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Review

Biohythane production from two-stage anaerobic digestion of food waste: A review

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ABSTRACT

The biotransformation of food waste (FW) to bioenergy has attracted considerable research attention as a means to address the energy crisis and waste disposal problems. To this end, a promising technique is two-stage anaerobic digestion (TSAD), in which the FW is transformed to biohythane, a gaseous mixture of biomethane and biohydrogen. This review summarises the main characteristics of FW and describes the basic principle of TSAD. Moreover, the factors influencing the TSAD performance are identified, and an overview of the research status; economic aspects; and strategies such as pre-treatment, co-digestion, and regulation of microbial consortia to increase the biohythane yield from TSAD is provided. Additionally, the challenges and future considerations associated with the treatment of FW by TSAD are highlighted. This paper can provide valuable reference for the improvement and widespread implementation of TSAD-based FW treatment.

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Introduction

Fossil fuels such as coal, oil, and natural gas have been the dominant energy sources worldwide for a considerable period. In 2018, fossil fuels represented more than 80% of the world's total primary energy supply (Bundhoo and Mohee, 2016; IEA, 2020). However, fossil fuels are being rapidly depleted, and their combustion leads to the emission of pollutants, which adversely impact the environment and human health (Soares et al., 2020). To mitigate these problems, renewable energy resources such as solar, wind, geothermal, and bioenergy are being widely investigated as alternatives to fossil fuels (Dinesh et al., 2018; Nazir et al., 2019). Bioenergy is an

eco-friendly and renewable alternative energy resource that is typically obtained from perishable organic wastes. Hydrogen (H₂) and methane (CH₄) are promising bioenergy sources owing to their high energy densities of 142 and 55.5 MJ/kg, respectively (Hassan et al., 2019; Singh et al., 2015). When the two gases unite, they form a mixture named biohythane, which is an advanced fuel with hydrogen fractions of 10%–30% (Meena et al., 2020). Biohythane inherits the benefits of both H₂ and CH₄, such as low production costs, environmental friendliness, and sustainability, which enhance its potential in the marketplace (Rawoof et al., 2021).

In addition to the energy crisis, the treatment of increasing amounts of municipal solid wastes (MSWs) has emerged as a notable global concern. The annual MSW production is expected to reach 2.3 billion tons by 2025 (Qin et al., 2019). Food waste (FW) is one of the most abundant and problematic MSWs, accounting for over 50% of the total MSW produced in

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China (Yun et al., 2018). FW includes uneaten food residues and food discarded during production, processing, retailing, and consumption (Zhou et al., 2018), which are typical perishable substances. Unless properly managed, FW may lead to severe water, air, and soil pollution through its leachate, odour, and rapid decomposition during collection, transportation, and storage (Kim et al., 2021). FW is generally treated and disposed of by composting, landfilling, incineration, or use as feed for animals. However, these methods have inherent disadvantages. For example, the landfilling of FW emits large volumes of CH₄ and carbon dioxide (CO₂), which are greenhouse gases and contribute to the global climate change (Giwa et al., 2019; Park et al., 2021). Moreover, the leachate generated by landfilling may pollute underground water sources, and the odour may have adverse environmental effects (Giwa et al., 2019).

Anaerobic digestion (AD) is an effective technique for treating organic wastes (Guimaraes et al., 2018). However, single-stage AD (SSAD) of FW involves many limitations, such as ammonia inhibition, system instability, insufficient buffering capacity, and accumulation of volatile fatty acids (VFAs) (Paillet et al., 2021). Two-stage AD (TSAD), in which the hydrolysis-acidogenesis and methanogenesis stages are separated, can overcome the disadvantages of SSAD through the provision of optimal conditions for bacterial activity in each stage. Therefore, TSAD is a promising technique to treat FW and recover bioenergy in the form of CH₄ and H₂ (Xiao et al., 2018). There are some reviews focusing on the TSAD of FW. For example, *Srisowmeya et al. (2020)* investigated the parameters influencing the TSAD performance, analysed the reported findings, and highlighted the importance of stage separation in treating perishable substrates. However, the previous publications mainly focused on the biohydrogen produced in the first stage instead of biohythane and did not discuss the main strategies, especially the regulation of microbial consortia, for enhancing the biohythane production from the TSAD of FW.

The objective of this study was to comprehensively examine the biohythane yield from TSAD of FW, in terms of the principle, influencing factors, research status, challenges, and future considerations. Therefore, the main characteristics of FW were analysed, and the basic principle of TSAD was clarified. Subsequently, the main factors influencing the performance of TSAD, i.e., the pH, temperature, substrate characteristics, organic loading rate (OLR), hydraulic retention time (HRT), carbon to nitrogen ratio (C/N ratio), inoculum, and reactor types, were identified, and an overview of the state-of-the-art in biohythane production from TSAD of FW was presented. Moreover, the main challenges and future considerations were specified. This review is expected to provide valuable insights into treating FW by TSAD to produce biohythane, and a reference for improving the TSAD process.

1. Main characteristics of FW

Large amounts of FW are produced every year. For example, in China, 125 million tons of FW was generated in 2020 (Liu et al., 2022), and this amount is expected to increase in the future (Song et al., 2018). FW generation is correlated with the gross domestic product, population, output of major agri-

cultural products, and animal husbandry and fishery activities (Li et al., 2019b). Larger amounts of FW are expected to be produced in regions with higher population densities and better living standards. FW generally includes catering and kitchen waste. Catering waste, which refers to the food residues of restaurants and unit canteens and the waste generated in processing fruits, vegetables, meat, oil, and pastry in back kitchens, mainly contains oil and solid-liquid mixtures. Kitchen waste, which refers to perishable organic waste such as leftover fruits, vegetables, food materials, and fruit skins discarded in daily life activities, mainly contains solid waste.

In terms of the physical characteristics of FW, the total solids (TS) content is 15%–25% (with a moisture content of approximately 75%–85%), and the ratio of the volatile solids (VS) to TS is 85%–95% (Yun et al., 2018; Zhou et al., 2018). The high water content of FW usually results in a low calorific value, and thus, solely FW cannot be incinerated (Wang et al., 2021). Additionally, FW has a low initial pH with a mean value of 5.3, which generates a low-pH phase and threatens the stability of the AD system. Therefore, the initial pH must be adjusted to realise the AD of FW (Li et al., 2019b). In terms of the biochemical characteristics, FW contains natural fibres, carbons, proteins, fats, lipids, vitamins, and minor minerals embedded in an organic matrix that is biodegradable. The chemical compositions of individual food components in FW differ owing to the diverse eating habits in different regions (Karthikeyan et al., 2018). Appendix A Fig. S1 shows the chemical compositions of several foods in FW (Musaiger, 2011). Compared with other countries such as the United Kingdom and South Korea, the FW in China is characterised by high water and salt contents, large amounts of lipids, and the presence of complex materials such as paper, plastic, metal, and glass (Giwa et al., 2019; Negri et al., 2020).

2. Basic principle of the TSAD of FW

The AD performed by a group of microbes can be divided into four successive stages: hydrolysis, acidogenesis, acetogenesis, and methanogenesis (Al-Rubaye et al., 2019). In the AD of FW, the organic matter in FW undergoes rapid hydrolysis, acidogenesis, and acetogenesis, resulting in an accumulation of VFAs, which can inhibit methanogenesis and lead to AD failure (Wang et al., 2018a). These phenomena occur because the hydrolysis-acidification bacteria readily convert the organic matter of FW to VFAs, but the methanogens cannot convert the VFAs to CH₄ at the same rate. In other words, in a single reactor, hydrolysis-acidification bacteria and methanogens are not synchronised with respect to their metabolic efficiencies towards FW. To fully exploit the functionalities of the two groups of microorganisms, strategies involving TSAD of FW have been developed, in which hydrolysis-acidogenesis and methanogenesis occur in separate reactors (Chatterjee and Mazumder, 2019). Fig. 1 illustrates the main steps in the TSAD of FW.

2.1. First stage

The first stage in the TSAD of FW involves hydrolysis, acidogenesis, and acetogenesis. In the hydrolysis process,

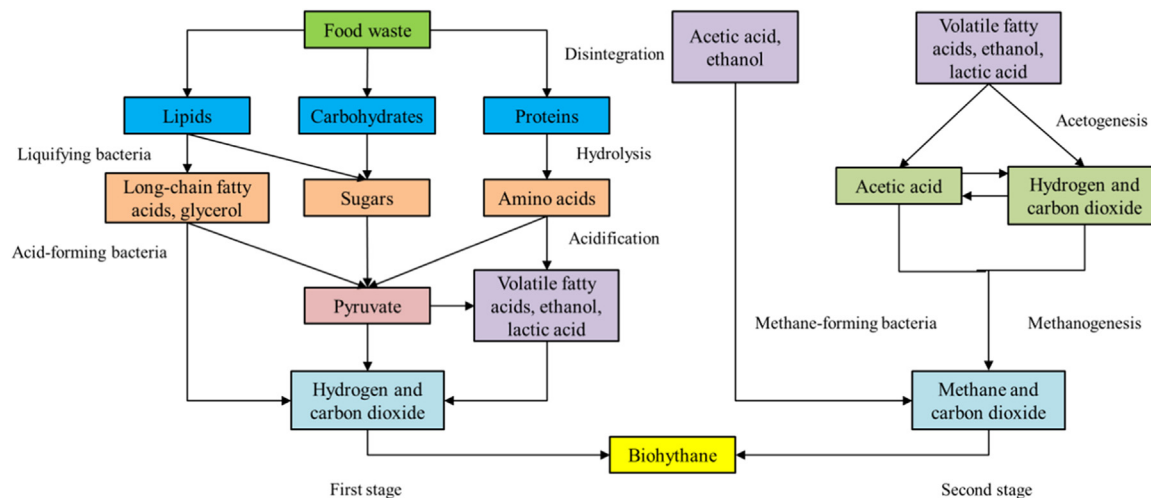


Fig. 1 – Schematic of biohythane production from food waste through two-stage anaerobic digestion.

the complex and high molecular weight compounds in the FW are decomposed into small soluble molecules (Okoro-Shekwaga et al., 2019). For example, carbohydrates, proteins, and lipids are converted to sugars, amino acids, and long-chain fatty acids and sugars, respectively (Gujer and Zehnder, 1983). During acidogenesis, the low molecular weight cultures obtained from hydrolysis are used by the acidogenic bacteria to excrete VFAs such as formic acid, acetic acid, propionic acid, butyric acid, and pentanoic acid; alcohols such as methanol and ethanol; aldehydes; and gases such as CO_2 and H_2 (Srisowmeya et al., 2020). The dominant VFAs are acetic acid and butyric acid, with proportions of 44.0% to 69.3% and 22.9% to 47.2% in the studies by Slezak et al. (2021). Hydrolysis occurs more rapidly than the other AD processes, and the composition of VFAs produced in this step considerably influences the succeeding phase. In the acetogenesis process, acetogenic bacteria (a restricted group of homoacetogens) convert a part of the VFAs and other intermediates generated during acidogenesis to acetic acid, ammonia, H_2 , and CO_2 (Kainthola et al., 2019; Li et al., 2019a). These products function as key substrates in methanogenesis.

2.2. Second stage

The second stage of TSAD involves methanogenesis, during which the products from the first stage are converted to CH_4 and CO_2 . Hydrogenotrophic and acetoclastic methanogens contribute to approximately 30% and 70% of the methane production, respectively (Jiang et al., 2019). Hydrogenotrophic methanogens convert H_2 and CO_2 to CH_4 , whereas acetoclastic methanogens convert acetic acid to CH_4 and CO_2 (Angelidaki et al., 2018; Kurade et al., 2019). A few methanogens such as *Methanosarcina* sp. can utilise both pathways for generating methane (Chandra et al., 2012). Moreover, methanol can be converted to CH_4 and H_2O during methylotrophic methanogenesis. In general, hydrolysis is considered the rate-limiting step during the AD of complex solid-state organic matters such as sewage sludge (Ma et al., 2013). However, because the hydrolysis-acidification of FW typically

occurs rapidly and methanogens are extremely sensitive to the accumulation of VFAs in the medium, the methanogenic process may often become the rate-limiting step in the AD of FW (Cremonez et al., 2021).

3. Factors influencing the performance of TSAD of FW

The generation efficiency of biohythane from FW through TSAD is influenced by many factors such as the pH, temperature, substrate types, and OLR, summarised in Table 1.

3.1. pH

The pH considerably influences the stability of the AD process, especially for FW. Extremely low or high pH values may inhibit TSAD (Panigrahi and Dubey, 2019). The TSAD of FW involves various microorganisms that perform optimally in different pH values. For example, fermentative bacteria are functionally active within a wide range of pH values between 4.0 and 8.0, whereas methanogens perform efficiently within a pH range of 6.5–7.5 (Srisowmeya et al., 2020). In TSAD, optimal conditions for the hydrolysis-acidification bacteria and methanogens can be provided by regulating the pH in two reactors (Dangol et al., 2022). Generally, the ideal pH range in the first stage of TSAD is 5.2–6.5, with 5.5 being the optimal pH (Elreedy et al., 2015; Li et al., 2013). The degradation rate of carbohydrates is approximately 70% at pH 4.0 and reaches 95% at pH 5.5 (Yu and Fang, 2002). Additionally, the VFA composition in the effluent of the first stage varies with the pH and can be classified as ethanol-type, mixed-acid-type, propionic-acid-type, and butyric-acid-type fermentation at pH 4–4.5, 4.5–5, 5–5.5, and 5.5–6.5, respectively (Ren et al., 2007; Zheng et al., 2015). The pH range of 6.8–7.8 is favourable for the second stage of TSAD (Chatterjee and Mazumder, 2019). Srisowmeya et al. (2020) narrowed this pH range to 6.5–7.2 as the ideal range for methanogenesis. To avoid sharp fluctuations in the pH in the first stage, recirculation from the second stage to the first stage can be implemented.

Table 1 – Factors influencing the hydrogen and methane production from food waste through two-stage anaerobic digestion.

Substrates	Inoculum source	Type of digestion	Reactor volume L)	Temperature (°C)	pH	HRT (day(s))	OLR	Removal efficiency	Biohythane yield		Refs.
									Hydrogen yield	Methane yield	
Sewage sludge and wine vinasse	Anaerobic sludge	Two-stage	3, 3	55, 35	5.44, 7.86	1, 4	28.66 g VS/(L·day), 5.43 g VS/(L·day)	8.92%, 53.48% COD	22.90 mL/g VS _{added}	212.56 mL/g VS _{added}	Tena et al. (2021)
Simulated food waste	Anaerobic sludge	Two-stage	3, 5 (working volume)	35, 35	6	8, 42 (SRT)	1.9 g/(L·day)	–	31.1 mL/g	460 mL/g	Feng et al. (2020)
Pineapple peel waste	Pig farm digestate	Two-stage	1.2, 5.0 (working volume)	37, 37	5–6, 7	4 hr, 3	328.78 kg COD/(m ³ ·day)	42.3%, 42.7% COD	4.1 mL/g COD	167.9 mL/g COD	Chu et al. (2020)
Food waste	Anaerobic sludge	Two-stage	4.5, 4.5 (working volume)	45, 45	5.5, 8.0	17 hr, 17 hr	20 g COD/(L·day), 4.5 g COD/(L·day)	76.6%, 74.1% COD	22.7 mL/g COD _{consumed}	30 mL/g COD _{consumed}	Hassan et al. (2020)
Food waste	Anaerobic sludge	Two-stage	0.5, 0.5	37, 37	6, 8	3, 21	–	–	212.2 mL/g VS	412.6 mL/g VS	Zhao et al. (2021)
Orange peel	Anaerobic sludge	Two-stage	4.3, 4.3	35, 35	5–6, 7–8	25.8	0.36 g COD/(L·day)	46%, 50% TVS	0.79 L/g TVS		Jimenez-Castro et al. (2020)
Fast food restaurant leftovers	Anaerobic sludge	Two-stage	4.6, 4.6 (working volume)	35, 35	4, 8	17 hr, 17 hr	–	58.3%, 47.5% COD	4.1 mL/g COD _{consumed}	5.7 mL/g COD _{consumed}	Hassan et al. (2021)
					5.5, 8.0	17 hr, 17 hr	–	62%, 93% COD	4.2 mL/g COD _{consumed}	7.4 mL/g COD _{consumed}	
					5.5, 8.0	34 hr, 17 hr	–	72.8, 94% COD	1.0 mL/g COD _{consumed}	4.3 mL/g COD _{consumed}	
Food waste	Anaerobic digestate	Two-stage, batch	1, 1	34, 55	5.5, 7.0	–	5.1 g VS _{FW} /L	–	53.5 mL/g VS	307.5 mL/g VS	Ghimire et al. (2021)
Artificial food waste	Anaerobic sludge	Two-stage, batch	5.3, 10	37, 37	6.0, 7.2	–	–	59.7% VS	30.3 L/kg VS _{added}	190.3 L/kg VS _{added}	Luo et al. (2021)
Citrus peel waste	Upflow-anaerobic-sludge-blanket reactor sludge	Two-stage, batch	0.25, 0.25	30, 30	8.5,-	–	–	–	13.29 mmol/L	50.2 mmol/L	Camargo et al. (2021)
Food waste	Anaerobic sludge	Two-stage, batch	0.2 0.2 (working volume)	55, 55	7, -	–	–	70–90% COD	176.10 mL/g COD	310.77 mL/g COD	Wongthanate and Mongkarothai (2018)
Household and restaurant wastes	Cow farm sludge	Two-stage with recirculation	1200, 4000	32, 35	7.4–7.5	5.5–7.2, 35–47 (SRT)	2.5–3.8 kg VS/(m ³ ·day)	83–87% tCOD	189–263 L/kg TCOD _{removed}		Phuoc-Dan et al. (2021)

(continued on next page)

Table 1 (continued)

Substrates	Inoculum source	Type of digestion	Reactor volume L)	Temperature (°C)	pH	HRT (day(s))	OLR	Removal efficiency	Biohythane yield		Refs.
									Hydrogen yield	Methane yield	
Food waste	Maize straw digestate and municipal sewage sludge	Two-stage with recirculation	2, 4.5 (working volume)	55, 35	5.1, 7.6	5, 9	18 kg VS/(m ³ ·day), 5.7 kg VS/(m ³ ·day)	29.2%, 56.6% TCOD	135 L/kg VS _{in}	510 L/kg VS _{in}	Algapani et al. (2019)
Food waste and activated sludge	Activated sludge	Two-stage with recirculation	3, 12 (working volume)	37, 37	5.54, 7.35	3, 11.9	14.6 kg TVS/(m ³ ·day), 2.5 kg TVS/(m ³ ·day)	32.3%, 54.5% TVS	8.6 NL/(kg TVS·day)	428.3 NL/(kg TVS·day)	Baldi et al. (2019)
Food waste					5.52, 7.43	3, 12.8	14.2 kg TVS/(m ³ ·day), 2.5 kg TVS/(m ³ ·day)	23.5%, 62.5% TVS	12.6 NL/(kg TVS·day)	482.1 NL/(kg TVS·day)	
Food and paper waste	Thermophilic sludge, anaerobic sludge	Two-stage with recirculation	3, 12 (working volume)	55, 37.5	5.31, 7.28	6, 24	4.34 kg COD/(m ³ ·day)	78.4% VS	79 NL/kg VS _{fed}	329 NL/kg VS _{fed}	Qin et al. (2019)
Food waste	Anaerobic sludge	Two-stage with recirculation	8.3, 10	–	6, 7	–	8.03 kg VS/(m ³ ·day)	30%–45% COD	22.35 mL/g COD _{feeding}	167.55 mL/g COD _{feeding}	Chakraborty et al. (2022)
Peach pulp waste	Anaerobic sludge	Two-stage with recirculation	5, 5	30, 30	5.5, 7.4	1, 5	21.2 g COD/(L·day), 0.9 g COD/(L·day)	97.6% COD	–	0.32 L/g COD	Carvalho et al. (2018)
Apple pulp waste				37, 37		1, 2.5	26.7 g COD/(L·day), 7.4 g COD/(L·day)	91.9% COD		0.3 L/g COD	
Food and vegetable wastes	Food waste slurry	Two-stage with recirculation, batch	1, 1.8 (working volume)	–	6, 7	–	–	54.1% COD	–	226.86 mL/g VS	Chakraborty and Mohan. (2018)
Organic market waste	Cow manure	Two-stage with water recirculation	1.34, 13.4 (working volume)	35, 35	5.5, 7.0	1.1–1.5, 11–15	19.39–26.84 g VS/(L·day), 1.27–1.95 g VS/(L·day)	–	0.05–0.07 NL/g VS	0.12–0.18 NL/g VS	Camacho et al. (2019)

HRT: hydraulic retention time; SRT: solid retention time; OLR: organic loading rate; COD: chemical oxygen demand; TCOD: total chemical oxygen demand; VS: volatile solids; TVS: total volatile solids; -: not available.

3.2. Temperature

The effectiveness of the functional microorganisms in the TSAD of FW depends on the temperature. Based on the temperature, the AD process can be classified as psychrophilic (below 20°C), mesophilic (20–45°C), thermophilic (55–70°C), or hyperthermophilic (above 70°C) (Divya et al., 2015; Kumar and Samadder, 2020). Mesophilic bands make up the average temperature of most tropical countries, which are widely used by the need of high methane levels. And greater diversity of active anaerobic microorganisms and better process stability are ensured by them (Kainthola et al., 2019). Luo et al. (2021) investigated the effect of leachate recirculation with various water replacement rates on the performance of TSAD (37°C for the first and second stages) using FW as the substrate, and the highest hydrogen and methane yields corresponded to TS contents of 15%. Compared with mesophilic AD, thermophilic AD offers various advantages, such as more efficient degradation of organic waste, more efficient killing of intestinal parasites and vegetative pathogens, higher reaction rates, and shorter retention times (Suryawanshi et al., 2010). However, thermophilic AD is characterised by low stability, a long startup phase, and large heat energy input (Suryawanshi et al., 2010). Some studies have highlighted the superior performance of mesophilic AD compared with thermophilic AD, attributable to exogenous additives. For example, Sunyoto et al. (2018) reported that with the addition of biochar, the hydrogen yield from the TSAD of FW in mesophilic conditions was higher than that in thermophilic conditions.

The concept of temperature-phased anaerobic digestion (TPAD) has been introduced, in which TSAD is implemented at different temperatures in different stages (Hagos et al., 2017). Specifically, thermophilic and mesophilic conditions can be applied in the first and second stages, respectively. Thermophilic conditions can promote biohydrogen generation, and mesophilic conditions can promote the generation of propionic acid during the hydrolysis-acidification process. Additionally, the stability and methanogenic diversity in the second stage in mesophilic conditions are higher than those in the thermophilic condition. Kanchanasuta and Sillaparassamee (2017) experimentally investigated the TPAD of decanter cake and crude glycerol. The first stage involved thermophilic conditions (55°C) for hydrogen fermentation, and the second stage involved mesophilic conditions (37°C) for methane production (Kanchanasuta and Sillaparassamee, 2017). High energy-recovery efficiency was achieved. Notably, methanogenic archaea are sensitive to the temperature, and the process may fail when the temperature change exceeds 2–3°C (Zhang et al., 2020).

3.3. Substrate

Carbohydrates, proteins, and lipids are the main components of FW and function as the main substrates in the AD process. The different types of substrates can be listed in descending order of their degradation rates as sugars (monosaccharides and disaccharides), starch, proteins, hemicellulose, lignin, waxes, and greases (Cremonez et al., 2021). Carbohydrates, which consist of easily fermentable sugars, can yield biohydrogen in a short period in the acidification phase, fol-

lowed by large amounts of biomethane production. In contrast, residues composed of lignocellulosic carbohydrates exhibit low degradability and must be pre-treated before the AD process (Jain et al., 2015). Additionally, proteins are not regarded as ideal substrates for TSAD because of their inhibitory effects due to production of toxic compounds such as ammonia (Braun et al., 2003; Ma et al., 2018). Lipids have the highest potential for biogas production amongst all the compounds and can be gradually transformed into solid fatty acids during the process. However, the accumulation of fatty acids and slow degradation of lipids in AD can adversely influence the AD system. First, the accumulation of macromolecular fatty acids can decrease the pH. Because FW contains high concentrations of lipids that degrade slowly, blocks may be generated by the accumulation and conversion of fatty acids. These blocks can surround the microorganisms, making it difficult for them to contact the degradable feed stock, thereby decreasing the biogas production. Moreover, energy conversion, extensive substance transfer, and information transmission occur between methanogens and hydrogen-producing bacteria when they are in direct contact. The limited contact between microorganisms owing to the lipids can inhibit the AD (Yue et al., 2020). Therefore, pre-treatments should be introduced to accelerate the process.

Moreover, nutrient element compositions, such as those of carbon, nitrogen, oxygen, and hydrogen, considerably influence the AD performance. Notably, the optimal C/N ratios differ across studies, and various ranges such as 20–35 (Khalid et al., 2011) and 15–30 (Neshat et al., 2017) have been reported. Extremely high C/N ratios may limit the inoculum renewal and new cell formation. Extremely low C/N ratios may lead to the presence of excessive nitrogen in the form of ammonium, which is toxic to the microbes (Esposito et al., 2012; Rocamora et al., 2020). The C/N ratio of mono FW as the substrate is typically high. Therefore, the co-digestion of FW and other organic wastes has been recommended to balance the C/N ratio (Zheng et al., 2021).

The metal elements in substrates influence the TSAD process of FW. Metal elements can be divided into light metals (i.e., sodium, potassium, and calcium) and heavy metals (i.e., copper, nickel, zinc, and cobalt), and some of them can facilitate the active functioning of microbes and serve as cofactors for enzyme activity and synthesis (Cirne et al., 2007; Silva et al., 2018). For example, magnesium is an important component of cellular walls and membranes, which are needed for building cellular protein and act as activator and cofactor of many enzymes (Bundhoo and Mohee, 2016). Sodium at low concentrations works as a micronutrient which is needed for bacterial growth plays a crucial role in building Na-K-ATP pump which transports substrate into cells (Blaustein et al., 2020). Calcium can enhance bacterial growth, strengthen microorganisms and cell aggregation (Tang et al., 2022). Nickel is crucial in forming [Ni-Fe]-hydrogenases which plays an important role in converting formate into biohydrogen (Salazar-Batres and Moreno-Andrade, 2022). Moreover, iron has been widely introduced in AD processes in various forms such as Fe²⁺ and Fe_xO_y nanoparticles (Chen et al., 2021). Pyruvate is the main intermediate product in a dark fermentation system and can be anaerobically oxidised to acetyl coenzyme A (acetyl-CoA) through the formate lyase (Pfl) pathway

or ferredoxin oxido-reductase (Pfor) pathway (Holladay et al., 2009). Ferredoxins containing iron and inorganic sulphur can assist in transporting electrons for the oxidation of pyruvate to acetyl-CoA under the Pfor pathway, in which CoA and ferredoxin oxidase oxidise pyruvate into acetyl-CoA, ferredoxin reductase, and CO₂ (Hallenbeck and Benemann, 2002). Iron can also promote bacterial growth by facilitating the biosynthesis of enzymes such as hydrogenases and reducing the inhibition caused by sulphides (Bundhoo and Mohee, 2016). However, when the concentration of these metals in the TSAD process is beyond certain thresholds, process failure may occur (Chen et al., 2014). Zhao et al. (2020) studied the dosing effect of nano zero-valent iron (NZVI) on dark fermentation from co-digestion of lake algae and FW, and reported that the addition of 10 mg/g TS of NZVI improved the hydrogen yield from 30.99 to 40.04 mL/g VS, whereas the addition of 20 and 40 mL/g VS decreased the hydrogen yield to 14.77 and 5.37 mL/g VS, respectively. Several strategies have been proposed to alleviate these inhibitory effects. First, the inhibition threshold must be determined, which varies with several factors such as the substrates fermented, inoculum used, and operating conditions (Ho and Ho, 2012; Wang and Wan, 2009). The reactor contents can be diluted to concentrations below the thresholds to minimise inhibition. Another useful technique is the precipitation of heavy metals (Jose Leal-Gutierrez et al., 2021). Activated carbon and organic ligands can be used as the adsorbent and chelating agent, respectively, for the removal of heavy metals (Chen et al., 2008).

3.4. OLR

The OLR significantly affects the stability of TSAD by influencing the stability of VFA production. By ensuring a proper OLR, adequate carbon can be provided (Wainaina et al., 2019). In general, a high OLR represents a high treatment ability of the system. However, extremely high OLRs may lead to the rapid formation of metabolic intermediates that cannot be consumed in time, resulting in VFA accumulation, which can cause process instability (Nagao et al., 2012). A high OLR is more harmful to the methanogenic phase than to the acidogenic phase, and thus, the optimal OLR range is different in the two stages of the TSAD process (Lay et al., 2019; Porpatham et al., 2007). In other words, the acidogenic stage can be implemented at a higher OLR than the methanogenic phase. Baldi et al. (2019) operated a TSAD system using FW as the substrate with OLRs of 14.2 and 2.5 kg TVS/(kg-day) for the first and second stages, respectively, and the hydrogen and methane yields were 12.6 and 482.1 L/(kg TVS-day), respectively, and the biohythane production was enhanced. Notably, in the literature, the OLR is typically presented in inconsistent units such as kg COD/(m³·day) or kg VS/(m³·day), and it is difficult to standardise the units because of the lack of conversion factors. Moreover, the optimal OLRs vary across studies owing to the different process conditions, and there is no consensus on the ideal range. For the first stage of TSAD, the OLR values have been reported to be 20–64 kg COD/(m³·day) or 8–38 kg VS/(m³·day) (De Gioannis et al., 2013). In the second stage, the apparent viscosity of the fermentation medium may increase when the OLR is higher than 5 g TS/(L·day) in

mesophilic conditions, thereby limiting the heat and mass transfers (Wainaina et al., 2019).

3.5. Retention time

Both the solid retention time (SRT) and the HRT are key parameters in biological treatment processes. The SRT is defined as the average time the biomass remains in the digester, whereas the HRT is defined as the average time the substrate remains in the digester (Chatterjee and Mazumder, 2019). In general, the SRT is longer than the HRT, although the two values are equal in a continuous stirred-tank reactor (CSTR) (Santiago et al., 2019). The HRT is calculated as the ratio of the volume of the reactor to the daily feed flow rate. The process operation conditions (especially the temperature) and type of substrates influence the HRT. The retention time in thermophilic conditions is shorter than that in mesophilic conditions, and thus, the AD performance is superior in thermophilic conditions. Additionally, the HRT values for the two stages of TSAD are different, usually 2–4 days and 12–16 days for the acidogenic and methanogenic stages, respectively. Owing to the shorter retention time, the TSAD process is generally more efficient than SSAD, which typically has an HRT of 18–30 days (Rajendran et al., 2020; Srisowmeya et al., 2020).

3.6. Inoculum

The appropriate inoculum should be selected to ensure the performance of the TSAD of FW (Cremonez et al., 2021). The TSAD process typically involves two types of inoculum. The dominant microorganisms in the two stages of TSAD are different (typically, *Thermoanaerobacterium* sp. for acidogenesis, and *Methanoculleus* sp. and *Methanosarcina* sp. for methanogenesis) because of the different operating parameters in the two stages (O-Thong et al., 2016). Additionally, inoculum can be obtained from the fermentation system in either pure or mixed form. The ability of pure cultures to produce hydrogen and methane has been extensively studied, and they have been noted to be effective in generating specific products. For example, *Clostridium* and *Enterobacter* are widely used for hydrogen production (Dinesh et al., 2018). Although pure cultures present a higher rate of biogas generation than mixed microbial species (Mamimin et al., 2019), mixed cultures exhibit several advantages. For example, sterile operating conditions are not required, there is no dependence on the strain of a specific microorganism, the microorganisms can consume a great diversity of substrates, and they can be obtained at low cost (Kleerebezem and van Loosdrecht, 2007; Lu et al., 2011; Wainaina et al., 2019). Consequently, mixed cultures are more suitable for the engineering application of AD systems. Mixed cultures contain anaerobic sludge, organic compost, and bovine manure (Bakonyi et al., 2014; Soares et al., 2020). Amongst mixed cultures, the sludge from treatment ponds and digesters for degrading residues having similar characteristics can be easily adapted as the inoculum to enhance the efficiency of AD systems (Jeihanipour et al., 2011; Yang et al., 2015). However, the inoculum may contain inhibitors, i.e., hydrogen-consuming bacteria that can limit the hydrogen production in the first stage of TSAD. Therefore, the

inoculum for the first stage of TSAD must be pre-treated to eliminate the inhibitors (Kan, 2013).

A suitable substrate-to-inoculum ratio (S/I) must be selected to buffer the capacity, control the accumulation of VFAs, and increase the digester stability. The optimal S/I values for the two stages are different. S/I ratios (based on the VS) of 7–10 are preferable for producing H₂, although the corresponding values for CH₄ production are much lower (Yun et al., 2018). Cappai et al. (2018) evaluated the influence of the S/I ratio on dark fermentation and demonstrated that the H₂ yield was maximised when S/I was 7.14. Additionally, recirculation influences the inoculum. The two-stage recirculation of the methanogenic reactor effluent can favour the growth of bacteria in the first reactor and archaea in the methanogenic reactor, thereby contributing to the hydrogen production (O-Thong et al., 2016).

3.7. Reactor

The types of bioreactors influence the growth and activity of microorganism. CSTRs have substrate blending systems, which can ensure a high level of mixing. In batch or continuous conditions, such reactors are preferable for the AD of FW as they can manage wastes with high solid loads. Consequently, CSTRs represent the most widely used reactor configuration in TSAD processes (Gianico et al., 2015). An up-flow anaerobic sludge reactor (UASB) consists of a feeding system in the lower part. The substrate flows upward and passes through a sludge blanket that contains a suspended bed and granules. UASBs exhibit several advantages such as a high OLR and low HRT and are thus often used in the second stage of TSAD (Li et al., 2020a; Tauseef et al., 2013). However, UASBs cannot effectively treat organic wastes with high solid loads (Cremonez et al., 2021). Several other reactor models are available, such as anaerobic packed-bed reactors, anaerobic baffled reactors, expanded granular sludge blanket (EGSB) reactors, leachate bed reactors, internal circulation (IC) reactors, or sequential batch reactors (SBRs) (Cremonez et al., 2021; Dinh Pham et al., 2020). The reactor should be selected based on the application requirements.

4. Research on biohythane production from FW through TSAD

4.1. Research status

Biohythane, a mixture of biohydrogen and biomethane with specific ratios, has gained interest as an alternative renewable energy source to reduce carbon monoxide emissions. For example, in the USA, biohythane is being used by many automotive manufacturing companies (Rena et al., 2020). The TSAD of FW is a promising route for biohythane production, and many researchers have focused on evaluating the corresponding potential of TSAD systems, for instance, by analysing the factors affecting the system stability, eliminating the inhibitors of the system efficiency, and formulating strategies to enhance the biohythane yield.

First, because optimal conditions can be provided to the TSAD systems for producing both biohydrogen and

biomethane, the energy efficiency of TSAD is higher than that of traditional AD. Alexis Parra-Orobio et al. (2020) compared the energy balance and carbon dioxide emissions associated with the TSAD and SSAD of FW, and the results showed that the energy efficiency of TSAD was 57.5% higher than that of SSAD. Second, several factors affect the stability of TSAD of FW, such as pH value, temperature, substrate type, and OLR. The optimal operational parameters are summarised as follows. The ideal pH range is 5.2–6.5 and 6.8–7.8 for the first and second stages of TSAD, respectively (Chatterjee and Mazumder, 2019; Elreedy et al., 2015; Li et al., 2013). In terms of the temperature, thermophilic and mesophilic conditions are preferred for the first and second stages, respectively (Chen and Chang, 2020; Li et al., 2020b; Wang et al., 2018b). In terms of the substrates, FW rich in carbohydrates is suitable for biohydrogen production in the first stage (Cremonez et al., 2021). The first stage is typically operated at a higher OLR than the second stage (Lay et al., 2019; Porpatham et al., 2007). The ideal HRT values for the first and second stages are 2–4 and 12–16 days, respectively (Srisowmeya et al., 2020). In terms of the inoculum, S/I ratios (on a VS basis) of 7–10 are preferable for producing H₂, and those for CH₄ production are considerably lower (Yun et al., 2018). Several reactor models (e.g., IC, UASB, and EGSB) are available for implementing TSAD processes (Dinh Pham et al., 2020), although the CSTR configuration is the most widely used (Gianico et al., 2015). Third, the inhibitors in TSAD systems have attracted considerable attention. The inhibitors can be divided into pre-process and in-process inhibitors. Pre-process inhibitors consist of inhibitors in mixed microflora, metal ions, and inhibitors from substrate pre-treatment such as furan derivatives and phenolic compounds. In-process inhibitors consist of ammonia, intermediate products, and end products (Bundhoo and Moehee, 2016). Many researchers have examined the mechanisms through which these inhibitors affect the TSAD process and recommended strategies for eliminating or alleviating the inhibitory effects. For example, Hou et al. (2021) studied the mechanism by which high salinity inhibited H₂ and CH₄ production from the TSAD of FW in different salt concentrations and examined the potential of air-nanobubble water addition in mitigating the inhibition, and the results showed that the two stages were severely inhibited when the salt concentrations were more than 20 g NaCl/L, because high salinity inhibited the growth of microorganisms by dehydration or increase in the osmotic pressure (Hou et al., 2021; Tsapekos et al., 2019). The addition of air-nanobubble water could enhance α -glucosidase, protease, and coenzyme F₄₂₀ activities and promote microbial electron transfer, thereby enhancing the performance of TSAD of FW in highly saline conditions (Hou et al., 2021). Fourth, many researchers have attempted to enhance the biohythane production through strategies such as pre-treatment, co-digestion, and regulation of microbial consortia. These methods are discussed in Section 4.2.

The economic aspects of the TSAD have been studied, with estimations based on the total capital investment, which consists of the working capital cost and fixed capital cost (Jarunglumert et al., 2018). Then, annual revenue and the annual production cost can be calculated after the optimal equipment type running in the best operating condition is selected (Jarunglumert et al., 2018). In addition, the profitability

performance indicators including the net present value, internal rate of return, and payback period are also needed to be determined (Lembo et al., 2022). Moreover, the profitability and economic feasibility of TSAD in terms of the biohythane production have been examined by analysing crucial factors such as the feedstock, process type, process scale, and storage costs of biohythane (Hans and Kumar, 2019). The results highlight that although FW as a substrate may be expensive owing to the pre-treatment, transportation, and microbial strain selection, TSAD of FW is a sustainable approach to attain higher economic profits with reduced environmental effects (Dangol et al., 2022; Nielsen et al., 2017; Yun et al., 2018). Several researchers have compared the economic performances of TSAD and traditional AD. For example, Yu et al. (2020) comparatively assessed the full-scale SSAD and TSAD of FW and reported that the economic benefits of TSAD were slightly superior. Lembo et al. (2022) studied the economic performances and greenhouse gas emissions of second cheese whey of SSAD and TSAD, and the results showed that the TSAD system exhibited a higher energy output and reduced 60% of greenhouse gas emissions than that of the SSAD system, and a detailed cost analysis also confirmed the TSAD was more economically viable with a payback period of 9 years, which was 3 years shorter than that of SSAD. Nevertheless, research on the economic aspects of TSAD, especially over a large scale, is limited, and the benefits must be systematically studied.

4.2. Strategies for enhancing biohythane production from TSAD of FW

4.2.1. Pre-treatment

The pre-treatment methods for FW can be divided into physical, mechanical, biological, and combination methods (Raj et al., 2022; Ren et al., 2018), as summarised in Table 2 along with the corresponding hydrogen and methane yields. Each method has advantages and disadvantages. For example, although pre-treatment methods such as ultrasound, microwave, hydrodynamic cavitation, autoclave, pulsed electric field, and pyrolysis based techniques can be rapidly implemented, they are energy intensive and generate free radicals (Rawoof et al., 2021). Table 2 indicates that hydrothermal pre-treatment is a promising technique. Jia et al. (2017) concluded that short-term hydrothermal pre-treatment is a green and feasible technique for bio-gasification from TSAD of FW. These findings were supported by those of Ding et al. (2017).

4.2.2. Co-digestion

Co-digestion techniques can balance the nutrients in FW and dilute the toxic substances, thereby improving the biohydrogen yield and energy recovery (Xu et al., 2018). Additionally, co-digestion can help maintain the pH in the TSAD system without using additional chemicals. The digestion time can be shortened by implementing the co-digestion of animal waste, phytomass, crop residues, and sewage sludge (Sole-Bundo et al., 2019). Table 3 summarises the hydrogen and methane yields associated with the co-digestion of FW and other organic wastes through TSAD. Zhu et al. (2022) studied the effects of co-digesting corn straw, FW, and chicken manure in TSAD processes on the microbial community composition and bioavailability of trace elements, and the results

showed enhanced abundance of obligate hydrogenotrophic methanogens and higher range of bioavailability of trace elements compared with those in the digestion of the mono-substrate.

4.2.3. Regulation of microbial consortia

The microbial communication can be regulated to enhance the efficiency of biohythane production from TSAD (Lin and Lu, 2021). The native microbial species in the TSAD system of FW are typically ineffective in generating biohythane, but their abilities can be improved through gene manipulation, gene inactivation, genetic recombination, and metabolic engineering (Salakkam et al., 2021). For example, Mario Lopez-Hidalgo et al. (2021) studied the effect of using genetically engineered *Escherichia coli* for hydrogen and ethanol co-production and noted that 30% higher yields could be obtained. Song et al. (2017) used four kinds of genetically modified bacteria including *Enterobacter cloacae/HycE*, *Enterobacter cloacae/HPP*, *Enterobacter cloacae/HoxEEUYH*, and *Enterobacter cloacae/HycG* to produce hydrogen via fermentation from pretreated water hyacinth, and they found that *Enterobacter cloacae/HPP* performed best in producing hydrogen among the four genetically modified bacteria. The highest hydrogen yield was 74.9 mL/g VS, and 50% higher than that of using the wild strain from pretreated water hyacinth as substrate (Song et al., 2017). Moreover, the effects of the strain behaviours on biohythane production must be clarified to design metabolically engineered strains, because the types of fermentation pathways depend on the strain characteristics (Zhou et al., 2018). In the TSAD process, several fermentation pathways (i.e., lactic acid pathway and propionic acid pathway) are not desirable as they produce by-products such as lactic and propionic acids that negatively impact the biohythane production (Bundhoo and Mohee, 2016). By blocking these undesirable pathways and introducing metabolically engineered strains, the biohythane generation can be enhanced. Bioaugmentation is another potential technique to promote the TSAD of FW by regulating the microbial communication. Representative strategies include inducing a strain with an adequate enzymatic arsenal and providing nutrients exogenously (Ishaq and Dincer, 2021). Bioaugmentation eliminates the need for pre-treating substrates and can inhibit VFA accumulation (Rawoof et al., 2021; Shanmugam et al., 2020). Nkemka et al. (2015) evaluated bioaugmentation with an anaerobic fungus called *Piromyces rhizinflata* YM600 in the TSAD of corn silage and cattail, and concluded that bioaugmentation with anaerobic fungus could increase hydrogen and methane production rates. Xie et al. (2021) explored the bioaugmentation of activated carbon and rumen cellulolytic bacteria in thermophilic digestion using cornstalk as the substrate, and the results showed that the methane production was improved when activated carbon and rumen cellulolytic bacteria were added, and the methane yield was maximised (144.9 L/kg VS) when both activated carbon (10 g/L) and rumen cellulolytic bacteria (30 mL) were introduced.

5. Challenges and future considerations

The production of biohythane from TSAD of FW has emerged as a research hotspot over the past decade (Rajendran et al.,

Table 2 – Hydrogen and methane yields associated with different food waste pre-treatment methods in two-stage anaerobic digestion.

Substrate pre-treatment	Substrates	Biohythane yield		Refs.
		Hydrogen yield	Methane yield	
Hydrothermal pre-treatment	Food waste and <i>Enteromorpha</i>	62.7 mL/g VS	590.6 mL/g VS	Wei et al. (2022)
Short-term hydrothermal pre-treatment	Food waste	–	–	Jia et al. (2017)
Hydrothermal pre-treatment	Food waste	43.0 mL/g VS	511.6 mL/g VS	Ding et al. (2017)
Hydrothermal pre-treatment	Source-separated organics	–	314 mL/g TCOD _{added}	Azizi et al. (2019)
No pre-treatment	Kitchen waste	–	109.1 L/(L-ton waste)	Shahriari et al. (2013)
Microwave pre-treatment			114.0 L/(L-ton waste)	
No pre-treatment	Vinegar residue and anaerobic sludge	23.8 mL/g VS	38 mL/g VS	Wang et al. (2015)
HCl pre-treatment		53.2 mL/g VS	192 mL/g VS	
Oxalic acid pre-treatment		46.7 mL/g VS	178 mL/g VS	
No pre-treatment	Pulp and paper sludge and food waste	52.16 mL/g VS _{fed}	383.8 mL/g VS _{fed}	Lin et al. (2013)
NaOH pre-treatment		78.35 mL/g VS _{fed}	90.41 mL/g VS _{fed}	
H ₂ SO ₄ pre-treatment		70.7 mL/g VS _{fed}	105.7 mL/g VS _{fed}	
In situ mild alkaline (pH 8) pre-treatment	Waste cooking oil and sewage sludge	–	0.71 L/g VS _{feed}	Yan et al. (2021)
N ₂ -and-air-nanobubble water supplementation	Food waste	27.31 mL/g VS _{added}	373.63 mL/g VS _{added}	Hou et al. (2021)
No pre-treatment	Glycerol trioleate and food waste	0.60 mL/g TVS	520.37 mL/g TVS	Yue et al. (2020)
Ozonation pre-treatment		22.55 mL/g TVS	946.45 mL/g TVS	

–: not available.

Table 3 – Hydrogen and methane yields from co-digestion of food waste and various waste materials through two-stage anaerobic digestion.

Substrate type	Anaerobic digestion	Biohythane yield		Hydrogen production rate	Methane production rate	Refs.
		Hydrogen yield	Methane yield			
Food waste, sewage sludge, and glycerol	Two-stage anaerobic digestion	140.2 mL/g VS	342 mL/g VS	17 mL/(g VS-hr)	21.7 mL/(g VS-day)	Silva et al. (2018)
Used disposable nappies and expired food products	Two-stage anaerobic digestion	1.93 L/L feed	14.26 L/L feed	–	–	Tsigkou et al. (2020)
Food waste and waste activated sludge	Two-stage anaerobic digestion	42.0 mL/g VS _{added}	329.8 mL/g VS _{added}	2.3 mL/hr	1.6 mL/hr	Liu et al., 2021
Food and paper wastes	Two-stage anaerobic digestion	–	0.38 L/g VS _{added}	–	1.27 L/(L-day)	Xiao et al. (2019)
Food waste and brown water	Two-stage anaerobic digestion	99.8 mL/g VS _{added}	728 mL/g VS _{added}	3.5 L/day	24.6 L/day	Paudel et al. (2017)
Garden and food wastes	Two-stage anaerobic digestion	46.2 mL/g	181.8 L/kg	21.6 mL/(g-day)	–	Abreu et al. (2019)
Food waste		16.5 mL/g	276.8 L/kg	2.3 mL/(g-day)		
Food waste, corn straw, and chicken manure	Two-stage anaerobic digestion	–	–	3.2 mL/hr	217.3 mL/day	Zhu et al. (2022)
Food waste and cow dung	Two-stage anaerobic digestion with digestate recirculation	–	–	3.3 L/(L-day)	3.1 L/(L-day)	Wang et al. (2020)

–: not available.

2020). The following limitations must be addressed to improve its capabilities and promote widespread use. First, the process parameters are challenging to optimise (Nabaterega et al., 2021). The key parameters such as the pH, temperature, OLR, and HRT vary between the two stages and significantly influence the microorganism communities. These parameters must be optimised to ensure a stable and efficient process. Owing to the interactions between parameters, a change in one or more of the parameters may lead to variations in the others. Therefore, the optimal parameters are inconsistent in different conditions and often vary over time. It is thus challenging to identify the optimal parameters of a particular system and maintain them in long-term operation. Second, the directional metabolic pathways of functional microorganisms in the TSAD of FW must be regulated. In the TSAD process, several pathways that produce by-products such as lactic and propionic acids, which negatively impact the biohythane production, are not desirable and must be blocked (Bundhoo and Mohee, 2016). Ethanol and butyric acid metabolic pathways are typically prevalent in the first stage of TSAD (Cao et al., 2022; Zhang et al., 2017). However, the accumulation of ethanol and VFAs may lead to increased toxicity and reduced pH, respectively (Zhou et al., 2018). It remains challenging to minimise the production of undesirable acids and maximise the yield of the target product in the second stage of the TSAD (Qureshi and Maddox, 2005). Therefore, methods to inhibit the by-products and enhance the metabolic pathways of the target product must be identified. Third, the reactors for the TSAD of FW must be able to achieve cooperative control of the first and second stages. Therefore, the reactors for full-scale TSAD must be equipped with intelligent real-time control and monitoring systems. Future studies may focus on the development of new reactors and the introduction of artificial intelligence technologies to TSAD systems. Fourth, novel methods for eliminating the main inhibitors in the TSAD of FW must be identified. The inhibitors (i.e., pre-process and in-process inhibitors) represent bottlenecks of TSAD systems, and their inhibition mechanisms must be systematically clarified. Fifth, several challenges remain in gas purification, storage, and transport. Gas purification methods such as chemical absorption, pressure-swing adsorption, purification through water or solvent scrubbing, cryogenic purification, and membrane separation rely on sophisticated equipment and are expensive to implement (Sasidhar et al., 2022). Additionally, the stage separation and management of pipelines carrying biohythane are expensive and limit the scaling up of TSAD systems.

To enhance the biohythane yield, future research must focus on the following aspects: (1) The adopted techniques must be widely applicable from lab-scale to full-scale, and biohythane must be commercialised. (2) Innovative low-carbon technologies must be developed to enhance the biohythane yield from the TSAD of FW in terms of the pre-treatment, co-digestion, and regulation of microbial consortia. (3) Bioreactors that can withstand the fluctuations in process parameters must be developed, and strategies to control and monitor the TSAD systems must be further explored. (4) The characteristics and biokinetics of microbial communities must be examined. (5) Because biohydrogen and biomethane are produced separately, techniques for gas purification and their *in*

situ blending into biohythane can be explored, and the storage of biohythane must be studied (Sasidhar et al., 2022). (6) Comprehensive techno-economic and lifecycle analyses must be performed for large-scale TSAD of FW (D'Silva et al., 2021; Sun et al., 2019). (7) Policies must be introduced to promote the use of biohythane as an energy source and expand its market share, such as pushing forward the implementation of biohythane energy industrial projects, providing investment subsidies for the use of biohythane production technology and assisting in the formation of supply-chain of biohythane.

6. Conclusions

The main parameters (e.g., pH values, temperature, and OLR) affecting the TSAD of FW are discussed. The optimal parameters of the first and second stages are considerably different. TSAD systems can simultaneously provide the optimal conditions for the two groups of microorganisms in two stages and stably produce biohythane. The existing studies and economic evaluations of TSAD-based FW treatment highlight the potential of TSAD in producing biohythane. Future studies may focus on the corresponding reaction mechanism, system enhancements, and economic analyses for large-scale TSAD systems. The performance of TSAD of FW can be enhanced by pre-treatment for hydrolysing FW, co-digestion of FW and other organic wastes to dilute inhibitors, supply of nutrients, synergism among inoculums, and regulation of microbial consortia by modifying or inducing new microorganisms. Nevertheless, several challenges remain to be addressed, in terms of identifying and sustaining optimal parameters; regulating the directional metabolic pathways; developing suitable reactors; eliminating the inhibitors; and purifying, storing, and transporting biohythane. Researchers must attempt to scale-up the system, develop innovative low-carbon technologies, combine specific bioreactors, formulate strategies for gas purification and *in situ* blending of biohydrogen and biomethane into biohythane, and clarify the characteristics and biokinetics of microbial communities. Furthermore, the construction costs, operating costs, payback, and feasibility of TSAD-oriented ventures for FW treatment must be comprehensively examined for large-scale frameworks, as the existing studies have mainly focused on economic considerations for lab- or pilot-scale systems.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2023.04.031.

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