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A review of high-solid anaerobic digestion (HSAD): From transport phenomena to process design

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ABSTRACT

High-solid anaerobic digestion (HSAD) is an attractive organic waste disposal method for bioenergy recovery and climate change mitigation. The development of HSAD is facing several challenges such as low biogas and methane yields, low reaction rates, and ease of process inhibition due to low mass diffusion and mixing limitations of the process. Therefore, the recent progress in HSAD is critically reviewed with a focus on transport phenomena and process modelling. Specifically, the work discusses hydrodynamic phenomena, biokinetic mechanisms, HSAD-specific reactor simulations, state-of-the-art multi-stage reactor designs, industrial ramifications, and key parameters that enable sustained operation of HSAD processes. Further research on novel materials such as bio-additives, adsorbents, and surfactants can augment HSAD process efficiency, while ensuring the stability. Additionally, a generic simulation tool is of urgent need to enable a better coupling between biokinetic phenomena, hydrodynamics, and heat and mass transfer that would warrant HSAD process scale-up.

1. Introduction

Sustainable management of municipal solid waste (MSW), sewage sludge, and food waste (FW) are of significant economic and

environmental consequences. The waste can be disposed of using various thermochemical and biochemical technologies including incineration, pyrolysis, gasification, composting, landfill, and anaerobic digestion (AD) [1,2], among which landfill remains one of the dominant

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ways. For example, landfill accounted for 61.4% of waste disposal in China, followed by incineration (15.84%) in 2011 [3]. In the USA, 54% of the MSW generated was landfilled, with recycling and composting only accounting for about 33%. Despite such features as simple operation and low cost, landfill suffers from various drawbacks, such as large land-area requirement, low energy recovery, and significant secondary pollution, which calls for the increasing exploration and exploitation of alternative technologies.

AD is a biochemical process in which microorganisms decompose and convert organic matter synergistically in an oxygen-free environment producing biogas and biosolids. Biogas is a mixture of methane (CH_4), carbon dioxide (CO_2), and other gases, such as hydrogen sulfide (H_2S) and ammonia (NH_3) [4]. AD has been considered one of the most sustainable, cost-effective technologies for energy recovery from organic wastes that are commonly featured by high moisture contents and make typical thermochemical approaches (e.g., gasification and pyrolysis) less appropriate [5]. Depending on the content of total solids (TS), AD can be divided into wet (<15% TS) and dry (\geq 15% TS) processes [6].

Dry digestion, also called solid-state anaerobic digestion (SSAD) or high-solids anaerobic digestion (HSAD), is receiving increasing attention because of various strengths such as lower waste handling volume and consequently, reduced waste transportation emissions, lower water requirement requiring smaller digester sizes, a higher flexibility for batch systems, and a higher organic loading rate (OLR) [7]. These features improve the biogas (or CH_4) generation capacity of HSAD while reducing the total cost of ownership (TCO). However, the application of HSAD is still limited due to several major drawbacks such as relatively low heat and mass transfer rates (i.e., long degradation time), and frequent process inhibition caused by the accumulation of toxic and inhibitory compounds (e.g., volatile fatty acids (VFAs) and NH_3) [8]. Indeed, HSAD processes are typically featured by a slow release of hydrolysed substances for further microbial conversion, low accessibility of substrates to the microbial community, and the dispersion of inhibitory compounds [9]. This poses significant challenges against their practical scale-up and industrialization. It is critical to understand and improve the heat and mass transfer of HSAD for process enhancement.

Recently, there has been a surge in the number of reviews relating to the technical principles and improvement methods of HSAD processes. For example, researchers have reviewed HSAD of crop waste [10] and animal manure [11], influence of scale of operation [12], feedstock rheological properties [13] and pre-treatment methods [14], benefits of co-digestion [13], the influence of process parameters on HSAD stability [15], and techno-economic benefits of HSAD [8]. However, these articles do not critically discuss the HSAD process design improvements from the viewpoint of thermo-hydraulic aspects and heat and mass transport phenomena. Such a review will be of great value for guiding the design of research to mitigate the issues caused by limited heat and mass transfer during HSAD processes. Hence, this review aims to fill this gap by presenting a detailed summary of existing research on the mass and heat transfer phenomena of HSAD, focusing on mechanisms, modelling and simulation, bioreactor design, and process optimization of HSAD.

2. Background

The AD process generally includes four steps, i.e., hydrolysis, acidogenesis, acetogenesis, and methanogenesis as shown in Fig. 1 [16]. In hydrolysis, large molecular compounds, such as protein, carbohydrate, and fat are broken into small molecules. In the acidogenesis step, long-chain fatty acids are converted to short-chain VFAs, while in the acetogenesis step, acetate, carbon dioxide, and/or hydrogen are formed via the fermentation of the VFAs and other products. In the last step, the produced acetate, carbon dioxide, and/or hydrogen are converted to CH_4 by methanogens.

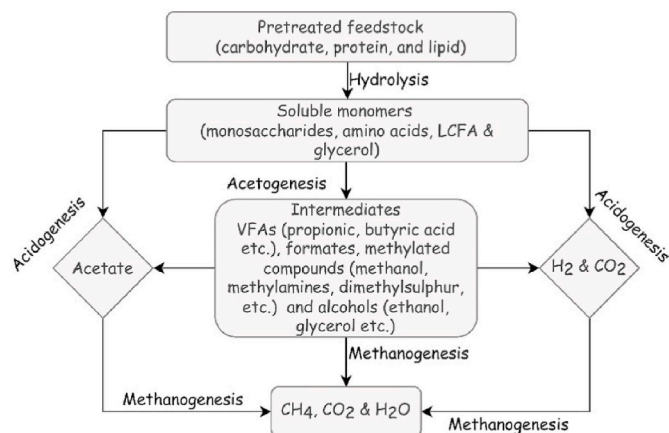


Fig. 1. Generalized schematic showing stages involved in anaerobic digestion; LCFA – Long chain fatty acids, VFAs – Volatile fatty acids. Reproduced with permission from Ref. [16].

2.1. Importance of mass transfer

Mass transfer results from either advection or diffusion of species due to a concentration gradient. Because of the complexity of the AD process, the mass transfer depends on a variety of factors such as inoculum, nature of substrate, temperature, moisture content, mixing efficiency, etc. Compared to wet AD processes, HSAD processes experience greater technical difficulty because of limited mass transfer. It is essential to achieve sufficient mass transfer to promote the interaction between biodegradable organic matter and microorganisms and to improve the overall process efficiency [17].

An HSAD system contains solid, liquid, and gas phases (see Fig. 2) corresponding to a solid substrate, seeding inoculum, water, biogas, and non-condensable gases (N_2 , CO_2 , NH_3 , and H_2S). The organic solid substrate is discontinuous and exhibits the basic characteristics of a porous media. The substrate's size, shape, and porous structure determine the void space while the gases in pores constitute the continuous gas phase. The accessible surface area for microbial growth and thus solid substrate utilization are affected by the porosity of the materials [18]. Hence, the mass diffusion of HSAD is synergistic with the porous media theory.

HSAD is generally initiated by a seed inoculum followed by the development of a multi-zoned reaction front which gradually advances until the stabilization of the whole waste mass [19]. A key determinant of this front zone mechanism is the minimum viable size for a seed body, which depends on the thickness of reaction zones. In the front zone, VFAs are produced, and diffuse to a methanogenic zone, with a passive buffer zone to mitigate the potential effect of organic acids inhibition [20,21]. Biomass digestion goes on as the front advances, which suggests that good inter-particle contact is beneficial for the process. The amount of inoculum and seeding patterns can determine the distance of the fronts and relevant process kinetics.

2.2. Importance of heat transfer

Heat transfer refers to the flow of thermal energy driven by a non-equilibrium and non-uniform temperature field. In an HSAD process, heat transfer is affected by various factors: (a) microbial inoculum (e.g., temperature, microbial physiology, specific growth rates, metabolic heat generation rates, etc.), (b) substrate morphology (e.g., particle size, shape, bed porosity, etc.), (c) equipment dimension (diameter, length, and internal device), and (d) operational conditions (inoculum to biomass ratio, pH, temperature, and humidity of the gas) [22–24]. A representative schematic for the heat transfer associated with a continuously stirred tank reactor (CSTR) is shown in Fig. 2. It illustrates

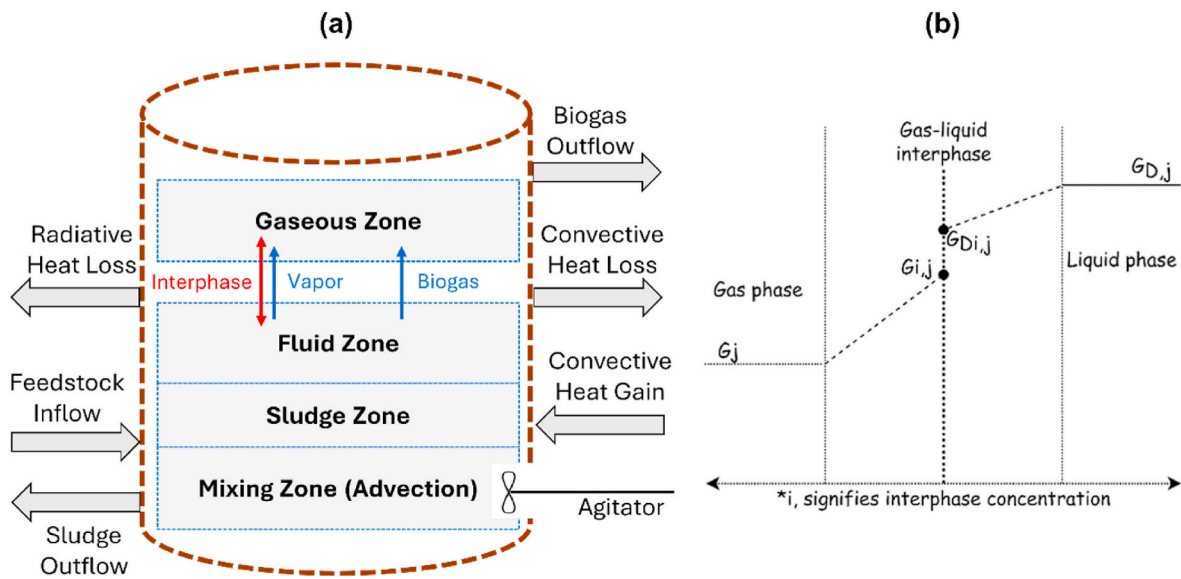


Fig. 2. (a) Schematic of salient heat and mass transfer processes during anaerobic digestion within a CSTR. (b) Interphase mass transfer of biogas from a liquid substrate (e.g., waste stream), G_j and $G_{D,j}$ are the j th undissolved and dissolved biogas species. Reproduced with permission from Ref. [16]. The corresponding interphase for a CSTR is shown in the left diagram in red color.

the convective heat gained using water jackets wrapped around the CSTR and the associated heat loss (convective and radiative) through the reactor walls. The heat transfer in an HSAD system is also closely associated with the strength of its metabolic activities. Therefore, the substrates with low thermal conductivities tend to cause lower heat transfer rates. Mechanical agitation and aeration with inert gas are two important methods to enhance material mixing and ensure effective heat and mass transfer during the process [25]. For mechanical agitation, the diameter of the impeller critically affected the radial mixing, with the axial mixing dictated by the distance between impellers [26,27]. Flow velocities were closely related to the rotating speed and had a lesser impact on expanding the mixing range. There have been limited studies on the specific effects of the mixing methods on the productivity of HSAD, except a few [26,27].

3. Decisive factors affecting heat and mass transfer

Process intensification or optimization is an important method for developing smaller, cleaner, and more energy-efficient technologies [28]. The process optimization or intensification of HSAD can alleviate the adverse impacts of inhibitory compounds, improve the rates of mass and heat transfer, and enhance the biogas yield and feedstock degradation efficiency. Various influential factors such as input waste properties (both chemical and rheological), TS, the ratio of substrate to inoculum, adsorbents (or surfactants) addition, and agitation are closely related to the mass and heat transfer of HSAD processes. Several kinds of waste biomass including OFMSW, FW, agricultural waste, animal manure, or organic industrial wastes can be used as substrates for HSAD. As a result, the different types of waste vary widely regarding physico-chemical properties and associated digestion performance. Even for the same kind of waste biomass, the properties can significantly differ by particle size, shape, and moisture content. Accordingly, compositions of substrates, crystallinity, porosity, particle size, surface area, structural characteristics, and homogeneity also affect AD processes.

3.1. Feedstock particle size

The particle size of feedstock is critical for the microbial conversion of biomass by affecting the specific surface area, density, heat/mass transfer, and microbial growth. In a wet AD system, reducing particle

size generally improves the bio-conversion kinetics and digestion performance [29]. The reduction of biomass particle size serves as a pre-treatment method to reduce the dosage of water and chemical substances, improve homogeneity, and avoid clogging. The effects of particle size variation on biogas production for HSAD processes are still not well understood [7]. A reduction in the particle size can enlarge the specific surface area and improve biological contact and reactions, promoting biogas production, for the substrates with a high fibre content and low degradability [7]. The rate and mechanisms of hydrolysis in AD are influenced by particle size and composition. Hydrolysis of protein was sharply enhanced by 776% from 0.034 day^{-1} to 0.298 day^{-1} , when the protein particle size decreased from $500 \mu\text{m}$ to $50 \mu\text{m}$. In addition, the corresponding specific surface area increased from $0.01 \text{ m}^2 \text{ g}^{-1}$ to $0.19 \text{ m}^2 \text{ g}^{-1}$, while the maximum methane production rate increased by 133% [30]. The large specific surface area is closely related to the growth and activity of microorganisms. During an AD process, the microbial conversion starts from the surface, penetrating the particle's interior. The porosity distribution of biomass is crucial for liquid flow through porous solid substrate in HSAD. Using the Water Retention Curve (WRC) analysis, it was found that macro-, meso-, and micro-pores ranged from 33–63%, 25–44%, and 7–16% for cattle manure, roadside grass, and corn stover, respectively [31]. When cattle manure was treated in a leach bed reactor (LBR), macro-pore volume decreased from 30.4% to 1.7%, leading to a considerable reduction in permeability and an increase in the solid bed compaction. The process stability of HSAD can be improved by particle downsizing. Particle size reduction increased the mass transfer and improved the distribution of metabolites by enlarging surface area and facilitating the access of biomass and metabolites to microbes [32]. Another research work investigated the dynamic influence of particle sizes (0.1, 0.7, and 1.4 mm in diameters), and revealed that the particle size for HSAD became an important factor after the start-up phase [33].

Although fine milling improves the accessibility to the substrate and increases the conversion rate, it increases the risk of acidification because of rapid production and accumulation of VFAs [34]. Hence, it is not necessary to enhance HSAD performance using smaller waste biomass particles because of the production and accumulation of inhibitory compounds on the available surface area. For particle size in the range of 1.4–2.0 mm, the highest methane yield of sunflower oil cake achieved was $213 \pm 8 \text{ mL CH}_4 \text{ g}^{-1}$ volatile solid (VS), while, for particles

0.355–0.55 mm, the highest methane yield was $186 \pm 6 \text{ mL CH}_4 \text{ g}^{-1} \text{ VS}$ [35]. An optimum size could be different for different kinds of substrates, e.g., 0.6 mm for FW or 0.2–0.3 mm for wheat straw, depending on the biodegradability and inoculum to substrate ratio [36,37]. Since the conclusions on the effect of particle size on biogas and CH_4 production are conflicting in the literature [38], it is instructive to include particle size distribution of the input waste while developing mechanistic and data-driven models for HSAD processes. In this regard, consideration of particle-size-dependent dimensionless quantities such as Biot numbers for (a) heat transfer $Bi_{heat} = hL/\lambda$ and (b) mass transfer $Bi_{mass} = k_m D/L$ helps to identify the competitive significance of internal resistance to external resistance. The length scale L in the expression for Bi is taken as the particle size, λ and D are the thermal and mass diffusivities of solids, and h and k_m are the heat and mass transfer coefficients of solids, respectively.

3.2. Total solids in feedstock

Waste handling and pumping are unavoidable challenges for HSAD processes due to their high solid contents. The solid content strongly affects the diffusion and reaction kinetics, which is based on the theory that higher water content leads to faster the diffusion and reaction rates [39]. Prior experiments suggested that as the TS was increased, the viscosity showed an exponential increase [40], while the diffusivity coefficient showed an exponential decrease [41]. The reduced mass transfer led to the accumulation of inhibitory compounds which could lead to the deterioration and even breakdown of an HSAD process. Under semi-dry conditions, an increase in VFAs concentration was found with the increase of VS content, inhibiting methane production [42]. For a pH range of 5–7, VFAs are in their undissociated form, which is toxic for microorganisms [43].

The diffusion coefficient, an important indicator for mass transfer, has been well-studied in wet processes. However, it is difficult to characterize the coefficient in dry AD media using experimental methods. Compared to wet processes, the diffusion coefficient sharply decreases in an HSAD process because of diffusion-controlled kinetic degradation [44,45]. The performance of HSAD media was completely different from that of a biofilm reactor. The diffusion of solutes in HSAD was extremely slow in the absence of agitation. When TS was greater than 15% and agitation was poor, substrates derived from waste biomass would have to react and diffuse to become available to the inoculum for further degradation, limiting the reaction kinetics if the biomass and inoculum were not homogenized. The transfer limit promotes the accumulation in 'clusters' degradations zones (also referred to as dead zones), which leads to inhibition and reduces the overall microbial efficiency [46].

A high TS content affects the physical properties of the mixture of inoculum and waste biomass. It affects the rheological behaviour of digestate which is in the form of a viscoelastic material characterized by increasing yield stress levels with an increasing TS content. A prior work [47] studied the effects of TS concentration on the AD of cardboard using experiments in a batch reactor. The work found that the methane production slightly reduced as the TS concentration increased from 10% to 25% and TS = 30% represented a threshold level above which methanogenesis would be strongly inhibited. Using Anaerobic Digestion Model No. 1 (ADM1), a detailed model for biokinetic simulation of AD, the work also found that low methane production at high TS was attributed to mass transfer limitation. Another research [48] investigated the impacts of TS concentration on the performance (i.e., methane yield, biogas production, VS reduction, etc) of HSAD of FW and cattle manure. It was shown that optimum methane yields of 0.18 and 0.21 $\text{m}^3 \text{ CH}_4 \text{ kg}^{-1} \text{ VS}$ were achieved for FW at TS = 25%/HRT = 41 days and TS = 30%/HRT = 31 days, respectively. The mechanisms underlying the impact of solid concentration (3%–15%) on methane production through AD were investigated through dewatered sludge experiments. It was found that 6% TS represented a threshold value differentiating low-solid and high-solid with the accumulative methane yield

decreasing exponentially as TS increased from 6% to 15% [49]. This was closely associated with the efficiency of mass transfer and as TS increased, sludge viscosity increased exponentially while the diffusive coefficient decreased exponentially. The reduced mass transfer led to the accumulation of VFAs and free ammonia, deteriorating the performance of the AD process, which can change the dominant microbial communities and metabolic pathways. When increasing the solid content from 4.4% to 17.6%, the AD process changed from the acetoclastic pathway to methylotrophic methanogenic pathway due to the altering of the microbial community (methylotrophic methanogens dominated in HSAD with the abundance of 82.6%) [50].

3.3. Reactor operating temperature

The operating temperature of an HSAD reactor plays an important role in dictating (a) biological activity within the feedstock and (b) heat loss between the reactor and surroundings. Generally, there are three temperature ranges for operating AD processes, namely psychrophilic (10–30 °C), mesophilic (35–40 °C), and thermophilic (55–60 °C), among which the mesophilic and thermophilic conditions are widely used [51]. It is essential to select an operating temperature condition based on the type of feedstock (e.g., food waste, straw-derived, sewage sludge, or woody waste), since the reaction kinetics, reactor stability, effluent characteristics, and biogas yield depend on that. Specifically, the proximate and ultimate compositions of feedstock are often considered to determine suitable conditions for HSAD processes.

Since the HSAD process consists of a high solid content within the feedstock and is generally prone to poor mass transfer, an intuitive choice will be the thermophilic temperature range to enhance the degradation kinetics, ultimately resulting in higher methane production [52]. However, the heat loss between a thermophilic reactor to its surroundings is relatively sensitive to environmental condition fluctuations. Additionally, under a thermophilic operational scenario, the hydrolysis process of HSAD will be negatively affected, which would eventually increase the VFAs accumulation and decrease the process stability [53]. A recent statistical literature review revealed that the median of CH_4 yield for the thermophilic HSAD process is 275 m^3 per tonne VS, while that for the mesophilic condition is 220 m^3 per tonne VS (i.e., ~20% less than thermophilic) [51].

3.4. Reactor hydrodynamics

HSAD is featured by its long digestion time and poor organic removal rates due to the mass transfer limitation, which can be overcome by mechanical agitation-based reactor homogenization, forced leachate recirculation, or digestate recirculation [13]. The mass transfer for such processes can be characterized by the Sherwood number $Sh = f(Re, Sc)$, which represents the ratio of advective mass transport to diffusive mass transport [54]. Since Sh is directly dependent on Reynolds number Re and Schmidt number Sc (ratio of kinematic viscosity to mass diffusivity), changing the flow velocity (or agitator frequency) is a promising means to make advective mass transfer from a purely diffusive regime. The input waste with a high solid content is featured by its rheological properties such as high viscosity, shear-thinning, and thixotropy [40]. Consequently, the value of Sc for HSAD is significantly higher than wet AD, which significantly limits mass diffusion. For a fixed fraction of TS in the waste for an HSAD process, the Sc will be constant, which suggests that the only means for augmenting Sh is to increase the Re . The Re for mechanical agitator-based mixing for HSAD with non-Newtonian fluid is given by $Re = \rho N^{2-n} D^2 / K$ [27]. Here ρ is the density of the fluid in kg m^{-3} , N is the rotational frequency in s^{-1} , D is the diameter of the agitator (or impeller) in m, K is the consistency in Pa s^n , and n is the rheological flow index.

For a single-stage process, agitation using mixing devices such as a scraper and piston is necessary to ensure tight contact between the

inoculum and substrate. The diffusion coefficient of input waste decreased sharply when the TS content increased [41]. Different from OFMSW, yard and agricultural waste and sludge are sticky semi-solid with high viscosity and the mixing is difficult as TS is greater than 10% [55]. The influence of solid concentration could be interfered by agitation. A comprehensive list of HSAD processes with various mechanical agitation arrangements is provided in Table 1. However, for FW, improvement of mass transfer is not always efficient in AD reactors without phase separation. Mechanical agitation can increase the tolerance of HSAD to a high concentration of ammonia and lead to a good distribution of digestate. Otherwise, free ammonia aggregates in the micro-regions inside sticky biomass can create a microenvironment with a high concentration free ammonia [49].

The mixing or homogenization can be achieved by a nozzle for flushing slurry or gas recirculation [63]. Recirculation of liquid digestate or percolate could effectively improve the homogenization of nutrients and organics in the HSAD of waste, in addition to the positive effects in inoculum replenishing and inhibitory compound removal via a washing effect [11,64,65]. HSAD digestate can contain more un-fermented organic matter, nitrogen, phosphorus, and inoculum microorganisms due to limited heat and mass transfer as compared to normal AD, and thus HSAD digestate recirculation serves as a promising way for enhancing organic material utilization and extending the residence time of digestate towards higher methane production. As an example, a prior work [66] showed that increasing the digestate recirculation ratio from 50% to 60% for the HSAD of corn straw and cow dung increased the cumulative methane yield from 70 L to 116 L, and the biogas productivity reached 1.6 L L⁻¹ VS day⁻¹ for the case of 60% digestate recirculation ratio. The higher recirculation ratio enhanced the VadinBC27 wastewater-sludge group and *Methanobacterium* proliferation, reducing VFAs while improving process efficiency.

3.5. Adsorbent or surfactant addition

Adding porous materials or surfactants is another method to improve the mass transfer in an HSAD process. The enhancement of HSAD efficiency and relevant mechanisms via adsorbent or surfactant addition are summarized in Table 2. In addition, the properties and structure of extraction media can increase the mass transfer, especially for the hydrolysed products in the micropores of biomass.

Non-ionic surfactants can be used as a low-toxicity medium to improve the hydrolysis of biomass to enhance hydrogen production, by modifying the substrate structure and making it hydrophilic and more accessible to enzymes [67]. As a cost-effective method, iron could be added into a digester to enhance the AD of sludge without pre-treatment. Fe⁰ powder can be used to increase the methane yield by 14.46%. Compared with Fe⁰ powder, Fe scrap is more effective to improve the reaction rate (by 21.28%) due to its higher mass transfer efficiency with liquid and sludge. Among Fe in powder form, clean scrap, and rusty scrap, the rusty scrap could induce microbial Fe³⁺ reduction. This ultimately led to the highest methane yield (~29.51% improvement) and largest VSS reduction (48.27%). Rusty scrap could also enhance the diversity of aceto-bacteria and iron-reducing bacteria, improving the degradability of complex substrates [78]. The methane production rate can be improved by adding limonite, improving the degradation of soluble organic metabolites, and increasing the abundance of microorganisms associated with protein or amino acid decomposition [80]. Like Fe, the addition of Ni can increase methane production by increasing the abundance of archaea communities and improving metal bioavailability [82]. Porous adsorbents such as biochar and activated carbon can be used to enhance the HSAD process by changing the microbial community composition, accelerating the bioconversion of macromolecular, increasing the buffering capacity, improving the thin-layer diffusion effects to augment mass transfer, and promoting electron transfer efficiency [73–76]. For example, the addition of biochar into the HSAD of FW at a dosage of 25 g L⁻¹ considerably enhanced VFAs degradation and

Table 1

Summary of hydrodynamic experiments and CFD simulations for investigating mechanical agitation in HSAD.

No.	Aim	Contribution	Ref.
1	Computational Fluid Dynamics (CFD)-based comparison between A-310 and helical ribbon impellers	<ul style="list-style-type: none"> • Re > 100 affecting the growth of bio-structure • Helical ribbon design suitable due to the associated low shear and does not affect growth of bio-structure 	[56]
2	CFD-based comparison between six mechanical mixing impellers: helical ribbon, anchor, curtain-types, counterflow, modified high solidity (MHS), and pitched blade	<ul style="list-style-type: none"> • Ribbon, counterflow, and MHS impellers showing better flow-field uniformity • MHS impeller requiring lowest pumping power to homogenize manure slurry 	[57]
3	CFD assessment for HSAD of sewage sludge with dynamic sludge rheology	<ul style="list-style-type: none"> • Sludge yield stress influencing HSAD mixing pattern • Sludge age affecting torque and power required to drive the agitator 	[58]
4	Hydrodynamic measurement of HSAD flow field using particle image velocimetry (PIV)	<ul style="list-style-type: none"> • Higher rotation speeds not significantly affecting the mixing area • Multiple impellers recommended for high mixing efficiency 	[59]
5	CFD simulation to compare between double-blade and ribbon impeller	<ul style="list-style-type: none"> • Ribbon impeller having 18 times shorter mixing period than double blade impeller • Ribbon impeller intensifying the overall HSAD process efficiency 	[60]
6	Simultaneous CFD and PIV measurements of double stage, three stage and ribbon impellers.	<ul style="list-style-type: none"> • Radial fluid mixing depending on impeller diameter and distance between multiple stages • Despite rotational speed affecting flow velocities, the mixing range nearly unchanged 	[27]
7	Laser-induced fluorescence-based hydrodynamic mixing investigation of HSAD	<ul style="list-style-type: none"> • Mass transfer not enhanced by prolonging the mixing time 	[17]
8	CFD and PIV-based investigation of HSAD hydrodynamics	<ul style="list-style-type: none"> • Concept of mixing-fluidity-energy proposed • Re-based HSAD scale up suggesting suitable rotational speed 	[26]
9	Comparative assessment of continuous and intermediate mixing via HSAD experiments, metagenomics, and CFD	<ul style="list-style-type: none"> • Continuous mixing inducing apoptosis and hindering methane production • Intermittent mixing strategy promoting syntrophic metabolism between bacteria and methanogen, while maintaining flow homogeneity • Genes in acidogenic and methanogenic pathways enriched via intermittent mixing 	[61]
10	CFD simulation of FW HSAD with variable rheology	<ul style="list-style-type: none"> • Evaluation of uniformity index, breakup number, global velocity gradient, and mixing energy • Mixing energy level being a key indicator for mitigating dead zone formation within the reactor 	[62]

increased the cell membrane integrity from 2.9% to 6.4%, leading to an accumulative methane yield of up to 251 mL CH₄ g⁻¹ VS [60]. The presence of biochar also enriched *Syntrophomonas* and methanogens *Methanosarcina* & *Methanocelleus*, which shifted the methanogenic pathways from acetoclastic/hydrogenotrophic to more metabolically

Table 2
Influence of adsorbent or surfactant addition on AD.

Media	Enhancement of AD	Mechanism	Ref.
Surfactant	<ul style="list-style-type: none"> Improving enzymatic hydrolysis of biomass and achieving a higher H₂ yield using non-ionic surfactants 	<ul style="list-style-type: none"> Modification of the substrate surface and making it more accessible to enzymes 	[67]
Surfactant	<ul style="list-style-type: none"> Adding Tween 80 increased the H₂ yield from POME by 59.1%. POME added with PEG increased the H₂ yield by 64.97%. 	<ul style="list-style-type: none"> Formation of adsorption/aggregation and micellization, extraction of hydrophobic part of OFMSW, and enhance the accessibility to enzymes 	[68, 69]
Adsorbent	<ul style="list-style-type: none"> When granular activated carbon (GAC) increased from 0 to 5.0 g, the methane yields the sludge reduction rate increased by 17.4% and 6.1%. 	<ul style="list-style-type: none"> Enriching methanogens promoting direct interspecies electron transfer Accelerating substrate consumption and methane production by enhancing the electron exchange between syntrophs and methanogens 	[70]
Adsorbent	<ul style="list-style-type: none"> For efficient anaerobic digestion of ammonium-rich swine waste using modified wheat rice stone (WRS). 	<ul style="list-style-type: none"> Enhancing the growth of microorganisms on the surface of the modified WRS and methanogens diversity, as well as toxic tolerance 	[71]
Adsorbent	<ul style="list-style-type: none"> The process is favored by adding natural zeolite with the optimum dose value at 0.10 g·g⁻¹ VSS. 	<ul style="list-style-type: none"> For piggery waste, process enhancement due to immobilization of microorganisms For synthetic waste, due to the immobilization of microorganisms and reducing the concentration of NH₃ 	[72]
Biochar	<ul style="list-style-type: none"> Increase in biological stability (554 mg O₂ kgVS⁻¹ h⁻¹ vs. 809 mg O₂ kgVS⁻¹ h⁻¹) Slightly increase in methane yield from 311.78 L kgVS⁻¹ to 366.43 L kgVS⁻¹ 	<ul style="list-style-type: none"> Changing the microbial community composition Decreasing digestate toxicity 	[73]
Biochar	<ul style="list-style-type: none"> Methane yield of 294 mL g⁻¹ VS added, 69% higher than that of the control 	<ul style="list-style-type: none"> Accelerating the transformation of macromolecular into dissolved substrates Increasing the buffering capacity 	[74]
Biochar	<ul style="list-style-type: none"> Increasing TS and VS reduction rates by 36.4% and 34.1% than control Cumulative methane production increased by 1.3%–7.8% 	<ul style="list-style-type: none"> Increasing formate electron donor methanogens Promoting electron transfer efficiency 	[75]
Zero valent iron	<ul style="list-style-type: none"> Electron transfer capacity and electroactive proteins of sludge increased by 5.4 and 2.3 folds Relative abundances of <i>Methanotrix</i> and <i>Methanosarcina</i> enriched to 67.5% and 27.2% 	<ul style="list-style-type: none"> Improving interspecies hydrogen transfer Promoting direct interspecies electron transfer 	[76]
Zero valent iron	<ul style="list-style-type: none"> Methane production 1.07, 1.24, 1.41, and 1.46 times as compared with the control Hydrolysis rates increased 3.5–8.21 times 	<ul style="list-style-type: none"> Improving hydrolysis rate and methane production 	[77]

Table 2 (continued)

Media	Enhancement of AD	Mechanism	Ref.
Fe scrap	<ul style="list-style-type: none"> Enriching the CH₄ yield by 14.46% and improving the digestion rate by 21.28%. The methane yield (29.51% higher), and VSS reduction rate (48.27%) were the highest with rusty scrap. 	<ul style="list-style-type: none"> Improving the mass transfer efficiency with sludge and liquid Inducing microbial Fe³⁺ reduction Enriching iron-reducing bacteria and enhancing diversity of aceto-bacteria for the degradation of complex substrates 	[78]
Fe ²⁺	<ul style="list-style-type: none"> Biogas yield of 465.24 mL g⁻¹ VS with a maximum rate of 16.72 mL (g VS·d)⁻¹, compared to the control sample with 6.78 mL (g VS·d)⁻¹ 	<ul style="list-style-type: none"> Increasing the abundances of Firmicutes and Euryarchaeota Enhancing the hydrolysis-acidification and methanogenesis process 	[79]
Limonite	<ul style="list-style-type: none"> The maximum methane generation rates increased by 30.5% at limonite concentration = 1%. Degradation efficiency = 38.1%–45.9%, greatly higher than the control 	<ul style="list-style-type: none"> Enhancing the degradation efficiency of soluble organic metabolites Increasing the abundance of microorganisms related to the decomposition of protein or amino acid 	[80]
Magnetite	<ul style="list-style-type: none"> Methane production accelerated by 26.6% 	<ul style="list-style-type: none"> Enhancing direct interspecies electron transfer Improving acetate-dependent methanogenesis 	[81]
Nickel	<ul style="list-style-type: none"> Methane yields increased by 11.6–31.8%. Metal bioavailability enhanced by 18.9–42.6% 	<ul style="list-style-type: none"> Increasing the abundance of archaea communities Improving metal bioavailability 	[82]

diverse. The dosage and types of biochar used affect its impact on HSAD. Another research work [83] showed that adding pristine wood biochar to the HSAD of chicken litter at the dosages of 0.25 and 0.5 g_{TS-char}/g_{TS-feed} did not considerably change the 90-day methane yield, while the one at the dosage of 1 g_{TS-char}/g_{TS-feed} increased the yield by 39% with a significant increase in propionate degradation and a 35% reduction in lag time. The addition of both pristine and re-used wood biochar recovered from a 90-day HASD process reduced the retention times, enabling more batches of operation and thus a high throughput.

4. Modelling and simulation tools

The AD process is a confluence of biochemical reaction kinetics, associated hydrodynamics of feedstock, and energy transport from feedstock to the output biogas, making mathematical modelling challenging. These models can be used as a tool to investigate *what-if* scenarios for system scale-up, perform rapid system optimization, design control algorithms for HSAD reactors, and integrate fault detection methods for smart operation of digesters. Three types of models can be applied to model HSAD processes: (a) biochemical kinetics models [16], (b) computational fluid flow (or hydrodynamics) models [84], and (c) data-driven models [85], as discussed below.

4.1. Biokinetic models

There exists a wide range of biochemical kinetic models for predicting biogas generation for AD processes among which some popular choices are Gaussian [86], Gompertz [87], multi-regression [88], acidogenesis-methanogenesis-two-steps model [89], and ADM1 [90]. ADM1 is the most sophisticated model that can capture the four-step AD

phenomenon shown in Fig. 3. It is a multi-step degradation model developed [90] to study the performances of wet-AD systems. It has proven to be robust and effective for process design, optimization, and up-scaling of AD processes. To apply ADM1 for HSAD systems, the mass transfer coefficients need to be altered and calibrated. For example, based on the ADM1 model, an HSAD-specific model was developed with experimental biological kinetic parameters and mass transfer coefficients of biodegradation products [91]. ADM1 was extended to model a two-stage HSAD system consisting of an HSAD and an upflow anaerobic sludge blanket (UASB) seed reactor with good accuracy in terms of predicting VFAs, pH and biogas production [92]. The model identified that recycling methanogenic seeds from the UASB reactor to the HSAD reactor increased the CH₄ concentration and decreased the H₂ concentration in the HSAD reactor. ADM1 was also used to model continuous mesophilic HSAD considering a variety of solids concentrations, sludge retention time, OLR, and sludge types [92]. The model was then used to evaluate the feasibility of replacing conventional AD units in a water resource recovery facility with HSAD. Table 3 summarises the ADM1 model applied to HSAD, SSAD and co-digestion.

ADM1 can be used to study the influences of process parameters on the performance of HSAD and associated transport phenomena. TS is one of the most important factors that influence AD digestion performance. The simulation using ADM1 showed that limited mass transfer at high TS led to low CH₄ production, and that the rate of hydrolysis decreased with increasing TS, especially for TS > 30% [47]. When the TS increased from 10% to 25%, there was a slight decrease in the total CH₄ production. When the TS was above 30%, methanogenesis was significantly inhibited. In an HSAD process, the volumetric mass transfer coefficient (correlating mass transfer rate, mass transfer area, and change in concentration) might be significantly reduced because of reduced solid-liquid-gas interface (see Fig. 2b).

Modelling of heat transfer is another important aspect of the ADM1 and modified ADM1, which is often neglected in related research works. A temperature model is extremely important for ADM1 toward robust evaluation of temperature-dependent parameters such as the mass transfer coefficient, pressure, microbial growth and death rates, and physicochemical properties of fluid. As evidenced in the literature [16, 104], a generic energy balance formulation can be applied to determine the digester temperature and associated heat transfer for AD processes as shown in Eqs. (1) and (2). The equations hold true for a CSTR-based AD system where the required reactor thermal energy is supplied via hot water supplied through an auxiliary heat exchanger.

$$\dot{m}^{FS} C_p^{FS} T_i^{FS} + \dot{m}^w C_p^w (T_i^w - T_o^w) = \left(\dot{m}^{dg} C_p^{dg} + \dot{m}^{bg} C_p^{bg} \right) T^{AD} + U^{AD} A^{AD} (T^{AD} - T^{amb}) \tag{1}$$

$$\dot{m}^w C_p^w (T_i^w - T_o^w) = U^{HX} A^{HX} \Delta T_{lm}^{HX} \tag{2}$$

where T is the temperature, \dot{m} denotes the mass flowrate, U depicts the overall heat transfer coefficient, and ΔT_{lm} denotes the log-mean temperature difference. Various subscripts and superscripts used are FS : feedstock, w : water, i : input, o : output, AD : anaerobic digester, dg : digestate, bg : biogas, and HX : heat exchanger. Simultaneously solving these two equations reveal the two unknowns *i.e.*, digester temperature T^{AD} and hot water outlet temperature.

The incorporation of a thermal model within the ADM1 (or modified ADM1) framework is highly recommended, which enables accurate quantification of heat exchange energy, heat lost to the ambient through reactor walls, heat generated within the highly viscous HSAD fluid due to reactor stirring, and contribution from heat addition due to biological reactions. As a result, the impact of various thermal parameters and fluid flow rates on the biogas generation can be quantified in a coordinated manner [104]. A representative schematic showing various heat transfer (convective and radiative) and mass transfer (liquid to gas phase) for a CSTR is shown in Fig. 2.

Macro-scale models considering the operation of reactors, can describe the mass and heat transfer across different substrate beds and reactors [105]. There are two types of macro-scale models, *i.e.*, balance/transport models and kinetic models, with the former modeling the mass and heat transfer within and between the various phases of a reactor while the latter considering those at cell and particle levels [106]. Typical kinetic models include the first-order kinetic model [16], Cone model [107] and modified Gompertz model [108]. The first two being widely used to predict the CH₄ production for conventional AD and co-digestion processes. The modified Gompertz model considers the maximum CH₄ production rate and lag phase. HSAD experiments based on dewatered sludge with antibiotics and kinetic modelling showed that the modified Gompertz model served as the best model in term of cumulative biogas yield prediction [109]. Kinetic modelling analysis based on the first-order kinetic model and the superimposed first-order kinetic model has been used to explore the kinetics mechanisms of the process where thermal treatment was used to enhance the HSAD of swine manure (>20% solid content) [110]. The thermal treatment was done by placing CSTRs in a water-bath at 70 ± 1 °C for 1–4 days. The model analysis showed that biodegradable organics was increased by the

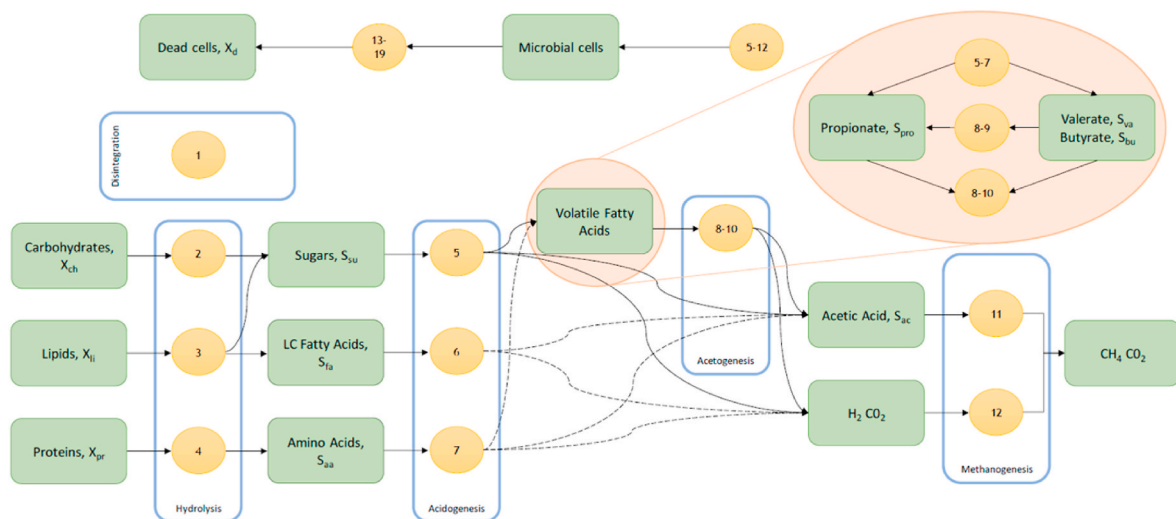


Fig. 3. Simplified overview of biological processes of ADM1. The blue boxes represent salient steps, the yellow circles signify multi-step biokinetic processes, while the variables inside green square are different output variables from the model.

Table 3
Summary of modified ADM1 deployed for HSAD.

Type	Remarks	Findings	Ref.
HSAD	<ul style="list-style-type: none"> Calibrated by mass diffusion described by the Fick equation. The diffusion molar flux is proportional to the gradient of concentration 	<ul style="list-style-type: none"> Useful information for enhancing CH₄ production 	[93]
HSAD	<ul style="list-style-type: none"> Modelling the effect of the inhibition of free ammonia on HSAD performance Considering the VFAs generation and ammonia accumulation in batch, semi-continuous and 30 m³ full-scale systems. 	<ul style="list-style-type: none"> Applicable to HSAD of sludge 	[94]
SSAD	<ul style="list-style-type: none"> Much lower hydrolysis rate constant and volumetric liquid/gas mass transfer coefficient than wet AD 	<ul style="list-style-type: none"> Limited mixing, and low biogas bubble formation and moisture content in SSAD reduced the mass transfer coefficient 	[45, 47]
SSAD	<ul style="list-style-type: none"> Adjustments made for "maximum acetate utilization rate" and half saturation constant 	<ul style="list-style-type: none"> Diffusion limitations decreased the saturation constant at high TS contents in SSAD Accessibility of the substrate reduced, and diffusion limitation aggravated in SSAD 	[95]
Co-digestion (High-solid)	<ul style="list-style-type: none"> Simulating the steady state co-digestion of municipal wastewater sludge and restaurant grease trap waste 	<ul style="list-style-type: none"> Results of biogas production, CH₄ and CO₂ contents, pH, alkalinity, COD and VSS under steady state Good consistency regarding the values of CH₄ production, pH, ammonia, alkalinity, and COD 	[96]
Co-digestion (High-solid)	<ul style="list-style-type: none"> Modelling two separate influent substrates with different biodegradation kinetics Sewage sludge degradation modelled based on ADM1 while organic fraction of municipal solid waste (OFMSW) disintegration with surface-based kinetics. 	<ul style="list-style-type: none"> Modelling the CH₄ production of the co-digestion of OFMSW and sewage sludge in a CSTR Suitable to predict the occurrence of process failure 	[97]
Integrated models (High-solid)	<ul style="list-style-type: none"> A model for a new two-stage HSAD system with an HSAD and UASB seed reactor. 	<ul style="list-style-type: none"> Recycled methanogenic bacteria increasing the CH₄ content and decreasing the H₂ content, but pH increased in the batch mode of the recycling rate 	[92]
HSAD	<ul style="list-style-type: none"> Calibrated using CH₄ production data from FW-AD at TS of 4.2%, 12.8% and 19.2%, and rice straw AD at TS of 4.8%, 14.8% and 23.4% 	<ul style="list-style-type: none"> The changes of kinetic parameters potentially caused by limited mass transfer of soluble products to bacterial sites 	[98]
ADM1-based dispersive model (High-solid)	<ul style="list-style-type: none"> Better describe UASB reactor performance by considering the hydrodynamics and bio-dynamics 	<ul style="list-style-type: none"> Early warning of reactor abnormalities Providing useful information for design and operation 	[99]

Table 3 (continued)

Type	Remarks	Findings	Ref.
Pre-treatment (High-solid)	<ul style="list-style-type: none"> Calibration procedures using batch tests showed the effects of pre-treatment 	<ul style="list-style-type: none"> Feasible to predict and assess the behaviour of digesters 	[100]
Integrated models (High solid)	<ul style="list-style-type: none"> Validated in a continuous AD pilot plant, treating mixed organic wastes at different HRTs at mesophilic conditions 	<ul style="list-style-type: none"> The feasibility of the blends estimated 	[101, 102]
HSAD	<ul style="list-style-type: none"> Novel modification of ADM1 for mass/volume dynamics for homogenized HSAD reactors for long operation 	<ul style="list-style-type: none"> Account for the effect of TS concentration on soluble species Model well-suited for operating in the wet- and dry-AD regimes of OFMSW 	[103]
Integrated models (High-solid)	<ul style="list-style-type: none"> Integration of modified ADM1 with heat transfer model to accurately quantify temperature in HSAD 	<ul style="list-style-type: none"> The model is suitable for OFMSW and the effects of various thermal parameters on biogas yield can be studied. Quantitate evaluation of rise in inlet hot water temperature per degree change of reactor temperature 	[104]

thermal treatment, contributing to the enhanced CH₄ production.

4.2. Computational fluid dynamic models

Since AD processes occur within a bioreactor over a prolonged period, the transport phenomenon is spatiotemporal. Although the multi-step biokinetics models (e.g., ADM1) have often been used to describe transient generation or extinction of species within the digester, they cannot quantify fine-grained spatial changes in the fluid flow field. This is due to the lumped or zonal nature (referred to as zero-dimensional zonal model) of the biokinetic models, which assumes the reactor to be spatially homogenous (or well-stirred). CFD simulation circumvents this drawback, and the literature cites various instances where it was used to resolve perplexing multi-phase flow fields for ADs. For HSAD, the CFD simulation is highly desired due to the existence of a high percentage of solids in the incoming waste stream, which results in all three phases (solid, liquid, and gas) within the digester. The literature pool contains a significant number of works toward CFD simulation of wet-ADs [84], among which only a smaller fraction focuses on resolving the flow-field for HSAD summarized in Table 1.

From the viewpoint of transport phenomenon, the HSAD process suffers from several problems such as (a) hydraulic short-circuiting, (b) dead volumes induced by low flow velocities, (c) deficit in the degree of mixing, (d) concentration gradients of contaminants and suspended solids, and (e) edge effects introduced by reactor baffles [111]. Hydraulic short-circuiting occurs when the flow velocity of incoming waste stream significantly increases. In this case, the waste stream experiences only a short residence time within the reactor, ultimately affecting the reactor performance and lowering biogas yields. In this case, the advection time scale of the flow is much longer than the reaction time scale. Dead volumes (or stagnation zones) are formed within AD in the regions where the flow velocities (or Reynolds numbers) are very low, which are usually seen at the bottom segment of the reactors. The problem is important for HSAD due to the high viscosity of the waste stream, leading to the formation of stagnant eddies. Such drawbacks can be mitigated by improving the agitation frequency of the reactor, which

increases the energy cost of the overall process. The diffusion of unwanted contaminant gases (such as N_2 , H_2S , and NH_3) largely alters the flow field, which is highly detrimental to biogas production. Although reactor baffles help to improve the flow field, an unoptimized baffled reactor creates numerous eddies and vortices, ultimately lowering the momentum of the fluid. This problem is more pronounced for HSAD due to the interaction between solids within the waste stream to the reactor baffles.

To carefully study the transport phenomenon discussed above, a popular choice of CFD models deploy Reynolds Averaged Navier-Stokes (RANS) equations with the $k-\epsilon$ turbulence model [27,112]. In addition, two-phase models such as volume of fluid or Eulerian-Eulerian approaches are also applied for multi-phase flow modelling [84,111], with very few exceptions where three-phase flow modelling was utilized [113]. Furthermore, since the input waste with a large solid content is a non-Newtonian fluid with characteristics such as high viscosity, shear-thinning, and thixotropy, accurate viscosity models are also needed within the CFD framework. Accurate implementation of the biokinetic models within the CFD framework is challenging due to the requirement of a prohibitively high number of parameters, model choices, and governing equation selection. This drawback has been addressed by integrating the ADM1 biokinetic model with the CFD framework [111,114–116]. Such hybrid models have enabled inter-relating kinetics mechanisms of AD, while accurately resolving the associated spatiotemporal flow fields that dictate the biogas and digestate production. An example of this model is the recently developed open-source ADM1Foam (*i.e.*, ADM1 with OpenFoam CFD solver) [111].

4.3. Data-driven black-box models

Data-driven black-box models become increasingly popular in process modelling when the detailed internal mechanisms (physical or chemical) of a process are not well understood or computationally expensive to implement. Modelling of AD is complex as it requires knowledge of population dynamics of the microbial community, biochemical reaction kinetics models, fluid flow and heat transfer models, and energy balance models. In this case, data-driven models built upon *a priori* experimental data or benchmark simulations have been evidenced to be promising for achieving accurate AD process modelling [117]. Data-driven AD models are of three types: (a) static regression models that predict biogas yield and compositions, (b) classification models that detect faults and process anomalies within an AD reactor, and (c) transient regression models of biogas yield and compositions that can be used for process control [117]. Nevertheless, there has been a much lesser effort toward developing HSAD-specific data-driven models, except in a few instances [118,119].

A static multi-linear regression model has been developed to predict specific CH_4 production from a pilot-scale HSAD of organic fraction of municipal solid waste based on 332 experimental observations, resulting in a coefficient of determination (R^2) of 0.91 [118]. The model utilizes various input parameters such as organic loading rate (OLR), lignin content, total VS, C to N ratio, hydraulic retention time (HRT), and total VFAs. However, the model did not include fluid flow parameters such as agitation frequency, viscosity of the input waste, etc., which can often become an issue for the scale-up design of HSAD reactor. Another work developed a recurrent neural network (RNN)-based time-series regression model for predicting the daily yield of biogas generated from HSAD of FW [119]. The work utilized a long-short term memory (LSTM) approach, an advanced class of RNN, and resulted in an R^2 of 0.82. However, the parametric input space for the data-driven model only included HRT, chemical oxygen demand (COD), total volatile fatty acids (TVFAs), total NH_3 , and free NH_3 , which excluded thermo-hydraulic parameters associated with HSAD. These initial efforts toward data-driven modelling for HSAD clearly indicate a wide opportunity for model development for different feedstocks, biological parameters, and reactor thermo-hydraulic parameters. Data-driven models should be

holistically integrated with model-agnostic explainability methods (*e.g.*, Shapley additive explanation and partial dependence analysis), multi-objective optimization problems for *what-if* scenario analysis, and dynamic model predictive control (MPC) frameworks. Physics-informed hybrid models (*i.e.*, comprising of both data and physics) can be developed by informing the data-driven models with differential equations of ADM1 and CFD simulations, and such models are often referred to in the literature as physics-informed machine learning and featured by higher interpretability [117,120].

5. Digester selection

Digester design and selection is one of the most vital aspects to ensure process efficiency and mitigate emissions from HSAD. The choice of design, scale of implementation, and modes of operation dictate the spatiotemporal hydrodynamics, biokinetics, and heat and mass transfer within the digester. HSAD reactors can be classified based on various aspects [121]: (a) orientation (horizontal and vertical), (b) scale of operation (lab-, pilot-, and full-scale), (c) number of stages (single- and multi-stage), (d) number of fluidic phases (single- and multi-phase), (e) operating temperature (psychrophilic, mesophilic, and thermophilic), and (f) substrate feeding (batch and continuous).

5.1. Standalone reactor

In a standalone reactor (*i.e.*, single stage), the four AD steps take place simultaneously. For an HSAD process, the suitable pH for hydrolysis and acidogenic microbes is 5.5–6.5, whereas the pH for methanogens is 6.8–7.2 [122,123]. The biogas yields of single-stage reactors are lower than that of multi-stage reactors because the pH values are different in different stages in the multi-stage reactors. The improvement of mass transfer and heat transfer is more important for single-stage reactors than for multi-stage processes or wet digestion processes [124].

In a plug-flow reactor, organic wastes and inoculum are not completely mixed but move as a plug through the reactor from the feed port to exit. High-solids system could support a 4–6 times higher OLR, leading to higher volumetric CH_4 production. Due to the limited mass transfer, when the free ammonia concentration was higher than 600 mg L^{-1} , it became the main factor influencing system stability [125]. For HSAD systems, mechanical mixing, slurry recirculation and gas recirculation are often used to eliminate the limitations of mass transfer and heat transfer. Compared with slurry recirculation and gas recirculation, mechanical mixing is more effective in increasing heat and mass transfer rates by reducing hydraulic dead space and enhancing homogenizing metabolite. For example, A 12 m^3 multiphase flow anaerobic digester was developed to achieve higher biogas production and constant temperature during winters. Experiments were run to compare between dynamic AD (with mixing) and static AD. It was shown that the biogas production for dynamic AD was 115.2 m^3 or 127.1% higher than that of AD with the same digestion temperature. Heat transfer performance experiments showed that the heat transfer rate of the system increased significantly in dynamic AD [126].

Another type of AD is the LBR, consisting of a batch digester for loading solid substrate and a liquid tank containing leachate that can be discontinuously sprinkled over the substrate (Fig. 4e–h). After an initial phase, leachate injection is less important, and the flush-rate could be reduced. While during the acidification phase, increasing the flush-rate of leachate is essential to increase degradation kinetics [127]. LBR has the advantages of high loadings, reduced water consumption, lower investment costs and high stability [128]. However, LBR requires frequent loading and unloading, resulting in incomplete degradation [129]. For CSTR, the digestate is relatively well mixed and partially digested. Washout of digestate negatively affects the performance and biogas yield because of the removal of undigested biomass.

Heat and mass transfer performance of HSAD can be also regulated

via mechanical mixing (as shown in Fig. 4a–d) or addition of porous media. For example, porous materials including loofah sponge, expanded clay (EC) and activated carbon (AC), have been used to improve the biogas production in fixed-bed bioreactors. Findings revealed that biogas yields were much higher (compared to loofah sponge) when using EC and AC as a support matrix with HRT within the range 0.5–5 h [130]. Furthermore, porous materials such as zeolite and activated carbon can be used to promote the diffusion-dominated mass transfer and microbial growth, and stabilize the process [70,71]. A porous media facilitates thin-layer diffusion by increasing the effective surface area, while conventional reactors have semi-infinite species diffusion during biokinetic reaction [39].

Another process enhancement strategy for HSAD adopted digestate or leachate recirculation (see Fig. 4b and d). Percolate/leachate recirculation in HSAD systems have also been applied to mitigate mass transfer limitations via homogenization of microbes, organics, and nutrients [10,63,64,129]. Another work studied the effects of digestate recirculation ratio on the biogas yield of continuous HSAD with the analysis of microbial community succession [66]. It was found that biogas production was increased at a 60% digestate recirculation ratio as compared to a 50% ratio. A biogas production rate of 1.6 L (L d)⁻¹ was achieved for the case of 60% digestate recirculation ratio, which enhanced both microorganisms (*Methanobacterium*) proliferation and VadinBC27 wastewater-sludge group.

The strategies discussed above has been the basis of several industrially-available full-scale HSAD systems, among which the following are notable (a) Kompagas, (b) Dranco, (c) Valorga, (d) Strabag, (e) Bekon, (f) Gicon, (g) Bioferm, and (h) Biocel (see Fig. 4), [11,64,65]. Among these Bekon, Biocel, Bioferm, and Gicon are batch HSAD systems, while Kompagas, Dranco, Valorga, and Strabag are continuous digesters. The continuous systems use impeller-based mixing in the

reactor, while the batch system use percolate spraying. These systems also differ by their TS feed range, operational parameters (e.g., temperature and sludge retention time) and waste loading capacity.

5.2. Coupled reactor

HSAD being a multi-step process can be conducted under variable reactor conditions to facilitate different stages of the process, thus enhancing the overall process efficiency. This can be achieved by coupling several bioreactors with different conditions (e.g., pH, temperature, HRT). Common multi-stage HSAD processes consist of two or three stages. In a typical two-stage HSAD process, the hydrolysis and acidogenesis occur in the first stage, while the second stage is responsible for acetogenesis and methanogenesis. In the first stage, the hydrolysis of complex carbohydrate limits the reaction rate, while in the second stage, the rate limiting factor is the growth of methanogenic bacteria. Methanogenic bacteria typically prefer neutral to slightly alkaline pH (7.0–8.5) environments, while acidogenic bacteria preferred slightly acidic pH. For a three-stage reactor, the hydrolysis and acidogenesis is further broken into two separate stages (see Fig. 5).

A number of prior works have leveraged multi-stage HSAD to enhance the overall process efficiency. For example, a thermophilic (*i.e.* reactor temperature 55–60 °C) three-stage HSAD of horticultural waste and FW achieved up to 45% improvement in methane yield and a 61% VS reduction efficiency, when compared to two-stage and single-stage reactors [131]. In another work, HSAD of corn stalk was investigated in a coupled reactor combining an up-flow anaerobic solid-state (UASS) reactor and an anaerobic filter (AF) reactor, which showed higher methane production rates than single-stage and thermophilic two-stage systems [132]. A novel, compact three-stage anaerobic digester (TSAD) with three separate chambers was developed to improve the efficiency

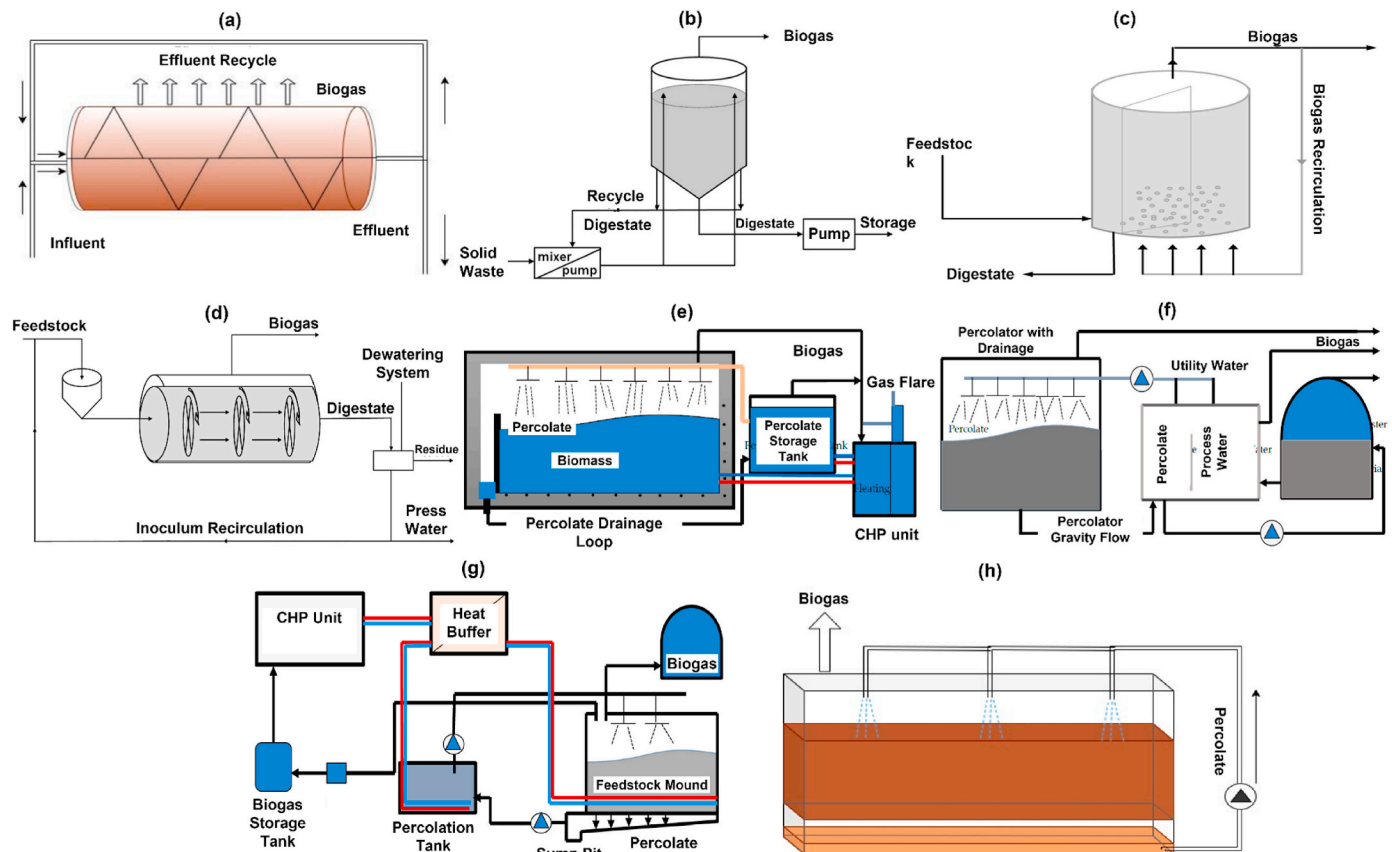


Fig. 4. Illustrative schematics of various industrially available full-scale HSAD reactors (a) Kompagas, (b) Dranco, (c) Valorga, (d) Strabag, (e) Bekon, (f) Gicon, (g) Bioferm, and (h) Biocel. Reproduced from the literature [11,64,65].

of FW AD as shown in Fig. 5a [123]. The TSAD has three separate chambers to improve hydrolysis, acidogenesis and methanogenesis. The methane yield for the design was up to 54% higher than conventional single- and two-stage designs for an OLR of 10 gVS/L FW. The TSAD also offered a relatively high VS reduction rate *i.e.*, $83.5 \pm 1.3\%$ compared to other reactors.

A hybrid anaerobic solid-liquid (HASL) bioreactor with a two-stage system consisting of a solid waste acidification reactor and a UASB reactor for methanogenesis has been developed [133]. The HASL is featured by (1) effective acidification by recycling the leachate of the acidification reactor, (2) washout of VFAs by circulating the effluent from methanogenic reactor to the acidification reactor, (3) efficient methanogenesis in the UASB reactor, and (4) a built-in mechanism to keep system stable through VFAs removal. In the methanogenic phase, the removal efficiencies of VFAs and COD were 77–100% and 75–95%, respectively. A two-stage HSAD process named Biopercolat was developed, which includes a liquefaction/hydrolysis reactor and a methanogenic UASB. In the hydrolysis stage, a limited amount of oxygen is supplied under high-solid and microaerophilic conditions, which enhanced the microbial growth and led to complete digestion within 7 days at a high OLR of $15 \text{ kgVS} (\text{m}^3 \text{ day})^{-1}$. In the two-stage process, the longer biomass retention with biofilm increased the resistance of methanogens to high ammonium concentrations [134].

6. Summary and future trends

This work systematically reviewed and summarized up-to-date knowledge on the developments of HSAD, with a central focus on highlighting the various aspects related to its heat and mass transfer. An in-depth discussion of underlying hydrodynamic transport phenomena, critical regulatory parameters, classical and new-age modelling/simulation methodologies, industrial HSAD reactor designs, and utilization of engineered materials to augment HSAD process efficiency were

provided.

Because of the discontinuousness of the solid phase and the mass transfer limitation, it is challenging to achieve representative measurements of the operational parameters of HSAD, which makes experiment-based HSAD process optimization difficult for the time being. For the *in-situ* characterization or visualization of the mass transfer, and analysis of metabolic compounds, sophisticated optical techniques (*e.g.*, PIV and LIF) should be developed and benchmarked to efficiently measure the transport phenomena of HSAD processes. Moreover, for HSAD systems, the studies on the enhancement of mass and heat transfer are mainly based on lab-scale experiments, and there is a limited understanding of the impacts of system scaling on performance. Lack of reliable pilot-scale data is among one of the most significant obstacles against engineering design, digester fabrication, and industrial application of HSAD systems, and poses a real limitation to the development and validation of associated HSAD models, especially the data-driven ones. It is crucial to standardize relevant experimental protocols and accumulate systematic datasets of HSAD development of different scales, which will lay a concrete foundation for promoting cross-study comparison and the development of systematic theories of HSAD. This will have strong implications for developing data-driven HSAD models such as rule-based systems, neural networks, and principal components, towards the innovation of control algorithms and what-if scenarios analysis tools. It will be also worth exploring the application of machine learning-based methods to specifically model the mass and heat transfer phenomena, which can be then incorporated with other methods (*e.g.*, CFD simulation) towards accurate modelling of HSAD processes. The “simplified” consideration of the mass and heat transfer, whilst not sacrificing the accuracy of accounting for the phenomena, will facilitate efficient and reliable modelling of HSAD processes on the whole.

Although mixing and mechanical agitation are effective for improving the mass and heat transfer in wet-AD systems, their effects on the overall system efficiency and energy cost of HSAD are seldomly

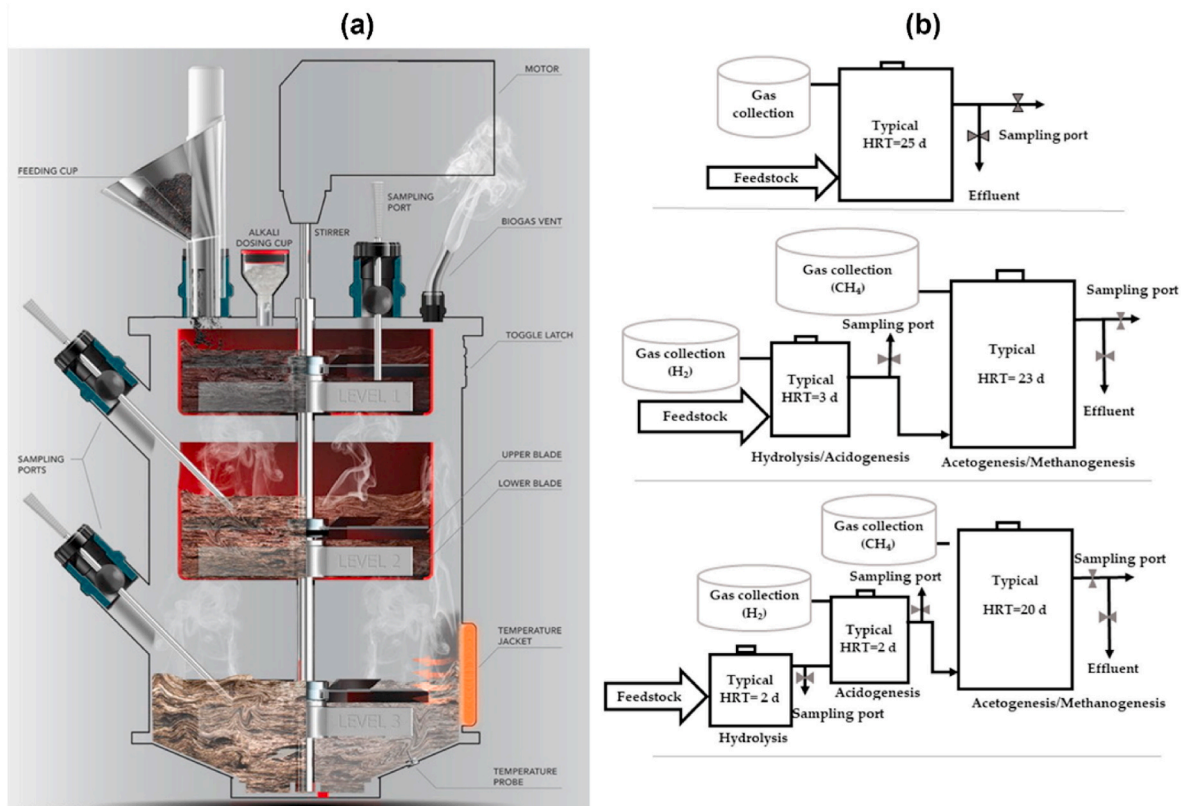


Fig. 5. (a) A compact three-stage anaerobic digester with three separated stages for hydrolysis, acidogenesis and methanogenesis reactions [123]. (b) Comparative schematic of typical process flows for one-, two-, and three-stage ADs.

investigated. Moreover, the effect of mechanical agitation (or flow perturbation) on the growth of methanogenic bacteria should be systematically studied for a wide variety of co-digestion scenarios. Integration of other renewable energy sources (e.g., solar and wind) or low-carbon emission technologies (e.g., heat pumps) with HSAD processes and thus the development of hybrid renewable energy systems could serve to reduce energy consumption-related expenditure and carbon footprint, which is an important direction worth much future research. It is important to note that for such integrated systems, essential 3E analysis (i.e., energy, environment, and economic) can be done via coupling process models, life cycle assessment, and cost-benefit models, which is important for holistically realizing the true benefits of the systems.

Dedicated and consistent research based on experimental and modelling methods is needed to improve the understanding of thermal and hydrodynamic transport phenomena concerning HSAD processes. This serves as one of the potential directions in which technological breakthroughs can be achieved for increasing the efficiency of HSAD. Furthermore, the development of advanced simulation tools based on, e.g., the combination of biokinetic models, CFD simulation, data-driven models, and experimental measurements will facilitate the understanding and implementation HSAD processes. A generalized theory for understanding the influences of process conditions (pH, nutrient, and moisture conditions, VFAs content, NH₃ content) and distribution of microbial community for various scales of the HSAD process is also required. However, systematic and consistent theories underlying the impacts have not been available, largely attributed to the lack of comparable research. This calls for a thorough investigation into the influences of the conditions for accurate and efficient control and management of HSAD processes toward higher economic, energy and environmental benefits.

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.

Data availability

A related statement was added in the Acknowledgements section.

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