

Opportunities and Challenges in Sustainable Treatment and Resource Reuse of Sewage Sludge: A Review

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ABSTRACT. Sludge or waste activated sludge (WAS) generated from wastewater treatment plants may be considered a nuisance. It is a key source for secondary environmental contamination on account of the presence of diverse pollutants (polycyclic aromatic hydrocarbons, dioxins, furans, heavy metals, etc.). Innovative and cost-effective sludge treatment pathways are a prerequisite for the safe and environment-friendly disposal of WAS. This article delivers an assessment of the leading disposal (volume reduction) and energy recovery routes such as anaerobic digestion, incineration, pyrolysis, gasification and enhanced digestion using microbial fuel cell along with their comparative evaluation, to measure their suitability for different sludge compositions and resources availability. Furthermore, the authors shed light on the bio-refinery and resource recovery approaches to extract value added products and nutrients from WAS, and control options for metal elements and micro-pollutants in sewage sludge. Recovery of enzymes, bio-plastics, bio-pesticides, proteins and phosphorus are discussed as a means to visualize sludge as a potential opportunity instead of a nuisance.

KEYWORDS: Waste activated sludge, Anaerobic digestion, Thermochemical treatment, Bio-energy, Resources recovery, Sustainable management

1. INTRODUCTION

Waste activated sludge (WAS) can simply be defined as the residual semi-solid material which is inevitably left over from municipal or industrial wastewater or sewage treatment processes. The rapid rise in population coupled with increasing industrialization has enhanced the production of sludge manifolds [1], which is speculated to increase further, in the near future. Engineering and design relative to wastewater treatment plants (WWTPs) encounters challenging issues related to processing, reuse and disposal of sludge [2].

Wastewater is usually treated via physical routes such as flotation, sedimentation, etc., chemical pathways such as flocculation, etc. and biological ways such as microbial treatment. In addition, wastewater treatment (WWT) processes are grouped into subsystems (for example, primary, secondary and tertiary treatments). The three-fold objectives are (i) to achieve the removal of contaminants, (ii) to ease the management of generated byproducts, and (iii) to meet the legislative standards about the quality of discharged water. As shown in **Fig. 1**, primary sludge is generated by post mechanical treatment after the primary stage whereas WAS is generated via biological treatment at secondary stage in WWTP. Usually WAS is used for resource recovery or energy generation. A typical WAS composition includes 59 – 88 % w/v biodegradable organic matters (OMs), composed of 50 – 55 % C, 25 – 30 % O, 10 – 15 % N, 6 – 10 % H with little amount of P and S [3].

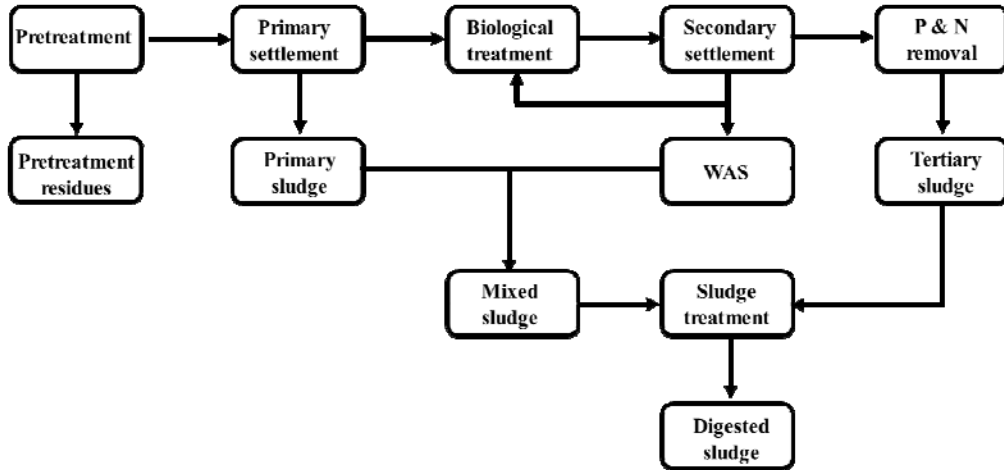


Fig. 1 Sludge generation in a WWTP [4].

Post-treatment WAS can be categorized as aerobic or anaerobic stabilized sludge. The structural composition, chemical properties and thermal behavior of WAS are the functions of stabilization techniques, engineering features of WWTP and pollution load of effluents. Sludge is an intricate blend of numerous materials such as OMs, inorganic matters, other substances, and microbes in dissolved or suspended states. Moreover, it also contains valuable inorganic ingredients such as N, P, K, Ca, S and Mg, along with pollutants and potential carcinogens such as heavy metals (HMs), dioxins, furans, pathogenic microbes [5, 6]. Bacterial constituents such as proteins, lipids, their decay products coupled with inorganic matter and cellulose form the chemical structure of a typical WAS [7].

WAS is the most vital by-product generated from WWTPs, whilst it causes human health problems as a potential source of secondary environmental pollution. Therefore, its proper disposal and treatment carries utmost significance. A diverse treatment range and valorization technologies are available for the safe disposal, resource recovery and power

generation from WAS. Commonly used technologies for energy / resource recovery include anaerobic digestion (AD), incineration, pyrolysis and gasification [3]. In addition to nutrients (N and P), enzymes, bio-plastics, bio-pesticides and proteins can be recovered from WAS employing diverse techniques. Moreover, this perspective of bio-refinery and resource recovery for WAS valorization will not only aid in holistic WAS management but will also lessen significant adverse environmental impact to a greater extent.

Thus, we have reviewed the scientific literature published on WAS treatment facilities during the last 5 - 7 years. In spite of these facts, there is no single review on sewage sludge treatment with the emphasis on treatment technologies, energy recovery, bio-refinery approach for resource recovery as well as treatment scenarios in major countries such as EU, USA and China. We consider that, in all these respects, this is timely contribution. We expect that this review will encourage research and development work and may aid in the scale-up of conventional and advanced technologies. This review article is aimed at providing an overview and discusses the leading sludge treatment technologies (anaerobic digestion, incineration, pyrolysis, gasification and microbial fuel cell integrated process) and their comparative analysis. The coproduction of bio-refinery products from sewage sludge, resource recovery and pollution control for sewage sludge are also reviewed as an opportunity towards sustainable sludge management. An account of treatment scenarios for EU, USA and China is given, which is then followed by conclusions and future perspectives.

2. SEWAGE SLUDGE TREATMENT AND ENERGY RECOVERY

Herein, anaerobic digestion (AD), incineration, pyrolysis, gasification and MFC integrated process are discussed as commonly employed routes for the safe treatment and energy recovery from WAS. There are some other pathways also such as wet air oxidation and hydrothermal treatment but they have very limited application and hence are not discussed here.

2.1 Anaerobic digestion

AD transforms sludge organic solids to biogas via the following biochemical reaction in an anaerobic condition (Eqn. 1).



where, $X = 1/8 (4c + h - 2o - 3n - 2s)$ and $Y = 1/4 (4c + h - 2o + 3n + 3s)$

Biogas comprises of 60 – 70 % methane, and 30 – 40 % of carbon dioxide, trace amounts of other gases (e.g. hydrogen, hydrogen sulfide and nitrogen), with a relative density of around 0.85, and calorific value of about 13 – 21 MJ kg⁻³, which is lower than that of coal (15 - 27 MJ kg⁻³), but equivalent to lignite's (12 – 16 MJ kg⁻³) [8].

Apart from biogas, AD yields digestate as the final product containing high amounts of nutrients (e.g. phosphorus, potassium and nitrogen), which can be further utilized as fertilizer and/or compost.

Methane produced from WAS can be utilized for various applications such as gas engines, electricity and/or heat. The energy acquired via biogas is likely to cover 50 % of the total operational cost of WWTP [9]. Currently, various parametric optimization studies were conducted to enhance the biogas yield and its quality (e.g. ratio of CH₄ to CO₂) [10-13]. Operating temperature is the most crucial parameter influencing the

quantity and quality of the biogas and digestion rate. Three dissimilar temperature ranges, recognized as psychrophilic (12 – 25 °C), mesophilic (35 – 38 °C) and thermophilic (50 – 60 °C), are applied depending on situations where the anaerobic fermentations occur, such as landfills, sediments, anaerobic digester [14]. Some studies have reported various biogas yields from AD of WAS (270 – 385 mL CH₄ gVS⁻¹) [15-17]. It is however known that OMs in WAS are immobilized; therefore, the OM hydrolysis or disintegration is known as rate-limiting step for the subsequent AD process [18]. Alternatively, various pretreatments such as thermal, chemical and mechanical have been applied to accelerate the conversion of persistent biomaterials present in WAS into soluble fractions, for which the main results are summarized in **Table 1** [10, 12, 19, 20].

Compared to above pretreatments, post/inter-stage treatments are considered to be more advantageous for higher methane production [21, 22]. Nielsen et al. [21] compared different pretreatments such as moderate thermal, high thermal and thermochemical prior to AD with inter-stage treatments under the same experimental conditions as described in **Table 1**. They found thermal or thermochemical treatments of WAS more efficient when applied as inter-stage treatment rather than a pretreatment. The inter-stage treatment conditions (170 °C, pH 10, KOH) led to improvement of methane yield by 28 %, whereas only 2 % methane yield was improved using conventional pretreatment. A full-scale simulation showed that methane production would be improved by 45 % using two anaerobic reactors with thermal inter-treatment, whilst only 20 % improvement in methane production could be achieved using conventional AD and pretreatments [22]. More recently, a comprehensive study conducted by Campo et al. [23] demonstrated the low-temperature (<100 °C) thermal and hybrid (thermal + NaOH and Ca(OH)₂)

intermediate and traditional pretreatments under the same experimental conditions. The intermediate treatments were found to be more efficient and produced 16 % higher methane compared to traditional pretreatment, mainly due to their capability of utilizing a 7-day digestate for AD process. The economic assessment conducted in a commercial scale WWTP (2,000,000 population equivalent) revealed that thermal or hybrid pretreatments would probably augment the profits from the electricity sale between 13 to 15 %, when compared with the existing setup. On the other hand, intermediate treatments on a 7-day digestate would offer a revenue of 26 % or 32 %, varying based on the operating temperature (70 or 90 °C). Albeit above methods could efficiently disintegrate WAS and promote methane production, almost all of them demand various chemical reagents and energy, which might be cost-intensive in application. Recently, Zhao et al. [19] (**Table 1**), utilized aged refuse (massively produced in landfills with high concentrations of enzymes and functional microorganisms) as a cost-effective alternative to accelerate the hydrolysis, solubilization and acidogenesis mechanisms and has exhibited promising results. Considering the vast production of WAS, this approach should have considerable economic and ecological benefits if established at landfill sites. However, more studies to examine the potential role of AR in AD is urgently needed. Moreover, the applications of AD process at landfill sites will improve methane capture as fuel source for electricity generation and/or heat and also decrease CO₂ emission from WWTPs operation. According to an analysis by United States Environmental Protection Agency (U.S.EPA), 2.3 million metric tons (Mt) of CO₂ is released per annum (corresponding to the emissions by 430,000 automobiles) from WWTP. This amount could be counterbalanced, if existing WWTPs (with volume above 5 million gallons day⁻¹) use AD

technology for electricity production [24]. More recently, Kretschmer et al. [25] reported that in WWTPs in Austria provided with AD could attain electric independence under optimum operation of WWT and cogeneration. Thus, energy from biosolids in the form of biogas ensures energy security, a reduced dependence on fossil fuels, and lowered greenhouse gas (GHG) emissions. However, further research is required for the enhancement of energy recovery, process optimization towards energy independence using novel technologies such as the integration of solar energy with AD and Fuel Cell systems.

Table 1. Summary of key information of WAS anaerobic digestion.

| Reactor configuration | Pretreatment conditions | Results | Remarks | Ref. |
|--|---|--|---|------|
| <ul style="list-style-type: none"> Equipment: CSTR*, Reactor volume: 2.5 L, AD T: 50 °C, OLR^a: 1.6 – 20.5, SRT^b: 35 – 3 days. | 314 – 348 mL CH ₄ -g VS ⁻¹ | Increasing content of volatile fatty acids (VFAs), decrease in concentration of partial alkalinity and pH were observed as a result of reactor instability. | Short SRT and high VS destruction efficiency led to enhanced biogas production and a positive energy balance for the process. | [10] |
| <ul style="list-style-type: none"> Equipment: CSTR*, Reactor volume: 0.9 L, AD T: 35 – 38 °C, SRT^b: 20 – 4 days. | 175 L CH ₄ -g chemical oxygen demand (COD) ⁻¹ | The considerable shift in bacterial population from 20 – 4 days was noticed: Chloroflexi and Syntrophomonas decreased from 28 to 4.5 % and 9 to 0 %, whilst Bacteroidetes augmented from 12.5 to 20 %. | The lower SRTs are proactive signs for defining rate limitation in AD process. | [26] |

| Reactor configuration | Pretreatment conditions | Results | Remarks | Ref. |
|--|--|---|---|------|
| <ul style="list-style-type: none"> Equipment: Batch reactor, Reactor volume: 1 L, AD T: 35 °C, pH: 4 – 13, carbon to nitrogen (C/N) ratios: 9/1 to 20/1, SRT^b: 30 days. | 310 mL CH ₄ -g VS ⁻¹ | Higher amounts of protein (167 g-COD/kg-TS) and carbohydrates (666 g-COD/kg-TS) were consumed. | The optimum conditions for CH ₄ yield were pH 12 and C/N ratio 17/1. | [17] |
| <ul style="list-style-type: none"> Equipment: Batch reactor, Reactor volume: 280 - 300 mL, AD T: 37 °C, Pretreatment: Moderate thermal (T: 80 °C), high thermal (130 – 170°C), thermochemical (170, pH 10), Post-treatment: at same conditions, Pretreatment SRT^b: 40 days, Post-treatment | | No effect of pretreatment on CH ₄ yield at 80 °C, whilst post-treatment offered a 20 % upsurge. Further, inter-stage treatment led to enhancements of 9 % (130 °C), 29 % (170 °C) and 28 % (170 °C/pH 10). | The thermal treatment appears more efficient when employed as an inter-stage treatment instead of pretreatment. | [21] |

SRT^b: 19 – 21 days.

| Reactor configuration | Pretreatment conditions | | Results | Remarks | Ref. |
|---|---|--|---|---|------|
| <ul style="list-style-type: none"> Equipment: Batch scale CSTR*, Reactor volume: 22 L, AD T: 35 °C, pH: 1 – 6, SRT^b: 12 days. | Acid pretreatment: Hydrochloric acid (HCL, 37 %), 1 day. | 124.5 mL CH ₄ -g COD ⁻¹ | Increase of CH ₄ yield (14.3 %) compared to untreated WAS. | A cost analysis showed that current method of pretreatment is not economically favourable at present. | [12] |
| <ul style="list-style-type: none"> Equipment: Batch reactor, Reactor volume: 1 L, AD T: 35 °C, pH: 7, C/N ratios: 9/1 to 20/1, SRT^b: 30 days. | Aged refuse: 0 to 1200 mg-g dry sludge ⁻¹ Note: Inorganic substances (e.g., sticks, stones and glass were manually. | 173.6 to 213.4 (AR = 0 – 400 mg-(g dry sludge) ⁻¹ | The increase of soluble COD from 1150 to 5240 g L ⁻¹ , when AR dosage was increased from 0 – 400 mg-g dry sludge ⁻¹ . | AR as a low cost alternative significantly enhanced the WAS hydrolysis, solubilization and acidogenesis. The results offer a promising method for the development of WAS AD systems in landfills. | [19] |

removed.

| Reactor configuration | Pretreatment conditions | Results | Remarks | Ref. | |
|--|--|---|---|---|------|
| <ul style="list-style-type: none"> ▪ Equipment: Batch reactor, Reactor volume: 400 L, AD T: 37 °C. | Chemical: 7 g NaOH/L, 1 day. Thermobaric: T: 121 °C, 1 day. | The CH ₄ was increased by 3.28% (chemical) and 8.32% (thermobaric). | Soluble COD was augmented from 16.3 % in the control to 20.84 % (thermobaric), 40.82% (chemical). Early inhibition was reduced by 100 % in the thermobaric group. | The initial cost analysis recognized thermobaric as the most feasible pretreatment method for industrial application. | [20] |
| <ul style="list-style-type: none"> ▪ Equipment: Air-tight bottle, Reactor volume: 160 mL, AD T: 35 °C, Shaking at 100 revolution per minute (rpm), 30 days. | Biological hydrolysis: T: 35 -55 °C, 3 days. | 200 mL CH ₄ -g COD ⁻¹ in 15 days compared to 30 days without treatment (150 mL CH ₄ -g COD ⁻¹) | Soluble COD increased from 175.2 ± 38.2 to 3314.5 ± 683.4 mg L ⁻¹ and the dominant VFA concentration was augmented from 41.5 ± 2.1 to 786.0 ± 133.2 mg L ⁻¹ | The pretreatment of WAS for 15-days at 42 °C attained comparable biodegradability as untreated WAS in 30-days, suggesting biological hydrolysis could decrease solid SRT by 5 to 15 days compared to conventional AD. | [27] |

| Reactor configuration | Pretreatment conditions | Results | Remarks | Ref. | |
|--|--|---|--|--|------|
| <ul style="list-style-type: none"> Equipment: Batch reactor, Reactor volume: 125 mL, AD T: 33 °C, SRT^b: 18 days. | <ul style="list-style-type: none"> Microwave: 50 to 175 °C. | <ul style="list-style-type: none"> Increase of CH₄ production (>31 %). | <ul style="list-style-type: none"> Soluble COD increased from 9 (unpretreated) to 35 % as a result of pretreatment. | <ul style="list-style-type: none"> Inoculum acclimation enhanced biogas yield as well as the extent of ultimate biodegradation of pretreated WAS. | [28] |
| <ul style="list-style-type: none"> Equipment: Batch reactor, Reactor volume: 25 mL, AD T: 35 °C, SRT^b: 100 days. | <ul style="list-style-type: none"> Mechanical (ultrasound): 20 kHz, 0.33 W mL⁻¹, 20 min. | <ul style="list-style-type: none"> CH₄ was increased from 143 (without treatment) to 292 g kg⁻¹ TS_{in}. | <ul style="list-style-type: none"> The existence of polyelectrolyte flocculants increased CH₄ yield during first 6 days of digestion but inhibited thereafter. | [29] | |

*Continuous stirred-tank reactor; ^aOrganic loading rates (kg VS m⁻³ day⁻¹); ^bSolid retention time

2.2 Incineration

Incineration is an exothermic oxidation process of biosolids resulting in the flue gas comprising of CO₂ and H₂O, ash and a certain amount of heat (Eqn. 2). Incineration reduces 90 % sludge volume with the simultaneous destruction of pathogens. The residual ash (about 30 wt. %) can be disposed into the landfills or to be utilized for building material production [30]. Whereas, the recovered heat produces steam, which is subsequently converted into power via steam turbines [3].



Recently, incineration of WAS has gained great attention in several countries [7, 31] on the account of restricted application of WAS in arable land for food production [32, 33]. However, the augmented HMs quantities and the low P plant-accessibility is often related with incineration [31, 34]. Therefore, several approaches are being developed to upgrade the incineration ashes or re-circulate WAS-P directly. **Table 2** illustrates the operating conditions and results of several new methods such as thermal treatment with polyvinylchloride (PVC) and magnesium oxide (MgO) [35], acid leaching [36, 37], and electro dialysis [38, 39]. Thomsen et al. [40] (**Table 2**), reported that high content of total P retained in char and ash with the lower HMs per unit of P makes sludge as a potential source of P-fertilizer for agricultural systems. More discussion on P recovery from incineration is presented in Section 5. Recently, WAS incineration processes are increasingly established to recover the energy in the form of electric power or heat [41-43].

The fluidized bed (FD) incinerators have been considered well efficient for WAS combustion in the form of dry or wet phase (with around 35 – 59 wt. % moisture content)

with high combustion efficiency (e.g. lower organic fraction in the fly ashes <0.3 %) and very low pollutants generation (SO_x and $\text{NO}_x < 200 \text{ mg Nm}^{-3}$) [44]. Murakami et al. [41] successfully demonstrated WAS incineration in a unique FD incinerator (capacity 4.32 t day^{-1}) provided with a turbocharger in Oshamanbe Cho in Hokkaido. Compared to conventional plants, less than half of emissions corresponding to CO, NO_x , and N_2O were observed in the flue gases. Nearly 50 % of energy savings was estimated at incineration plant with capacity of 100 t day^{-1} . Additionally, CO_2 emission and the costs related to supplementary fuel and electricity consumption can be lowered by more than 40 % and 0.2 million dollars, respectively. As reported by the National Association of Clean Water Agencies (NACWA), metro WWTP in St. Paul, Minnesota, U.S. has successfully installed incineration technology of 4.7 megawatt (MW_e) electricity generation capacity. This plant has achieved approximately 20 % of decrease in GHG emissions and about \$1 million savings each year [45]. Also, 0.2 and 0.8 MW_e incineration facilities are being established in Cleveland Ohio (U.S.) and in Hartford, Connecticut, respectively, which will substantially help to meet the energy requirement of plants [3, 45]. In another example, in Cyprus, Vassiliko cement plant, the sludge co-incineration with pet coke has successfully been employed [46]. The major advantages of WAS co-incineration in existing coal-based power plants can be summarized as below [47, 48]:

- To preclude the high cost of an incineration technology;
- To reduce the GHG emissions;
- To advance the energy retrieval efficacy as well as public acceptability.

In addition, low-caloric surplus heat of exhaust gases released from power plant can be effectively employed to enhance the sludge drying process [5]. Therefore, to ensure

the simultaneous energy exploitation and efficient recovery of nutrients, further research is needed on the co-incineration of WAS with other feedstocks (such as coal, wood, etc.), the design of energy efficient incinerator, and the management of ashes with improved and optimized methods.

Table 2. Summary of key information of WAS incineration.

| Experimental conditions | Results | Remarks | Ref. |
|---|--|--|------|
| <ul style="list-style-type: none"> ▪ Equipment: Fluid bed reactor, Plant scale: full scale, T: 850 °C, Air Equivalence ratio (ER): > 1, Particle retention time: 5 sec | <p>Main elements in ash (wt. % dry): Al (4.3), Ca (14.4), Fe (9.2) K (1.5), Mg (1.6), Na (0.5), P (10.3), S (1).</p> <p>HMs in ash (mg kg⁻¹): Cd (63), Cr (1200), Cu (7500), Ni (1000), Zn (25000).</p> | <p>The P-concentration in ash and char was very high making them potential P-fertilizer material.</p> | [40] |
| <ul style="list-style-type: none"> ▪ Equipment: - Plant scale: lab scale, T: 850 °C, Incineration time: 4 h, P extraction: HCl and NaOH (0.01 – 0.8 mol L⁻¹), HM removal: cation exchange resin (CER, 0 – 0.2 g mL⁻¹). | <p>Main elements in ash (wt. % dry): Na₂O (1.51), MgO (3.94), Al₂O₃ (9.89), SiO₂ (43.1), P₂O₅ (27.4), K₂O (3.38), CaO (10.3).</p> <p>HMs in ash (mg kg⁻¹): Cr (89), Cu (787.4), Ni (53.8), Zn (3318), Hg (0.23).</p> | <p>More than 95 % of the total P recovery was achieved at 0.5 mol L⁻¹ HCl at a liquid/solid ratio of 50 mL g⁻¹.</p> <p>2. HMs were effectively removed at 0.04 g mL⁻¹ CER.</p> <p>0.43 kg·kg⁻¹ of struvite production was obtained, indicating a profit of > \$5 kg⁻¹ WAS ash based on the struvite value.</p> <p>The proposed method provides a sustainable way for P recovery from WAS incineration ashes.</p> | [36] |

| Experimental conditions | Results | Remarks | Ref. |
|---|--|--|------|
| <ul style="list-style-type: none"> Equipment: Rotary furnace, Plant scale: lab scale, T: 800 – 950 °C, Air: 5 L min⁻¹, Thermal treatment for HMs removal: PVC and MgO, Post-treatment: Chemical (sulfuric and phosphoric acid). | <p>HMs were significantly removed in the temperature of range of 800 – 950 °C by PVC as Cl-donor.</p> | <p>As a result of thermal treatment ash with PVC and MgO, the P solubility augmented from 14.6 – 36.3 %. The post-treatment considerably improved the P-solubility to 50.2 % and 54.3 %.</p> | [35] |
| <ul style="list-style-type: none"> Equipment: FD Incinerator, Plant scale: Pilot scale, Ca-based bead sorbents (SL-1 to SL-4) for removal SO₂ and particulate matter (PM), T: 850 °C, Particle retention time: 6 sec | <p>HMs (mg kg⁻¹) in the PM collected via the bead sorbents at different times (60/120 h). As (1.1/1.3), Cd (13.2/15.1), Cr (266/288), Cu (480/522), Hg (0.88/2.52), Pb (144/180), Zn (2420/3245).</p> | <p>SL-4 showed 99 % removal efficiency for SO₂, probably due to higher specific surface area for sorbent. The collection of fine PM simultaneously eliminates HMs WAS incineration. The sorbent was found to be an effective material for simultaneous removal of SO₂ and PM from WAS incineration and therefore, it could be exploited for industrial settings.</p> | [49] |
| <ul style="list-style-type: none"> Equipment: Circulating FD | <p>Gaseous products (%): H₂ (13.87), CO (6.5), CO₂</p> | <p>The combustible concentration in the fly ash</p> | [50] |

reactor and Down flow (15.84), CH₄ (2.37), NH₃ (271 mg N m³), HCN collected from the DFC was 2.8 %, corresponding to combustor (DFC)-A combined (592 mg N m³). an overall combustion efficiency of 99.2 % in the approach, Plant scale: Pilot scale, Residue from gasification analysis (wt.%): system.

T: 1150 °C, Air (N m³ h⁻¹): 7.5 moisture: 1.4, volatile matter (VM): 22.3, fixed The study demonstrates that a combined gasification (primary flow rate), 6.4 carbon: 13.8, ash: 62.5 The concentration of NO_x and incineration seem to be viable option for WAS (secondary flow rate) and 12 in the incineration was 220 ± 6 mg N m³. management in an environmentally friendly manner. (tertiary flow rate).

The syngas together with solid residue were fed into the DFC (incineration).

2.3 Pyrolysis

Pyrolysis is the thermal conversion (350 – 900 °C) of WAS performed in an oxygen deficient environment generating vapors or pyrolytic gases. To separate the liquid or oil, the vapors need to be condensed through cooling, leaving behind a solid product namely char [51]. The quantity and quality of liquid, gas and char products depend on the process conditions including operating temperature, reaction time and pressure as well as the WAS characteristics.

Pyrolysis is generally characterized based on heating rate (HR), temperature, and gas residence time (GRT). A pyrolysis carried out at lower HR (0.1 – 1 °C s⁻¹), lower operating temperature (300 – 400 °C), and higher GRT (5 – 30 min) is called slow pyrolysis. In contrary, fast pyrolysis takes place at higher HR (10 – 200 °C s⁻¹), higher temperature (450 - 600 °C), and a shorter GRT (0.1 – 0.3 s). Fast pyrolysis is a suitable approach for conversion of WAS to liquid or gaseous products. Carver-Greenfield technology (C-GT), Oil-from-Sludge technology (OFS) and the Siemens Schwell-Brenna technology (SSBT) are advanced applied pathways to generate renewable fuel (bio-oil) via pyrolysis of WAS [4]. C-GT uses the principle of multi-effect dehydration of water-bearing WAS and centrifugation to produce valuable products such as oil, animal feed and fertilizer [52, 53]. OFS based pyrolysis enhances bio-oil production at ~450 °C for over 30 min under atmospheric pressure to produce straight chain hydrocarbons, which are then condensed into oil [4]. SSBT includes co-pyrolysis of WAS and crushed wastes at 450 °C in a rotary kiln [54].

Compared to the highly exothermic incineration, most of the pyrolytic reactions are endothermic consuming energy of around 100 kJ kg⁻¹ [55]. **Table 3** shows the varying

operating conditions and results of WAS pyrolysis carried out under various methods such as slow pyrolysis [56], fast pyrolysis [56-58], and microwave assisted [59] pyrolysis. As a result of using distinct technologies, operating conditions, and raw materials, WAS pyrolysis product characteristics and distribution differ noticeably. Atienza-Martínez et al. [58] (**Table 3**), reported a maximum liquid yield for anaerobically digested and thermally dried WAS fast pyrolysis (32 %) at 275 °C in FD reactor, when pretreatment (Torrefaction) and post-treatment (catalytic) were investigated. Gao et al. [60] investigated pyrolysis of dried WAS under the operating conditions described in **Table 3**, using a tubular reactor. They reported 46.14 % of maximum tar yield at 550 °C. One of the benefits of WAS pyrolysis is the low operating temperatures, thereby avoiding melting and evaporation of the HMs [4, 61-63]. Unlike incineration, WAS pyrolysis mainly yields a high amounts of char (approximately 50 % of the mass of sludge), as shown in **Table 3**. These chars can be potentially used as solid fuel for heat or can be employed to adsorb HMs or organic contaminants [56]. Therefore, the emission of HMs into the environment triggering serious ecological and health effects could be significantly reduced [64]. Moreover, pyrolysis char has been explored as a cost-effective catalyst or catalyst support and an option for soil conditioning [65]. Nonetheless, the risk associated with the behavior of HMs contained in WAS pyrolysis char is still a serious concern, when considering its utilization or disposal.

Currently, several studies have been conducted mainly focusing on the distribution of HMs from pyrolysis of WAS. For instance, Gao et al. [56] studied the distribution of HMs in char and bio-oil produced from dried WAS under the experimental conditions presented in **Table 3**; it was noticed that most of the HMs were retained in the char,

whilst very low concentration of HMs was observed in the bio-oil. HMs propensity in the char and the bio-oil were found to be comparable with the data reported by Trinh et al. [66], albeit the pyrolysis temperature was different. In addition, Jin et al. [67] (**Table 3**), suggested that majority of the HMs that accumulated in the char at 600 °C caused a major decline in bioavailability, indicating a tremendously low eco-toxicity/risk related to char subjected to the environment. Comparable results were also reported by Devi and Saroha [68] and Chen et al. [69], however different pyrolysis temperatures were used. The economic feasibility of pyrolysis is another challenging factor due to the relative complexity of processing equipment. However, it can be substantially supported by enhancing oil yields and production of high valued products from pyrolysis char [4]. The products produced from WAS such as oil, gas and char can be utilized for various application such as raw materials for producing chemicals and biofuel [4]. Moreover, pyrolysis flue gases also require lesser clean-up to satisfy emission limits than those from incineration. Samolada and Zabaniotou [70] compared WAS incineration, gasification and pyrolysis. Pyrolysis was identified as a zero waste technology, thus offering a potential solution to the pollution associated with WWT, compared to other technologies.

On the whole, it could be inferred that the technology of pyrolysis for WAS holds potential to satisfy the environmental criteria of sustainable development and economic and social concerns [70, 71]. However, pyrolysis of WAS is still in infancy stages. Therefore, the introduction of more effective strategies (based on the type and content of HMs in the WAS) such as pre-/post-treatment, sorbents, and optimization of pyrolysis temperature towards high bio-oil production and the immobilization and/or minimization of HMs transfer to the products is needed.

Table 3. Summary of key information of WAS pyrolysis.

| Experimental conditions | Results | Remarks | Ref. |
|--|---|---|------|
| <p>▪ Equipment: Horizontal tubular furnace reactor, Plant scale: Lab-scale, Mode: slow pyrolysis (8 °C min⁻¹) and fast pyrolysis (100 °C min⁻¹), Sludge loading: 120 – 200 g.</p> | <p>Main elements in pyrolysis residue (wt. % dry): Al (1.22), Ca (1.79), Fe (1.62) K (1.5), Mg (0.35), Na (0.24), P (4.3), and S (1.3). HMs in pyrolysis residue (mg kg⁻¹): Cr (661), Ni (311), Mn (514), Ba (469), La (231), Ce (208), and Sr (47). Tar yield (46.14 %), Non-condensable gas yield (28.64 %), Char yield (47.07 %).</p> | <p>Fast pyrolysis gave higher yield of tar compared to that of slow pyrolysis. With increasing temperature, total gas yield increased and char decreased in both cases.</p> | [60] |
| <p>▪ Equipment: FD reactor, Plant scale: Lab-scale, Sludge pretreatment: Torrefaction (T: 250 °C, 13 min), Mode: Fast pyrolysis, T: 250 – 275 °C, Sludge feeding rate: <1 kg h⁻¹, Hot pyrolysis vapors post-treatment: γ-Al₂O₃ catalyst (Auger reactor, T: 480 °C, SRT: 6.5 min)</p> | <p>Liquid yield (32 %), Non-condensable gas yield (21 %), Char yield (47 %).</p> | <p>The combination of the treatments (torrefaction and catalytic) did not enhanced the properties of liquid for use as fuel, concerning O/C molar ratio and nitrogen content. The treatment combination benefits were (i) the saving of catalyst ensuing from the lower quantity of vapors produced via the pyrolysis torrefied WAS and (ii) the minor decline in the coke yield.</p> | [58] |

| Experimental conditions | Results | Remarks | Ref. |
|---|---|---|------|
| <ul style="list-style-type: none"> Equipment: Homemade pyrolysis and carbonization furnace, Plant scale: Lab-scale, T: 400 - 600 °C, Sludge loading: 500 g. | <p>The proportion of char yield decreased gradually from 60.6 to 53.1 %, whilst ash content increased by 15.6 % with respect to increase in pyrolysis temperature from 400 to 600 °C.</p> | <p>The results show WAS pyrolysis as auspicious technology for HM immobilization in chars.</p> <p>Moreover, the results indicate the potential to enhance the quality and minimize the toxicity and associated effects of char via controlling the pyrolysis temperature.</p> | [67] |
| <ul style="list-style-type: none"> Equipment: FD reactor, Plant scale: Lab-scale, Sludge pretreatment: Torrefaction (T: 220 - 320 °C, 3.6 – 10.2 min), Mode: Fast pyrolysis, T: 530 °C, Sludge feeding rate: <1 kg h⁻¹, SRT: 5.7 min | <p>Liquid yield (12 – 34 %), non-condensable gas yield (10 %), char yield (48 %).</p> | <p>Torrefaction did not showed any clear advantages towards improving the pyrolysis liquid properties, but decreased the amount of oxygen-containing aliphatic compounds, thus it seems that this pretreatment could improve the stability of the liquid.</p> | [57] |

| Experimental conditions | Results | Remarks | Ref. |
|--|---|--|------|
| <ul style="list-style-type: none"> Equipment: continuous screw-feeding reactor, Plant scale: Lab-scale, T: 500 - 800 °C, Sludge feeding rate: 4.35 – 100 g min⁻¹, RT: 6 – 46 min | <p>Main elements in char (wt. % dry): SiO₂ (25.76), P₂O₅ (16.64), Fe₂O₃ (10.82), Al₂O₃ (11.42), CaO (8.67), SO₃ (5.72), CuO (4.92), K₂O (4.67), MgO (4.02), ZnO (1.3).</p> <p>HMs in char and bio-oil (mg kg⁻¹, presented as Char/bio-oil): Cu (1500/129), Zn (2996/38), Mn (2388/33), Cr (680/10), Ni (337/5), Pb (134/4), Cd (2/0).</p> <p>Bio-oil yield (16.69 %), gas yield (16.49 %) and char yield (65.2 %)</p> | <p>The maximum bio-oil yield was obtained at 700 °C and SRT of 23 min.</p> <p>The increase in temperature and SRT decreased the char yields.</p> <p>A great proportion of HMs retained in the chars.</p> | [56] |
| <ul style="list-style-type: none"> Equipment: microwave (MW) oven, Plant scale: Lab-scale, MW power: 900 Watt at 2450 MHz frequency, T: 400 - 800 °C, Sludge loading: 200 g, Holding time: 30 min | <p>Bio-oil yield (14–20 wt. %), gas yield (15–29 wt. %) and char yield (57–69 wt.%). The heating value of bio-oil varied from 8700 – 9200 kcal kg⁻¹. Maximum energy recovery of 54 % at 600 °C.</p> | <p>The elevated temperature decreases the yield of bio-oil and chars and increases the gaseous products.</p> <p>MW assisted pyrolysis of wet sludge is a promising approach for enabling pyrolysis oil as fuel for various applications.</p> | [59] |

2.4 Sludge gasification

Gasification converts dried WAS into combustible gases known as syngas (mainly composed of H₂, CO, CO₂ and CH₄) under partial oxidation at elevated temperatures of 700 – 1000 °C [72]. Gasification reduces the volume and fixes the HMs in the solid residues [73]. Calorific value of syngas varies from 4 to 6 MJ Nm⁻³ and is mainly appropriate as a fuel for heating, steam turbine and gas engines etc., [74, 75]. WAS gasification mechanism includes four stages: (i) drying, (ii) pyrolysis or devolatilization, (iii) combustion, and (iv) char gasification or reduction. Major reactions occurring during gasification are shown (Eqns. 3 – 11) [76, 77].

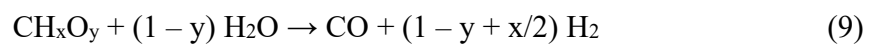
Oxidation zone reactions:



Reduction zone reactions:



Reforming reaction occurs between hydrocarbons and steam (water vapor), which results in rich H₂/CO mixture in gaseous products (Eqn. 9) whereas the hydrogen is further enhanced by water-gas shift (WGS) reaction (Eqn. 8) [78].



The end-products of WAS gasification depend on its properties (such as moisture, VM, ash, and thermal conductivity) and experimental conditions (such as temperature, HR, and feeding ratio) [79].

Table 4 shows experimental conditions and results corresponding to sludge air gasification [72, 80, 81], steam sludge gasification [72, 81, 82] and supercritical sludge gasification [83] reported in literature. These studies have shown considerable results for WAS gasification for producing hydrogen-rich syngas. However, implementation of WAS gasification confronts several challenges. For instance, higher moisture content of approximately 80 wt. % and lower heating value (LHV) of WAS are the major challenges, which results in lower gasification efficiency (GE) [84]. Whereas the dewatering process is energy-intensive (i.e. kilogram of H₂O requires about 2,260 kJ of unrecoverable energy [74]) and thus, overall cost of WAS disposal becomes considerably higher. Moreover, high tar production from WAS gasification is also another main obstacle which requires additional treatment because it can probably hinder the gasifier operation through blocking the tubing or fouling the downstream apparatus [85]. The tar removal can be accomplished through two attempts: i) the ‘primary method,’ where tar treatment is performed inside the gasifier by considering various parameters (i.e., gasifier design, temperature, gasifying agent and catalysts), and ii) the ‘secondary method,’ where tar is removed from gaseous products by means of installing equipment after the gasifier [85]. The primary method has been proven more economically appealing than the secondary one, because it avoids high costs of installing additional technology such as gas scrubbers [86]. Choi et al. [80] (**Table 4**), studied the effect of ER and activated carbon (AC) to sludge ratio on tar production. With increasing ER from 0.22 to 0.5, tar content, H₂ yield,

and HHV of producer gas were significantly decreased from 296 to 76 mg Nm³, 28.1 to 24.4 vol %, and 6.25 to 5.27 MJ Nm³, respectively. This trend has also been widely reported in literature [85, 87]. Similarly, Roche et al. [72] (**Table 4**), investigated the effect of throughput (TR), the gasifying agent, and the dolomite catalyst on tar production and gas composition. The results showed that the increasing throughput reduced the H₂ production and greatly increased the tar production. On the other hand, air plus steam, mainly in the presence of dolomite, improved the H₂ production by 20 – 30 %, reaching tar removal efficiency of ~71 %. Typical design of gasifier is also an alternative approach at primary tar removal. A recent study conducted by Molino et al. [88] highlighted an effective operation of the FD gasifier in biomass gasification, emphasizing special characteristics of FD gasifier (i.e. high mixing potential and a high tar conversion), particularly in presence of catalysts. Currently, numerous trials using the novel type or modified gasifiers have been used for tar reduction inside the gasifiers. Choi et al. [80, 89] (**Table 4**), conducted the sludge gasification with a three-stage gasifier (i.e. auger reactor, FD reactor, and tar-cracking reactor connected in sequence) and reported the least tar quantities of 22 and 27 mg Nm³ in producer gas, respectively. Another exclusive gasifier is that in which the catalytic filter components were integrated into the freeboard of the FD steam gasifier to enable the efficient tar and removal of trace elements (TEs) [90]. Moreover, Lurgi-Ruhrgas and the ChemChar waste gasification processes are considered as advanced technologies for sludge gasification, because they manage to achieve significant retaining of metals in the char matrix and high conversion of carbonaceous char residues from WAS into combustible gas [4].

On the other hand, WAS co-gasification with other carbonaceous materials entitles various benefits which advocate its utilization as a feedstock for gasification. Currently, WAS and woody biomass co-gasification have been extensively studied [73, 91-95]. The high VM, low ash and moisture content (in particular), are some of the major attributes of woody biomass that makes it an appropriate to be blended with moist WAS for gasification.

Peng et al. [73] investigated the co-gasification of wet WAS (76 % dry basis moisture content) and woody biomass (8.6 % dry basis moisture content) in a lab-scale fixed bed gasifier. The effects of different sludge mixing ratios (0 %, 30 %, 50 %, 70 % and 100 % in the test) and reactor temperatures (700 to 900 °C) were assessed on product distribution and gasification performance. The results indicated that the sludge moisture participated in the gasification with char. The addition of woody biomass to WAS compensated the moisture content and improved the VM content in the blends. An optimum H₂ and CO yield of 35.8 % and 28 % were obtained at a WAS ratio of 50 % and temperature of 800 °C. The LHV and carbon conversion efficiency (CCE) varied from 11.89 to 12.72 MJ Nm⁻³ and 57.86 to 72.21 %, respectively.

Ong et al. [94] co-gasified woody biomass with WAS in a fixed-bed downdraft gasifier. The effects of different WAS ratios (0 – 33 wt. %) and operating temperature (700 – 1000 °C) on the product distribution were studied. The results indicated that 20 wt. % of WAS ratio in the feedstock was successfully gasified to generate gaseous products comprising of >30 vol. % of syngas (H₂ and CO) with an average LHV of 4.5 MJ Nm⁻³. Further increase in the WAS ratio to 33 wt. % resulted in gasifier blockage due to the ash agglomeration.

Furthermore, Seggiani et al. [93] co-gasified dehydrated WAS (20 % w/w moisture content) blended with woody biomass in a pilot-scale updraft fixed-bed gasifier operating at the atmospheric pressure. Authors found it viable to co-gasify WAS with woody biomass. However, at high content of WAS (70 % w/w), slagging and extreme residue development in the oxidation zone could occur due to high ash content and low ash fusion temperatures of WAS, which makes the whole process unstable. It was found that increase in the ER leads to higher gas yields and cold gas efficiencies (CGE).

Similarly, Lee et al. [95] determined the optimum process parameters and their influence on gas composition via co-gasification of WAS blended with woody biomass in a FD gasifier. The WAS and woody biomass were mixed at different ratios of 50 - 100 wt.%, respectively. The temperature was varied from 600 to 900 °C. By adding the wood, H₂ and CO increased from 4 to 7 % and 11.5 to 19 % at WAS mixing ratios from 100 to 50 % and 100 to 60 %, respectively. Temperature also supported gaseous products at elevated temperatures.

Table 4. Summary of key information of WAS Gasification.

| Experimental conditions | Results | Conclusions and remarks | Ref. |
|--|---|---|------|
| <ul style="list-style-type: none"> ▪ Equipment: FD reactor (continuous feeding and ash removal mode), Plant scale: Lab-scale, T: 770 – 850 °C; Cyclone T: 450 °C; Gasifying agent: steam and air, g gasifying agent/g sludge (daf, dry and ash-free): 0.8 – 1.1, ER: 0.12 – 0.32. | <ul style="list-style-type: none"> Gas yield (0.89 – 1.32 m³ kg⁻¹ sludge), solid yield (35 – 41 wt.%), H₂ and CO (20 – 52 and 137 – 414 g kg⁻¹ sludge daf), GE (39 – 66 %), H₂/CO molar ratio (1.46 - 3.25), gas LHV (4.12 – 6.2 MJ m³). | <ul style="list-style-type: none"> Increasing reaction temperature enhanced the gas yield and the GE and reduced the tar quantities. The steam presence supported LHV of gas. | [81] |
| <ul style="list-style-type: none"> ▪ Equipment: FD reactor, Plant scale: Lab-scale, T: 800 °C, Gasifying agent: air + steam, TR: 110 – 322 kg h⁻¹ m³, Steam-to-biomass ratio (S/B): 1, Catalyst: dolomite (10 %) ▪ ER: 0.3 | <ul style="list-style-type: none"> Gas production (2.8 – 3.2 N m³ kg⁻¹ sludge daf), gas products (% dry): H₂ (9.6 – 14), CO (5.3 – 9.7), CH₄ (2.2 – 3.2), N₂ (58.5 – 63.3), Gravimetric tar yield (3.7 – 17.4 mg g⁻¹ sludge daf), Char (30 – 58.1 g kg⁻¹ daf), GE (35 – 48.8 %), LHV (2.9 – 3.9). | <ul style="list-style-type: none"> Increasing TR reduced the H₂ production and enhanced the tar yield. The catalyst increased H₂ content from 12 to 26 %. The obtained results offer enough insight on the potential development of current method to a large scale. | [72] |

| Experimental conditions | Results | Conclusions and remarks | Ref. |
|---|---|--|------|
| <ul style="list-style-type: none"> Equipment: The auger, FD and tar-cracking reactors (Three stage gasification process), Plant scale: Lab-scale, Auger reactor T: 650 °C, FD reactor T: 810 – 830 °C, Gasifying agent: air, Sludge loading: 800 g for each test, AC to sludge ratio: 0.5 – 2, ER: 0.22 to 0.5 | <ul style="list-style-type: none"> Gas production (65.2 – 77 wt.%), gas products (vol.%): H₂ (14.1 – 29), CO (9.2 – 12.8), CH₄ (3.8 – 6.4), N₂ (43.4 – 51.6), tar yield (0.2 – 1.1 wt.%), Char (11.7 – 24.5 wt.%), CGE (66.79 – 107 %), CCE (60.30 – 108 %), LHV: 5 – 6.8 MJ N m³. | <ul style="list-style-type: none"> The increasing ER significantly decreased impurities in gaseous products. The H₂ production increased by increasing AC to sludge ratio and the optimum ratio turned out to be 2/1. | [80] |
| <ul style="list-style-type: none"> Equipment: FD reactor, T: 650 °C, Pressure: 30 MPa, Alkali catalysts: KOH, NaOH, K₂CO₃ and Na₂CO₃ | <ul style="list-style-type: none"> Gas products (mol kg⁻¹): H₂ (15.49), CO₂ (9.45), CH₄ (2.2), GE (50.1 %), CCE (45 %). | <ul style="list-style-type: none"> The highest H₂ yield was achieved with KOH at 540 °C, whilst K₂CO₃ improved the GE. The increasing temperature and alkali catalyst promoted WGS reaction. | [83] |
| <ul style="list-style-type: none"> Equipment: Fixed bed gasifier, Plant scale: Lab-scale, T: 700 - 1000 °C, Gasifying agent: steam, steam flow rate: | <ul style="list-style-type: none"> Maximum syngas yield (1.14 g_{gas} g⁻¹_{solid}), Gas products (%): H₂, CO, CO₂ and CH₄ (46.9 % to 56.09, 10 to 18, 30 to 15 and 9 to 5 from | <ul style="list-style-type: none"> The increasing reactor temperature increased H₂ production. WAS produced more H₂ compared to food | [82] |

0.3 g min⁻¹, Sludge loading: 35 g for 700 to 1000 °C).

each test

[96] and paper [97] waste under same experimental conditions.

Steam gasification of WAS yielded about three times higher H₂ than that of air gasification [98].

2.5 Integration of microbial fuel cells with WAS treatment

Bioelectrochemical systems (BES), such as microbial fuel cells (MFCs) are a fascinating technology that biologically converts OM into electricity [99, 100]. MFCs are advantageous on account of their ability to directly convert OMs into electricity and their efficient operation under ambient temperature, normal pressure and neutral pH. These characteristics distinguish them from all other existing bioenergy approaches. In a MFC, electrons produced through bacteria from the OM are switched to the anode and flow to the cathode connected via a conductive substance comprising a resistor [101]. In the cathode, terminal electron acceptors (e.g., oxygen) consume the electrons to complete the electrochemical reaction. Recently, various studies have reported the successful application of MFC technology for producing bioelectricity from organic wastes (e.g., WAS) [102-104]. MFCs significantly decrease the adverse environmental impacts associated with conventional WAS treatment (e.g., incineration and landfill) [105]. Dentel et al. [106] first demonstrated the direct generation of electricity from sludge via MFC. They used anaerobically digested sludge and achieved a maximum voltage of 517 mV [106]. Thenceforth, the concern of addressing an enhanced power generation from WAS has become the key research direction [99, 102, 103, 107, 108], and the power output densities increased from 2004 to 2017 (0.1 to 227 W/m³) [108]. In addition to power generation, sludge digestion is also of utmost significance to make MFC technology competent. Ghadge et al. [109] have reported the simultaneous sludge degradation of 81 % and power generation (8.7 W m³) in MFC with sodium hypochlorite as catholyte within short RT of 8 days. Moreover, as compared to other fermentable materials (e.g., glucose, acetic acid and lactic acid) typically exploited in MFCs, WAS

composition is complex, slowly biodegradable, and the persistent biomaterials/organics are challenging to be consumed efficiently [110]. Thus, to make these refractory organics more susceptible to effective microbial degradation pretreatment of WAS is absolutely imperative [111]. Today, many pretreatment methods such as alkaline pretreatment [112-114], ultrasonic pretreatment [103, 111, 113], aerobic digestion [107], thermal pretreatment [107], microwave digestion [115], ozonation [115] have been explored to enhance the electricity generation via WAS in MFCs. Jiang et al. [103, 111, 113] investigated the ultrasonic and alkaline pretreatments. Both methods accelerated the OM dissolution leading to remarkable total COD removal rate. Xiao et al. [107] and Yuan et al. [116] studied the base and heat pretreatments as an effective alternatives to accelerate the hydrolysis, and thereby increasing electricity production. However, recent findings have shown the possibility of destruction of the original microbial consortia via base and heat pretreatments, indicating that inoculation would be essential in BES [107]. More recently, Oh et al. [117] evaluated the influence of ultrasonic and heat/alkaline pretreatments with varied sources of sludge (such as primary sludge, WAS and AD sludge) for electricity production in MFCs. Although the aforementioned pretreatments are efficient in liquefying WAS and promoting overall system performance, they also increase the operational cost of MFCs. Thus, more cost-effective approaches as recently adopted by Suor et al. [99] and Cia et al. [118] should be further researched to increase electricity generation from WAS via MFCs.

3. COMPARATIVE STUDY OF THE LEADING TECHNOLOGIES FOR SLUDGE DISPOSAL

The comparison among well-known technologies for WAS is depicted in **Table 5**. On the account of stringent flue gas emission requirements, WAS incinerators require advanced emission control systems [70]. Additionally, WAS co-firing with coal and/or biomass appears a favourable and encouraging option. The pyrolysis of WAS is a more complex technology when compared with incineration. However, compared to incineration, pyrolysis entitles potential benefits such as zero waste method, the lower costs, less environmental impact and much lower dependency on ever diminishing fossil fuel reserves [70].

Moreover, these versatile benefits endow WAS pyrolysis with adequate capacities to be stepped up for commercial applications in the near future. Nonetheless, enhancement of energy conversion efficiency is the major consideration and an area of concern for WAS pyrolysis. Conversion technologies like WAS pyrolysis and gasification are still in embryonic stages. Several of the claims such as pollution-free and economic viability for these technologies are based on laboratory and pilot scale demonstration projects. The real problem with these approaches is that the plants fail to meet the increasingly stringent air quality and emissions standards when scale up to real world applications or outside of the laboratory [70, 119, 120]. In addition, some other limitations on WAS technological advancement are as follows:

- Improper matches among the risk bearing capabilities of suppliers, clients, and investors. Generally, they are not able or reluctant to realize/accept any levels of

the probable risks associated with a project if the technology applied is unguaranteed or unproven.

- The obstacles to secure safe investment for technologies with restrict operating experience and/or without any proved track record.
- For WAS treatment and disposal, low gate fees are enumerated as one of the major inhibitors for all types of waste management projects and the inhibition is mostly linked to the utilization of capital intensive plant and equipment, indeed.
- Considering unreal expenses of scarce resources and other benefits as the priority rather than actual effort and focus on developments that might result in real advantages.

Table 5. Comparative analysis of leading sludge treatment technologies [4, 14, 23, 70, 104, 121-125].

| Technology | Factors | Recognized benefits | Limitations |
|---------------------|--------------------------|---|---|
| Incineration | Technological | <ol style="list-style-type: none"> 1) Well-established and reliable 2) About 90 % WAS volume reduction 3) Almost complete destruction of toxic organic components and pathogens 4) Heat recovery to power the steam turbine and WAS drying) | <ol style="list-style-type: none"> 1) Dewatering of the sludge is required (41–65 wt. % content of dry material) 2) Incineration can be energy deficient 3) Far from Zero Waste method |
| | Social- Environmental | <ol style="list-style-type: none"> 1) Low GHG emissions compared to open burning 2) Co-combustion with other conventional fuels such as coal can reduce GHG emissions and public acceptability thereof | <ol style="list-style-type: none"> 1) Strong Public’s opposition 2) Mono-incineration: 232.2 kg CO₂ eq./t sludge 3) Co-incineration with municipal solid waste (MSW): (–15.4 kg CO₂ eq./t sludge) 4) NO_x and SO₂ emissions, slag, TEs, fly ash and ash disposal |
| | Economic | <ol style="list-style-type: none"> 1) Energy savings 2) Existing infrastructure of energy kilns 3) Co-firing with other conventional fuels | <ol style="list-style-type: none"> 1) Strict and expensive emission control 2) High cost of the flue gas cleaning and ash disposal problems (HMs etc.) |

| Technology | Factors | Recognized benefits | Limitations |
|-------------------|---------------------|---|--|
| Pyrolysis | Technological | <ol style="list-style-type: none"> 1) It converts both raw and digested WAS into useful bioenergy. 2) Non-burning and zero waste process 3) Bio-oil: used to produce various chemicals and even as a fuel 4) Biochar (HHV of WAS is $\sim 5 \text{ MJ kg}^{-1}$) as a byproduct (holds potential for carbon sequestration and soil conditioning) | <ol style="list-style-type: none"> 1) Complex when compared with incineration 2) Dewatering requirements limits the WAS pyrolysis applications 3) Products have not very well established markets 4) Pyrolysis gas needs less treatment to meet emission limits than incineration 5) Limited technological acceptance by the low economic value of the produced oil |
| | Socio-Environmental | <ol style="list-style-type: none"> 1) Lowest GHG emissions than incineration due to the lower temperatures and oxygen absence 2) Lower temperatures are also responsible for the HMs absence in the pyrolysis gas 3) Turns a waste into a valuable raw material | <ol style="list-style-type: none"> 1) Air pollution 2) HMs remain trapped in the resulting char, those require costly treatments, without which their landfilling is not possible due to legal constrains |
| | Economic | <ol style="list-style-type: none"> 1) Viability is proven only in large scale plants ($>20,000 \text{ tons year}^{-1}$) 2) Sustainable development and green entrepreneurship | <ol style="list-style-type: none"> 1) High investment cost 2) Unstable economic environment |

| Technology | Factors | Recognized benefits | Limitations |
|---------------------|---------------------|---|---|
| Gasification | Technological | <ol style="list-style-type: none"> 1) Turn various types of waste into useful energy 2) Potential of co-feeding with biomass 3) High energy efficiency 4) Marketable products: WAS conversion to a combustible gas (i.e. syngas), which significantly contributes to produce methanol. 5) Char or slag, oils and water as byproduct 6) Avoids problems such as supplementary fuel, SO_x and NO_x emissions, fly ash and HMs etc. 7) Energy self-sustaining | <ol style="list-style-type: none"> 1) WAS water content required (<10 % dry solids (DS) content) 2) WAS dewatering and drying is essential 3) Gas cleaning for syngas applications 4) HHV of WAS syngas reaches ~4 MJ m⁻³, which is suitable for electricity/heat |
| | Socio-Environmental | <ol style="list-style-type: none"> 1) Lower environmental impacts 2) Turn a waste into energy | <ol style="list-style-type: none"> 1) Heavy organic pollutant compounds in the exhaust stream |
| | Economic | <ol style="list-style-type: none"> 1) Economies of scale and automation favor large scale operations 2) Green Entrepreneurship 3) Sustainable development | <ol style="list-style-type: none"> 1) High investment and operation cost |

| Technology | Factors | Recognized benefits | Limitations |
|---------------------------------|---------------------|---|--|
| Anaerobic digestion (AD) | Technological | <ol style="list-style-type: none"> 1) Accepts organic feedstocks with high moisture content (80 – 90 %), thus suitable for WAS. 2) Methane-rich and high calorific value biogas 3) Combined heat and power plants. | <ol style="list-style-type: none"> 1) Lengthy reaction steps 2) Low conversion efficiency by microbe and/or enzyme 3) The HMs and Persistent organic pollutants (POPs) contained in WAS cannot be alleviated via AD |
| | Socio-Environmental | <ol style="list-style-type: none"> 1) Significant reduction in carbon emissions 2) Creates jobs and increases skilled labor force 3) Replaces chemical fertilizer with organic fertilizer for arable land 4) Decreases use of firewood, helping to combat deforestation | <ol style="list-style-type: none"> 1) Digestate resulted from AD would have impacts on the public health and environment if not treated properly. |
| | Economic | <ol style="list-style-type: none"> 1) Digestate is energy profitable due to its organic matter 2) Reduces transport and disposal costs 3) Cost-effective fertilizer 4) Green Entrepreneurship 5) Sustainable development | <ol style="list-style-type: none"> 1) High capital and maintenance costs |

| Technology | Factors | Recognized benefits | Limitations |
|---|-------------------------|---|---|
| WAS- microbial fuel cell | Technological | 1) It is a clean technology (no combustion) 2) Direct conversion of a great variety of organics into electricity 3) Efficient operation under ambient temperature, normal pressure and neutral pH. 4) Sludge stabilization and volume reduction 5) Noise free operation | 1) Low power output. 2) Pretreatment requirement due to complex structure of WAS 3) The scale-up is still a big challenge 4) Biological fouling 5) In some cases, (i.e. phototrophic MFCs) artificial illumination is required 6) The lack of electrolytes buffer capacity drastically impediment scaling up of MFCs for WWT |
| | Socio- Environmental | 1) Considerable reduction of environmental impacts associated with WAS treatment. 2) Bioenergy in the form of electricity. 3) It diminishes the competition with food production as is the concern with conventional biofuels. | |
| | Economic | 1) Energy saving. 2) Reduced maintenance cost since not moving parts. | 1) High operational (i.e. pretreatment) and material costs (i.e. membrane materials, Pt catalyst and |

| | | | |
|--|--|-----------------------------|---|
| | | 3) Sustainable development. | bipolar plates etc.). 2) Additional costs for artificial illumination. |
|--|--|-----------------------------|---|

AD is the most prevalent sludge disposal route as it is not affected by high water content of the sludge. However, it suffers from long processing time, improper removal of the OM and low efficiency. Incineration has high efficiency and can be employed to generate power. However, flue gases require expensive treatment unlike AD before they are vented out to the atmosphere. Other thermal treatments such as gasification and pyrolysis have important benefits as they generate syngas and bio-oil, respectively, which can be used for diverse applications, but they suffer from low efficiency and high costs of WAS pretreatment to reduce moisture content. Developments in the technologies such as AD coupled with pyrolysis, co-combustion and co-incineration are needed to diminish the energy penalty and improve the process economics. This will in turn aid in the scale up of pilot plant studies to commercial level applications.

4. CO-PRODUCTION OF BIOREFINERY PRODUCTS FROM SEWAGE SLUDGE

4.1 Enzymes

Three major biochemical families (carbohydrates, proteins and lipids) of WAS represent approximately 80 % of the OM [3]. Thus, the recovery of various enzymes (e.g. lipases, dehydrogenase, glycosidase, peroxidase, and aminopeptidases) from WAS is essential on account of their high value. Nabarlitz et al. [126] studied various enzyme extraction methods. The obtained results proved ultrasonication alone or supported with additives as an effective approach for enzyme recovery methods at the operating conditions listed in **Table 6**. They also suggested that enzymes help to enhance the bio-digestibility of OMs towards higher biogas production during AD and so forth [126]. Likewise, according to

Guanghui et al. [127] (**Table 6**), using ultrasonication supported by ethylenediaminetetraacetic acid recovered various enzymes from WAS. The method demonstrated an effective recovery of the hydrolytic enzymes (e.g. lipase and protease).

More recently, Ni et al. [53] (**Table 6**), utilized WAS to extract the hydrolytic enzymes. According to observations, the produced enzymes appear to be an appropriate compound enzyme for feed.

Generally, enzymes are biological catalysts that upsurge the rate of chemical reactions and are employed commercially for various industrial applications such as pharmaceutical, food, diagnostics, cosmetics, detergent and fine chemical industries. With regard to their wide commercial significance, reliable and economical supply of these enzymes is a key endeavor on account of the fact that 30 – 40 % of the production costs in aforementioned industries attribute to the cost of preparing culture medium [128]. Therefore, WAS-derived enzymes could potentially lessen the industrial processing time and cost. Therefore, holistic research is required to optimize the enzyme recovery along with techno-economical assessment of the processes. The enzymes produced are appropriate for certain industrial applications (e.g., as additives for detergents or bio-catalysts in esterification reactions); however, such applications demand high purity and specific enzymatic activity, and therefore further purification and concentration procedures are usually required.

4.2 Bio-plastics

The plastics are commonly derived from petrochemicals. However, the environmental and human health problems related to the application of conventional plastics

(petrochemical derived plastics) have motivated researchers to develop the biodegradable alternative.

Besides, other polymers such as polyhydroxyalkanoates (PHAs), which exhibit properties comparable to petroleum based plastics, are a group of biodegradable polymers (linear polyesters) produced via microorganisms using carbon as a substrate [129]. Poly-beta-hydroxybutyric acid and its co-polymer poly (3-hydroxybutyrate-co-hydroxyvalerate [P(3HB-co-HV)]) are very common PHAs [130].

Several microorganisms in WAS have the ability to accumulate PHAs (0.30 to 22.7 mg polymer g-sludge [131]); however, high production cost (i.e. \sim USD 4 – 6 kg⁻¹ compared to USD 0.6 – 0.9 kg⁻¹ for conventional plastics) [129, 132] has limited their widespread uses. The current advances in bacterial fermentation and WAS use are likely to improve the yield and diminish the process costs of PHAs production [129, 132-135].

The excess carbon and limitation approach under ‘feast and famine’ conditions has been observed promising for the effective cultivation of PHA yielding organisms. In addition, Longo et al. successfully exploited nitrogen removal strategy to obtain higher yield of PHA during the PHA production step [136]. However, being aerobic, these processes are energy intensive (1 kg PHA production requires \sim 39 MJ), when the aerobic accumulation step is used [137]. **Table 6** presents the experimental approaches applied for the extraction of PHAs. Jiang et al. [138], Morgan-Sagastume et al. [139], and Pittamnn et al. [135] (**Table 6**) examined the potential for the enhancement of PHA producing communities and the PHA accumulation capacity of fermented sludge under varying experimental conditions. The obtained results demonstrated the successful enrichment of PHA-storing communities by means of the fermented sludge as feedstock,

albeit excessive nutrient loads did control the maximum level of PHA accumulation [139]. Morgan-Sagastume et al. [139] also scrutinized a novel concept for the operation of WWTP, where the variety of PHA accumulating organisms were successfully supplemented based on the conversion of the readily biodegradable COD from municipal wastewater without its pre-conversion to VFAs [140]. On the other hand, post-treatment was necessitated, especially for higher nutrient removal of the treated effluent [141].

Recently, a pilot-scale study conducted by Bengtsson et al. [140] demonstrated PHA accumulation potential integrated with biological nitrogen removal through pre-denitrification, nitrification and post-denitrification. The obtained PHA accumulation potential was comparable to what has been formerly reported by employing a completely aerobic ‘feast and famine’ process [141]. Therefore, based on above results, nitrogen removal could be assimilated into the enrichment process without diminishing the PHA accumulation potential. But, this is directly contrary to what has been reported in literature [142]. Moreover, Bengtsson et al. [140] also confirmed previously reported findings of high PHA accumulation using conventional domestic wastewater regardless of low to negligible concentrations of influent VFAs [141]. Alternatively, municipal WWT has been using streams with high VFAs resulting from fermentation of solids to produce enhanced biomass along with high PHA accumulation [135, 143, 144], which has been reported as unnecessary [140]. Therefore, the conversion of highly concentrated VFA-media into only polymers seems to be a good choice rather than biomass and polymers. This will in turn lead to a higher overall PHA potential. Despite the clear technical potential of PHAs in sludge, it is not yet economically appealing [129, 145].

Therefore, to make sludge-derived PHAs more attractive in future, holistic research approach aiming to improve the yields through process integration, parametric optimization, and PHAs storage capacity, etc., are necessary. Such aspects will boost the confidence and effectiveness in the industrial fraternity about the economic and commercial viability of the technology.

4.3 Bio-pesticides

Currently, *Bacillus thuringiensis* (*Bt*) is by far the most effective bio-pesticide, which owns the ability to produce delta endotoxins (δ -endotoxin, known as cry and cyt toxins), being utilized widely in agronomy, forestry and public sector [3, 146]. Moreover, bio-pesticides are highly target specific compounds with zero toxic residues, and therefore have a much lesser impact on the environment compared to chemical pesticides. However, high cost associated with raw materials has impeded the commercial application of *Bt* to a great extent [147]. The conventional fermentation medium for *Bt* based bio-pesticides production carries a significant portion of ~40 – 60 % of the total production cost [148, 149]. Therefore, it has become important to explore a new raw material considering some criteria which should include i) renewable in nature, ii) high yielding potential, iii) cost-effectiveness, and iv) year round availability for *Bt* production. With these, WAS appears to be highly nutritional and cost-effective medium for *Bt* production [150, 151], which will substantially reduce the *Bt* production cost and will contribute to sustainable WAS utilization and management. WAS based bio-pesticide production undergoes three steps namely, i) WAS fermentation, ii) harvesting and/or product recovery, and iii) product formulation. The use of sludge for *Bt* production has been widely studied as shown in **Table 6** [147, 152, 153]. These studies have

demonstrated that *Bt* based bio-pesticides production highly depends on fermentation conditions such as pH, C/N ratio, dissolved oxygen concentration, solids concentration and inoculum sludge type. Montiel et al. indicated that more than 50 % of net savings can be achieved by substituting commercial medium for producing *Bt* with WAS (\$0.25-0.34 L⁻¹ sludge medium vs \$0.75 L⁻¹ commercial medium) [152, 154]. Attending a higher entomotoxicity (Tx) level could lead to further decrease the *Bt* production cost [152]. Zhuang et al. [147] (**Table 6**), showed higher yields of Tx corresponding to different substrates (i.e. sludge + wheat and commercial medium) compared to sludge, indicating the reliance of Tx production efficiency on the type of medium. The lower Tx productivity in sludge sample could be attributed to the complex nature of sludge and thus less biodegradability. Vidyarthi et al. [155] indicated the availability of biodegradable OMs in the substrate was very crucial limiting factor for obtaining a high yield of Tx and δ -endotoxin. Therefore, the suitable pretreatment to alter less biodegradable materials into more easily degradable ones is highly recommended [156, 157]. Ultrasonication as sludge pretreatment has been introduced as a potential approach to improve the sludge biodegradability [158]. However, recently, Fenton oxidation pretreatment was proven to be more effective for growth of sludge based *Bt* production compared to ultrasonication pretreatment, proposing that further research on *Bt* production using above pretreatment in large scale reactor are needed [153]. Zhuang et al. [147] also demonstrated the feasibility of producing *Bt* based bio-pesticides using WAS as raw materials under solid-state fermentation technology. The effect of HMs [Pb (0 – 300 mg L⁻¹), Cu (0 – 150 mg L⁻¹), Cd (0 – 30 mg L⁻¹), Cr (0 – 300 mg L⁻¹)] on *Bt* production was examined. The highest *Bt* production corresponding to Pb, Cu, Cd and Cr

was found to be 227, 82, 15, and 263 mg L⁻¹, respectively. The high contents of HMs had considerable negative influence on the growth, crystal formation and toxicity of *Bt*. The extent of metal toxicity to *Bt* production caused 50 % inhabitation in total cell (TC) biomass of *Bt*. Among the examined metals, *Bt* exhibited the utmost tolerance against Cr (III), followed by Pb(II), Cu(II) and Cd(II).

Thus, sludge based *Bt* production followed by its use to forests and in particular, agro-crops for pest control appears to be entirely compatible with up-to-date sludge disposal practices (e.g. zero or minimal transportation cost). However, the aforementioned methods mostly suffer from low *Bt* productivity and complex nature of sludge. Therefore, further studies focusing on downstream processing (i.e., parametric optimization, process control, and product separation/purification) are required to enhance *Bt* production into the global pesticide market. This will also contribute to sustainable sludge utilization and management by developing the final *Bt* formulation, and consequently expand the repertoire of commercial *Bt* product types.

4.4 Proteins

The major organic constituents of WAS are polysaccharides, proteins and lipids [159, 160]. Noticeably, due to the higher portion of proteins (~50 % dry weight of bacterial cells), WAS has high potential to be used as a protein source [3, 161]. On another side, protein is an essential component used in animal feed for supplying energy and nitrogen [160]. Thus, protein recovery from WAS offers various benefits over traditional protein recovery sources. For instance, destruction of microbes can result in increased dewaterability, and the protein removed WAS would take less volume and inhibit limited OMs.

Many significant studies on protein extraction and analysis from WAS have been published [160-164], as listed in Table 6. Hwang et al. [163] disintegrated WAS (5,330 mg L⁻¹) for the intracellular protein recovery assisted by an alkali treatment coupled with ultra-sonication under the parametric conditions elucidated in **Table 6**. They reported protein extraction rate as high as 80 % at an optimum pH of 3.3, with comparable nutrimental composition as of commercial protein feeds and thus suitable as an animal feed supplement. Similarly, Jimenez et al. [161] (**Table 6**) investigated the efficiency of different colorimetric methods such as Lowry, modified Lowry, and the Bicinchonic Acid (BCA) to characterize the OM (e.g. protein, carbohydrates and lipids) contents of the WAS. The protein was found as a main component representing 50 % of biochemical portion.

Table 6. Summary of key information of WAS bio-refinery.

| Bio-refinery | Experimental conditions | Results | Remarks | Ref. |
|--------------|--|--|---|---------------------------|
| Enzymes | <ul style="list-style-type: none"> ▪ Methods: (1) Stirring: 200 mL sample in reactor, T: 5 °C, stirrer rpm: 500 – 1500; (2) Ultrasonication: 200 mL sample in vessels, T: 5 °C, power intensity: 8 W cm², frequency: 20 kHz and extraction time: (1 – 10 min), enzymes: protease and lipase, additives: non-ionic detergent (NID), resin and buffer. ▪ Method: Ultrasonication: T: 4 °C, power intensity: 138 – 690 W g⁻¹ TSS, frequency: 20 and 40 kHz, extraction time: 2 - 20 min, enzymes: protease, α-amylase, α-glucosidase, alkaline phosphatase and acid phosphatase | <p>Ultrasonication produced 52.9 units g⁻¹ VSS of protease (NID: 2% v/v, 10 min) compared to 57.4 units g⁻¹ VSS (NID: 2% v/v, 1 h stirring) with stirring method. In case of lipase, ultrasonication produced 21.4 units g⁻¹ VSS of lipase (NID: 2% v/v, 20 min) compared to 15.5 units g⁻¹ VSS (10 mM buffer, pH 7.5 + 0.48 g mL⁻¹ CER + 0.5 % NID) with stirring method.</p> <p>An optimum extraction corresponding to all enzymes was obtained at an intensity of 552 W g⁻¹ TSS, a frequency of 20 kHz, and a time of 10 min.</p> | <p>Ultrasonication method was found to be appropriate for extraction of enzymes from WAS. Moreover, this method could be scaled up for industrial application.</p> <p>The optimized extraction parameters can disintegrate cells and extricate both the extracellular and intercellular enzymes from WAS.</p> | <p>[126]</p> <p>[127]</p> |

| Bio-refinery | Experimental conditions | Results | Remarks | Ref. |
|--------------|--|---|---|-------|
| | <ul style="list-style-type: none"> Method: Ultrasonic combined with stirring: 25 g sample suspended in phosphate-buffered saline (25 mL, pH 6.8) or distilled water, T: 4 °C, 10 – 60 min, enzymes: protease, lipase, amylase and cellulases, Triton X-100: 0.5 – 0.2 %. | <p>An enzyme product of 2534, 1, 1150 and 2340 units g⁻¹ corresponding to protease, lipase, amylase, and cellulases activities was obtained.</p> | <p>The obtained enzyme product has high market potential as an animal feed.</p> | [53] |
| Bio-plastics | <ul style="list-style-type: none"> Method: Aerobic: T: 21 °C, plant scale: lab, mode: batch, working volume: 4 L, oxygen source: air, WAS alkaline fermentation liquid to tap ratio: 1:4, Influent pH: 7, sludge retention time: 2 h | <p>The maximum PHA concentration was reached 73 %.</p> | <p>The presence of N and P did affect the PHA production, indicating that the removal of N and P is an unnecessary, while producing PHA from fermented WAS.</p> | [138] |
| | <ul style="list-style-type: none"> Method: Aerobic or feast-famine: T: 35 °C, plant scale: lab, mode: batch, operating time: 90 days, working volume: 3 g TSS L⁻¹, oxygen source: air (10 – 20 L min⁻¹), organic loading rate: 6 – 12 g COD L⁻¹ d⁻¹ | <p>A maximum PHA: ~25 g-g TSS⁻¹</p> | <p>The fermented WAS based PHA production seems viable. However, more studies on parametric optimization are required to enrich the accumulation of PHA.</p> | [139] |

| Bio-refinery | Experimental conditions | Results | Remarks | Ref. |
|----------------|---|--|---|-------|
| | <ul style="list-style-type: none"> Method: Aerobic: T: 15 - 30 °C, plant scale: lab, mode: batch, reactor operating time: 90 days, substrate concentration (SC): 1200 - 2000 mgVFA L⁻¹, pH: 6 – 9, cycle time (CT): 24 – 72 h. | <p>A highest PHA production of 25.9 % (20 °C, 1200 mgVFA L⁻¹, pH 7) and 28.4 % cell dry weight (24 h, pH 8) was achieved.</p> | <p>The PHA production was highly affected by temperature, pH, SC and CT. Lower SC, 20 °C, neutral pH-value and 24 h CT were found best for high production of PHA.</p> | [135] |
| Bio-pesticides | <ul style="list-style-type: none"> Method: Fermentation: STR bioreactor, <i>Bt</i> fermentation profile: volume: 15 L, time: 3 – 48 h, air flow: 5 – 20 L min⁻¹, agitation rate: 150 – 1350 rpm, DO: 95 – 35 % | <p>A maximum Tx: 16,552 SBU μL⁻¹</p> | <p>The Tx was directly associated with the protease activity.</p> | [151] |
| | <ul style="list-style-type: none"> Method: solid state fermentation: Fermentation: substrate (4 types): (i) sludge (SL), (ii) sludge + wheat roughage (SW); (iii) sludge + straw powder (SS); (iv) commercial culture (CM), pH: 7.2, CT: 80 h. | <p>The mixture of SW (ii) gave the maximum production of viable cells, spore counts, toxin and Tx of 5.98 10¹⁰ CFU g⁻¹, 5.26 10¹⁰ CFU g⁻¹, 7.14 mg g⁻¹ and 4758 IU mg⁻¹, respectively.</p> | <p>The solid culture media (SL, SW SS and CM) supported <i>Bt</i> production under solid state fermentation.</p> <p>The highest Tx during the fermentation of SW and CM indicated that Tx potency highly dependence on the type of substrate.</p> | [147] |

| Bio-refinery | Experimental conditions | Results | Remarks | Ref. |
|--------------|--|---|--|-------|
| | <ul style="list-style-type: none"> Methods: (1) Fenton oxidation (FO): sludge: 300 mL, acidification pH: 3 with 10 N H₂SO₄, H₂O₂: 30 % v/v and FeSO₄ 0.01 g L⁻¹, rpm: 150; (2) Ultrasonication: sludge: 400 mL, autotune homogenizer: 750 W, frequency: 20 kHz. | <p>The highest biodegradability of 74 % was achieved. Whilst, a maximum TC count and sporulation of <i>Bt</i> growth in FO was achieved to be 1.63×10^9 and 96 %, respectively.</p> | <p>FO method using sludge was shown to be rather more efficient for <i>Bt</i> production using sludge when compared with ultrasonication.</p> | [153] |
| Proteins | <ul style="list-style-type: none"> Methods: (1) Enzymatic: sludge + distilled water ratio (1:6) + alkaline protease (2 % w/w), pH: 8; (2) Acid: pH sludge 0.5 with HCl (2.0 M), sludge agitation at 121 °C for 5 h; (3) Alkaline: sludge pH 13 (NaOH, 1.0 M) and sludge agitated at 140 °C for 3 h. | <p>A maximum of 58.7 % protein was recovered. The alkaline was found to have highest contents (860.1 ng g⁻¹) of polycyclic aromatic hydrocarbons (PAHs), in proteins compared to acid (451.9 ng g⁻¹) and enzymatic (213.7 ng g⁻¹) methods.</p> | <p>Within all methods applied, enzymatic method was an efficient and eco-friendly to remove the PHAs contained in protein for the production of animal feed.</p> | [160] |
| | <ul style="list-style-type: none"> Method: (1) Alkali combined with ultrasonication: sludge pH: 12, NaOH h (1.0 M), stirred: 2 h, frequency: 20 kHz, input energy: $0 - 3.84 \times 10^{10}$ kJ kg⁻¹ VSS. | <p>A maximum protein extraction rate of 80.5 % was reached.</p> | <p>The recovered protein contained comparable nutritional composition to those of Brewer's yeast and bone meal, indicating that it can</p> | [163] |

technically be used as animal feed.

- Methods: colorimetric methods (Lowry, The protein was found as a main The selected methods for [161] modified Lowry, and the BCA): sludge: 0.5 component representing 50 % of characterization of OM exhibited to and 0.1 mL, linearity for Lowry and the biochemical portion. be appropriate for WAS analysis modified Lowry: 0 to 100 mg_{BSA} L⁻¹, with a good recovery of the VMs. linearity for BCA: 0 – 2000 mg_{BSA} L⁻¹.
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5. RESOURCE RECOVERY AND POLLUTION CONTROL FOR SEWAGE

SLUDGE

5.1 Nutrients

WAS contains considerable amounts of nutrients, particularly P total solids (TS, 0.5 – 0.7 %) and N (2.4 – 5.0 % TS) [3], these nutrients exist primarily in the form of proteinaceous material. The disintegration and solubilization of WAS and its later transition to ammonia and phosphates could possibly lead the way to produce magnesium ammonium phosphate (struvite), which is an excellent plant fertilizer and can be employed directly for land application [165-167].

About 1 kg N and 1 kg P in commercial fertilizer costs approximately US \$ 1.3 and US \$ 2.6, respectively. The DS of WAS possesses ~ 1 kg P and 1.2 kg N inhabitant⁻¹ year⁻¹ corresponding to a cost of nearly US \$ 4.2. Regarding P recovery from WAS, the cost has been reported around 1.6 to 3.0 times higher than the commercial P in different studies [168, 169].

On the other hand, P is a limited non-renewable resource as there will be no P left in apatite mines within 150 years [3]. Therefore, it must be considered as a highly valued asset/product and recovered efficiently from WAS.

Recently, significant efforts have been made toward P recovery from WAS based on the precipitation of phosphoric minerals in the form of struvites, hydroxyapatites or calcium phosphates [170, 171]. These P-recovery technologies have been developed and put into operation largely in the Japan and Netherlands [170, 172]. However, their high operational cost US \$ 52,100 year⁻¹ (**Table 7**) for struvite recovery with low

reimbursement level of US \$ 16,100 – 21,200 year⁻¹ has declared them outdated and economically unjustifiable [173, 174]. Some researchers claim that struvite could be extracted via biological nutrient removal process before WWT, resulting in 80 – 85 % of P recovery [174]. While others claim that struvite extraction will never be reasonably efficient because of the high ammonia demand [175]. However, recently, Wang et al. [176] studied MW and NaOH techniques for WAS treatment to release and recover the P, for application as a fertilizer. The MW and NaOH released 34.2 – 43.73 % of total P. At pH 9.5 and an Mg:P ratio of 1.5:1, 23.48 – 32.07 % of the total P was recovered as a result of crystallizing struvite. The total process cost to treat WAS was found to be \$85 – 103 DT⁻¹ WAS. Therefore, more research on struvite extraction is required to counter aforementioned uncertainties.

The P-recovery from WAS incineration ashes is also under investigation nowadays [177-180]. These ashes contain high amounts of P, mainly due to the considerable volume reduction of up to 90 % [181]. Generally, the P content in WAS varies from 1 – 5 % (may reach up to ~15 % if any suitable technology is employed), whereas its content in ashes varies from 5 – 11 % (more than 20 % in specific case) [34, 169]. In addition, Cieřlik et al. [174] stated that P-recovery could be 5 – 10 times higher from ashes compared to that from WAS directly, but unfortunately viability of such technologies is proved only in large scale treatment plants, primarily because of the need to incur large capital costs associated with building a facility which meets all the environmental standards for WAS incineration. Le et al. [180] demonstrated the reuse of P resources from WAS ash in a pilot scale circulating FD kiln . The sample was mixed with 2 – 10 wt. % of calcium oxide (CaO). The temperature was varied from 750 – 950 °C. The P

released from WAS was deposited in bottom ash (89.29 %) and fly ash (5.61 %), respectively. Moreover, results suggested that higher temperature and more dose of CaO were advantageous for the conversion of non-apatite inorganic phosphorus to apatite P.

On the other hand, Shih et al. [171] treated synthetic wastewaters containing high concentrations of P via crystallization of struvite in a FD crystallizer. The higher phosphate removal and crystallization ratio of 96 % and 93.5 %, respectively, were obtained at optimum conditions, namely pH of 9.5, molar ratio Mg/N/P of 1.3/4/1, struvite seed dose of 30 g L⁻¹, total flow rate of 12 mL min⁻¹, and reflux of 120 mL min⁻¹.

Similarly, other investigations have also reported a remarkable P content of >95 % and ammonia >50 % solubilization via various combined thermochemical treatments [182-184]. Guedes al. [181] and Couto et al. [185] presented a viable method for P-recovery by an electro dialysis approach from WAS. The process gave a total yield of 30 – 85 % including some amounts of gypsum, thus necessitating further research to evaluate the opportunity of utilizing residues as dopants for construction materials.

The technological advancement refereeing to commercial scale P-recovery includes Aqua Reci (AR), OSTARA, SUSAN and KREPO, which are based on physicochemical and thermal treatment to dissolve P and then recover it by precipitation. The AR process, a commercial technology, has been established in Sweden to recover both P and energy using combined Supercritical Water Oxidation technology. The process can efficiently recover ~100 % of P by means of HCl and H₂SO₄ with 2 h of reaction time and at 90 °C [186]. Another process called OSTARA has been employed to recover P from WAS stream using magnesium chloride (MgCl₂) at the city of Edmonton, Canada. The process was established in 2007, and is expected to produce 200 – 250 Mt year⁻¹ of struvite [3,

187, 188]. A commercial scale plant (45,000 m³ day⁻¹ treatment capacity) with the technology for P-recovery (> 90 %) as struvite (about 550 kg day⁻¹; equivalent to 0.01 kg struvite m⁻³) has been in operation at the Lake Shinji East Clean Center in Japan [189]. Moreover, the Crystalactor[®] technology, was commercialized in Netherlands. However, this technology was challenged by high cost of P-recovery and thus considered as uneconomical [190]. The high cost of P-recovery from WAS is considered the major obstacle in the scaling up of the explored technologies. As a result, it is likely that a new technology should be proposed that may allow not only the recovery of valued phosphoric material, but also all-inclusive management of all wastes produced during WAS thermal treatment. The limitations and benefits of presented P recovery approaches from sludge are presented in Table 7.

Table 7. Recognized benefits and limitations of selected methods for P recovery from sludge [174, 191-195].

| P recovery methods | Process sub-classification | Recognized benefits | Limitations |
|--|--|--|---|
| Direct use of WAS in agricultural land | <ol style="list-style-type: none"> 1) Stabilization via earthworms 2) Composting 3) Stabilization in pools 4) Drying and pellet production | <ol style="list-style-type: none"> 1) Low cost technologies, even if the small quantities of WAS is used. 2) The management of all sludge with lower HMs concentration is possible if earthworm stabilization is employed. | <ol style="list-style-type: none"> 1) Long stabilization periods at lower temperature. 2) Risk of environmental pollution through high organic carbon loads, pathogens and parasites, aromatic hydrocarbons and HMs. 3) Limited applications (i.e. soil conditioning and fertilizers). 4) Based on soil remediation strategy, these technologies are opposed by the EU. |
| WAS incinerated ashes | <ol style="list-style-type: none"> 1) Incineration 2) Acidic extraction 3) Thermochemical treatment 4) Cementing | <ol style="list-style-type: none"> 1) Partial reimbursement of expenditures. 2) Highly efficient WAS management with inspiring P recovery. 3) Substantial savings related with sludge disposal, with | <ol style="list-style-type: none"> 1) High operational cost 2) Risk of environmental pollution through HMs, organic pollutants and chlorinated species. 3) Complex procedures. 4) Obstacles with achieving high strength building materials. 5) The technology is economical only if large amounts |

| | | | |
|-----------------------------|--|--|--|
| | | wide variety of applications. | of WAS are used. |
| | | 4) Facility of simultaneous HMs treatment. | |
| | | 5) Energy recovery. | |
| | | 6) Processing is less odours. | |
| Sewage sludge and leachates | 1) Precipitation of P in form of struvite and hydroxyapatite | 1) Partial reimbursement of expenditures. 2) Low possibility of emitting HMs. 3) High probability of resolving the issues of blocking tubes/pipes. 4) High efficiency of N removal. | 1) High operational cost 2) Risk of environmental pollution through high organic pollutants, pathogens and parasites. 3) Limited use as fertilizers only 4) Not enough efficient to fully recover the P (high demand of magnesium and ammonia). 5) Inefficient approach for sludge management. |

5.2 Reduction and immobilization of heavy metals in sewage sludge products

WAS has been under vigilance due to the high concentration of HMs such as chromium (Cr), copper (Cu), zinc (Zn), lead (Pb), cadmium (Cd), nickel (Ni), arsenic (As) mercury (Hg) and selenium (Se). **Table 8** shows the comparison of HMs content from different sources and a group of hazardous inorganic compounds examined in WAS. The presence of such metallic species in high concentrations can be a problematic case for value-added chemicals and extraction of biochemical materials as well as for energy recovery from WAS [196].

Referring to WAS thermal conversion via gasification and pyrolysis technologies, TEs typically exist in the gaseous stream, liquid pyrolysis products, and residual biochar. It is reported that the volatilization of TEs is clearly associated to their boiling point [197]. Consequently, higher gasification temperatures opposed to pyrolysis result in an enhanced gaseous product contamination risk and henceforth the significance of monitoring the HMs distribution in the gasification is more. Various gasification studies have demonstrated the potential routes of HMs to various process products [197]. He et al. [198] conducted the pyrolysis with a sequential extraction procedure in electric furnace, mainly focusing on fractionation of HMs (e.g. Zn, Pb, Cu and Cd) in WAS and its residues which were generated after pyrolysis at temperatures ranging from 250 to 700 °C. The Cd was volatilized in the off-gas at 700 °C and decreased in the residues. In addition, Cu, Pb and Cd in the WAS and pyrolysis residues, were primarily bounded to OM and sulphides, whereas Zn was bounded to Fe and Mn oxides

Table 8. Heavy metal contents in different biomass materials [199, 200].

| Feedstock | Heavy metals (mg kg ⁻¹ dry basis) | | | | | | | |
|-------------------|--|----------------|-------------|------|-----------|------------|----------------|------------|
| | Cd | Cr | Cu | Hg | Ni | Pb | Zn | As |
| WAS | < 1 - 3410 | 10– 990,000 | 80– 2300 | 2.7 | 2– 179 | 13– 465 | 101– 49,000 | 3 – 230 |
| Paper sludge | <0.4 | 110 | 310 | 1000 | - | 160 | 470 | 8 |
| Paper sludge | 350 | 100 | 450 | - | 480 | 480 | 170 | - |
| Wheat straw | 1.0 | 25 | 0.06 | 6 | - | - | - | 0.18 |
| Beech wood | 1.0 | 2.5 | 43 | 0.12 | - | 33 | 15 | 3.5 |
| Recovered fuel | 24 | 1020 | 2800 | - | 209 | 1100 | - | 37 |

Hwang et al. [201] investigated the distribution of HMs such as Cr, Cd, Zn, Pb, Cu, K, Na and Mg present in pyrolysis residue of WAS (obtained at 500 °C) in column experiments under simulated landfill conditions and the bottom ash of WAS incineration (achieved at 900 to 1000 °C) to compare the environmental loads of the leachates depending on thermal treatment. Most of the HMs in the WAS remained immobile in the residues without being volatilized except Cd, Pb, and Zn. Although the residues comprised more of surplus OM than that of ash, their carbon emission into the leachate under aerobic conditions were comparable to that of ash under anaerobic conditions. Therefore, it was concluded that pyrolysis of contaminated WAS limited to 500 °C temperatures reduces the emissions of HMs from produced char in landfill, unlike other

pyrolysis studies [201-203] conducted at higher temperatures, making it a promising method of treating WAS before landfilling.

Contemplating the consequences of potentially harmful HMs during gasification, it should be noticed that ash dust is a chief carrier of (specially Cd and Pb) in turbulent waste gasification routes; therefore, if the gasification occurs under non-turbulent conditions, particulate ash emissions are almost eliminated [197]. Currently, Saveyn et al. [197] have studied the fate of metallic species during WAS gasification, indicating that HMs such as Cu, Pb, and Zn were nearly retained in the char, whilst Hg and Cd were depleted from the WAS and ended up in the different downstream sections like char residues, condensate liquids, and particles in filters. Similar trend for Cd was observed during co-gasification of solid recovered fuel and coal [204]. The transformation of HMs (Zn, Pb, Ni, Cu, Cr and Cd) from gasification of WAS to by-products and their major accumulation in solid and liquid residues, i.e. ash, char and tar, have been reported by several studies [75, 200, 205]. On the other hand, Hg and As due to high volatility at high temperatures can be entirely mobilized with aerosol particles [4].

Generally, the type, speciation and concentration of HMs depend on the sources of raw sludge, which majorly pass into products at high operating temperatures used for thermochemical conversion processes. The scheme of linking WAS and biofuel production can be introduced as a promising approach. However, implementation of appropriate thermochemical processes and its optimum operating conditions are highly desirable based on the type and content of HMs in the WAS, as well as, reactor design and the presence of probable pre-/post-treatment techniques [206]. For example, pyrolysis process at lower temperatures around 400 to 600 °C has been found safer and

efficient compared to gasification, liquefaction and incineration with higher temperatures/pressure, thereby increasing the emission of HMs into gaseous phase. Moreover, low temperature pyrolysis could contribute a considerable reduction on contamination level, bioavailability and environmental risk of HMs [67, 206, 207].

Even though, thermochemical processes can be suggested as a promising pathway for clean bioenergy recovery from WAS contaminated with HMs, still they are in infancy stages in terms of implication of pre-/post-treatment towards the immobilization and/or minimization of HMs transfer to the products [71]. For example, in some research studies, the implication of hydrothermal treatment prior to pyrolysis has been introduced as an effective method to accumulate the major amount of HMs in the biochar [71]. Besides, other studies have shown the considerable effect of operating reactors equipped with post-treatment technologies, such as hot-gas filter, [208, 209] cyclone, [210, 211] etc., to minimize the HMs emission from the thermochemical conversion of other contaminated biomass. Moreover, the blends of sewage sludge with other clean biomass, such as algae, sawdust, etc., has been also introduced to enhance the processes efficiency, to improve the characteristics of biofuel, and to reduce contamination resulted from diluting inorganic and toxic compounds [4]. In addition, the toxicity/risk of HMs, particularly Cu and Zn, could be reduced through dewatering the sludge prior to utilization by pyrolysis or liquefaction technologies [212]. Therefore, it is anticipated that the implementation of aforementioned policies, i.e. operating reactor at optimum conditions and equipped with post-treatment technologies, could be practical to prevent or minimize the emission of HMs during thermochemical conversion of WAS. As outlook, the implication of proper pretreatment techniques using potential sorbents, hydrothermal and leaching pretreatment

could be a good policy to hinder the transfer of HMs to the products, which to our best knowledge has not been sufficiently addressed.

5.3 Removal of micro-pollutants

Apart from HMs, sludge is also comprised of a wide range of micropollutants (MPs). MPs are mainly classified into three categories: (i) pharmaceutical products (PHPs) such as anti-inflammatory drugs, antibiotics, X-ray contrast media and β -blockers; (ii) personal care products (PCPs) such as steroids, analgesics, synthetic hormones, fragrances, cosmetics, sun screens, lipid regulators and shampoos; and (iii) endocrine disrupting chemicals (EDCs) such as estrone (E1), 17β -estradiol (E2) and 17α -ethinylestradiol (EE2) [213, 214]. The serious environmental and human risks associated with the contamination of WWTP sludge (through biosoild amended soils, and groundwater) have been widely reported in literature [33, 215-217]. This results mainly due to the hydrophobicity or propensity of pollutants to be adsorbed on particles (sludge) during primary and biological treatments in WWTPs.

Therefore, effective removal strategies for OMPs are urgent and arduous task to limit the contamination of environment. Today, various efforts have been made for removal/transformation of OMPs during sludge treatment via AD. Example include Nonylphenol ethoxylates and steroid estrogens via a lab scale reactor running under mesophilic (35 °C) and thermophilic (55 °C) trials [218], pharmaceutical products via two semi-continuous lab reactors under mesophilic (38 °C) and thermophilic (55 °C) conditions [219]. The results obtained from above studies concluded that the temperature (mesophilic or thermophilic) is not an important parameter, except for carbamazepine and nonylphenol, whilst naproxen, trimethoprim and sulfamethoxazole are substantially

removed. On the contrary, the removal efficiencies of several studies for OMPs removal are rather contentious; they have shown low (<25 %) or no removal of E1, E2, EE2 (hormones) [219, 220], musk fragrances [221, 222], diclofenac [219, 223], ibuprofen [219, 221] and triclosan [223]; whereas other researchers disagree. For example, a study conducted by Carballa et al. [224] showed that hormones and musk fragrances were effectively removed up to ~70 % and ~95 %, respectively; also, Samaras et al. [214] reported removals more than 90 % for diclofenac and ibuprofen, and within the range of 60 – 80 % for triclosan. The reasons of these inconsistencies remain vague. Further research in this area is highly valuable to deeply assess the impact of WAS treatments on the fate of MPs.

6. PREFERENCE OF WAS VS. LIGNOCELLULOSIC BIOMASS

Herein, the preference and/or comparison of WAS over other lignocellulosic biomasses (LBs) is discussed concisely. The pros and cons of WAS compared to LBs (i.e. agricultural residues and forestry waste) are mainly attributed to their basic structural and compositional differences. As previously described, WAS possesses a semi-solid structure with 59 – 88 % w/v biodegradable OMs [1, 3]. Whereas, generally most of the lignocellulosic biomass comprises of cellulose (30 – 50 %), hemicellulose (20 – 35 %) and lignin (10 - 27 %) [149]. Firstly, lignocellulosic biomass is known to be non-biodegradable due to its complex chemical composition [225], making it difficult to produce high yield and quality biofuels, value-added products and chemicals [226]. In order to cope with these challenges, these biomasses require various pretreatments, such as crushing, cracking, drying, preheating [227, 228] and more costly and sophisticated (thermo-)chemical operations to enhance the accessibility of biomass for conversion

reactions [229-231]. For instance, various thermochemical processes have been investigated to produce bio-oils or other form of biofuels from LBs, but the quality of biofuels still remains a big challenge, which needs further catalytic, separation or purification reactions to be upgraded [232-235]. Likewise, challenging fractionation procedure to separate cellulose from lignin and hemicellulose can be remarked to emphasize the controversial practicability of an expensive and/or hazardous pretreatment approaches, like ionic liquids, eutectic/organic solvents, steam explosion, acid and alkaline methods, etc., to facilitate the conversion and saccharification processes or materials recovery from agricultural wastes [230, 236, 237]. Secondly, regardless of the high potential of LB for thermochemical biofuel production [238], relatively a low biological methane yield [239], limited value-added products [239, 240] and chemicals recovery [241].

On the contrary, as comprehensively discussed in this paper, the soft structure and rich OMs content endow WAS with adequate potential for the recovery of energy [3, 21, 72], nutrients [36, 165, 174], and value-added materials [126, 136, 147] using various thermochemical and biological etc., processes with/without implementing costly pretreatment or post-treatment stages. The WAS ashes can also be reused in cement mixtures to produce diverse construction materials, including brick, mortar, etc. [242, 243]. However, regardless of aforementioned advantages of using WAS over LB, there are some limitations associated with each one, which can be compensated using their co-processing, particularly for energy recovery. For example, LB on account of the high C/N ratio and bio-recalcitrant compounds cannot reach to higher biogas yield [239, 244]. Whereas, WAS can be anaerobically biodegraded with high biogas yield of 270 – 385

mL CH₄ gVS⁻¹ [3, 15-17]. Therefore, the anaerobic co-digestion of WAS (holding low C/N ratio) and lignocellulosic biomass (with medium or high C/N ratio) under mesophilic and thermophilic condition seems to be a rational option to increase biogas yield[244]. Similarly, Chiang et al. [245] have elucidated that a mixed feedstock with higher ratio of WAS causes an enhancement of thermal reaction activity. The reason is also attributed to the less fixed carbon content of WAS, so that it could decrease the activation energy and consequently the significant increase of thermal reaction. On the other hand, WAS has a lower bio-oil yield of 32 wt. % and heating value of about 13 – 21 MJ kg⁻¹ [8, 57, 58], compared to the lignocellulosic biomass with higher bio-oil yields of 40 – 50 wt. % and heating value of 16.79 – 19 MJ kg⁻¹ [238, 246, 247]. In addition, another major problem linked to the WAS are higher moisture content and lower syngas calorific value of 4 – 6 MJ Nm⁻³ [58, 74], which can be compensated through co-pyrolysis, [248] -gasification, [73] –incineration [249, 250] and –liquefaction [251] of WAS with lignocellulosic biomass to high yield and quality usable energy in an environment friendly manner.

7. SLUDGE TREATMENT SCENARIOS IN EU, USA AND CHINA

Global WAS production is on a relentless growth curve and environmental quality requirements for WAS are becoming increasingly stringent, therefore disposal outlets are decreasing and economic pressures require low-cost solutions [252]. EU, USA and China represent a vast fraction of present world in terms of population, growth, technological advancements and, rules and strategies for environmental protection. The other countries in the world follow them as benchmarks especially in the arena of waste disposal and reuse.

7.1 European Union-15 countries

Among EU-15 countries, over 76 % of total WAS production belongs to Germany, UK, Italy, France and Spain as depicted in Fig. S1 (see Electronic Supporting Information, ESI). Figs. S1 and S2, WAS quantity in the EU-15 countries was around 10 million of DT in 2010 and it is estimated to reach to 10.5 million DT by 2020 [7, 70, 253].

According to EU landfill Directive, [254] landfill practice of wastes are forbidden. As a result, landfilling in EU-15 countries presenting a continuing decline between 2010 and 2020, from 11 % to 4 % (Fig. S3). Furthermore, WAS recycling has been a prevailing technology in EU-15 countries for soil conditioning and fertilization [7]. On the other side, HMs, pathogens and POPs often outweigh the soil's nutrients leading to the risk of eutrophication, acidification and GHG emissions [70, 255]. Therefore, WAS application for arable land will remain almost steady among EU-15 countries (43 % in 2010) expecting to attain 45 % in 2020, owing to imposed legislative restrictions (Fig. S3) [70]. The highest proportion of WAS recycling in 2010 was observed in Luxemburg (90 %), followed by UK (75 %), Ireland (75 %), France (65 %), Spain (65 %), Portugal (50 %), Denmark (50 %) and Italy (25 %), indicating that the recycling was most preferred disposal practice in EU-15 countries except Netherlands (Fig. S2). The change in WAS recycling has been estimated for 2020 as follows: for Italy and France (10 %), Spain (5 %), Austria and Luxemburg (-10 %), and for UK and Germany (-5 %).

Although incineration is considered as a cost intensive process with potential environmental problems [256], most of EU-15 countries seem to invest in incineration facilities (Fig. S2 (A) and Fig. S3). The Netherlands demonstrates 100 % incineration of the WAS produced, followed by Belgium (90 %), Germany (50 %), Denmark (45 %),

Portugal (30 %) UK (20 %), Italy (20 %), France (15 %) and Spain (10 %) (Fig. 2S (A)). The incineration shares based on 2020 estimation are predicted to increase as follows: Austria (45 %), Greece (40 %), Spain (15 %), Luxemburg (15 %), Italy, Ireland and Portugal (10 %) (Fig. S2 (B)). The significant variations in incinerated WAS among EU-15 countries are closely related to the incineration methods. WAS co-incineration with MSW, coal-fired power plants or cement kilns are preferred.[257, 258] Mono-incineration practice is widely expanding in Germany, UK, and Denmark with 23, 11 and 5 dedicated WAS incinerators, respectively [7]. Mono-incinerators have proved to be a reliable approach to recover phosphorus from incineration ashes [258]. Moreover, modern incinerators should be capable of using resultant heat to generate their own electricity so that WAS treatment becomes energy-neutral.

7.2 United States of America (USA)

The USA employs WAS (biosolids) typically for arable land application as shown in Fig. S4. It adds the nutrients into soils, thereby providing more OMs and improving its holding capacity [32, 259]. Referring to Fig. S4, over 8 million DT of WAS is annually produced (~ 23 kg of WAS person⁻¹ year⁻¹) in USA, 55 % applied to arable land, 30 % landfilled, and 15 % incinerated [32]. The cost of arable land application, landfilling, and incineration are estimated to be 300 – 800, 100 – 650 and 300 – 500 \$ DT⁻¹, respectively. Moreover, local ordinances have forbidden WAS land use to curb the associated odors, pathogens and pollutants [32].

In terms of nutrients (N and P) recovery, the reported data [260] of 8 million DT of WAS recycled in arable land (55 %) (Fig. S4), with N and P contents of 3.4 and 2.3 %, respectively [32]. Consequently, the potential annual net flux is respectively 134,640 t N

year⁻¹ and 91,080 t P year⁻¹. Based on the data presented in the literature [261] regarding the annual nutrients consumption, aforementioned values could provide only 1.1 % and 5.3 % of the net annual N and P fertilizer consumption. Thus, recovered N and P do not seem significant enough to rely upon them as imperative nutrients source for land application. However, their removal from wastewater and prevention of eutrophication phenomenon in water bodies are still in the first priority. In terms of energy, around 16,000 WWTPs are operating in U.S.A. However, only 544 of WWTPs have adopted AD technology for biogas production, and merely 106 of these are able to use the biogas produced for various applications such as electricity and/or thermal energy [262].

7.3 China

Over the last 20 years, China has been investing considerably in WWTPs. Currently, China has become the world's second largest WAS producer with around 5,797 WWTPs in 2014, generating 6.25 million DT equivalent to ~ 4.5 kg person⁻¹ year⁻¹, with an average annual growth of 13 % [52]. On the other hand, more than 80 % of the WAS has not been treated or disposed properly, and subsequently poses a great threat to both the public health and the environment [263]. As a result, China government has set a goal to start treating 5.18 MM of DT year⁻¹ [52]. Fig. 2 illustrates the detailed WAS treatment/disposal options indicating that arable land application, sanitary landfill, AD, and incineration are the four most commonly employed routes in China. Hitherto, the mainstream routes applied in China are thickening, conditioning, and dewatering [52]. The thickening route reduces over 5 % WAS moisture and the major pathways include flotation, mechanical and gravity thickening [52, 264]. Moreover, the organic polymer

electrolytes and inorganic coagulants (chemical conditioning) have been frequently applied routes for enhancement of WAS dewatering [52].

The accurate official data on WAS disposal is scarce, rather conflicting sources with dissimilar estimations [52, 265]. According to Yang et al. [52] over 84 % of WAS is disposed by improper dumping. Concerning the proper disposal, sanitary landfill has been most widely used method, which accounts for 13 %, followed by land application (2 %), incineration (0.4 %) and building material production (0.2 %). Data reported by Wang et al. [265] showed that the most commonly employed disposal method is landfilling (63 %), followed by agricultural applications (14 %), and incineration (2 %). Moreover, the disposal method for approximately 21 % of WAS is unknown.

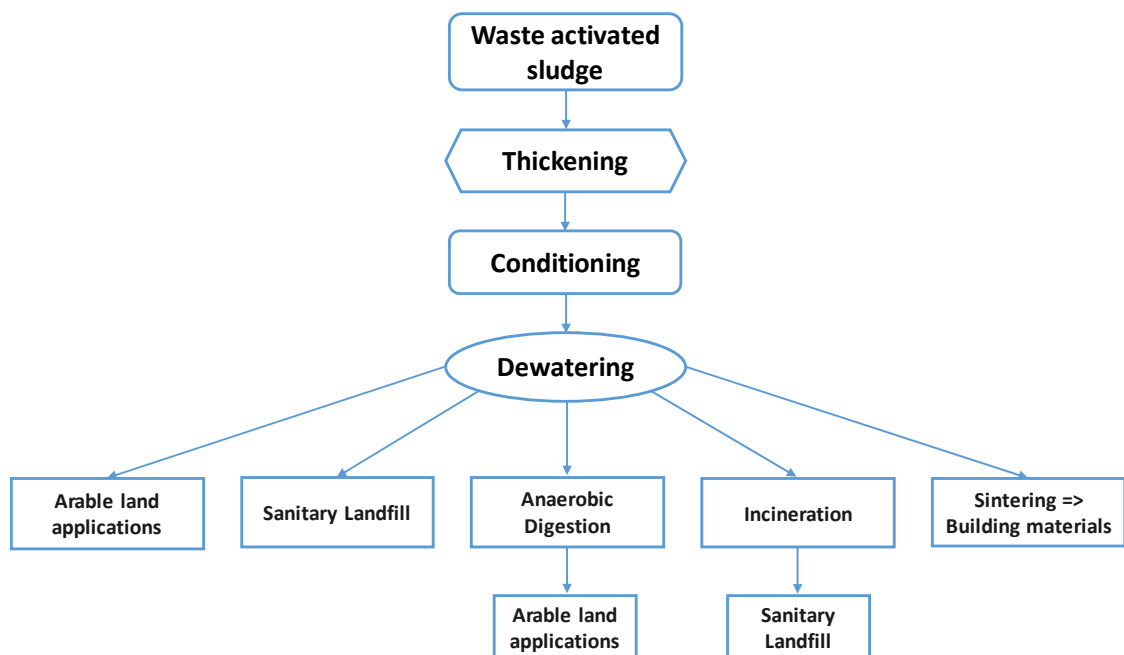


Fig. 2 Technical routes of WAS treatment and disposal in China [52].

The significant variations of data are attributed to WWTPs in China, because they do not send proper reports on the treatment and final disposal of their WAS [266]. Moreover,

the cost of sanitary landfill, arable land application, incineration, building materials are estimated to be 72 – 96, 240 – 280, 240 – 260 and 240 – 260 \$ DT⁻¹, respectively, whereas improper disposal only costs around 16 – 24 \$ DT⁻¹ [52]. Therefore, WWTPs and associated organizations typically opt for cheap disposal routes. In China only 50 % of WWTPs adopted AD to stabilize WAS. However, only 30 % of those 50 % plants are operating the AD apparatus properly due to insufficient operational expertise and insufficient funding [52, 267]. Furthermore, the treatment efficiency of WAS by AD process is not sufficient due to high sand content and low volatile organic compounds [267]. According to Yang et al. [52] thickening-dewatering-AD and subsequent arable land application can be employed as the major technological route in the future. This route appears to be promising for large and moderate WWTPs due to the limited land availability in the cities and scarcity of energy supply in economically poor countries.

8. FUTURE PERSPECTIVE

Rapid increase in population and societal living standards will undoubtedly enhance the sludge generation to a vital extent. In the wake of lesser land-fill sites and stringent disposal regulations, a shift is required from conventional disposal to advanced valorization strategies. Numerous pathways are employed nowadays to explore the possibilities of energy extraction from sludge. However, complex composition of sludge such as the presence of very high amount of moisture, bacterial constituents (proteins, lipids, etc.), HMs and other contaminants (e.g. polycyclic aromatic hydrocarbons (PAHs), dioxins, furans) reduces the efficiency and safety of these processes and enhances their

overall costs. In addition, it also demands special equipment / plant design to cater such a need.

Biological route such as AD is considered as the most appropriate sludge valorization technique but it suffers for poor efficiency and long processing time. Thermochemical pathways such as incineration, pyrolysis and gasification have improved efficiencies with faster processing and versatile end applications. However, they all are energy intensive for high moisture containing feedstock such as wet sludge. In addition, flue gases (in incineration) and syngas (in gasification) require conditioning and expensive clean-up prior to usage and emissions. Same is the case with pyrolysis derived bio-oil which also needs upgrading prior to its application. Therefore, innovation is needed in the existing pathways to eliminate these limitations. AD coupled with pyrolysis, co-combustion and co-incineration can be the viable routes provided the process economics is enhanced via proper process design and reduction in energy requirements. This may lead to the scale up of pilot plant studies to commercial level applications.

There can be other sustainable solutions for efficient and cost-effective disposal of sludge with simultaneous energy and materials recovery. An integrated bio-refinery system could be designed focusing on reuse options rather than disposal pathways to generate power and recover all the possible resources. It will not only valorize the sludge but will also aid in reducing adverse environmental impact to a significant extent. This integrated system can be designed based on the local circumstances such as composition and amount of sludge, economy, topography, and weather conditions. In addition, sludge disposal and application models given by EU and USA can serve as references for other developing nations. On the whole, application of advanced technologies for WAS

management must transform from treatment of a liability toward a promising opportunity (i.e. recovery of embedded energy, metals, and nutrients), while enduring to protect the environment and public health.

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ABBREVIATIONS

| | |
|------|------------------------------|
| AD | Anaerobic digestion |
| AC | Activated carbon |
| Bt | Bacillus thuringiensis |
| BES | Bioelectrochemical systems |
| BSA | Bovine Serum Albumin |
| BCA | Bicinchonic Acid |
| COD | Chemical oxygen demand |
| C/N | Carbon to nitrogen ratio |
| CER | Cation exchange resin |
| CCE | Carbon conversion efficiency |
| CGE | Cold gas efficiency |
| C-GT | Carver-Greenfield technology |
| CT | Cycle time |
| DFC | Down flow combustor |
| ER | Equivalence ratio |

| | |
|-----------------|-----------------------------------|
| FB | Fluidized bed |
| GRT | Gas residence time |
| GHG | Greenhouse gas |
| GE | Gasification efficiency |
| HM _s | Heavy metals |
| HR | Heating rate |
| LHV | Lower heating value |
| LBs | lignocellulosic biomasses |
| MW | Microwave |
| MW _e | Megawatt |
| Mt | Metric tons |
| MSW | Municipal solid waste |
| MFCs | Microbial fuel cells |
| OMs | Organic matters |
| OLR | Organic loading rates |
| OFS | Oil-from-Sludge technology |
| PHA | Polyhydroxyalkanoates |
| POPs | Persistent organic pollutants |
| PVC | Polyvinylchloride |
| RPM | Revolution per minute |
| SSBT | Siemens Schwell-Brenna technology |
| Tx | Entomotoxicity |
| TEs | Trace elements |
| TS | Total solids |
| TD | Tons of dry solids/dry |
| VFAs | Volatile fatty acids |
| VM | Volatile matter |
| WAS | Waste activated sludge |
| WGS | Water-gas shift |

| | |
|------|----------------------------|
| WWTP | Wastewater treatment plant |
| WWT | Wastewater treatment |

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