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Research article

Integrated review of resource recovery on aerobic granular sludge systems: Possibilities and challenges for the application of the biorefinery concept

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ABSTRACT

Aerobic Granular Sludge (AGS) is a biological treatment technology that has been extensively studied in the last decade. The possibility of resource recovery has always been highlighted in these systems, but real-scale applications are still scarce. Therefore, this paper aimed to present a systematic review of resources recovery such as water, energy, chemicals, raw materials, and nutrients from AGS systems, also analyzing aspects of engineering and economic viability. In the solid phase, sludge application in agriculture is an interesting possibility. However, the biosolids' metal concentration (the granules have high adsorption capacity due to the high concentration of extracellular polymeric substances, EPS) may be an issue. Another possibility is the recovery of Polyhydroxyalkanoates (PHAs) and Alginate-like exopolymers (bio-ALE) in the solid phase, emphasizing the last one, which has already been made in some Wastewater Treatment Plants (WWTPs), named and patented as Kaumera® process. The Operational Expenditure (OPEX) can be reduced by 50% in the WWTP when recovery of ALE is made. The ALE recovery reduced sludge yield by up to 35%, less CO_2 emissions, and energy saving. Finally, the discharged sludge can also be evaluated to be used for energetic purposes via anaerobic digestion (AD) or combustion. However, the AD route has faced difficulties due to the low biodegradability of aerobic granules.

1. Introduction

Wastewater Treatment Plants (WWTPs) are indispensable for human health and environmental quality. However, although these systems contribute to a better quality of life, they request high rates of energy and chemical products and generate various types of solid wastes that need to have the correct treatment and disposal (Al-Obaidi et al., 2020).

The municipal wastewater treatment is reported to be responsible for around 5% of the electric demand in some countries. In the USA, this consumption is equal to 30.2 billion kWh per year (Shen et al., 2015). A paper evaluating the sustainability of decentralized WWTPs reported that the energy consumed in sewage treatment could reach up to 20% of the total energy used in a city (Wang and Li, 2012). Such a demand can be duplicated in the next 15 years due to population growth, persistent contaminants, and the water discharge standards becoming more rigorous every time. Considering the WWTP's economic management, electric energy consumption and solid waste management represent more than 60% of the operational costs (OPEX, Operational Expenditure). In some cases, only sludge management is responsible for about 50% of the OPEX (Shen et al., 2015).

Most of the WWTPs treating sewage are designed only to remove contaminants. In another perspective, in the last years, a new approach has been considered. The sewage is considered a resource containing many high aggregated value products that can be recovered, such as water, nutrients, energy, heat, and chemical products. Verstraete and Vlaeminck (2011) defend that the classical WWTPs (without considering resource recovery) should be redesigned as a biorefinery, which is an alternative to a petroleum-based refinery, where non-edible biomass is used as a raw material in the production of biofuels, chemical industries, and biomaterials (Mohan et al., 2016). The biorefinery allows the recovery of high aggregated value products/resources, such as phosphorus, nitrogen, biopolymers, methane, hydrogen, amongst others (Batstone et al., 2015). Therefore, the new plants must aggregate the removing contaminants systems associated with the recovery of high aggregated value resources to obtain a higher economic sustainability.

Until the '90s, anaerobic digestion was considered as the heart of the

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biorefinery. In the fermentation process, usually in traditional anaerobic WWTPs, the main products are carboxylic acids, alcohol, hydrogen, and methane (Verstraete and Vlaeminck, 2011; Metcalf, 2003). The emergence of new sewage treatment technologies has allowed the recovery of many other commercial value products, increasing the possibilities of new treatment routes focused on resource recovery. Among the emerging technologies that allow contaminants removal and resource recovery, the aerobic granular biomass can be considered as a very interesting option (Verstraete and Vlaeminck, 2011; Metcalf, 2003; Rollemberg et al., 2020).

The granular sludge is the third modality of microbial aggregation in sewage treatment, different from activated sludge and biofilm-based systems (Rollemberg et al., 2018). Such modality includes the anaerobic granular sludge and the aerobic granular sludge (AGS). The anaerobic system is applied worldwide, and its potential is already consolidated. On the other hand, the aerobic system presents several advantages like simultaneous carbon, nitrogen, and phosphorus removal in a single reactor (Pronk et al., 2015; Świątczak and Cydzik-Kwiatkowska, 2018).

Two decades after its discovery, AGS has been the main subject of different researches, which tested not only domestic wastewater treatment but also bioremediation of toxic aromatic compounds (including phenol, toluene, pyridine), industrial wastewater treatment (textile, dairy, brewery), application on removing nuclear wastes, adsorption of heavy metals, and recovery of high aggregated value resources. However, there are a few reports analyzing resource recovery in AGS reactors.

In this sense, this paper presents a systematic review of resources recovery such as water, energy, chemicals, raw materials, and nutrients from AGS systems, also analyzing aspects of engineering and economic viability. To the best of the authors' knowledge, this is the first review giving this perspective considering the aerobic granular biomass.

2. AGS technology

The AGS technology simplifies the process design because the biological treatment and biomass separation occur in the same reactor, not demanding secondary clarifiers in the treatment flowchart. Around 50 full-scale AGS wastewater treatment plants are currently operating worldwide (Rollemberg et al., 2020; Li et al., 2014). Therefore, AGS treatment should be considered alongside other compact treatment alternatives such as activated sludge (AS), moving bed biofilm reactors, and membrane bioreactors (Metcalf, 2003; Rollemberg et al., 2020).

In comparison with AS, numerous advantages can be found in the AGS, such as the simultaneous removal of organics and nutrients in the same tank, better settling ability of the formed biomass, higher biomass concentration, and significant reduction in footprint (∼75%) and power consumption (30–50%) (Thwaites et al., 2018). In relation to membrane bioreactor (MBR), the AGS process had an estimated electricity consumption that was 35–70% lower (Bengtsson et al., 2018). Owing to these advantages, AGS is considered one of the most promising biological wastewater treatment technology in the 21st century. Some authors predict that this technology will overtake AS and provide more sustainable wastewater treatment over a century or more (van Loosdrecht and Brdjanovic, 2014).

The AGS systems' flowchart is very similar to conventional technologies (activated sludge, for example). The pre-treatment includes different screening (medium and fine, for instance), unities of sieving, and grit removal. The concern about the presence of fat in AGS reactors has been frequently highlighted in several papers because this kind of material causes granules washout by flotation due to oil and fat presence (Wagner et al., 2015; Rollemberg et al., 2018). Thus, unities focused on removing fat upstream of the AGS reactor should always be considered in the design.

The conception of WWTP with AGS does not need to include primary or secondary clarifiers. The treatment process may need one equalization tank, mainly when a Sequencing Batch Reactor (SBR) is used. Nevertheless, the WWTPs are composed of multiple modular reactors (usually three or more) operated in parallel. In cases when the amount of wastewater is relatively low (decentralized WWTPs), the treatment could occur in a single reactor preceded with an equalization tank. In this sense, Fig. 1 shows different design conceptions involving AGS technology.

This system has evolved into a mature solution for large-scale WWTPs, but it has presented high implementation costs, including a high investment for continuous monitoring. In the WWTP operated by Pronk et al. (2015), each reactor had devices to measure dissolved oxygen, redox potential, temperature, water level, dry material, and turbidity. The ammonium and phosphate were measured semi-continually (every 5–10 min) during the cycle by an automatic sampling and analysis device (Hach Lange, Filtrax, Amtax, and Phosphax). Sometimes there is one supervision system operating from outside the country. One great example is located in the Netherlands, where a treatment plant is remotely operated in the Gansbaai, South Africa, and Frielas, Portugal (Pronk et al., 2015). So, although the technology is promising, it is possible to present economic sustainability problems, especially in developing countries, which explains that approximately 70% of WWTPs of Nereda® are concentrated in Europe.

3. Possibilities of resource recovery from aerobic granules

The resource recovery in full-scale AGS reactors is still in the beginning (van Loosdrecht and Brdjanovic, 2014). Initially, the resource recovery was limited to water reuse (liquid fraction) and the use of excess sludge (aerobic granules) as inoculum to start new AGS reactors. Phosphorus recovery from AGS was firstly reported in 2007. So, the granules were cultivated in Enhanced Biological P-Removal (EBPR) conditions, which made the recovery after the P-release in the anaerobic phase possible, followed by the P-precipitation in the liquid phase (Yilmaz et al., 2007). In sequence, Lin et al. (2008) reported the possibility of alginate-like exopolysaccharides (ALE) extraction from aerobic granules in a lab-scale reactor treating synthetic domestic sewage. In the last decades, research has allowed the discovery of several resources with high added value in the aerobic granule biomass, mainly compounds extracted from the Extracellular Polymeric Substances (EPS), whose production is higher in AGS systems.

The possible resources to be recovered from aerobic granules are: (i) Phosphorus; (ii) Polyhydroxyalkanoates or PHAs; (iii) Alginate-like exopolymers (bio-ALE); (iv) Tryptophan; (v) Polysaccharide-based biomaterial; (vi) Methane from anaerobic digestion of the excess sludge; (vii) Water reuse; (viii) Use of excess sludge granules as cheap adsorbents etc. (Lu et al., 2016; Wang et al., 2014; Van Leeuwen et al., 2018; Wang et al., 2018b).

4. Liquid phase – water reclamation

Apply technologies that enable water reuse is the first step to sustainability in a WWTP (Lema and Martinez, 2017). In this regard, AGS reactors are an interesting option when the objective is to reuse the treated wastewater due to (i) the high organic matter and nutrients removals (Table 1), which are necessary for many industrial applications. This is mainly due to the SNDPR mechanism - simultaneous nitrification, denitrification, and phosphorus removal in the granules (Nancharaiah and Reddy, 2018); (ii) metals removal, which has been highlighted in some papers due to the high amount of EPS in the granules, which allows the biosorption of metals and subsequent removal in the excess sludge Wang et al., 2018a; (iii) removal/biotransformation of pharmaceuticals and personal care products, which is more efficient than conventional Activated Sludge (AS), due to a combination of both biotic and abiotic processes (Amorim et al., 2016).

However, experiences in full-scale AGS reactors showed concentrations of suspended solids (SS) higher than 20 mg/L, which did not enable

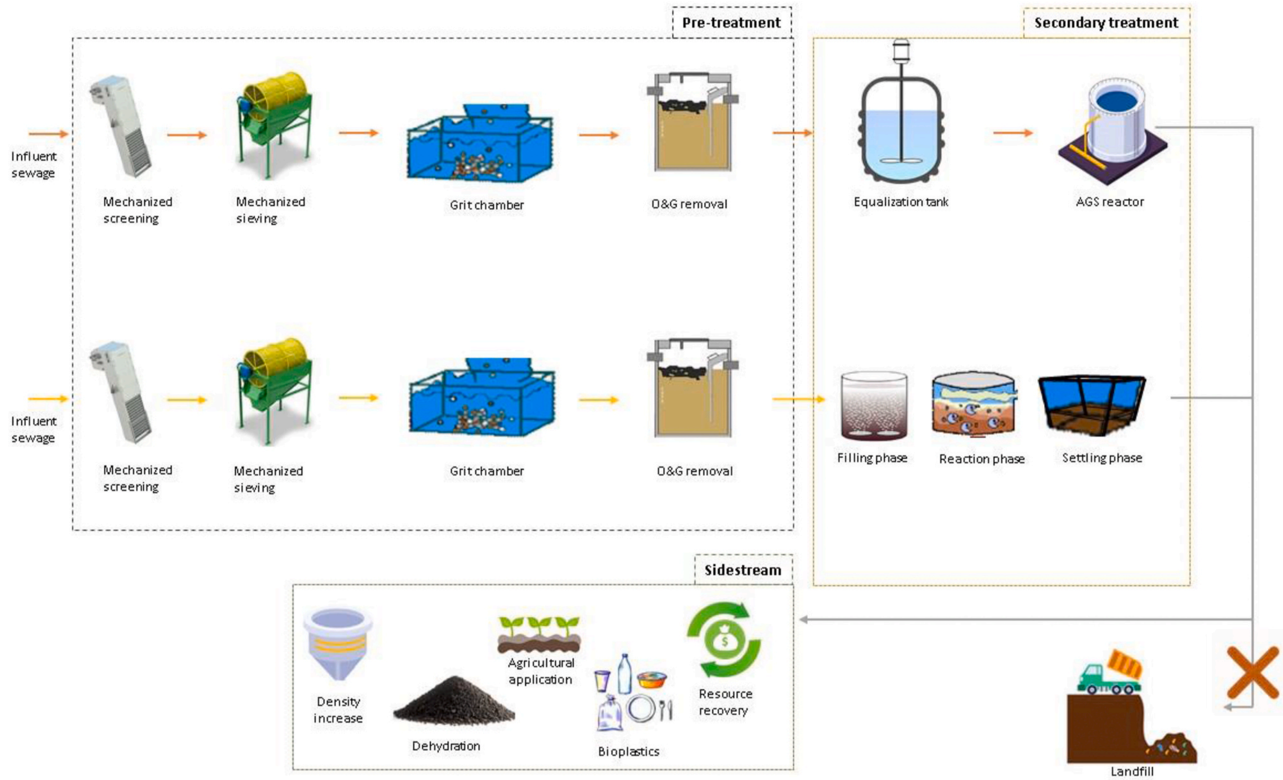

Fig. 1. Design conceptions with AGS technology.

Table 1
WWTP with AGS technology and quality of the final effluent.

Ref.	WWTP	Type of wastewater	Volume (m^3)	Cycle	Granule diameter (mm)	Characteristic of wastewater	Efficiency of removal	Effluent Quality
Giesen et al. (2013)	Full-Scale	Municipal	–	–	> 0.5 mm	COD: 1265 mg/L NH_4^+-N: 75 mg/L P total: 19 mg/L	COD: 97% NH_4^+-N > 98% P total: 82%	COD: 40 mg/L TSS: <5 mg/L PT: 3.2 mg/L NH_4^+-N: <1 mg/L NT: <10 mg/L
Li et al. (2014)	Full-Scale	30% (municipal) and 70% (industrial)	12,540	6 h	> 0.5 mm	COD: 56,000 mg/L NH_4^+-N: 39–93 mg/L	COD: 85% NH_4^+-N: 95.8% NT: 59.6%	NH_4^+-N: 28.2 mg/L TO: 34.5 mg/L BOD_5: 85 mg/L
Pronk et al. (2015)	Full-Scale	Municipal	9600	3–6:30 h	> 1.0 mm	COD: 560 mg/L NH_4^+-N: 39 mg/L TN:49,4 mg/L. PO_4^{3-}: 4.4 mg/L P total: 6.7 mg/L	N total >90% P total >90%	COD: 64 mg/L TSS: 20 mg/L PT: 0.9 mg/L TN: 6.9 mg/L NH_4^+-N: 1.1 mg/L BOD_5: 9.7 mg/L
Rollemberg et al. (2019)	Pilot-Scale	Municipal	0.14	6 h	> 0.9 mm	COD: 461 mg/L NH_4^+-N: 36.9 mg/L TN: 43.0 mg/L PO_4^{3-}: 4.8 mg/L P total: 5.1 mg/L	COD: 96% NT: 65.2% P total: 80%	COD: <20 mg/L TSS: 11 mg/L PO_4^{3-}: <1 mg/L NH_4^+-N: 4 mg/L NO_3^--N: 2 mg/L NH_2^--N: 4 mg/L BOD_5: 4 mg/L

attending restrictive limits in some water reuse legislations (Pronk et al., 2015). In this perspective, some modifications were proposed in the reactor design (sludge retention system at the top of the reactor) to obtain a SS concentration lower than 5 mg/L (Van Dijk et al., 2018). In the literature, only a few papers evaluated water reuse in AGS systems. For example, Karakas et al. (2020) studied the applicability for irrigation and found low metal concentration in the effluent, which is good for reuse in agriculture. On the other hand, the high nutrients removal in AGS is disadvantageous for irrigation applicability. In this regard, Pronk et al. (2015) showed that the effluent of a Nereda® AGS WWTP in Garmerwolde, in the Netherlands, had a concentration of dissolved PO_4^{3-}-P lower than 0.5 mg/L and Total Nitrogen lower than 7.0 mg/L.

As previously mentioned, one of the main issues related to effluent quality in the AGS reactor is SS concentration, sometimes higher than 50 mg/L (Rollemberg et al., 2020; Liu et al., 2005). Activated sludge systems can provide SS concentration in the effluent much lower than this value, and even lower than 10 mg/L (Parker et al., 2001). The low settling time, flotation of granules due to fat, the presence of filamentous micro-organisms with low settling velocity, and degasification due to denitrification during the feeding period are some reasons for the high effluent SS concentration. The last factor is more pronounced in a simultaneous fill/draw SBR because, in addition to gas ascension (denitrification), there is the contribution of the upflow velocity during the feeding (Van Dijk et al., 2018; Franca et al., 2018).

To decrease SS concentration in the effluent, some authors proposed the use of outer membrane (microfiltration or ultrafiltration) (Karakas et al., 2020), sand filter (Rollemberg et al., 2020), or vertical baffle on the top of the AGS reactor (Van Dijk et al., 2018). A vertical baffle introduction could reduce the SS concentration from 23 to 7 mg/L, allowing the reuse after disinfection and without the use of membranes or filters (Thwaites et al., 2018; Van Dijk et al., 2018).

Pharmaceuticals and personal care products (PPCPs) removal in AGS reactors is another aspect that makes the reuse of treated effluent of this system possible. Efficient methods to remove PPCPs include Activated Sludge, Advanced Oxidation Process (AOP's), adsorption on activated carbon (AC), and membrane technologies (Zhao et al., 2010). The pollutants can be adsorbed on suspend solids and be biodegraded or removed by the discharged sludge (Suarez et al., 2008). PPCPs removal in AGS was evaluated in some papers (Kong et al., 2015), and it was shown that granules have more potential than flocculent biomass to remove these substances. Amorim et al. (2016) observed that PPCPs increased EPS content in the granules, possibly because the drugs stimulate micro-organisms' metabolism.

The explanation for the high capacity of pollutants removal in AGS systems are: (i) High EPS concentration that acts like a "biological glue," that favor the adsorption of the compounds; (ii) High Solids Retention Time (SRT), which favor slow-growing micro-organisms related to the biodegradation/biotransformation; (iii) Presence of functional groups, including amino, carboxyl, phosphate, among others, on the surface of granular sludge, which can act on the co-metabolic reactions involved (Gao et al., 2010; Amorim et al., 2016).

Metal removal is another important aspect to be evaluated when the aim is the reuse in irrigation. For instance, Kong et al. (2015) observed that the removal mechanism in AGS systems was related to biosorption and EPS. Researchers showed that aerobic granules have a surface negatively charged, which causes a high affinity for cations, thus forming an external layer with an affinity for anions (Wang et al., 2018a). Therefore, the use of aerobic granular biomass to the removal/recovery of metals (Nancharaiah and Reddy, 2018) can be considered as an alternative instead of traditional methods for metals removal, for example, precipitation, coagulation, ion exchange, electrochemical process, and membranes (Gutnick and Bach, 2000).

Barrios-Hernández et al. (2020) evaluated pathogens removal in AGS system compared to conventional AS. The WWTP Garmerwolde, in the Netherlands, had values between 60 and 80% in both systems, therefore lower than 1 log removal. These results indicate that, like AS, a disinfection step is necessary to accomplish the discharge limits (Thwaites et al., 2018). However, the experiments showed that no effluent polishing step is required (membrane, filtration etc.), but just conventional disinfection (chlorination, UV, ozonation etc.).

Regarding operating cost, considering only biological processes, the AGS reactor has significantly low values than conventional aerobic systems for water reuse. Pronk et al. (2015) observed 0.17 kWh/m^3 and Rollemberg et al. (2020) 0.25 kWh/m^3, which is approximately 50% lower than the average in conventional AS for water production with a low concentration of organic matter, nutrients, SS, and turbidity.

5. Solid-phase – recovery of value-added products

The handling and disposal of excess sludge from WWTP is a major issue. In most developing countries, biological sludge is simply disposed of in open dumps or landfills. On the other hand, legislation in Europe requires energy recovery or the production of high-value compounds from this sludge (Metcalf, 2003).

AGS and other aerobic technological routes will produce a higher amount of sludge compared to anaerobic processes such as high-rate systems like upflow anaerobic sludge blanket (UASB) reactors. Therefore, ways to decrease sludge production, concentrate, resources recovery, and reuse of the biosolids have attracted some studies' attention (Zhang et al., 2019), especially because the strategies are not completely described in the literature.

Aerobic granular systems are a better alternative than conventional AS systems because they have a lower biomass yield coefficient (Val Del Río et al., 2013). The theoretical growth yields of aerobic granules were estimated as 0.2 (Liu et al., 2005) and 0.3 g VSS/g $COD_{removed}$ (Rollemberg et al., 2019), respectively, which is following the results obtained by Mosquera-Corral et al. (2005), operating an aerobic granular reactor fed with synthetic wastewater containing acetate. This range involves reducing sludge production by around 30% compared with conventional AS, characterized by a sludge growth yield of around 0.45 g VSS/g $COD_{removed}$ (Metcalf, 2003).

In terms of quality, AGS systems are suggested to produce a stabilized sludge (Pronk et al., 2015). However, Ali et al. (2019) showed a stratification similar to the one formed in reactors with anaerobic granules. Full-scale WWTP with the AGS technology shows microbial aggregates with different sizes (flocs, and small, medium and big granules) in the same space. Therefore, the biomass is not homogeneous, forming flocs on the top of the sludge blanket and granules on the bottom (Winkler et al., 2011). Thus, selective sludge discharge is important, controlling the SRT, as usually applied in AS, and sludge characteristics.

Two types of sludge discharge in AGS reactors can be distinguished (Adam et al., 2009): mixed sludge removed in every cycle, named selective discharge, and aims to remove biomass with low sedimentability (flocculent sludge), therefore applying a biological selection pressure (Ahmad and Idris, 2014); bottom discharge, when the removal takes place from the sludge blanket. This strategy aims to avoid high biomass concentration in the reactor, controlling SRT. It is important to note that the sludge initially discharged has low SRT and PAO's concentration, different from the one discharged in a second moment. Therefore, even if it is one reactor, it is possible to recover different resources depending on the discharge approach.

5.1. *Recovery of excess sludge as inoculum or catalyst material to accelerate granulation*

Studies showed that granulation could be achieved quickly (<1 month) with synthetic wastewater (especially acetate and propionate) (Liu et al., 2010). However, it can take months or a year with real domestic/municipal wastewater. Li et al. (2014) obtained complete granulation of his system after 337 days of operation in a WWTP treating municipal wastewater. Therefore, it is possible to use the excess sludge of AGS as inoculum to start up a new system or, when necessary, increase aerobic granular biomass in a reactor. This approach is done in Nereda® plants (Nereda, 2021), for example.

Some other strategies can be used to accelerate granulation, such as using EPS from biomass as a process catalyst. The main form of applying granular sludge as inoculum are crushed aerobic granules (Pijuan et al., 2011), aerobic granules storage for long periods (Liu et al., 2005), and mature granules (Long et al., 2014).

The first strategy (crushed granules) consists of dehydrating and crush the granules to make powder. Li et al. (2015) used this strategy and could reduce the granulation in two weeks. Another possibility is the natural drying process and material use as a catalyst. Such an approach was applied in some plants, showing to be an interesting option of sludge management (Stickland et al., 2013). Literature observed that natural processes favor EPS production and sludge aggregation because dehydration and a non-nutritive environment make micro-organisms produce more EPS for self-protection (Sheng et al., 2010). After some drying days, which depends on temperature, a rich-agglomerate in EPS is formed. Such compound can act as a biological glue, thus accelerating the aerobic granulation (Liu et al., 2004a; Sheng et al., 2010).

Even though the reuse of dry granules has shown good results, the storage and reuse of wet granules is the main route used (Wan et al., 2014b). Long et al. (2014) showed that applying 25% of mature aerobic granular sludge as inoculum reduced granulation for less than three weeks. The addition of this biomass with high sedimentability reduces the effluent SS loss and accelerates granulation (de Kreuk et al., 2010).

Some studies have evaluated the use of excess sludge of aerobic granules and the methods to allow this practice. Zhu and Wilderer (2003) reported that a long time of storage in a humid environment could favor sulfate-reducing bacteria (SRB), which use organic matter inside the granule in endogenous respiration, damaging the stored granules. On the other hand, Wan et al. (2014) found that the hostile environment with high agitation and the presence of DO stimulates cell secretion of various substances beneficial for granules maintenance for long periods, allowing their reuse as inoculum. Although granular sludge storage is not adequate, some studies have shown that it can be restored in a few weeks (usually two weeks) after aeration as inoculated sludge (Yuan et al., 2012). Besides, it is better if the sludge is stored correctly to maintain biomass active and stable to be used as inoculum.

The results indicate that AGS is an important biological resource, and it has an important role in reducing the reactor start-up period. In this sense, Fig. 2 shows the possibilities of AGS reuse in new plants. Besides, some authors mentioned that AGS could be used as an inoculum for conventional activated sludge (Peeters and Lu, 2013). It would be a way to feed AS with granules continually. Several studies showed that this practice could improve: (i) sludge sedimentability in AS, (ii) possibility to treat wastewater with higher OLR due to a high VSS concentration in a reactor; (iii) nutrients removal due to the presence of nitrifying and denitrifying bacteria, and also PAOs (Peeters and Lu, 2013). In this sense, Świątczak and Cydzik-Kwiatkowska (2018) studied a conventional activated sludge plant after cultivation with AGS. The authors observed that the COD removal increased from 75% to 92%, total nitrogen efficiency from 78% to 87%, and total phosphorus from 87% to 95%. After AGS addition, biomass concentration increased from 5.2 g/L to 9.2 g/L.

5.2. Phosphorus recovery in aerobic granular biomass

Every year, thousands of tons of mineral phosphate are taken to fertilizers' fabrication due to population increase and food demand, and the need to produce agrofuels (Martí et al., 2017). Because it is a finite resource, phosphorus scarcity has been a concern at the global level. It is estimated that, between 2070 and 2099, the phosphorus reserves will be depleted (Egle et al., 2016).

Approximately 20% of all mineral phosphorus consumed in the different activities (agriculture, industry etc.) is excreted by humans and hence recoverable (Batstone et al., 2015), from which 99% is present in urine and feces. The estimated annual load is about 3 million tons, which transform the wastewater treatment plants (WWTP) in "phosphorus factories". In this regard, many countries have taken initiatives to remove phosphorus in WWTPs, like the Swiss government that imposed, in 2016, a new standard to obligate the recovery and recycling of phosphorus in the inorganic products form of all sludge from sewage (Martí et al., 2017). van Kawenbergh (2010) reports that phosphorus recovery in all sewage treatment plants worldwide may last phosphorus reserves for more than four centuries, of course depending on the

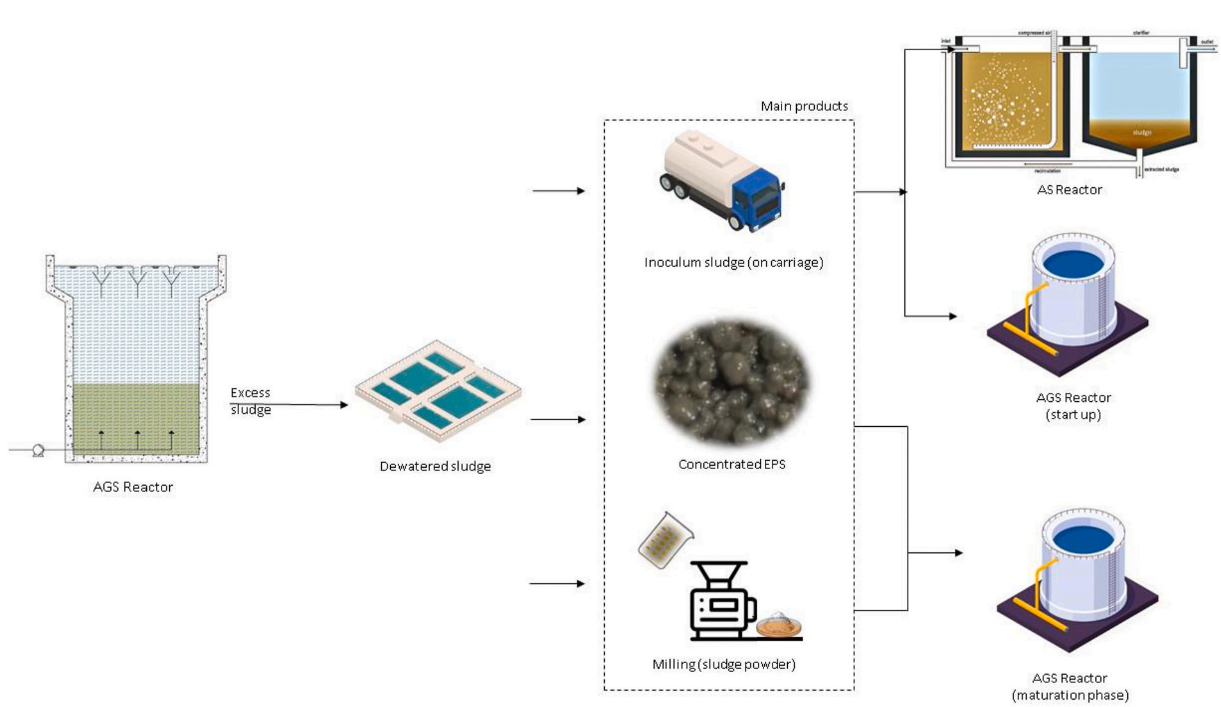

Fig. 2. Schematic for the reuse of excess AGS in WWTP.

recovery efficiency in WWTPs.

If Enhanced Biological P-Removal (EBPR process) is employed, up to 95% of the wastewater phosphorus can be concentrated in the sludge (Balmér, 2004). The literature reports that for each 3g of sludge (enriched with P), it is possible to obtain approximately 1g of phosphorus (Paul et al., 2001). Therefore, P recovery is obtained via the excess sludge rich in phosphorus discharged from the system. This process is economically beneficial to recover a high aggregated value product (phosphorus) and decrease disposal costs. The sludge transport and final destination costs may represent almost 50% of the WWTP's operational costs (Campos et al., 2009).

The biological P removal in wastewater involves incorporating phosphate by the micro-organisms (Reginatto et al., 2007). In this way, it is necessary to stimulate heterotrophic bacteria growth, named polyphosphate-accumulating organisms (PAO). The ordinary bacteria cells have stored about 2.6% in mass of phosphorus, while in the PAOs, this percentage increases up to 38% (Metcalf, 2003). So, the abundance of PAOs is essential when it aims to have a sludge with a high phosphorus concentration.

In this regard, PAOs bacteria are essential to phosphorus removal, but they are also important to stabilize the aerobic granules. Researches show that this microbial group, also known as slow-growing heterotrophic bacteria, assists in forming and maintaining good AGS granules during long periods of operation (Rollemberg et al., 2018). Different studies proved the high potential of aerobic granules to accumulate phosphorus. Li et al. (2014) obtained a P-rich aerobic granule during wastewater treatment, indicating that AGS can be prospectively used for P recovery in the excess sludge. He et al. (2020) have noted that aerobic granules may accumulate over 90% of the influent sewage P load. Pronk et al. (2015) operated a full-scale AGS and obtained phosphorus removal around 87%. Regarding phosphorus accumulation in activated sludge flocs (ASF) and aerobic granules (AGS), Rollemberg et al. (2019) have obtained values next to 0.05 and 1.4 mg P/mg VSS for ASF and AGS, respectively, indicating that aerobic granules had almost 30-times more phosphorus in the sludge.

The biological P removal process occurs due to the dynamic of phosphorus release and absorption by micro-organisms when subjected to changes from anaerobic to aerobic/anoxic environments, respectively (Zheng et al., 2014). Besides, the presence of volatile fatty acids (VFAs) in the anaerobic phase is necessary to make this process take place (Reginatto et al., 2007). Furthermore, it is possible to obtain phosphorus removal through anaerobic-anoxic cycles due to the denitrifying ability of DPAOs. These organisms use nitrate or nitrite instead of oxygen as an electron receptor, simultaneously promoting phosphorus removal and denitrification (Weissbrodt et al., 2017).

The abundance of PAOs could be affected by other microbial groups, such as GAOs (glycogen accumulating organisms) and OHOs (ordinary heterotrophic organisms), because these groups can directly compete with PAOs for the substrate. However, OHO's suppression occurs only with the proper maintenance of the anaerobic period (Weissbrodt et al., 2017). GAOs have a similar metabolism to PAOs, and they can obtain the energy necessary to capture acetate through glycolysis (Weissbrodt et al., 2017). Thus, it can be considered harmful to the EBPR process since these bacteria can store VFA, such as PHA in anaerobic conditions, but cannot promote the release or subsequent phosphorus absorption. These bacteria have been the cause of many failures detected in EBPR systems (Zheng et al., 2014).

As mentioned, in well-operated full-scale AGS systems, phosphorus removal can be higher than 85%. Phosphorus recovery from the granular sludge occurs mainly in 4 (four) ways: (i) crystallization followed by chemical precipitation (Lu et al., 2016); (ii) direct application to the soil (Nancharaiah and Reddy, 2018); (iii) phosphorus recovery through biosorbent using natural polymers (Dall'agnol et al., 2020); (iv) incineration and phosphorus recovery from sludge ash (Adam et al., 2009), as shown in Fig. 3.

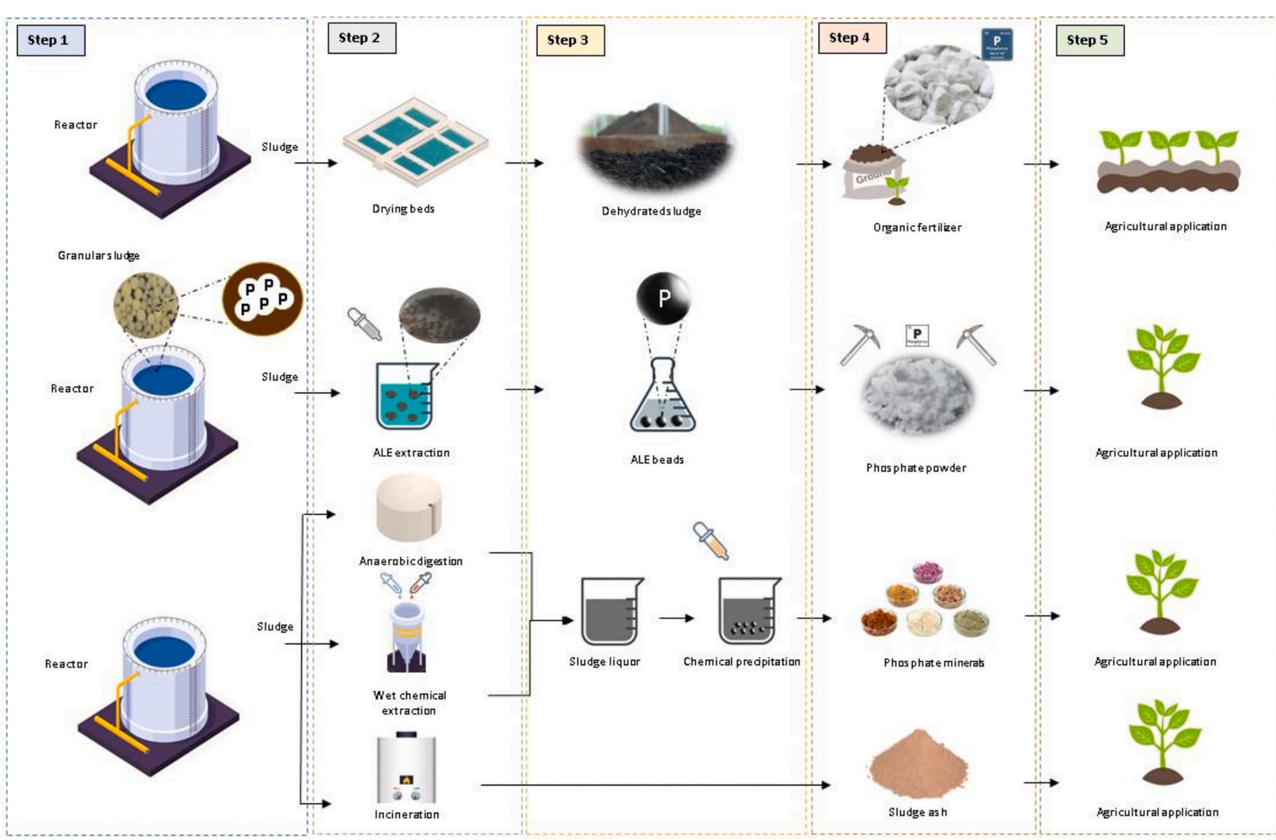

Fig. 3. Different possibilities for phosphorus recovery in AGS systems.

5.2.1. Phosphorus recovery by struvite crystallization or precipitation of hydroxyapatite - calcium phosphate

Struvite, used as a fertilizer, has the following advantages over the other fertilizers found on the market (Wang et al., 2005): (i) its dissolution is slow and, therefore, the applications of the mineral should be less frequent. So the plants can absorb the nutrients before they are washed away by surface runoff and are less subject to problems related to nutrient hyper saturation; (ii) fertilizers with mineral origin usually have heavy metals concentration (10^1 to 10^2) higher than that found in struvite; (iii) the essential macronutrients N, P, and Mg are introduced simultaneously on the soil, without the application of unnecessary components for the plants; (iv) due to being a slow-release fertilizer, eventual pollution of groundwater by the supply of nutrients or water bodies by the diffuse load is more difficult to occur. Controlled crystallization of struvite ($MgNH_4PO_4 \cdot 6H_2O$) is a way to recycle nutrients by extracting struvite from sludge digesting liquors because of its high concentrations of phosphorus, ammonium, and magnesium (Forrest et al., 2008).

Phosphorus recovery by crystallization occurs in three chemical phases: supersaturating (enrichment of P in the environment), nucleation (crystal formation), and crystal growth. Among the crystals formed, struvite and hydroxyapatite stand out due to their applicability in agriculture (Peng et al., 2018). The main types of phosphorus crystallization are biological induced crystallization and chemical crystallization (Kataki et al., 2016).

For the crystallization process to occur, it is necessary to have ammonia, phosphate, and metals (e.g., Mg, Ca, Fe, among others). It is worth mentioning that in the composition of AGS EPS, there are metals, standing out calcium and magnesium, which, combined with free phosphate, will form hydroxyapatite and struvite, respectively (Angela et al., 2011). This process normally occurs under alkaline pH from 7 to 11 and at a magnesium/ammonia/orthophosphate ratio of 1.5/1/1 (Münch and Barr, 2001).

In chemical crystallization are occurring reactions similar to those involved in biologically induced precipitation. However, the environmental conditions and the necessary components for crystallization are controlled with the addition of chemical reagents (Metcalf, 2003).

The precipitation of struvite is inefficient when applied to the solid fraction of wastewater due to the difficulty to separate struvite crystals from other solid materials, thus requiring phosphorus solubilization. Besides, it is important to mention that phosphorus is available as orthophosphate. For this reason, the precipitation involves an initial stage where the stored phosphorus by biomass must be released into the liquid phase, usually achieved in an anaerobic digestion reactor (Ahmad and Idris, 2014). The process may occur even faster if there is organic matter in the VFA form, facilitating the release of PO_4^{3-} (Yuan et al., 2012). It is important to highlight that when the digestion process is prolonged, there may be endogenous respiration with consequent release of organic P (through cell lysis). This compound will then be hydrolyzed and converted into inorganic P, thus increasing phosphorous' concentration in the liquid phase.

Xia et al. (2014) showed that the addition of COD in the form of acetate improved the phosphorus release process in the Phosphorus Accumulation Reactor (PAR) located before the precipitation reactor. The P was recovered from the liquid phase, while the solid fraction returned to the SBR used to treat the wastewater.

Some studies have assessed phosphorus recovery from excess sludge of AGS reactors, but several problems have been reported. Lu et al. (2016) studied the phosphorus release of AGS biomass and subsequent crystallization. The aerobic granules showed high phosphorus concentration, and more than 90% of the phosphorus accumulated in the biomass was released in the PAR. However, the authors observed that the excess granules used for P release disintegrated after the process, and it was not possible to recirculate to the reactor. Another issue studied is the low biodegradability of aerobic granules, which has made it difficult to release N into the liquid phase (Guo et al., 2020).

Another problem that makes the crystallization of struvite in aerobic granules harder is that a portion of the P accumulated in the granules is already precipitated in the sludge as phosphorus (mineral), functioning as a nucleus for the aerobic biomass (Daumer et al., 2008). Some studies have shown that several inorganic P species were present in aerobic granules, such as hydroxyapatite, struvite, or Ca–Mg phosphate. Thus, these minerals would already be readily available in the sludge for application, and there is no need to use the crystallization process mentioned (Angela et al., 2011). However, these studies pointed out that the greater or lesser content of these species in the granules directly depends on operational conditions, wastewater characteristics etc.

Struvite crystallization has high nutrient recovery rates, especially for recovering precious phosphate resources, and it is also economically viable. It is estimated that a WWTP with an influent flow rate of 0.1 m^3/s can produce struvite in the amount to generate a revenue of £ 8400–20,000 per year (Jaffer et al., 2002) even though there are problems associated with crystallization. The unintentional formation of struvite can block valves, tubes, centrifugation tanks, and pumps (Münch and Barr, 2001), and lead to decrease flow capacity and eventual equipment failure. Other issues include the high cost of chemical reagents for pH adjustment and to increase the amount of magnesium (Pastor et al., 2010). The phosphate concentration in sludge water from the digester can be quite high, like 85–95 gP/m^3 (Jaffer et al., 2002; Münch and Barr, 2001). The theoretical potential for struvite crystallization is close to 67,000 tons of P_2O_5 fertilizer per year in the UK and 270,000 tons in Western Europe (Gaterell et al., 2000).

5.2.2. Phosphorus recycle: biosolids application on agriculture

From an agronomic perspective, granular sludge is a material rich in organic matter, with large amounts of nutrients that can be used as fertilizer (Nancharaiah and Reddy, 2018). The application of biosolids into the soil involves spreading biosolids on the soil's surface, or incorporating or injecting biosolids into the soil. They are generally treated by at least one of the following processes, depending on the final use: (i) digestion, (ii) alkaline treatment, (iii) composting, and (iv) hot drying.

Biosolids obtained from biodigestion or alkaline stabilization can be used to recover degraded areas, soil applications (e.g., in agriculture as a fertilizer), among others. Composting produces highly organic and soil-like biosolids for use in horticulture, hatchery, and landscape. In addition to soil conditioning and reducing fossil fertilizers use, the biosolids application in the soil also prevents the excess of nutrients entering the environment due to its low nutrient content, compared to fossil-origin fertilizers. The main concerns with the use of biosolid involve health and safety, odor, and public acceptance.

In addition to the high content of nutrients, the EPS present in aerobic granules can assist in biomass application as a fertilizer. On the other hand, some authors have highlighted the problem of metals' content in biomass, given the high potential of granules to adsorb these materials (Nancharaiah and Reddy, 2018). Therefore, although aerobic granules have up to 30-times more phosphorus than activated sludge flocs (conventional systems), metals concentration in this biomass may be an issue, which can cause problems in agricultural reuse (Raheem et al., 2018).

The main disadvantage of using biosolids instead of sewage nutrients, as struvite crystals, is because the latter do not present pathogenicity or biodegradability. With the successful crystallization process, struvite salts have a high degree of purity, not requiring further processing. However, studies have shown that simple dehydration followed by mixing has been sufficient to stabilize excess sludge and apply it in agriculture (Świątczak and Cydzik-Kwiatkowska, 2018).

The use of excess sludge from granular biomass as biosolids becomes even more interesting because part of P minerals is already precipitated inside the granules (Daumer et al., 2008). The bacteria that accumulate phosphate can also be inside the granules (Lemaire, 2007). Therefore, the phosphate release could cause phophorous' precipitation inside the nucleus of the micro-organisms, making subsequent crystallization difficult. The phosphate precipitation in a granular sludge process was shown elsewhere (Yilmaz et al., 2007; de Kreuk et al., 2005).

A study conducted by de Kreuk and van Loosdrecht (2006) with aerobic granules verified that about 46% of the P removal occurred through precipitation, while the other part (i.e., 54%) occurred due to the accumulation of polyphosphate. In this sense, de Kreuk et al. (2005) used extraction techniques and verified that up to 5.0% of the sludge mass was due to precipitates (P/VSS). These values show that the excess sludge, in addition to having considerable accumulated intracellular P value, also contains a considerable amount of precipitated P. Therefore, the application of excess sludge as biosolids can be consolidated as an important way of reusing the solid phase, despite the concern with metals concentration.

5.2.3. Phosphorus recovery using natural biosorbent

Recently, an integrated approach to recover nutrients and biomaterials from wastewater treated in AGS systems has been studied (Rollemberg et al., 2020; Dall'agnol et al., 2020). In addition to conventional biological removal methods that involve enrichment of PAOs, some alternative phosphorus adsorption methods have been studied (Wang et al., 2018b). Different types of adsorbents have been proposed in the literature, but biological origin materials, named biosorbents, have been highlighted. They occur naturally, are renewable, and have lower costs than synthetic adsorbents (see item 5.3). Biosorbents can be converted into post-sorption added value products that can be applied to other uses, such as fertilizers in agriculture (Reddy, 2017).

Among the biosorbents, ALE has stood out, as it can be recovered from AGS, and after synthesis, it could be used again for phosphorus recovery. Dall'agnol et al. (2020) showed that ALE spheres from aerobic granules could be used for phosphorus adsorption from the liquid phase. Therefore, using ALE as a phosphorous adsorbent material, it is possible to find a way to reuse a sludge bio-product, reduce the amount of residual sludge, and recover phosphorus from the liquid phase. The overall results encouraged further studies on the possible application of this biomaterial enriched with phosphorus as a secondary source of this post-sorption added value nutrient.

5.2.4. Incineration and phosphorus recovery from sludge ash

Incineration is another possibility suggested for phosphorus recovery. Mono-incineration stands out among the techniques used, which consists of sludge incineration separated from the other residues. The residual ashes are rich in phosphorus in the form of calcium phosphate, aluminum phosphate, and iron phosphate (Melia et al., 2017). The problem with this technique is due to the impurities present in the ash, such as trace metals, preventing its direct use in the soil and requiring chemicals purification processes (Adam et al., 2009).

One of the techniques for removing phosphorus from ashes is the acidic dissolution of phosphorus (pH less than 2) to transfer phosphorus and part of the heavy metals from the ash to the liquid phase. After this, dissolution techniques are carried out to extract phosphorus, using acids such as HCl, HNO_3, and HF (Adam et al., 2009). To purify phosphorus in relation to the presence of heavy metals, techniques such as sequential precipitation, liquid-liquid extraction, sulfide precipitation, cation exchange, and nanofiltration are used (Petzet et al., 2012).

In addition to acid dissolution, sludge ash can be subjected to a thermochemical process to remove heavy metals from the ash. This technique basically consists of adding chlorine-based salts (KCl or MgCl) to a concentration of 5–15%, in which these compounds will react with heavy metals, decreasing their boiling temperature. The mixture must be heated to 900–1100 °C to volatilize the heavy metals (Mattenberger et al., 2010). This process has high efficiency for removing compounds such as Pb, Cd, Cu, and Zn. On the other hand, the removal efficiency of Ni and Cr is unsatisfactory. Additionally, depending on operational conditions, a loss of phosphorus (up to 30%) may occur during the process (Donatello and Cheeseman, 2013).

It is noted that the P recovery from the ashes has significant disadvantages compared to other forms of recovery due to the need to apply additional techniques to purify the phosphorus, thus requiring more energy and technology. Furthermore, it is a complex operation technique requiring specialized labor.

5.3. Tryptophan recovery

Tryptophan is a hydrophobic amino acid, and hydrophobicity is the main driving force of cell adhesion (Liu et al., 2004b; Wang et al., 2014). Because of this, it is usually present in AGS (Zhang et al., 2018). For its production, it is necessary substrates that have phosphoenolpyruvate and erythritol-4-phosphate as metabolic intermediates (e.g., glucose and glycerol) (Wen et al., 2017).

Besides having a fundamental role in granulation, tryptophan also has several applications in the chemical industry, agriculture, and, especially, in the pharmaceutical industry. Tryptophan is considered an essential component of the human diet, as it cannot be synthesized in the human body. Besides, it is the precursor to serotonin and melatonin (Mustafa et al., 2018).

The production of this compound can take place in three ways, chemical synthesis, biochemical synthesis, and microbial fermentation (Liu et al., 2009). In AGS, this production can be related to micro-organisms, such as Thauera and Paracoccus (Zhang et al., 2018). According to Sanchez and Demain (2009), the need for tryptophan in the world is more than 50 thousand tons per year, with an increase of 10% per year. In this sense, new methods to produce this compound are required, and AGS could be an alternative to this demand.

Rollemberg et al. (2020) related concentrations of tryptophan about 60 mg TRY/gVSS. The author observed that C/N ratio had an influence on TRY concentration. The higher the N concentration, the higher the amount of TRY. Furthermore, granules with a diameter between 0.5 and 1.0 mm had more TRY. Another factor that can influence tryptophan yield in AGS is the SRT. According to Zhang et al. (2019), SRT around 6 days is ideal to TRY production.

In terms of production rates, Rollemberg et al. (2020), in an AGS system fed with acetate, observed values of 4.0 gTRY/kgCOD·day. On the other hand, Rollemberg et al. (2020), cultivating AGS with municipal wastewater, had a production rate of 2.5 gTRY/kgCOD·day, confirming the substrate's effect on TRY concentration in the granule. Some authors believe that more fermentable effluent, for example, dairy wastewater and similar, could increase TRY in the granules (Roager and Licht, 2018).

Tryptophan recovery from AGS is considered disadvantageous due to its low concentration, and the process of extraction and purification is not clear yet. In the literature survey, any method of TRY recovery or purification was found. Regarding low concentration in granules, Rollemberg et al. (2021) observed an ALE/TRY ratio of 5 during municipal wastewater (COD $\approx$ 600 mg/L and TN $\approx$ 50 mg/L in the influent) treatment in pilot-scale AGS reactors. Additionally, TRY has a lower market value than the other products that can be recovered from AGS. It worth mentioning that, although this product has been identified as a resource, the studies are mainly focused on its role in the granulation process and sludge biodegradability (Zhang et al., 2018; Wang et al., 2018a).

5.4. Alginate-like exopolysaccharides (ALE) recovery

Alginates are natural polymers consisting of linear (unbranched), non-repeating copolymers of β-(1–4) linked d-mannuronic acid and β-(1–4)-linked l-guluronic acid units, which exist widely in brown seaweeds such as species of Ascophyllum, Durvillaea, Ecklonia, Laminaria, Lessonia, Macrocystis, Sargassum, and Turbinaria. Alginic acid exists as a salt, sodium, calcium, or magnesium alginate, for example (Lee and Mooney, 2012). The blocks are composed of consecutive G residues (GGGGGG), consecutive M residues (MMMMMM), and alternating M and G residues (GMGMGM). Additionally, the type of source affects M and G contents and the length of the blocks (Lee and Mooney, 2012).

Besides the algal, two bacterial genera, Pseudomonas and Azotobacter, have been described to produce alginate as an exopolymeric polysaccharide during their vegetative growth phase. The biological function in brown algae is related to structure, while in bacteria is more diverse. In the Gram-negative soil bacterium *Azotobacter vinelandii*, it is related to maintain structural integrity under adverse environmental conditions, and in *Pseudomonas aeruginosa*, it seems to be an important virulence factor during the infectious process of human epithelia (Moradali et al., 2017). Importantly, algal and bacteria alginates differ substantially from each other concerning their composition, modifications, molecular mass, viscoelastic properties, and polydispersity (Moradali et al., 2017).

In biofilms, extracellular polymeric substances (EPS), a complex mixture of polysaccharides, proteins (structural proteins or exoenzymes), nucleic acids, (phospholipids, humic substances, and intercellular polymers), keep micro-organisms together (Lotti et al., 2019). Chen et al. (2007) described for the first time the distributions of EPSs (proteins, α- and β -polysaccharides, and lipids) and cells (total and dead) in aerobic granules. After that, studies found the capability to form a gel of some EPS (Seviour et al., 2009). These polymers were called alginate-like exopolysaccharides (ALE) because their characteristics were similar to those of commercial alginate (Lin et al., 2015). In a mixed consortium as exists in aerobic granular sludge, possibilities may exist that alginates produced by both Pseudomonas and Azotobacter or other bacteria are exposed to those extracellular epimerases simultaneously, resulting in ALE with a high proportion of GG blocks (Liu et al., 2010). The difference between ALE and alginate is in the reactions with acid ferric sulfate, phenol-sulfuric acid, and Coomassie brilliant blue G250, which might be attributed to the appearance of O-acetylated substitution groups (Lin et al., 2015).

Lin et al. (2015) explored the recovery of polysaccharide-based biomaterial from AGS for application as a surface coating material. The recovered biomaterial contained both polysaccharides, and lipids showed amphiphilic character. The biopolymer was found to form a water-resistant film on hydrophilic surfaces such as paper. Another applicability is as flame-retardant materials for coatings (Kim et al., 2020). According to this latter research, Flax fabric coated with EPS recovered from aerobic granular sludge meets the flame retardancy requirements in the US Federal Aviation Regulation standards.

The extractable ALE is a major structural polymer and accounts for approximately 15–25% of the organic fraction in AGS (Felz et al., 2016). It is considered a valuable biobased product that can be extracted from waste AGS and can be a raw material for applications in sectors like chemical, paper, and textile industries. This polymer can also be used as a soil conditioner to improve water retention in semiarid environments (Wang et al., 2018).

Furthermore, ALE can be applied in agriculture as a matrix for fertilizer production in the construction industry to improve building materials' characteristics (e.g., fire resistance), or as a thickener for inks. Also, it can be used as a sustainable biosorbent for dye removal from aqueous solutions. For instance, Ladnorg et al. (2019) studied the removal of methylene blue in synthetic wastewater and found 69% and 79% while using ALE beads and alginate beads, respectively. The adsorption capacity of ALE was also verified for Pb^{2+} containing wastewater (Wang and Li, 2012).

Another applicability that has been studied is phosphorus removal. Dall'agnol et al. (2020), using ALE beads in AGS, showed phosphorus removal of 72.5% compared to that obtained using alginate beads (91.0%). This study also showed that ALE and alginate beads presented a similar capacity compared with other materials. They also suggested after detailed studies on toxicity and nutritional composition that ALE enriched beads could be used in agriculture or animal feed.

Alginate is used in the food industry as a stabilizer and moisture retention agent. Furthermore, it is used as an emulsifier in the cosmetical industry and as a thickener of paints and paper (Dhargalkar and Pereira, 2005). Lin et al. (2013) reported that ALE extracted from aerobic granular sludge showed similar sizing effects with commercial sizing chemicals. More knowledge about ALE composition and properties will help find more applicability in the industry (Lotti et al., 2019).

Rollemberg et al. (2020) and Schambeck et al. (2020), evaluating the presence of ALE in pilot reactors treating municipal wastewater, found ALE production rate of 10 gALE/kgCOD·day and 9 gALE/kgCOD·day, respectively. On the other hand, Rollemberg et al. (2020) found an ALE production rate of 17 gALE/kgCOD·day when AGS was cultivated with synthetic wastewater. Such a difference may be related to the carbon source because the works that used acetate presented higher ALE content in the granules than the studies that used domestic/municipal wastewater. Meng et al. (2019) cultivated AGS in a lab-scale reactor using synthetic sewage (a mixture of acetate and glucose), obtaining values of 6.9 gALE/m^3·day. Therefore, the substrate may have a significant impact on ALE production. As it is known, carbon source impacts the abundance of microbial groups (Rollemberg et al., 2019). Acetate, for example, is known to favor the presence of PAOs and GAOs (Bassin et al., 2012). In this regard, studies using propionate are recommended to assess whether the formed granules have a higher ALE content because this substrate is known to be one of the best substrates to support PAOs' growth (Rollemberg et al., 2018).

A Life Cycle Assessment (LCA) of ALE recovery from Nereda® systems was conducted, considering aspects such as influent characteristics, chemical use (7–14 L HCl and 0.2–1% Na_2CO_3), extraction temperature (long time at 40 °C vs. short time at 80 °C), separation technique (centrifugation vs. filtering) and ALE yield (16–29%) (Report, 2016–22, STOWA).

ALE recovery from AGS systems appears to be a trend. As it is known, alginates are produced from seaweeds, and the availability and costs of alginate seaweeds are beginning to be a concern of alginate producers. Higher costs have been driven by higher energy, chemicals, and seaweed costs, reflecting seaweed shortages (Bixler and Porse, 2011).

In a field test in Zutphen, The Netherlands, it was demonstrated that 18 kg bio-ALE could be produced from 80 kg of Nereda granular sludge, i.e., 22.5% bio-ALE recovery (Van Leeuwen et al., 2018). This result agrees with the value of 23% bio-ALE recovery obtained in a pilot-scale AGS SBR treating municipal wastewater (Rollemberg et al., 2020). In December 2020, a second factory started the operation in Epe, The Netherlands. Both factories can produce up to 500 tons of Kaumera annually. The term "Kaumera" is also used to represent the different biopolymers (including ALE) that can be extracted from AGS originated from the Nereda wastewater treatment process. So, depending on the AGS characteristics, a different biopolymer for a specific commercial application can be much more interesting to be produced (Royal HaskoningDHV, 2021). The total Dutch production is estimated at 85,000 tons per year from 2030. The market price depends on the quality and subsequent application. However, the total value in the Dutch market is currently estimated at € 170 million per year from 2030 (Van Leeuwen et al., 2018).

The analysis of the values reported above leads to the conclusion that one ton recovered from ALE (after extraction and refining costs) generates a final revenue of € 1000–2000. These results show that a WWTP with AGS technology treating domestic wastewater (COD $\approx$ 600 mg/L), with a flow rate of approximately 3.0 m^3/s, would be able to produce approximately 1 ton of ALE/day, generating a revenue of approximately € 365,000–730,000/year, which could decrease the OPEX of the treatment plant.

According to the study conducted by Oliveira et al. (2020), which analyzed the yield of Na-EPS and H-EPS, for 4 months, of a full-scale AGS system treating municipal influent, it is possible to produce 7–10 and 1–2 tons per day, respectively. This is taking into account that the aerobic biomass yield is 0.4 g MLVSS/g COD, and the organic loading rate is 6 tons COD/day.

The protocol for ALE extraction from AGS biomass and possible uses

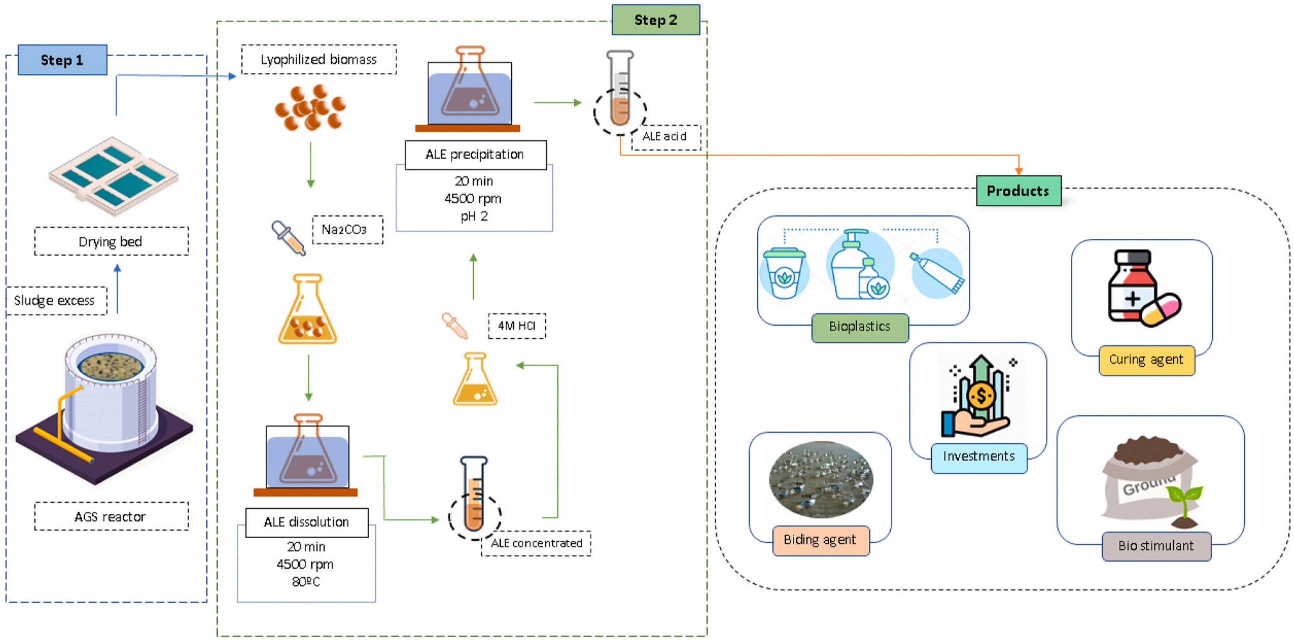

Fig. 4. ALE extraction from AGS biomass and possible uses.

is shown in Fig. 4.

5.5. PHA from aerobic granules

Annually, the world produces more than 350 million tons of plastic (Plastics Europe, 2018). Despite being an essential input to modern society, excessive demand has environmental impacts, for example, depletion of oil reserves, accumulation of plastic wastes in terrestrial and aquatic ecosystems, and release of toxic gases into the atmosphere caused by its incineration (Koller, 2017).

As an option to reduce these problems, green plastic or bioplastic is gaining commercial interest due to its production from renewable sources and biodegradability and biocompatibility properties (Fernández-Dacosta et al., 2015). Among them, polyhydroxyalkanoates (PHA) form a group of biopolyesters with mechanical properties similar to thermoplastic, such as polyethylene and polypropylene. Due to structural changes in its monomers, this material can be homo (for instance, poly-3-polyhydroxybutyrate, PHB) or copolymers (for instance, poly (3-hydroxybutyrate-*co*-3-hydroxyvalerate, PHBV), which are different in terms of chemical properties and composition (Li et al., 2016). Thus, the industrial application has a range of possibilities such as the manufacture of packaging, fibers, inks, biofuels, paper finishing, pesticide carriers, and moisture containment layers in sanitary products (i.e., diapers, absorbents), in addition to the biomedicine sector (Mathuriya and Yakhmi, 2017).

Since 1990, PHA production has been made on a large scale by pure microbial culture and commercialized by companies all over the world, such as Biomer (Germany), Bio-On (Italy), Danimer Scientific (USA), Kaneka (Japan), TianAn Biologic (China) e PHB Industrial (Brazil) (Chen, 2010). However, the sector faces difficulties due to the high operational costs, as sterile conditions, pure substrates, and extraction and purification methods are needed (Fernández-Dacosta et al., 2015). As a result, PHAs are 3 or 4 times more expensive (US$ 5–6/kg) than synthetic plastic (US$ 1.3–1.9/kg) (Kourmentza et al., 2017).

The synthesis of PHA takes place under conditions where growth is restricted. For example, when nutrients (nitrogen and phosphorus) are limited, dissolved oxygen is low and organic matter is in excess, some micro-organism can store carbon in the form of an intracellular inclusion so that it can be used for cell growth and to maintain the metabolism during the starvation period (de Kreuk and van Loosdrecht, 2004). Based on that, it is possible to impose operational conditions to select bacteria that can store PHA, thus optimizing the production. Sequencing Batch Reactor (SBR) used in the cultivation of AGS allows subject the biomass to unbalanced conditions, such as substrate feast and famine, as well as alternating aerobic and anaerobic periods, which is different from continuous conventional activated sludge (Qin et al., 2005). When the system has an anaerobic feeding period (feast) followed by a long aerobic famine period, bacteria groups that store PHA are selected: polyphosphate accumulating organisms (PAOs) and glycogen-accumulating organisms (GAOs) (Rollemberg et al., 2018). Because they have stored substrate, the growth rate is slower, which favors the development of dense and stable aerobic granules with good sedimentability (Sludge Volume Index, SVI < 50 mL/g) (de Kreuk and van Loosdrecht, 2004). However, the amount of PHA accumulated by these groups of micro-organisms is generally less than 20% of dry cell weight (CDW) (Satoh et al., 1996).

Mature granules rich in PHB have regular morphology with a diameter between 1.0 and 3.0 mm and a porous internal structure that probably facilitates the transport of substrate and nutrients (Wang et al., 2014). In general, smaller granules tend to store more PHA because the bigger surface area improves substrate diffusion, producing a higher amount of PHA (Gobi and Vadivelu, 2015).

PHA in granules can reach 40–70% of cell dry weight (CDW), in g PHA/g VSS, showing the high capacity of aerobic granular biomass to store this component (Wang et al., 2017a). In terms of volumetric yield, the amount of PHA produced using AGS, due to high density, is higher than activated sludge. For example, Waller et al. (2012) obtained 0.36 mg PHA/mg COD in activated sludge, with a yield of 39% of CDW. On the other hand, Gobi and Vadivelu (2014) obtained 0.66 mg PHA/mg COD and a yield of 68% of CDW in aerobic granular sludge. Additionally, the EPS concentration accompanied the increase in PHB, probably due to the stress conditions imposed in the cycle (Wang et al., 2014; (Li et al., 2019a).

One strategy to enhance PHA production in AGS is to apply an anaerobic followed by an aerobic period because it is possible the selection of PAOs and GAOs. During anaerobic feeding from the bottom of the reactors, all biodegradable organic matter is completely consumed, accompanied by glycogen and PHA production, and phosphate release. In the subsequent period, aerobic famine, glycogen is restored, and PHA is oxidized to support cell growth and maintenance in both micro-organisms, while phosphate is absorbed by PAOs cultivated aerobically and anoxically (Bassin et al., 2012). Johnson et al. (2009) reported a PHA's stock of 89% for activated sludge treating synthetic wastewater (acetate) during a long AN/AE process cycle. In contrast, Karakas et al. (2020) obtained a maximum yield of 10.8% of PHA in an AGS system treating municipal wastewater under AN/AE process.

The production of PHA can be associated with a biological WWTP designed to remove organic matter and nutrients. For example, Morgan-Sagastume et al. (2015), in a pilot-scale SBR, integrated an enrichment step for PHA production by applying aerobic dynamic feeding with municipal wastewater. The reactor was fed with plug-flow filtered influent municipal wastewater (influent: 290–570 mg COD/L, 35–60 mg N/L, and 4.3–7.6 mg P/L) operated under aerobic feast-famine at high organic loading rates (OLRs = 3.0 ± 0.8 gCOD/L·d) and short hydraulic retention times (HRT ~ 3 h). The cycle consisted of feeding (aeration and mixing: 10 min), reaction (aeration and mixing: 15 min), settling (25 min) and effluent discharge (10 min), and further reaction (aeration and mixing: 45 min). The SBR was operated with total suspended solids (TSS) controlled by sludge retention times (SRT) to sustain a stable specific OLR. The system obtained removal efficiency of 70% COD, 24% nitrogen, and 46% phosphorus. Besides, the granules have, on average, 0.14 g PHA/g VSS.

The advantage of AGS to produce PHA is the possibility to have all the required processes to produce a bioplastic in one reactor, which reduces area demand and operational costs related to the treatment process. According to Gobi and Vadivelu (2014), the longer the feast period, the higher the PHA concentration found in the AGS, suggesting then the sludge discharge period.

The extraction of PHA consists of two steps. The first is cell digestion, and the second is solvent extraction (Tan et al., 2014). According to Gobi and Vadivelu (2015), the extraction of PHAs from AGS is different from activated sludge and pure culture because EPS is present in higher quantity, leading to a higher resistance to the extraction process. Some studies evaluated using different solvents to extract PHA from aerobic granular biomass (NaOCl, C_3H_6O, NaOH, and NaCl). NaOCl had the best PHA recovery and EPS removal, avoiding adverse effects in the dissolution step with solvents. After cell lysis, usually colored compounds are used as solvents (e.g., chloroform) to capture the PHAs from the solution, which can be further recovered by solvent evaporation or by precipitation, adding compounds like methanol (Samorì et al., 2015).

5.6. Biogas recovery from aerobic granules

Another possibility for resource recovery in AGS reactors would be anaerobic digestion or co-digestion of excess sludge to generate biogas. In this sense, some studies have shown that the excess of aerobic granules could be sent to an anaerobic reactor for methanization (Val Del Río et al., 2013; Bernat et al., 2017), the digestate would return to the treatment, and the sludge could be applied in agriculture (already stabilized). However, some studies have reported problems with the digestibility of aerobic granules. Bernat et al. (2017) demonstrated that biodegradation was difficult to occur due to the granules' chemical composition. The content of lignocellulosic substances comprised about 54% of the fibrous materials, showing that the granules' biogas potential was about two times lower than activated sludge. In terms of production, the authors obtained values between 0.3 and 0.4 m^3/kg Total Solids. It was also observed that the methane content was about 56.7–59.5%.

Val Del Rio et al. (2013) tested the biogas potential of aerobic granular sludge from the SBR pilot-plant fed with the liquid fraction of swine sludge. It was observed a production of 0.35 m^3 of biogas/kg VSS fed, assuming 60% of the methane content, lower than the range of 0.5–0.75 m^3/kg of VSS reported in the literature (Van de Velden et al., 2008).

The high protein content in the granules, which is essential for AGS stability (Rollemberg et al., 2018), and also the high content of fiber (more than 18% of TS), especially lignin, which hamper biodegradation, contributed to the low methane yields in the BMP tests (Bernat et al., 2017; Val Del Río et al., 2013). In the activated sludge, it is possible to find up to about 20% of fiber, but this is usually composed of hemicellulose, which is easier to biodegrade than lignin (Bernat et al., 2017). In all of these papers, a methane content below 60% was observed.

Some studies have shown that after crushed, the granules could exhibit greater biodegradation kinetics, which means that destroying the structure of aerobic granules may, in fact, accelerate the rate of degradation of rapidly degradable organics (Val Del Río et al., 2013). However, mechanically destroying the AGS's compact structure does not significantly affect biochemical methane potential (BMP); it only accelerates the rate of degradation of rapidly biodegradable organics and releases a fraction of the slowly biodegradable organics.

Guo et al. (2020) showed the occurance of two different types of biomasses in AGS reactors: the mature aerobic granules located in the sludge bed (bottom) and the biomass containing granules of low sedimentability and filamentous sludge located in the sludge blanket. Rollemberg et al. (2020) evaluated two types of sludge discharge from AGS reactors (Adam et al., 2009): sludge that is removed at each cycle, which is named selective sludge discharge (SD). This is the most flocculent sludge with a slower sedimentation rate than aerobic granules. So, when removing this sludge, a biological selection pressure is applied to faster settling granules, which have a higher retention time (Ali et al., 2019; Ahmad and Idris, 2014); the excess granular sludge present in the reactor bed, which have a high state of maturity, is also removed (MG). They have a high SRT and are discarded to control solids' retention time in the AGS reactor (daily discharge). The authors observed that the biomass removed by the selective sludge discharge (AGS-SD, method 1) presented physical characteristics similar to activated sludge. The AGS-SD was characterized by a high BMP value, close to 0.25 m^3 CH_4/kg VSS. On the other hand, the BMP value of mature granules (AGS-MG, method 2) was close to 0.20 m^3 CH_4/kg VSS.

These results indicated that the biomass removed in the selective sludge discharge (removed in each cycle) has greater viability for anaerobic digestion and methane generation. On the other hand, studies have shown that it is worth investigating in the future whether changes in the operational parameters of the Nereda® reactor can affect the characteristics and digestibility of the different biomass fractions taken from the AGS reactors (Pronk et al., 2015). For example, AGS reactors with lower SRT are believed to have much higher BMP (Ali et al., 2019).

Regarding the biogas obtained, it can be used to obtain heat, energy, or cooking. The combined heat and energy systems (CHPs) stand out among the existing systems, which use biogas produced from anaerobic digestion to generate heat and electricity on-site (Mathiasson, 2020). The electricity produced by CHPs is reliable and consistent. However, the installation requires relatively high capital costs (around $ 2000/kW for an internal combustion engine, $ 7500/kW for a fuel cell, and $ 4500/kW for microturbine). Besides, the operation of CHPs requires a large volume of biogas, which restricts its implementation in small wastewater systems. CHPs have been reported to be economical only for WWTPs with a flow rate above 19,000 m^3/day (EPA, 2008). Of all the WWTPs operating in the USA, most of them being aerobic processes, less than 0.6% use biogas to generate electricity (EPA, 2007). The low application rate is due in part to the predominance of small wastewater systems in the USA. About 94% of WWTPs in the USA have a flow rate of less than 20,000 m^3/day (EPA, 2008). So, the wastewater treatment conception in terms of centralization level will be a determining factor for biogas recovery from the digested sludge in AGS WWTPs.

5.7. Other value-added by-products (recent discoveries)

Recently, new polysaccharides with added values were detected in EPS, such as glycosaminoglycans (GAGs) (Felz et al., 2020). Operational conditions in AGS, such as shear stress and feast/famine periods, favor GAGs yield by the micro-organisms (e.g., Streptococcus and Pasteurella). The special interest in GAGs is due to its use in the food, biomedical, and pharmaceutical industries (Felz et al., 2020; Liu et al., 2011). According to Chong et al. (2005), it is estimated that the world market for hyaluronic acid, a type of GAG, moves around 1 billion dollars per year. Therefore, this compound should be studied in future research due to its high market value and concentration in aerobic granules' EPS (Felz et al., 2020).

Other polysaccharides that were reported in aerobic granules are cellulose, xanthan, and curdlan. Cellulose is a light-dense biopolymer, insoluble in water, and has high mechanical resistance. The product can be used in biomedicine, electronics, and food industries (Esa et al., 2014). On the other hand, xanthan is more complex, and it can be applied in food, chemical (hydrocolloid production), pharmaceutical, and oil recovery (Freitas et al., 2017). They are produced by *Xanthomonas* spp, reported in AGS systems (Zhang et al., 2019). Regarding curdlan, it is a water-insoluble polymer with high molecular weight, and it was reported in experiments conducted by Li et al. (2019b).

In addition to tryptophan, other amino acids present in the EPS of aerobic granules (e.g., tyrosine and phenylalanine) have industrial applicability. Tyrosine is an aromatic amino acid with applications as food supplements, in the production of medicines, in agriculture, and as raw material for other chemical products (Patnaik et al., 2008). Additionally, tyrosine could be one of the main molecules for granulation (Wang et al., 2017b). Regarding phenylalanine, it is a basic component of sweetener, and it can be used as a food supplement (Cuellar et al., 2010). The metabolic pathway for these products is similar to tryptophan, and they have been found together in EPS (Niu et al., 2019).

Although the recovery chain of some resources that can be recovered from aerobic granules is already known, it is worth highlighting several by-products recently discovered in aerobic granules, which have high added value and high commercial demand. However, the literature lacks a more detailed characterization and the factors that influence their production in AGS systems.

6. Conclusions

This paper presented a systematic review of resources recovery such as water, energy, chemicals, raw materials, and nutrients from AGS systems, also analyzing aspects of engineering and economic viability. Reuse of water and the use of sludge as inoculum in AGS WWTPs or to improve the characteristic of biomass in activated sludge systems are the most applied method of reuse. Phosphorus recovery, especially to apply to the soil, seems to be a possible alternative. ALE recovery has been the main alternative within all solid by-products, providing a reduction in OPEX of up to 50%. Resources recovery in AGS systems could be a solution to reduce the treatment costs, making the technology more feasible and decentralized.

Credit author statement

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Declaration of competing interest

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

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