 55 °C to facilitate methanogenic activity and Yamada et al. (2013) operated a multi-staged UASB at 55 °C as well. In our ECSB system, the highest and lowest temperatures in the record were 40.8 and 26.5 °C, which were in the range of our setting in the mesophilic condition (30–40 °C). The

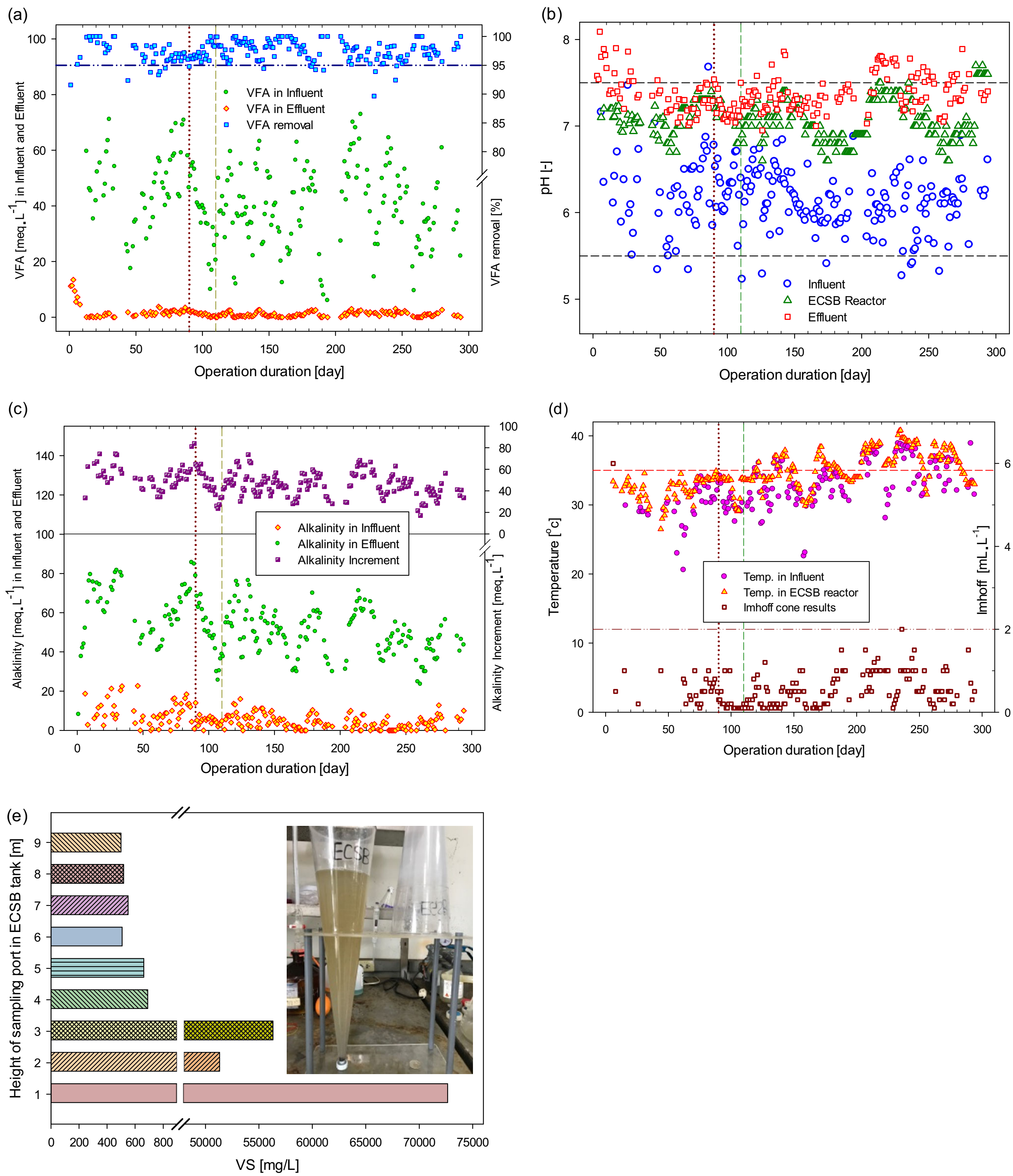

Fig. 4 **a** VFA in the influent, effluent, and removal; **b** pH in the influent, ECSB tank, and effluent; **c** alkalinity in the influent, effluent, and increment; **d** temperatures in the influent and ECSB tank; **e** VS of sludge from nine sampling ports and the Imhoff cone test apparatus

lowest influent temperature (20.6 °C) occurred during a cold current in Ilan County between February and April 2017 (corresponding to the 40th–100th day). Sudden temperature drops can affect the integrity of granular sludge in UASB, EGSB, and other reactors. The methanogenic activity of anaerobic microorganisms would slow

Table 2 Anaerobic systems for treatment of malt whiskey distillery or other high-strength wastewater and biogas production

System name	Feed type	Scale and capacity	Operation time, HRT, and temp.	Influent water quality	OLR (kg COD m^{-3} day^{-1})	Biogas rate (BPR)	Effluent water quality	Ref.
UASB	Pot ale from a commercial malt whiskey distillery	Two lab-scale reactors (Q = 0.5 L day^{-1}, $V_{work.}$ 1.05 L, I.D. 62 mm)	327 days, HRT: 2.1 days, 35 °C	COD: 3.5–47.9 g L^{-1}	1.7–22.8 (15 at stable operation)	BPR: 0.5–12.8 L day^{-1}	COD: ~ 90%	Goodwin and Stuart 1994
UASB	Alcohol distillery wastewater (cane-molasses vinasse) diluted with tap water	Pilot scale, $V_{tol.}$ 140 L, a column (H 4 m, I.D. 20 cm, $V_{col.}$ 126 mL) with a G-S separator ($V_{gss.}$ 14 mL) on the top	430 days, HRT: 47.1 h, 55 °C	COD: 309–3286 g L^{-1}	28	BPR: 0.29 Nm^3–CH_4/kg COD_{remov}	BOD: 80%, COD: 39–67%	Harada et al. 1996
UAF	Non-diluted whiskey pot ale supernatant	Pilot scale, volume of UAF 2 m^3, Q = 80 L day^{-1}	60 days, HRT < 2 h	COD: 63 g L^{-1} in pot ale, 48.2 g L^{-1} in the supernatant	20	BPR: 0.75 m^3/kg COD_{Cr}, CH_4: 65–75%	COD: 76%, TN: 70%	Tokuda et al. 1999
GR ABBR	Diluted whiskey distillery wastewater (DWW)	Lab-scale reactor, $V_{total.}$ 35 L, 10 equal compartments inside the reactor	9 months, four HRTs: 10, 7, 4, and 2 days, 37 °C	COD of raw: 16.6–58.0 g L^{-1} COD in influent: 9.5 g L^{-1}	0.99, 1.33, 2.37, and 4.75	10–22 L day^{-1}	BOD: 90%, COD: 80%	Akunna and Clark 2000
UASB	Pot ale from an experimental malt whiskey distillery	Lab scale (Q = 0.5 L day^{-1}, $V_{work.}$ 1.05 L, I.D. 62 mm)	279 days, HRT: 2.1 days, 35 °C	COD: 2.86–32.86 g L^{-1}	1.36–10.0 2	5.0 → 0.3 L day^{-1}	COD: 80%	Goodwin et al. 2001
UASB	Wine, (unsettled and settled) grain DWW	Two lab scale (H 160 cm, I.D. 9 cm, $V_{work.}$ 10.2 mL, Q = 4–12 L day^{-1})	414 days, 35 °C	1. 20–30 gL^{-1} 2. 25–30 g L^{-1} 3. 20–25	From 4.2 to 21.9	N.A.	COD: > 80%	Laubscher et al. 2001
BFFB (AR + MR)	Spent wash distillery	Two lab-scale reactors: 1. Acidogenic reactor (AR), H 134 cm, I.D. 9 cm, V_{work} 3 L 2. Methanogenic reactor (MR), H 152 cm, I.D. 18 cm, V_{work} 10 L), one aerobic flask (V 10 mL)	HRT of AR 6 days, HRT of MR 20 days	BOD: 50–60 g L^{-1}, COD: 110–190 g L^{-1}, pH 3.0–4.5	OLR of AR 33.3–80, OLR of MR 8–19	CH_4 of MR: 26.9–12.3 m^3 m^{-3} kg^{-1}	COD removal of AR: 20–4.1%, MR: 81.3–51%	Acharya et al. 2011
UASB	Wine distillation wastewater	Full scale, N.A	Three periods: 1998, 1999, and 2000	COD (g L^{-1}): 1. 19.6–34.9 2. 14.9–50.9 3. 13.0–35.8	2.0–18.0	N.A.	COD: 90%	Wolmarans and Villiers 2002
UASB (An/Ae SBR)	malt whiskey distillery wastewater	Lab scale: two anaerobic UASB columns connected in series (H 50 cm, I.D. 2.5 cm, V 113 mL), one aerobic flask (V 500 mL)	83 days (anaerobic); 15 days (aerobic), 35 °C	BOD: 15.6–22.1 g L^{-1}, COD: 30.1–50.7 g L^{-1}	39	0.019–0.020 m^3 kg^{-1} COD_{remov}	BOD: 99.5%, COD: 98.2%	Uzal et al. 2003
UASB + filter (anaerobic hybrid reactor)	Spent wash distillery	Lab scale: H 150 cm, $L \times W_{(internal)}$ 10 × 10 cm, a hopper in the bottom,	9 months, four HRTs: 4, 5, 6, 7, and 8 days	COD: 134.8 g L^{-1}	11.13, 8.70, 7.13, 5.73, and 4.53	~ 6.8m^3 m^{-3} day^{-1}, $Yield_{max,\ (CH4)}$: 0.334 m^3 COD kg^{-1}	COD: 60–79%, sulfate: 60–80%	Kumar et al. 2007

Table 2 (continued)

System name	Feed type	Scale and capacity	Operation time, HRT, and temp.	Influent water quality	OLR (kg COD m^{-3} day^{-1})	Biogas rate (BPR)	Effluent water quality	Ref.
		a G-L-S separator on top						
UASB	Distiller grains wastewater	Lab scale: H 190 cm, I.D. 8.2 cm, V_{work} 8.18 L, a G-L-S separator on top	420 days, HRT: 82–11 h, 37 °C	COD: 16.5–22.5 g L^{-1}, pH 3.3–4.3	5, 12.4, 25.7, 33.7, 41.6, 48.3	0.318 L CH_4 g^{-1} COD	COD: 80–97.3%	Gao et al. 2007
Multi-staged (MA) UASB	Barley-based Shochu distillery wastewater	Pilot scale (V 2.5 m^3, H 6.3 m, I.D. 0.8 cm) with six internal G-S separators	860 days; HRT: 8, 12, 20, and 24 h; 55 °C	COD: 86.9 g L^{-1}, pH 3.9, SS: 3.9 mg L^{-1}, TKN: 4.1 $g_{(as\ N)}$ L^{-1}	34.8, 41.7, 45.2	N.A.	Removal: COD 83.8%	Yamada et al. 2013
CSTR, ASBR, FBR	Simulated thin stillage from DDGS was diluted with tap water	Lab scale: V_{CSTR} 5 L; V_{FBR} 12.9 L; V_{ASBR} 13 L.	CSTR:758 days, HRT 33 → 6 days, FBR: 294 days, HRT 33 → 6 days, ASBR: 712 days, HRT 40 → 6 days; all in 38 °C	Total solid of 92.8% TS	CSTR: 2 → 10, FBR: 2 → 10, ASBR: 1 → 10,	64.5%, 64.4%	N.A.	Schmidt et al. 2013
UASB	Combination of domestic waste activated sludge and distillery's spent wash	N.A.	71 days, HRT of 5 days	N.A.	N.A.	Biogas: 2 L day^{-1}	COD: 65%, BOD: 40.4%, SS 87%	Prajapati and Chaudhari 2015
UASB	Hot spent wash without dilution from a distillery industry	Bench scale: H ~ 60 cm, I.D. 92.1 cm, V 5 L, one aerobic flask (V 500 mL)	635 days, 37 °C, HRT 47.1 h	COD: 68–110 g L^{-1} (mean ± SD 83.3 ± 9.7 g L^{-1}), pH 3.4–5.8	8.1, 10.2, 12.8 (opm), 15.3, 17.8, 19.0, 22.9, and 25.9	CH_4: 65–75%, 0.38 m^3 kg^{-1} COD_{remov}	Removal: BOD 68.5%, COD 89%	Saner et al. 2016
UASB + An/Ae-MBR	Winery distillery wastewater	Bench scale: H 75 cm, D 22 cm, V 24 L, followed by one anoxic-aerobic tank with UF membranes (V_{work} 0.85 L/1.0 L)	35 days, 37 °C, HRT 16 h	COD: 44.6 g L^{-1}, pH at 3.8	3.0, 4.5 (opm), 6.2, 10.5, and 11.5	BPR: 2.6 m^3 m^3 day^{-1}	Removal: COD 95%	Petta et al. 2017
EGSB	Supernatant from settled distillery wastewater	Pilot scale (1st stage: 45 L, 2nd stage: 60 L)	~ 20 days	COD: ~ 5, ~ 10, ~ 20, and ~ 30 g L^{-1}	~ 3, 5, 7, and 9	Biogas: 22–32%	Removal COD 33–42%	Ghorbanian et al. 2014a, b
ECSB in full scale (and two EGSBs in lab scale)	Scottish whiskey distillery	A full-scale ECSB (FSB, V_{work} 425 m^3, two internal G-L-S separators), and two EGSB lab-scale reactors (1-D and 3-D, both V_{work} 20 L, S-L separator on top each), D:H ratios of the three reactors are 7:12, 1:15, and 1:4, respectively	FSB: 85 days, 36.1 ± 1.0 °C; 1-D and 3-D: 70 days, both 37.0 ± 0.6 °C, HRT 16 h	COD for FSB: 6.53 ± 0.95 g L^{-1}; COD for 1-D and 3-D: 4.71 ± 0.73 g L^{-1}	FSB: 9.9 ± 3.0 g COD· $L_{reactor}$ day^{-1}, 1-D and 3-D: 6.2 ± 0.4 g COD $L_{reactor}$ day^{-1}	$BPR_{(1-D)}$, $BPR_{(3-D)}$: 2.95, 3.01 L^{-1} day^{-1}; CH_4 ratio for 1-D and 3-D: 74.5 and 72.8%	Removal of COD: FSB: 70.7%, 1-D: 91.1%, 3-D: 88.1%	Connelly et al. 2017

Table 2 (continued)

System name	Feed type	Scale and capacity	Operation time, HRT, and temp.	Influent water quality	OLR (kg COD m^{-3} day^{-1})	Biogas rate (BPR)	Effluent water quality	Ref.
ECSB	Pot ale mixed with spent wash from a distillery	Full scale, $Q = 300$ m^3 day^{-1}, D 6.55 m, H 15.9 m, V 490 m^3, U (upflow) 5.05 m^3 h^{-1}	349 days, 34.6 ± 2.7 °C,	COD: 53 ± 0.8 g L^{-1}	OLR: 9000 kg COD day^{-1}, VLR: 18.4 kg COD m^{-3} day^{-1}	CH_4: > 75%, ave. 1169 m^3 day^{-1}, (729–3466 m^3 day^{-1})	Removal: COD 95.3%	This study

1-D and 3-D one- and three-dimension reactor, *ABR* anaerobic baffled reactor, *ASBR* anaerobic sequencing batch reactor, *BFFB* biphasic fixed film bioreactor, *CSTR* continuous stirred tank reactor, *D:H* ratio reactor diameter to height ratio, *FBR* fixed bed reactor, *GRABR* granular bed anaerobic baffled reactor, *G-L separator*, gas-liquid separator, *G-S separator* gas-solid separator, *G-L-S separator* gas-liquid-solid separator, *IC + EC* internal circulation reactor with external circulation system, *MS-UASB* multi-staged UASB, *N.A.* not available, *PDL* pulsed discharge in liquid, *UAF* upflow anaerobic filter

down due to the instability of system alkalinity and temperature and eventually leading to system failure (Metcalf and Eddy Inc. et al. 2014). Therefore, the system applied heat generated from biogas burning, which is not only saving the fuel cost of the boiler but also reducing the carbon footprint of methane (biogas).

Granular sludge sampling and analysis

Granulation was affected by a number of extrinsic factors in UASB series reactor, including temperature, pH, alkalinity, OLR, upflow velocity, nature, and strength of substrate, nutrients, multivalent cations (e.g., Ca, Mg, Fe, Cu, Co, and Al), salinity and heavy metals, microbial ecology of seed sludge, extracellular polymeric substances (EPS), and addition of natural and synthetic polymer (Tiwari et al. 2006; van Lier et al. 2016). Although anaerobic granulation currently is still not well understood (Lim and Kim 2014), four hypothetic steps and explains were made. In this study, nine sampling ports distributed 1 m each with a vertical height of the ECSB reactor (from 1 to 9 m) which enabled demonstrated spatial differences of the granules from the expanded sludge bed. Although molecular biological analysis of microbial community physiology and taxonomy was not investigated as Connelly et al. (2017), we monitored settling property of the sludge spatially and temporally. The spatial distribution of sludge analyzed the samples from each port via an Imhoff cone test, using 1 L and observed the final volume of settled sludge after 5 min. Results and apparatus were shown in Fig. 4e, where VS of the three bottom ports (P1: 72,857, P2: 51,317, and P3: 56,332 mg L^{-1}) was two orders higher than the top six ports (P4–P9: 693–502 mg L^{-1}). Such results indicated that most of the sludge was very easy settled within 3-m height. Excellent solid-liquid separation was achieved in the top of the reactor. In addition to the investigated spatial distributions of sludge property, the temporal difference of sludge property was monitored via the same Imhoff cone tests throughout the whole operation period (294 days). As shown in Fig. 4d, results were always below 1.0 mL L^{-1} with an average of 0.5 ± 0.4 mL L^{-1}. Such results indicated that most of the formed anaerobic granular sludge (biomass) was kept well in the reactor without flowing out as a generally acceptable value (only below 3.0 mL L^{-1} in a 5-min Imhoff cone test). In phase 1, seeding sludge was about 3500–4000 kg, where roughly 1000 kg came from a sugar plant and others were from a pulp and paper mill. A total of 6200 kg of sludge in the ESBC system was estimated on February 27, 2017. Accordingly, accumulated anaerobic granular sludge was about 2200–2700 kg and total COD reduction percentage amount was more than 274,000 kg for 294-day

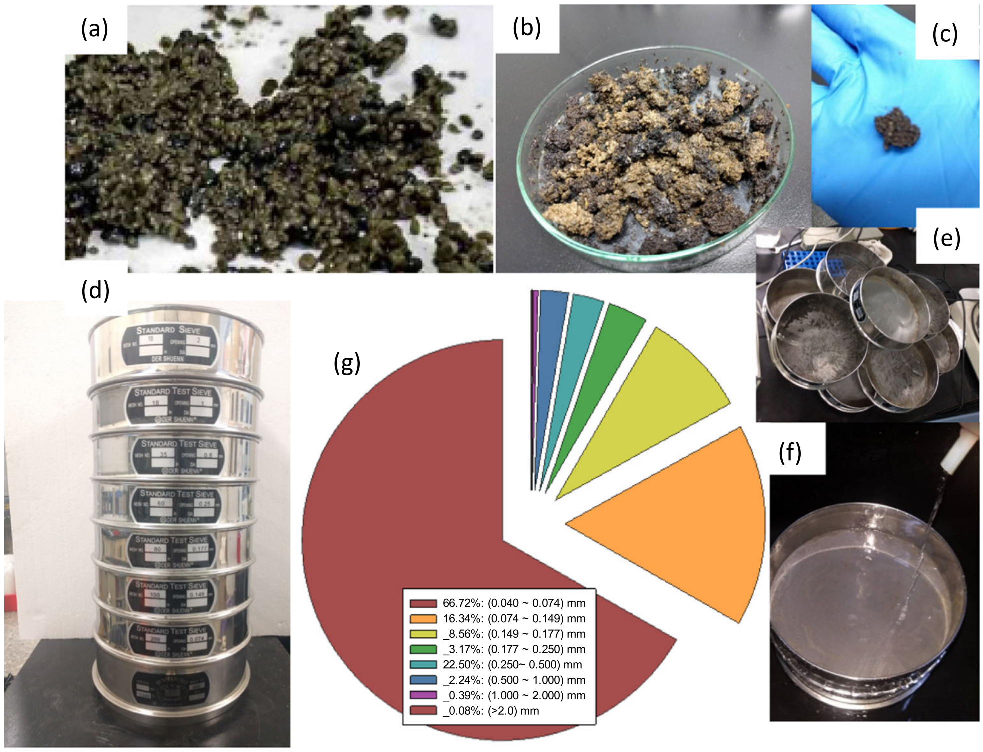

Fig. 5 **a** Granule sludge obtained from the ECSB system; **b** dried granule sludge; c single granule sludge on hand; **d** standard sieves; **e**, **f** filtration using each sieve by DI water rinse; **g** size distribution of dried granule sludge

continuous operation. Based on a growth yield of 0.02 kg mLVSS/kg COD, it is reasonable that anaerobic granular sludge increased to 2200 kg in phase 3 (190 days continuously operation).

Collected sludge samples, as shown in Fig. 5a, were dried in room temperature for 1 week, as shown in Fig. 5b. We further applied six standard sieves with openings of 2.0 mm (no. 2), 1.0 mm (no. 18), 0.5 mm (no. 35), 0.25 mm (no. 60), 0.177 mm (no. 80), 0.149 mm (no.100), 0.074 mm (no. 200), and 0.040 mm (no. 350), as shown in Fig. 5d. Results showed that most of the sludge (66.7%) was smaller than 0.04 mm (passing through the sieve no. 350), following a bigger part (16.3%) between 0.074 and 0.149 mm (retaining on the sieve no. 350 and passing through the sieve no. 200), then subsequently decreasing to 0.08% as the biggest part (retaining on the sieve no. 2).

Morphological observation of the granular sludge was performed by a field emission scanning electron microscopy (FE-SEM, S-4800, HITACHI, Japan) in Feng Chia University with energy dispersive X-ray spectroscopy (EDS) analysis its elemental composition. Two granular sludge with a size bigger than 2.0 mm (retained on the sieve no. 2) were firstly taken at magnitudes of × 30 under FE-SEM. Cutted inner core and outside surface of the two granular sludges can be observed, as respectively shown in Fig. 6a and d. Then, both of them were taken in bigger magnitudes of × 500 and × 2000 to take an insight of each structure, as respectively shown in Fig. 6b, c, e, and f. Even under the × 2000 magnitude, the cutted inner core was rough and the latter was smooth. Comparing with the "good quality granular sludge" proposed by van Lier et al. (2016), the size of our granular sludge generally might be smaller than the diameter (0.1–8 mm) of methanogenic granular sludge grown in paper mill wastewater. It could be interesting to further investigate the mechanism of this granular grown in whiskey wastewater and ECSB reactor. From the pie chart made for elemental analysis of EDS, as shown in Fig. 5g, the main elements in the cut inner core were carbon (44.4%) and oxygen (41.4%) with minor elements of nitrogen (7.2%), sulfur (3.8%), and iron (2.2%).

In general, settling properties of sludge and clinic inspection of granular sludge were critical to monitor the anaerobic sludge in the ECSB reactor. These results provide important information, such as how much anaerobic biomass was retained in the reactor, and can be a good index to check the operation in treating such high-strength wastewater. For simultaneously sustainable managing solid and liquid waste from distilleries, strategies performed by Weber and Stadlbauer (2017) can be further considered.

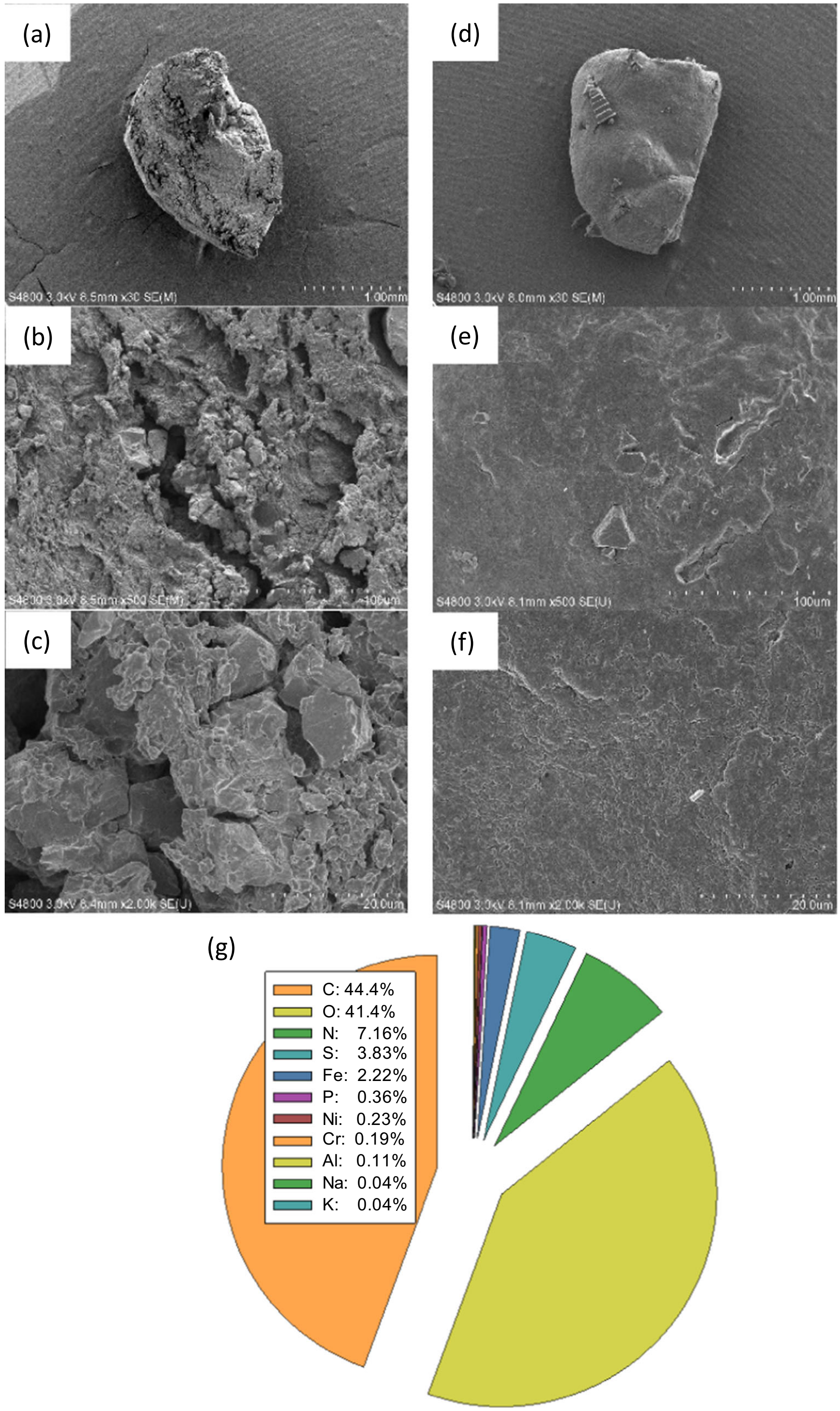

Fig. 6 FE-SEM of granular sludge (> 2.0 mm): internal structure of granular sludge in magnitude of **a** × 30, **b** × 500, **c** × 2000; outer shell of granular sludge in magnitude of **d** × 30, **e** × 500, **f** × 2000; **g** elemental percentage of internal granular sludge (EDS result)

Conclusions

This study presented technical feasibility of applying the third-generation anaerobic digester, i.e., external circulation sludge bed (ECSB), on whiskey distillery wastewater treatment, which was categorized as one of the high-strength wastewater and difficult to biodegradable by the conventional aerobic process. Simultaneously producing green energy from biogas during the treatment was achieved. Successful start-up of the system and continuously running the system for 200 days, which even suffered extreme loading fluctuations, were demonstrated. Comparing with previous anaerobic treatment processes and performances for whiskey distillery wastewater, the cost-effective wastewater treatment using the ECSB system was illustrated in this study as well as potential contributions on sustainability (e.g., utilizing green energy and reducing carbon footprint) was prospective.

Acknowledgments Efforts of employees in the Kavalan distillery during the system operation and intensive monitoring as well as supports from HydroThane during the constructing the ECSB system were both highly appreciated. Additional water and sludge analysis from Prof. Wen-Hsing Chen in National Ilan University was acknowledged.

References

Abbasi T, Tauseef SM, Abbasi SA (2012) Anaerobic digestion for global warming control and energy generation—an overview. Renew Sust Energ Rev 16:3228–3242

Acharya BK, Pathak H, Mohana S, Shouche Y, Singh V, Madamwar D (2011) Kinetic modelling and microbial community assessment of anaerobic biphasic fixed film bioreactor treating distillery spent wash. Water Res 45:4248–4259

Agarwal R, Lata S, Gupta M, Singh P (2010) Removal of melanoidin present in distillery effluent as a major colorant: a review. J Environ Biol 31:521–528

Akunna JC, Clark M (2000) Performance of a granular-bed anaerobic baffled reactor (GRABBR) treating whisky distillery wastewater. Bioresour Technol 74:257–261

Anonymous (2016) Bio-energy plants transform whisky distillery sector. Filtration+Separation. Nov–Dec. 2016.

Arimi MM, Zhang Y, Götz G, Kiriamiti K, Geißen S-U (2014) Antimicrobial colorants in molasses distillery wastewater and their removal technologies. Int Biodeterior Biodegrad 87:34–43

Barrena R, Traub JE, Gil CR, Goodwin JAS, Harper AJ, Willoughby NA, Sánchez A, Aspray TJ (2018) Batch anaerobic digestion of deproteinated malt whisky pot ale using different source inocula. Waste Manag 71:675–682

Bathgate GN (2016) A review of malting and malt processing for whisky distillation. J Inst Brew 122:197–211

Bilad MR, Declerck P, Piasecka A, Vanysacker L, Yan X, Vankelecom IFJ (2011) Treatment of molasses wastewater in a membrane bioreactor: influence of membrane pore size. Sep Purif Technol 78:105–112

Chowdhary P, Raj A, Bharagava RN (2018) Environmental pollution and health hazards from distillery wastewater and treatment approaches to combat the environmental threats: a review. Chemosphere 194:229–246

Connaughton S, Collins G, O'Flaherty V (2006) Psychrophilic and mesophilic anaerobic digestion of brewery effluent: a comparative study. Water Res 40:2503–2510

Connelly S, Shin SG, Dillon RJ, Ijaz UZ, Quince C, Sloan WT, Collins G (2017) Bioreactor scalability: laboratory-scale bioreactor design influences performance, ecology, and community physiology in Expanded Granular Sludge Bed bioreactors. Front Microbiol 8:664 15 pages

Enitan AM, Adeyemo J, Swalaha FM, Bux F (2015) Anaerobic digestion model to enhance treatment of brewery wastewater for biogas production using UASB reactor. Environ Model Assess 20:673–685

Fang HHP (ed) (2010) Environmental anaerobic technology: applications and new developments. Imperial College Press, London

Fatih Demirbas M, Balat M (2009) Progress and recent trends in biogas processing. Int J Green Energy 6:117–142

Fuess LT, Garcia ML (2014) Implications of stillage land disposal: a critical review on the impacts of fertigation. J Environ Manag 145:210–229

Fuess LT, Garcia ML (2015) Bioenergy from stillage anaerobic digestion to enhance the energy balance ratio of ethanol production. J Environ Manag 162:102–114

Gao M, She Z, Jin C (2007) Performance evaluation of a mesophilic (37^{o}C) upflow anaerobic sludge blanket reactor in treating distiller's grains wastewater. J Hazar. Mater 141:808–813

Ghorbanian M, Lupitskyy RM, Satyavolu JV, Berson RE (2014a) Impact of supplemental hydrogen on biogas enhancement and substrate removal efficiency in a two-stage expanded granular sludge bed reactor. Environ Eng Sci 31:253–260

Ghorbanian M, Lupitskyy RM, Satyavolu JV, Berson RE (2014b) Impact of hydraulic retention time at constant organic loading rate in a two-stage expanded granular sludge bed reactor. Environ Eng Sci 31:317–323

Goodwin JAS, Stuart JB (1994) Anaerobic digestion of malt whisky distillery pot ale using upflow anaerobic sludge blanket reactors. Bioresour Technol 49:75–81

Goodwin JAS, Finlayson JM, Low EW (2001) A further study of the anaerobic biotreatment of malt whisky distillery pot ale using an UASB system. Bioresour Technol 78:155–160

Graham J, Peter B, Walker G, Wardlaw A, Campbell E (2012) Characterisation of the pot ale profile from a malt whisky distillery. In: Walker GM, Goodall I, Fotheringham R, Murray D (eds) Distilled spirits: science and sustainability, vol 43. Nottingham University Press, Nottingham, pp 1–7

Guo K, Shang Y, Gao B, Xu X, Lu S, Qi Q (2018) Study on the treatment of soybean protein wastewater by a pilot-scale IC-A/O coupling reactor. Chem Eng J 343:189–197

Harada H, Uemura S, Chen A, Jayadevan J (1996) Anaerobic treatment of a recalcitrant distillery wastewater by a thermophilic UASB reactor. Bioresour Technol 55:215–221

Hosseini SE, Wahid MA (2014) Development of biogas combustion in combined heat and power generation. Renew Sust Energ Rev 40:868–875

Hutnan M, Hornak M, Bodik I, Hlavacka V (2003) Anaerobic treatment of wheat stillage. Chem Biochem Eng Q 17:233–241

Khanal SK, Giri B, Nitayavardhana S, Gadhamshetty V (2016) Anaerobic bioreactors/digesters: design and development In: Lee D-J, Hallenbeck P, Ngo H, Jegatheesan V, Pandey A (eds) Current developments in biotechnology and bioengineering: biological treatment of industrial effluents, vol 10. 1st edn. Elsevier, pp 261–279. https://doi.org/10.1016/B978-0-444-63665-2.00010-2

Kharayat Y (2012) Distillery wastewater: bioremediation approaches. J Integr Environ Sci 9:69–91

Krishnamoorthy S, Premalatha M, Vijayasekaran M (2017) Characterization of distillery wastewater – an approach to retrofit existing effluent treatment plant operation with phycoremediation. J Clean Prod 148:735–750

Kumar GS, Gupta SK, Singh G (2007) Biodegradation of distillery spent wash in anaerobic hybrid reactor. Water Res 41:721–730

Laubscher ACJ, Wentzel MC, Le Roux JMW, Ekama GA (2001) Treatment of grain distillation wastewater in an upflow anaerobic sludge bed (UASB) system. Water SA 27:433–444

Lim SJ, Kim T-H (2014) Applicability and trends of anaerobic granular sludge treatment processes. Biomass Bioenergy 60:189–202

Mallick P, Akunna JC, Walker GM (2010) Anaerobic digestion of distillery spent wash: influence of enzymatic pre-treatment of intact yeast cells. Bioresour Technol 101:1681–1685

Mao C, Feng Y, Wang X, Ren G (2015) Review on research achievements of biogas from anaerobic digestion. Renew Sust Energ Rev 45:540–555

Melamane XL, Strong PJ, Burgess JE (2007) Treatment of wine distillery wastewater: a review with emphasis on anaerobic membrane reactors. S Afr J Enol Vitic 28:25–36

Metcalf & Eddy Inc., Tchobanoglous G, Stensel HD, Tsuchihashi R, Burton F (2014) Anaerobic suspended and attached growth biological treatment processes. In: Wastewater engineering: treatment and resource recovery, vol 10. 5th edn. McGraw-Hill Education, New York, pp 1060–1103

Meyer T, Edwards EA (2014) Anaerobic digestion of pulp and paper mill wastewater and sludge. Water Res 65:321–349

Mohana S, Acharya BK, Madamwar D (2009) Distillery spent wash: Treatment technologies and potential applications. J Hazard Mater 163:12–25

Onodera T, Choeisai P, Khodphuvieng J, Prammanee P, Yoochatchaval W, Kaewpradit W, Kubota K (2013) Development of appropriate technology for treatment of molasses-based wastewater. J Environ Sci Health A 48:1114–1121

Pant D, Adholeya A (2007) Biological approaches for treatment of distillery wastewater: A review. Bioresour Technol 98:2321–2334

Petta L, De Gisi S, Casella P, Farina R, Notarnicola M (2017) Evaluation of the treatability of a winery distillery (vinasse) wastewater by UASB, anoxic- aerobic UF-MBR and chemical precipitation/adsorption. J Environ Manag 201:177–189

Prajapati AK, Chaudhari PK (2015) Physicochemical treatment of distillery wastewater—a review. Chem Eng Commun 202:1098–1117

Russell I, Stewart G (eds) (2014) Whisky: technology, production and marketing, 2nd edn. Elsevier Science Publishing Co Inc, United States

Saner AB, Mungray AK, Mistry NJ (2016) Treatment of distillery wastewater in an upflow anaerobic sludge blanket (UASB) reactor. Desalin Water Treat 57:4328–4344

Schmidt T, Pröter J, Scholwin F, Nelles M (2013) Anaerobic digestion of grain stillage at high organic loading rates in three different reactor systems. Biomass Bioenergy 55:285–290

Stout D (2015) You won't believe where the world's best whiskey comes from. TIME. http://time.com/3753768/world-best-whisky-taiwan-kavalan-world-whiskies-awards/ Accessed 30 May 2018.

Sun H, Wu S, Dong R (2016) Monitoring volatile fatty acids and carbonate alkalinity in anaerobic digestion: titration methodologies. Chem Eng Technol 39:599–610

Tauseef SM, Abbasi T, Abbasi SA (2013) Energy recovery from wastewaters with high-rate anaerobic digesters. Renew Sust Energ Rev 19:704–741

Tiwari MK, Guha S, Harendranath CS, Tripathi S (2006) Influence of extrinsic factors on Granulation in UASB reactor. Appl Microbiol Biotechnol 71:145–154

Tokuda M, Fujiwara Y, Kida K (1999) Pilot plant test removal of organic matter, N and P from whisky pot al. Process Biochem 35:265–267

Uzal N, Gokcay CF, Demirer GN (2003) Sequential (anaerobic/aerobic) biological treatment of malt whisky wastewater. Process Biochem 39:279–286

Uzukwu CC, Barraclough MJ, Dionisi D (2017) Experimental investigation of a new process for treatment and valorisation of pot ale wastewaters. Water Sci Technol 75:1194–1203

van Lier JB (2008) High-rate anaerobic wastewater treatment: diversifying from end-of-the-pipe treatment to resource-oriented conversion techniques. Water Sci Technol 57:1137–1148

van Lier JB, van der Zee FP, Frijters CTMJ, Ersahin ME (2016) Development of anaerobic high-rate reactors, focusing on sludge bed technology. Adv Biochem Eng Biotechnol 14:681–702

Weber B, Stadlbauer EA (2017) Sustainable paths for managing solid and liquid waste from distilleries and breweries. J Clean Prod 149:38–48

Wilkie AC, Riedesel KJ, Owens JM (2000) Stillage characterization and anaerobic treatment of ethanol stillage from conventional and cellulosic feedstocks. Biomass Bioenergy 19:63–102

Wolmarans B, de Villiers GH (2002) Start-up of a UASB treatment plant on distillery wastewater. Water SA 28:63–68.

Yamada M, Harada H, Yamauchi M, Yamaguchi T, Ohashi A (2013) Reduction of alkali supplement in a pilot-scale thermophilic multi-staged UASB reactor treating alcohol distillery wastewater. Int J Environ Res 7:823–830

Zero Waste Scotland, Sector study on beer, whisky and fish (2015) 87 pages, http://www.zerowastescotland.org.uk/BeerWhiskyFish. Accessed 30 May 2018.