



Biological processes modelling for MBR systems: A review of the state-of-the-art focusing on SMP and EPS

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ABSTRACT

A mathematical correlation between biomass kinetic and membrane fouling can improve the understanding and spread of Membrane Bioreactor (MBR) technology, especially in solving the membrane fouling issues. On this behalf, this paper, produced by the International Water Association (IWA) Task Group on Membrane modelling and control, reviews the current state-of-the-art regarding the modelling of kinetic processes of biomass, focusing on modelling production and utilization of soluble microbial products (SMP) and extracellular polymeric substances (EPS). The key findings of this work show that the new conceptual approaches focus on the role of different bacterial groups in the formation and degradation of SMP/EPS. Even though several studies have been published regarding SMP modelling, there still needs to be more information due to the highly complicated SMP nature to facilitate the accurate modelling of membrane fouling. The EPS group has seldom been addressed in the literature, probably due to the knowledge deficiency concerning the triggers for production and degradation pathways in MBR systems, which require further efforts. Finally, the successful model applications showed that proper estimation of SMP and EPS by modelling approaches could optimise membrane fouling, which can influence the MBR energy consumption, operating costs, and greenhouse gas emissions.

1. Introduction

Membrane bioreactors (MBR) are widely known as reliable elements of water resource recovery facilities (WRRFs) in terms of effluent quality, compliance with strict regulation limits, low sludge production, well-arranged operation, and low spatial requirements (Mannina et al., 2020, 2021; Zuthi et al., 2017; Zheng et al., 2018). Several studies were performed in the past years to ensure that MBR could become more

mature and widespread (Bozkurt et al., 2016; Krzeminski et al., 2017). Indeed, their full-scale applications have been registered very often (Attiogbe, 2013; Xiao et al., 2014; Meng et al., 2017). However, managers and researchers still present membrane fouling issues, module blocking, high energy consumption, and, by a consequence, high operating costs as significant obstacles to an ever more spread application of this technology (Tang et al., 2022). Although practical examples show that significant reduction in energy consumption and a long membrane

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lifetime are possible (Tao et al., 2019; Brepols et al., 2020), still, finding solutions to the obstacles above demands comprehensive studies.

Studies focusing on experimental data can be complemented by others using mathematical modelling to obtain predictive possibilities with less time-consuming routines and lower cost of implementation (Sun et al., 2016; Charfi et al., 2017; Mannina et al., 2018, 2019). In the past years, several works have been developed in view of demonstrating how mathematical modelling could be applied to MBR systems (Naesens et al., 2012; Nadeem et al., 2022) and their utilisation has been contributing to updating the knowledge of the technology (Krzeminski et al., 2017; Robles et al., 2018). In particular, the activated sludge model (ASM) family (Henze et al., 2000), formerly developed for conventional activated sludge (CAS) systems, has been expanded to consider the specific biomass kinetics related to MBR bioprocesses. These models are known as biomass kinetic or hybrid models (Mannina et al., 2021).

The biomass kinetic models are modified versions of ASMs with the ability to account for the formation and degradation processes of soluble microbial products (SMP) and extracellular polymeric substances (EPS), either as stand-alone models or as part of the ASMs (Zuthi et al., 2012). The need for hybrid models is due to the particular characteristics of MBR systems, e.g., higher concentration of mixed liquor suspended solids (MLSS) in the reactor and/or higher solids retention time (SRT), which contribute to the formation of microbial products in the MBR (Lu et al., 2001). These microbial products are known to cause membrane fouling, which has been one of the main constraints of MBR technology (Liu et al., 2018; Lin et al., 2014; Wang et al., 2022). The permeability of the membrane decreases due to fouling and leads to an increase in energy consumption caused by filtration and aeration (Juang et al., 2013). Mannina et al. (2017) showed the interlinkages between fouling, operational costs, and greenhouse gas emissions (GHG) from MBR systems. Fouling also increases chemical cleaning frequency (Wang et al., 2020). Therefore, minimising fouling would decrease energy and chemical consumption and eventually environmental footprint of the MBR system (Ioannou-Ttota et al., 2016). Thus, considering the formation/degradation of SMP and EPS is a reasonable approach while assessing the biomass and bulk properties that influence the MBRs filtration performance (Lu et al., 2001). Indeed, these hybrid models are particularly important in developing an integrated MBR model (i.e., a combination of hybrid and physical models) to fully understand MBR behaviour from a modelling standpoint.

Several studies in the past (Lu et al., 2001; Zarragoitia-González et al., 2008; Janus and Ulanicki, 2010; Mannina et al., 2011-2021; Zuthi et al., 2012) have examined the bioprocesses related to MBR modelling, mainly focusing on the correlation between biomass kinetics and membrane fouling. This review aims to facilitate a re-evaluation of findings from past studies by providing a current state-of-the-art in biomass kinetic process modelling, with special attention to the novel approaches to modelling SMP and EPS formation and degradation processes. Therefore, this work presents an overview of the concepts of SMP and EPS formation/degradation processes, followed by an overview of the biomass kinetic models. Then, the past and present applications of hybrid models to MBR are presented with a focus on updates related to bioprocesses. Finally, the main outlooks and conclusions retrieved from the review are presented.

2. General characterisation and mechanisms of SMP/EPS formation and utilization in MBR

The SMP concept was first introduced by Luedeking and Piret (1959) by studying glucose metabolism. Two new components were introduced, including UAP for utilisation associated products (growth-associated products) and BAP for biomass associated products (by-products of cell lysis). The following equation was used to translate the dynamic approach where X_B stands for active biomass.

$$\frac{dS_{SMP}}{dt} = \alpha \underbrace{\frac{dX_B}{dt}}_{S_{UAP}} + \underbrace{\beta X_B}_{S_{BAP}} \quad (1)$$

The existence of organic compounds generated by microbial cultures involved in wastewater treatment has been recognized in the 1960s (Barker and Stuckey, 1999). Nowadays, SMP and EPS are substances that cause fouling (Meng et al., 2017).

Prior to the presentation of SMP/EPS main concepts, some aspects must be introduced to ensure the full understanding of their formation and degradation processes. First, the organic substrates with high molecular weights (MW) are used by microorganisms for growth and become available due to a series of enzymatic reactions, collectively named hydrolysis. The hydrolysis allows slowly particulate biodegradable compounds (X_S) (with high molecular weight) to be converted into readily biodegradable substrates (S_S). The hydrolysis reactions related to the formation/degradation processes of SMPs, may occur in aerobic, anaerobic, and anoxic conditions. In the biomass growth process, the readily biodegradable substrate is directly used for growth or stored for internal processes. On the other hand, biomass decay/lysis and floc dissolution/degradation processes occur during the treatment processes. Most of the processes above, that may release SMP/EPS as by-products, are described by kinetic rate expressions and are detailed in modelling approaches that can account for such compounds.

It is generally believed that SMP are primarily formed during substrate utilisation, biomass decay, and hydrolysis of EPS (Fenu et al., 2010). They are released during cell lysis, lost during synthesis, excreted for some purpose, or diffuse through the cell membrane (Laspidou and Rittmann, 2002a; Le-Clech et al., 2006). In other words, SMP could be defined as the pool of organic compounds that are released into the solution due to microbial metabolism during growth and decay of biomass (Barker and Stuckey, 1999). It is now widely accepted that the SMPs could be divided into two groups, as originally proposed by Namkung and Rittmann (1986), UAPs and BAPs. The differences between both groups rely upon their production mechanisms, i.e., the bacterial phase from which they are derived (Lu et al., 2001). The UAPs are produced during substrate metabolism and biomass growth, with a production rate proportional to substrate utilization (Namkung and Rittmann, 1986; Barker and Stuckey, 1999). On the other hand, BAP can be defined as a by-product of endogenous respiration of cell mass and its production is independent of the cell growth rate (Zuthi et al., 2012). Indeed, their production mechanisms include either decay of the active biomass, hydrolysis of bound EPS, turnover of intracellular components, or a combination of those processes (Zuthi et al., 2013a; Liu et al., 2018).

MW of microbial products is important since it affects the specific filtration resistance which is an index to represent the fouling propensity of a foulant (Teng et al., 2020). It is important to note that the chemical structure of microbial products is as essential as their MW. According to Meng et al. (2011), the primary component of the high-MW compounds (>100 kDa) found in both the sludge supernatant and the biofilm of an MBR was predominantly polysaccharides. The high tendency of polysaccharides to cause fouling is not only due to their large size but also because of their significant gelling properties (Meng et al., 2017). The presence of humic substances and proteins adds complexity to fouling in MBRs. Hydrophobic humic substances adsorb to membranes, reducing pore size and altering their surface properties that facilitate the accumulation of hydrophilic biomolecules, predominantly polysaccharides (Kimura et al., 2015). Furthermore, proteins and polysaccharides form non-covalent interactions, creating a network that promotes fouling (Neemann et al., 2013). Zhou et al. (2012) determined that the biopolymers that are associated with the fouling present in the biofilm were primarily comprised of slowly biodegradable polysaccharides, which originated from SMP. Schiener et al. (1998) showed that MW of SMP showed bimodal distribution with 30% >1 kDa and 25% > 100 kDa. The SMP with low MW is associated with UAP and high MW is with BAP (Urban et al., 1998; Medina et al., 2020). Ni et al. (2011) showed that

the UAPs exhibit the characteristics of carbonaceous compounds with a low MW (<290 kDa) compared to the BAPs (>290 kDa) which consist mainly of macromolecules. Jiang et al. (2008) distinguished two types of UAPs (with lower and higher MW) and their classification depends on the utilisation of storage associated products. Regardless of different MW, chemical composition, and degree of biodegradability, it is now generally accepted that both UAPs and BAPs are biodegradable and recycled to become a substrate for microbial growth (Laspidou and Rittmann, 2002a, 2002b; Jiang et al., 2008; Menniti and Morgenroth, 2010; Zuthi et al., 2013a). Fenu et al. (2010) noted that the UAP fraction could predominate when the substrate degradation rates were high, while the BAP fraction could typically dominate over the UAP fraction at higher SRTs or under steady-state conditions. Indeed, BAPs have been more assessed in the literature over the years due to the lack of consensus regarding their production and degradation mechanisms (Zuthi et al., 2012).

EPS summarises numerous types of organic macromolecules, such as polysaccharides, proteins, nucleic acids, phospholipids, humic substances, and other polymeric compounds (Patsios and Karabelas, 2010; Gkotsis et al., 2014). They are usually bound at or outside the cell surface (regardless of the origin), surrounding cells and ensuring the stability and cohesion of the microbial aggregates, such as flocs, granules, and biofilms. The EPS provide a surrounding protection barrier, adhesion properties, and water retention around the bacteria (Laspidou and Rittmann, 2002a). The EPS can originate from several processes, e. g. active secretion, pouring of cell surface material, cell lysis, and adsorption from the mixed liquor suspended solids (MLSS) (Wingender et al., 1999). Polysaccharides in EPS have a higher fouling propensity compared to protein fractions when hydrophilic membranes are used, because the nature of proteins is hydrophobic and polysaccharides are hydrophilic (Li et al., 2012). Therefore, the protein-to-polysaccharide ratio in EPS is important for membrane fouling, particularly in cake layer formation in MBRs (Chang et al., 2002).

The EPS can be divided into two fractions, including bound EPS (bEPS) and soluble EPS (sEPS). The bEPS are bound to the sludge flocs, whereas the sEPS can move freely between sludge flocs and the surrounding liquor. sEPS is often included as part of the SMP fraction, since it is difficult to distinguish from one another (Fenu et al., 2010; Judd, 2010). The major difference between SMP and EPS is that SMP is usually present as suspended in the supernatant, while the EPS are bound to the floc (Drews, 2010; Zuthi et al., 2012). Moreover, Ramesh et al. (2006) compared the physicochemical characteristics of SMP and sEPS from different sludges. Their results did not support the hypothesis that SMP is identical to sEPS. Modellers may assume that they are identical to simplify their models. Fig. 1 presents a schematic representation of the relation between SMP (UAP and BAP) and bEPS.

SMP and bEPS are biological macromolecules with particular physical properties, such as a three-dimensional structure, high porosity with an interconnected pore structure which provide an appropriate surface structure for cell attachment, proliferation, and differentiation (Liu et al., 2018). Recognising their existence and characteristics transformed the mathematical modelling of MBRs since they play an important role in the initial and late fouling stages, respectively (Meng et al., 2017). In particular, some studies had revealed that SMP exert a significant influence before the jump of the transmembrane pressure (TMP) (Zhou et al., 2015; Liu et al., 2019b), while the bEPS originated from the deposited microbial cells contribute after the TMP jump (Luo et al., 2014; Zhou et al., 2015). These facts confirm that the inclusion of SMP and EPS (i.e., biomass biokinetics) in the assessment of MBR's bioprocesses is of utmost importance and leads to the development of biomass kinetic or hybrid models. Despite their importance in membrane fouling, it should be noted that the analytical determination of these compounds is challenging and often inaccurate. For example, Felz et al. (2019) showed that currently used colorimetric methods are not capable of accurately characterising EPS.

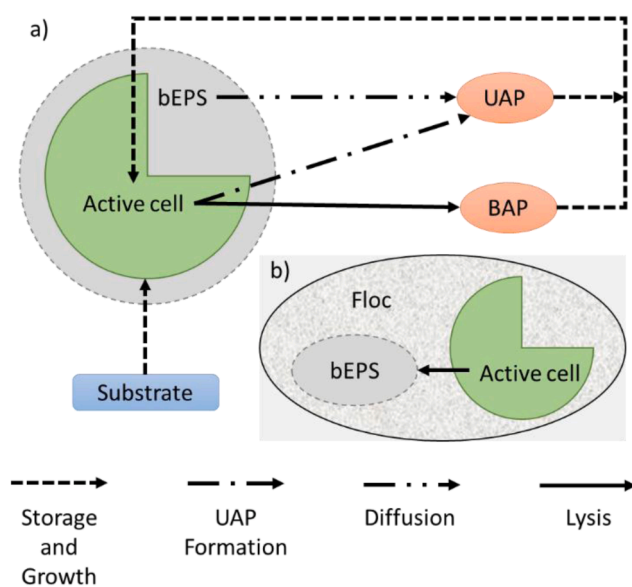


Fig. 1. Schematic representation of the SMP and bEPS and their links, where a) represents bEPS bound at the cell surface, while b) represents bEPS bound outside the cell surface.

3. Conceptual models of SMP/EPS formation and utilization

The biomass kinetic or hybrid models can be defined as expanded versions of the ASMs, in which the formation and degradation of SMP and EPS are inserted (Zuthi et al., 2012). The need to expand the ASM for application in MBRs is based on two rationales: (i) the ASMs were originally designed to address issues related to CAS systems, considering their specific features (e.g., lower SRT and low organic load compared to MBR); (ii) they were based on the Monod equations, which predict that the effluent concentration of the rate-limiting substrate should be independent of the influent substrate concentration (Barker and Stuckey, 1999). In the latter case, studies have demonstrated that soluble materials in the effluent were proportional to those in the influent. Thus, there was a demand for a new model that could describe the bioprocess complexity and account for the biomass characteristics that can affect membrane filtration performance (Patsios and Karabelas, 2010). According to Zuthi et al. (2012), a basic model of biomass kinetics in MBR should at least provide estimations of EPS concentration in the activated sludge flocs and SMP concentrations outside the flocs, which is not addressed by the original ASMs.

Fenu et al. (2010) recommended the use of ASM extensions with the EPS/SMP concepts in three cases, specifically when (i) linking biology with membrane fouling, (ii) predicting soluble COD, (iii) modelling systems with long SRTs. Additionally, this approach can be applied in modelling systems where heterotrophic activity is observed despite the absence of organic carbon in the influent. For example, Mehrani et al. (2022) modelled heterotrophic denitrification on SMP to describe the dominant abundance of heterotrophs in a system fed only with inorganic carbon and trace elements.

The first application of the original ASMs to model an MBR (Chaize and Huyard, 1991) was unsuccessful since the kinetics considered by the ASMs did not fully represent the reality of the MBR under assessment. The kinetics considered in an MBR model must be adapted to specific sludge characteristics that are influenced by different operating conditions (high SRT and MLSS concentration), which have a significant impact on the biomass metabolic pathways such as microbial product formation (Furumai and Rittmann, 1992). In this case, considering SMP and EPS formation avoids over-parametrization and overestimating biomass growth rates, which could lead to a severe error in predicting the effluent COD (Jiang et al., 2008). Neglecting SMP and EPS may thus

lead to erroneous estimations of membrane fouling. On this behalf, several hybrid models have been developed and described in the literature over the years (Barker and Stuckey, 1999; Zuthi et al., 2012; 2013a). For this reason, a brief historical review of their conceptual approaches is presented in the following section, with a particular attention to the latest progress.

3.1. Historical overview regarding SMP/EPS modelling

Different concepts have been developed for the formation and degradation of SMP/EPS over the past few decades, summarized in Fig. 2.

The first modelling attempt to estimate SMP was proposed by Luedeking and Piret (1959) (Fig. 2a). The purpose was to define the relationship between lactic acid formation and biomass growth in lactic acid fermentation. They observed that the lactic acid formation rate correlates with the biomass growth rate and amount. Baskir and Hansford (1980), considering the lactic acid in Luedeking and Piret's (1959) study

is SMP, concluded that SMP are related to (a) UAP that is proportional to the rate of biomass growth and (b) BAP that are not associated with growth but proportional to the concentration of biomass (associated with cell autoxidation or degradation).

The modelling SMP accumulation gained attention, especially in determining the source of effluent organic matter (EfOM), and several models have been proposed by different researchers (Baskir and Hansford, 1980; Namkung and Rittmann, 1986; Furumai and Rittmann, 1992; de Silva and Rittmann, 2000). Baskir and Hansford (1980) incorporated the Luedeking and Piret (1959) model into suspended activated sludge and showed that by-products of biological activity contribute to organic effluent concentration. Namkung and Rittmann (1986) presented a model for SMP growth in biofilm reactors to describe the fraction of SMP in the soluble EfOM. In the Baskir and Hansford (1980) and Namkung and Rittmann (1986) models, the UAP formation is correlated with the substrate utilization rate and the BAP consists of the direct by-products of substrate utilisation and microbial growth (Fig. 2b). On the other hand, the BAP formation is independent of

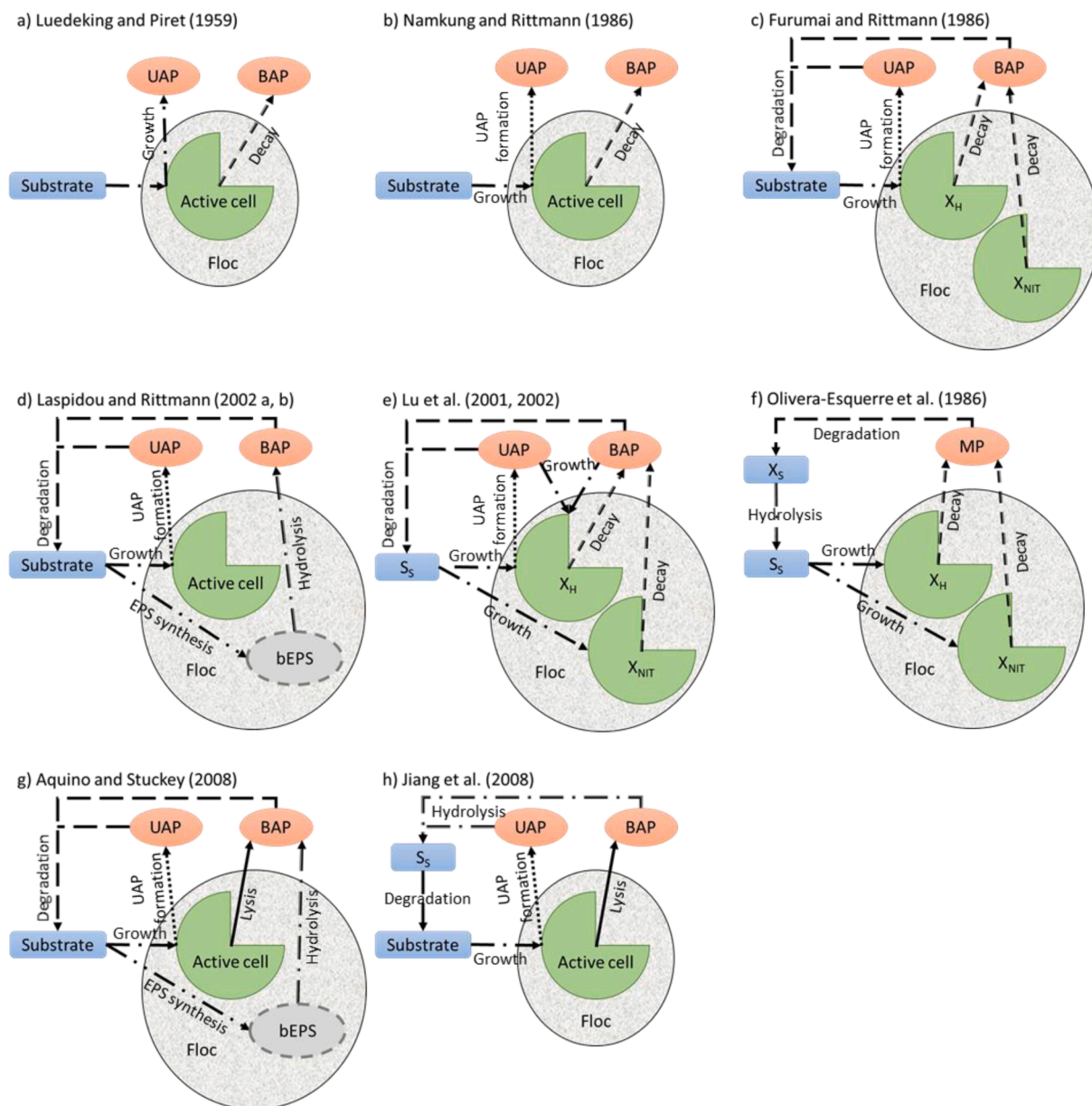


Fig. 2. Conceptual models of the formation and degradation of SMPs used in typical modelling studies - partially adapted and modified from Zuthi et al. (2013a). The acronyms are detailed in the text.

microbial growth, and the formation rate is proportional to the concentration of active biomass. However, the formation rate of BAPs may be proportional to the biomass decay rate with a stoichiometric coefficient, since BAPs are considered decay products of the overall active biomass (Jiang et al., 2008). The model proposed by Namkung and Rittmann (1986) is still considered a reference for modelling SMP formation. Before that work, only SMP production was studied in activated sludge systems, as it was believed to be inevitable due to its production from biomass decay and low biodegradability (Gaudy and Blachly, 1985).

Furumai and Rittmann (1992) focused on the interaction between heterotrophs and nitrifiers in terms of the exchange of organic matter and modelled SMP produced by nitrifiers (X_{NIT}) as an energy and carbon source for heterotrophs (X_{H}) (Fig. 2c). The degradation of SMP was studied later by Noguera et al. (1994), who developed a model using experimental results from a glucose-fed anaerobic chemostat. The results of Noguera et al. (1994) has validated by Aquino and Stuckey (2008) showing that most of the SMP accumulation corresponded to BAP and presented that BAP have slower degradation rates compared to UAP, suggesting that the decrease in acidogenic biomass was due to SMP formation rather than oxidation to carbon dioxide. It is important to note that quantitative formation of SMP may differ between anaerobic and aerobic systems and distinguishing SMP from fermentation products (volatile fatty acids (VFAs) is crucial (Mesquita et al., 2010). However, Ni et al. (2011) indicated that SMP/EPS modelling theories developed for aerobic systems are valid for anaerobic systems. Noguera et al. (1994) also proposed Monod constants for the storage of BAP and UAP from the growth kinetics of SMP as a substrate (Janus and Ulanicki, 2010).

In the meantime, an attempt to model SMP and EPS kinetics in activated sludge systems was made by Hsieh et al. (1994a, 1994b), who proposed a simple biokinetic model in which EPS and SMP production were measured in a single bacterial culture. That work was later tested and validated by Laspidou and Rittmann (2002a, 2002b), who used the prior works as a foundation for their model. In this regard, Laspidou and Rittmann (2002a, 2002b) differentiated bEPS from the active biomass and EPS hydrolysis as the sole mechanism of BAP formation, while no SMPs were assumed to be formed from the decay of the active biomass (Fig. 2d). They also hypothesised in their “unified theory” that SMP and soluble EPS are identical in systems where particle organics are not important, the growth-associated part of soluble EPS is identical to UAP, soluble EPS polymerizes to bEPS, the formation of bEPS is growth-associated and in direct proportion to substrate utilisation.

The simple concepts of SMPs concepts were incorporated into the ASMs by including non-biodegradable soluble products (equivalent to BAPs) produced during hydrolysis of slowly biodegradable organic compounds (X_{S}) (Orhon et al., 1989) and UAPs (Artan et al., 1990). However, ASM extensions incorporating SMP/EPS concepts became more common than the SMP/EPS stand-alone models since Lu et al. (2001, 2002) proposed the combination for MBRs. Lu et al. (2001, 2002) were the first to combine the concepts of SMP presented by Namkung and Rittmann (1986) with the ASMs for MBR studies. They highlighted that since biomass concentration and SRT are high and the F/M ratio is low, microbial products in MBR cannot be ignored. They initially modified the ASM1 (Lu et al., 2001) and then the ASM3 (Lu et al., 2002). Consequently, the overall active biomass was differentiated into X_{H} and autotrophic (X_{AUT}) biomass (Fig. 2e). In the modified ASM1, the UAPs are formed directly by the metabolism of readily biodegradable substrate (S_{S}). The soluble biodegradable organic compounds, derived from biomass decay, are classified as the BAPs. Both UAPs and BAPs can be reused directly by heterotrophs for their growth. Although the simulation results agreed with the experimental data, the model was subsequently questioned regarding COD and charge imbalances (Jiang et al., 2008). Oliveira-Esquerre et al. (2006) proposed a modification of ASM3 (ASM3-MP) by lumping the UAPs and BAPs together into a general term MP (microbial product), for which only the decay products of the

biomass were considered (Fig. 2f). Active biomass was considered by Furumai and Rittmann (1992) (i.e., X_{H} and X_{NIT}), and their growth was based on the prior hydrolysis of the slowly biodegradable substrate (X_{S}) into the readily biodegradable substrate (S_{S}). They also pointed out that the link between MPs and the fouling process must be evaluated.

Moving forward, Aquino and Stuckey (2008) disagreed with the unified theory proposed by Laspidou and Rittmann (2002a, 2002b) that soluble EPS and UAP are identical since Ramesh et al. (2006) demonstrated that the physicochemical characteristics of these components are different. They proposed a new approach to model EPS formation under anaerobic conditions as a non-growth associated process (Fig. 2g), while EPS degradation was modelled similarly to Namkung and Rittmann (1986). Differently from Laspidou and Rittmann (2002a, 2002b), they assumed that soluble EPS is not UAP and soluble EPS and cell decay products are the sources of BAP (Table 1). Concerning BAP formation, the model combined the approaches of the previous two models, where both decay of active biomass and hydrolysis of the bound EPS are the sources of BAP (Fig. 2g). Unlike Laspidou and Rittmann (2002a, 2002b), the EPS formation was considered as a mechanism independent of the microbial growth rate but related to biomass concentration and described by a first-order equation for the active biomass concentration (Table 1). Aquino and Stuckey (2008) emphasized that incorporating the SMP formation mechanism from the decay of the active biomass was a significant advantage in capturing SMP kinetics over a wide range of operational conditions (specifically SRTs) in the studied MBR, similar to Lu et al. (2001, 2002). Zuthi et al. (2013b) further confirmed that the model was flexible enough to predict the dynamic changes in bEPS and SMP production. Distinguishing soluble EPS and SMP formation in MBR models can be useful when testing different fouling control strategies since they have other factors that can affect their production and accumulation on the membrane surface.

Meanwhile, Jiang et al. (2008) criticized the SMP modelling effort of Lu et al. (2001, 2002) because of its complexity and over-parameterization. Additionally, they modelled BAP degradation not as a direct process (e.g. Lu et al., 2001; 2002; Laspidou and Rittmann, 2002a, 2002b; Oliveira-Esquerre et al., 2006; Aquino and Stuckey, 2008) but after the hydrolysis process yielding S_{S} (Fig. 2h). The rationale of that approach was based on the experimental observation that most BAP had an MW larger than 20 kDa and such large molecules would not be able to pass the cell membranes directly. That approach was adopted in future studies by Fenu et al. (2011) and Mannina et al. (2011, 2018). Jiang et al. (2008) also argued that previous SMP modelling studies were lacking proper calibration due to limited measurements and the validity of these models were questionable. They collected BAP and SMP data separately in their modelling study and validated their model with independent MBR steady-state measurements. The following section addresses the novel approaches presented by these works.

3.2. New development of conceptual approaches regarding SMP/EPS modelling

This section contains the most recent information regarding modelling SMP and EPS in MBR systems during past ten years (Fig. 3). The conceptual models, shown in Fig. 3, are related to the rate of formation and degradation of each process. For more details about the parameters used in the model, readers can refer to the publications (Janus and Ulanicki, 2010; Mannina et al., 2018; Al-Hazmi et al., 2020).

Zuthi et al. (2013b; M.F.R. 2015) proposed a novel approach for estimating SMP and bEPS from an MBR system. They argued that there was no unambiguous SMP/EPS measurement method to characterize the biomass and that the biomass viability could provide a better estimate of these components. They assumed that SMP affects biomass viability and serves as the binding sites for cake formation on the membrane surface, based on observations by Lee et al. (2003) and Rojas et al. (2005). They used the specific oxygen uptake rate (SOUR) as a reference to explain

Table 1

Expressions for the formation and degradation of UAP, BAP, and EPS in selected models – adapted and modified from Fenu et al. (2010) and Zuthi et al. (2013a).

Equation*	Process	Reference
$\alpha \frac{UAP}{dt} + \beta \frac{BAP}{dt}$ $k_1 \mu X + k_2 X$	SMP production	Luedeking and Piret (1959) (Fig. 2a)
$k_{f,UAP} \frac{S_s}{K_S + S_s} X_b$	UAP formation	Lapidou and Rittmann (2002a, 2002b) (Fig. 2d)
$f_{UAP}(\mu_H X_H + \mu_A X_A)$ $- k_{d,UAP} \frac{S_{UAP}}{K_{UAP} + S_{UAP}} X_b$	UAP degradation	Lu et al. (2001) (Fig. 2e) Lapidou and Rittmann (2002a, 2002b) (Fig. 2d)
$- k_{d,SMP} \frac{S_{SMP}}{K_{SMP} + S_{SMP}} X_H$ $k_{STO,UAP} \frac{S_{UAP}}{K_{UAP} + S_{UAP}} X_H$		Lu et al. (2001) (Fig. 2e) Janus and Ulanicki (2010) (Fig. 3a)
$k_{EPS} X_{EPS}$	BAP formation	Lapidou and Rittmann (2002a, 2002b) (Fig. 2d)
$k_h X_S + k_{h,EPS} X_{EPS}$		Aquino and Sruckey (2008) (Fig. 2g)
$f_{BAP}(b_H X_H + b_{PAO} X_{PAO} + b_{AUT} X_{AUT})$ $f_{BAP} b X + (1 - f_s) k_{h,EPS} X_{EPS}$		Jiang et al. (2008) (Fig. 2h) Janus and Ulanicki (2010) (Fig. 3a)
$- k_{d,BAP} \frac{S_{BAP}}{K_{BAP} + S_{BAP}} X_b$	BAP degradation	Lapidou and Rittmann (2002a, 2002b) (Fig. 2d)
$- k'_{d,BAP} S_{BAP} X_H$		Jiang et al. (2008) (Fig. 2h)
$k_{STO,BAP} \frac{S_{BAP}}{K_{BAP} + S_{BAP}} X_H$		Janus and Ulanicki (2010) (Fig. 3a)
$f_{p,EPS} r_s$	EPS formation	Lapidou and Rittmann (2002a, 2002b) (Fig. 2d)
$k'_{EPS} X_B$		Aquino and Sruckey (2008) (Fig. 2g)
$- k_{h,EPS} X_{EPS}$	EPS degradation	Lapidou and Rittmann (2002a, 2002b) (Fig. 2d)
$f_{p,EPS} \mu X$		Aquino and Sruckey (2008) (Fig. 2g)
$f_{p,EPS} r_s$		Janus and Ulanicki (2010) (Fig. 3a)
$- k_{h,EPS} X_{EPS}$	EPS hydrolysis/ dissolution	Janus and Ulanicki (2010) (Fig. 3a)

* Monod terms for nutrients and electron acceptors are not shown in the table (b_H): Lysis rate constant for heterotrophs; b_{AUT}: Lysis rate constant for autotrophs; b_{PAO}: Lysis rate constant for PAOs; f_{BAP}: Fraction of BAP generated as a product of cell lysis; f_{p,EPS}: Part of the substrate electrons shunted to EPS formation; f_s: Fraction of SS produced from XEPS hydrolysis; f_{UAP}: UAP formation yield; K_{BAP}: BAP affinity constant; k_{d,BAP}: BAP degradation rate constant; k_{d,SMP}: SMP degradation rate constant; k_{d,UAP}: UAP degradation rate constant; k_{EPS}: EPS formation rate constant; k_{f,UAP}: UAP formation rate constant; k_{h,EPS}: EPS hydrolysis rate constant; k_h: Hydrolysis rate constant; K_S: Substrate affinity constant; K_{SMP}: SMP affinity constant; k_{STO,BAP}: BAP storage rate constant; k_{STO,UAP}: UAP storage rate constant; K_{UAP}: UAP affinity constant; r_s: Substrate utilization rate; S_{BAP}: BAP concentration; S_{SMP}: SMP concentration; S_{UAP}: UAP concentration; X_A (X_{AUT}): Active autotrophic biomass; X_b (X_H): Active heterotrophic biomass; X_{EPS}: EPS concentration; α (k₁): Formation coefficient for UAP; β (k₂): Formation coefficient for BAP; μ (μ_H): Maximum growth rate for heterotrophs; μ_A: Maximum growth rate for autotrophs.

quantitatively the correlation between the SMP or bEPS and the biomass viability based on the trace of soluble or colloidal components (soluble or colloidal COD) in the effluent. They calibrated their model with 50-day of operating data for the results of SOUR and the concentrations of MLSS, its volatile fraction (MLVSS), SMP, and EPS, and later tested the model validity with another data set.

Janus and Ulanicki (2010, 2015) began modelling SMP and EPS from MBRs around 2010, and their work provided novel aspects until recent days. Initially, they were looking for the best approach to model SMP/EPS formation and degradation to propose an integrated MBR model. In particular, they presented ASM-based models that could account for the formation of SMP and EPS. They applied the unified SMP/EPS approach provided by Lapidou and Rittmann (2002b) to ASM-based models. UAP was considered as the fraction produced as a by-product of substrate utilisation and cell growth. BAP was assumed to originate from biomass decay and hydrolysis/dissolution of bEPS (Fig. 3a).

The model has been calibrated manually with data from biopolymer production from pure culture (Hsieh et al., 1994a, b) and SMP/EPS production from a pilot scale MBR system (Yigit et al., 2008). However, it needs to be validated with a different set of data to confirm the extent to which it accurately describes them. They also highlighted the SMP and EPS modelling limitations: (i) although SMP is divided into UAP and BAP based on their metabolic origin, the chemical compositions of UAP and BAP are important from a fouling perspective; (ii) apart from SMP and EPS, floc size distribution also affects fouling; (iii) SMP and EPS production are affected by parameters that the models do not consider, such as temperature and salinity.

The works of Janus and Ulanicki (2010, 2015) inspired a new model proposal by Mannina et al. (2018), which presented a comprehensive integrated MBR model to assess the organic matter, nitrogen and phosphorus biological removal, and greenhouse gas (GHG) formation. The model considers SMP formation and degradation (dividing SMP into BAP and UAP) and MLSS concentration as interactions between the biological and physical processes. In that model, the heterotrophic biomass was divided in phosphorus accumulating organisms (PAO) (X_{PAO}), ordinary heterotrophic organisms (X_{OHO}), while the autotrophic biomass was divided into ammonia-oxidizing bacteria (X_{AOB}) and nitrite-oxidizing bacteria (X_{NOB}). As shown in Fig. 3b, UAP and BAP are utilised by heterotrophic biomass for storage, growth, and respiration. The production of BAP is proportional to biomass decay and its reduction is related to the hydrolysis process. On the other hand, the production of UAP is related to biomass growth (except the X_{AOB}). Mannina et al. (2018) also considered the denitrification process to be responsible for the release of UAP, which in the model is performed by X_{PAO} and X_{OHO} following the four-step denitrification approach of Hyatt and Grady (2008). It should be noted that Hyatt and Grady (2008) did not consider X_{PAO} in their work. Fig. 4 shows the four-step denitrification with the release of UAP.

During step one, the NO₃⁻ is the main substrate of the processes and is reduced to nitrite (NO₂⁻). In this step, the X_{PAO} stores polyphosphate (X_{PP}) and utilizes organic accumulating products (X_{PHA}), while X_{OHO} use organic fermentable products (S_F) and acetate (S_A) as a substrate. In step two, NO₂⁻ is reduced into nitric oxide (NO), then to N₂O in step three, and finally, to nitrogen gas (N₂) in step four. Both X_{PAO} and X_{OHO} release UAP during the denitrification, and all related-processes are included in the model. The calibrated simulation results were compared to the data from an existing pilot plant treating real wastewater, which adds to the reliability and applicability of the integrated approach used by the authors.

This link between denitrification and SMP production was also found regarding the significant heterotrophic growth that takes place in anammox and deammonification systems (fed with no organic carbon). In this case, the SMPs were found to be the sole organic carbon and energy source for denitrifying heterotrophs. With this regard, Liu et al. (2016) developed a theoretical model for the biological processes

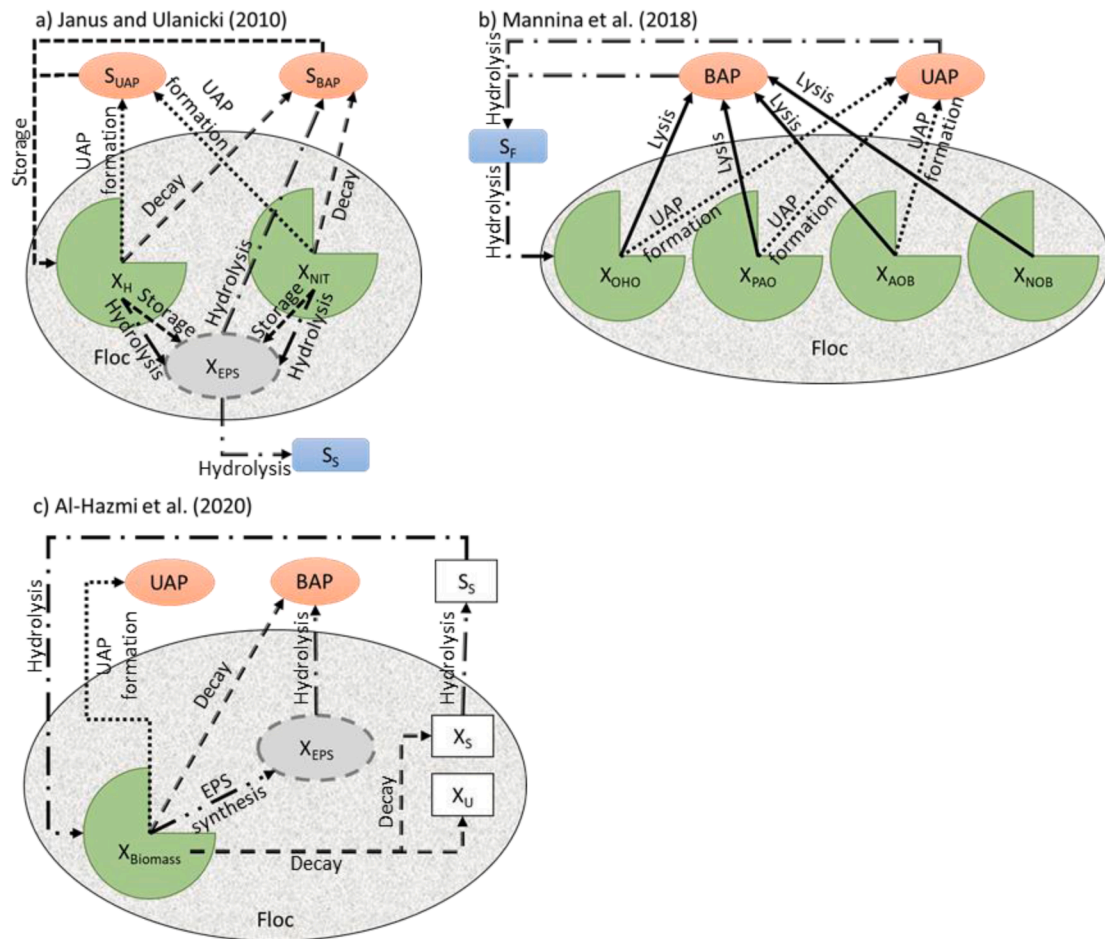


Fig. 3. Conceptual models of the formation and degradation of SMPs used in recent modelling studies. The acronyms are detailed in the text.

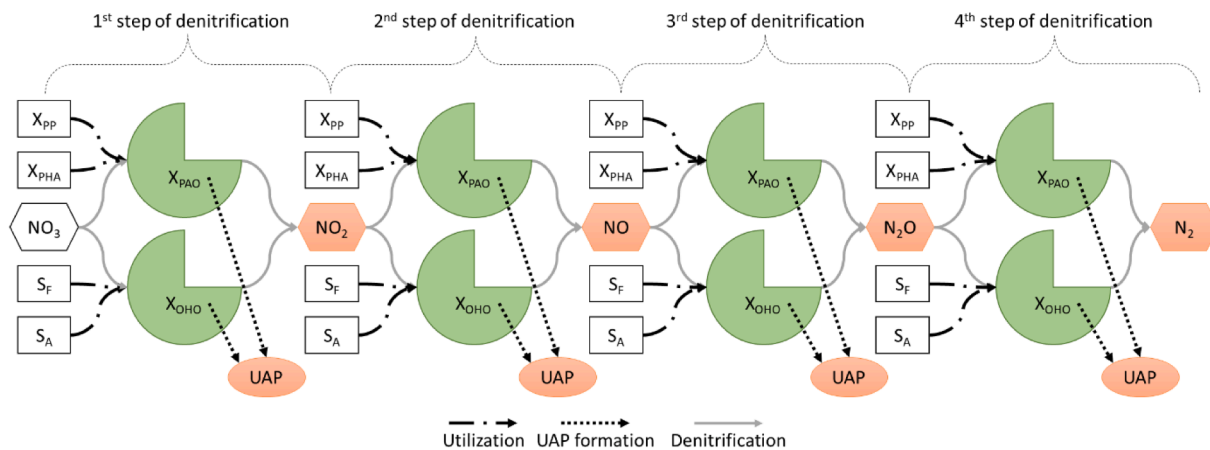


Fig. 4. Four steps of denitrification process considered by Mannina et al. (2018).

occurring in an anammox biofilm system and they validated their model with experimental data. Organic carbon for the growth of the heterotrophic bacteria was exclusively derived from three internal sources: anammox/heterotrophic growth (UAP), biomass decay (cell decay products and BAP), and hydrolysis of EPS (BAP). Subsequently, Lu et al. (2018) and Al-Hazmi et al. (2020) adopted the concept of Liu et al. (2016) to expand the ASM1 in view of predicting aerobic/anoxic growth of heterotrophic biomass from a laboratory-scale deammonification system. In both studies, it was assumed that the formation of microbial

products (UAP, BAP, and S_S) was not only derived from the activity of anammox and heterotrophs, but also from both groups of nitrifiers (AOB and NOB). The S_S utilization and BAP/UAP degradation were exclusively attributed to the growth of heterotrophs (Fig. 3c). Liu et al. (2016) applied a stepwise calibration procedure including sensitivity and uncertainty analysis and model validation. The conceptual deammonification model of Al-Hazmi et al. (2020) is presented in Fig. 5.

All the three models emphasised the significant role of autotrophic and heterotrophic bacteria on SMP formation. For further details, the

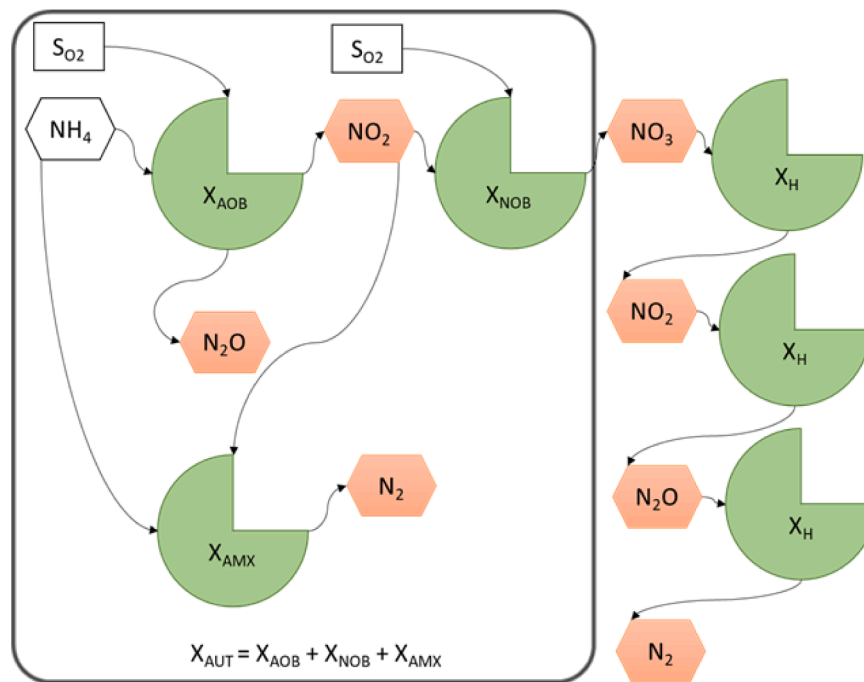


Fig. 5. The conceptual model of a deammonification system fed with inorganic substrates (Al-Hazmi et al., 2020).

reader is referred to Liu et al. (2016), Lu et al. (2018), and Al-Hazmi et al. (2020).

4. Kinetic models for the formation and utilization of SMP/EPS

Table 1 summarises the expressions for the SMP/EPS formation and degradation in selected models. The terms α and β (Eq. (1)) represent the formation coefficients for UAP and BAP, respectively (Luedeking and Piret, 1959). According to Janus (2013), different values can be assigned to α and β due to different kinetic dynamics present in mixed bacterial cultures (e.g. Berry et al., 2004). SMP/EPS dissolution was not considered due to the simplicity of the model.

In the Laspidou and Rittmann (2002a, 2002b) model (Fig. 2d), the UAP and bound EPS formation rates are described by the Monod-type equations. The rate of bounded EPS degradation due to hydrolysis is described by a first-order relationship with respect to the EPS concentration (X_{EPS}). The UAP and BAP degradation rates are described by similar Monod-type equations. However, the subsequent experimental observations revealed that the hypothesis of BAP formation only related to EPS hydrolysis was weak for two reasons (Fenu et al., 2010; Zuthi et al., 2013a). First, the BAP/UAP kinetics were not flexible enough, especially for predicting dynamic changes of the bound EPS to BAP. Secondly, the physicochemical properties of the hydrolysed (soluble) EPS and BAP were different. Laspidou and Rittmann (2002b) calibrated, Lu et al. (2001) calibrated and applied sensitivity analysis to model parameters. Lu et al. (2001) found that the maximum specific growth rate of SMP for heterotrophs (μ_{SMP}), UAP formation constant of heterotrophs ($\gamma_{\text{UAP,H}}$), and heterotrophic yield coefficient from SMP (γ_{SMP}) were sensitive to effluent COD and TN concentrations.

Jiang et al. (2008) defined the stoichiometric parameter f_{BAP} as a fraction of BAP generated as a product of cell (X_{H} , X_{PAO} , X_{AUT}) lysis. Janus and Ulanicki (2010) defined BAP is originated from biomass decay and hydrolysis of EPS. They also defined processes for aerobic and anoxic storage of UAP and BAP. They added a limiting factor η_{NO} for anoxic storage of UAP and BAP.

5. SMP/EPS model applications and kinetic parameter values in MBRs

The previously discussed works represent some of the most recent approaches to estimate SMP and EPS production in MBR systems. However, other recent modelling applications have also correlated bioprocesses (i.e., SMP and EPS) with MBRs. For physical model, the resistance-in-series model is usually used as it simulates fouling process with an increase in transmembrane pressure (TMP) due to the accumulation of deposited material on both the membrane surface and inside the membrane pores (Wintgens et al., 2003). Lee et al. (2002) combined SMP production/degradation model of Lu et al. (2001) with a physical model (resistance-in-series) to simulate fouling. However, Lee et al. (2002) did not calibrate their model by experimental data. Zarragotia-González (2008) integrated the unified theory of Laspidou and Rittmann (2002a) (as SMP model) and physical model. Their model predicted system performance under different MLSS concentrations, filtration cycles, and aeration strategies. However, it overlooks the possible influence of the dynamic deep-bed filtration which acts as a secondary filter, of cake on the organic removal (Mannina et al., 2011). Later, Di Bella et al. (2008) implemented the deep-bed theory to their physical processes in their integrated MBR model for MBR systems. They applied their model on a pilot-scale MBR system and showed the linkage between SMP and fouling. The downside of their modelling study is the assumption of uniform distribution of the cake deposition on the membrane surface which is not the case in real situations. Gabarrón et al. (2015) used a dynamic ASM2d-based model to test optimisation strategies to an MBR system in terms of effluent quality, energy, and cost. Then they applied the optimum operation strategy that was determined from the modelling study (dissolved oxygen concentration at 0.8 mg/L) to a full-scale plant and monitored sludge characteristics. They find out that there were no significant changes in SMP/EPS production. Zuthi et al. (2017) applied a simplified integrated modelling approach to a lab-scale sponge-submerged membrane bioreactor (SSMBR) to account for pore blocking and cake formation by taking into consideration the combination of aeration and backwashing effects. The integrated MBR model used SMP and MLSS concentration as a link between biological and physical models, mainly considering SMP as a

cause of pore blocking. The model described the effect of pore size reduction due to the adsorption of particles within the pores. According to the authors, the model could predict fouling development well, but the further assessment of the model is required by operating MBR systems under different MLSS concentrations and at different operating conditions.

Despite the significant results provided by these works regarding MBR performance and optimisation in both laboratory- and full-scale, the use of site-specific data hampers the replicability of such model approaches in future works, as no relationship between plant performance and SMP and EPS was provided. This hindrance may be observed in the number of model applications in the literature that applied comprehensive MBR models without coupling the conceptual expressions for the formation/degradation of SMP and EPS.

To address this issue, Mannina et al. (2020) proposed a process-based plant-wide model to assess a semi-hypothetical MBR plant in terms of effluent quality, energy consumption, and GHG emissions. In this model, the SMP concentration inside the MBR was considered a by-product of biological processes and estimated using a mathematical relationship obtained from Mannina et al. (2018). The relationship between SMP concentration and SRT was obtained by performing 2000 Monte Carlo simulations varying the SRT (Mannina et al., 2020). In spite of the fact that this model application was based on a semi-hypothetical MBR case study, the correlation applied was based on a comprehensive dynamic model based on the ASM-family with a significant data set as a baseline. Results of the model application showed a direct correlation between SMP concentrations and fouling, which also contributed to an increase in the energy consumption and, consequently, an increase in the GHG emissions. In other words, for that specific case, one may say that the SMP represented a significant influence over the model outputs that are considered the main obstacles to the spread of MBR as a wastewater treatment technology (Capodici et al., 2015; Qin et al., 2018). However, it is worth mentioning that the relationship between SMP concentration and fouling depends on multiple parameters such as SRT, organic loading rate (OLR), and F/M ratio of the system and MLSS and dissolved oxygen concentrations in the reactor (Drews, 2010) (Table 2).

6. Discussion and perspectives

The main outcomes of this review highlighted the modelling of SMP and EPS in MBR systems under a common frame. Indeed, SMP-based models are spread in the literature and have been improved and

updated since the late 1950s until the present day. In this section, the improvement of and updates on SMP and EPS models and the strengths and weaknesses of these models in MBR systems are summarised. Furthermore, suggestions to improve MBR models have been given.

Concerning the novel conceptual approaches hereby presented, Liu et al. (2016), Al-Hazmi et al. (2020), and Mannina et al. (2018) proposed modelling approaches that can be considered an evolution of those represented in Fig. 2, except for Namkung and Rittmann (1986) which did not attribute the formation of SMPs to the biomass. For this reason, their work could be applied to other MBR-related studies, even though Liu et al. (2016) and Al-Hazmi et al. (2020) did not direct the model efforts to MBR systems. As far as the authors are aware, the recent model applications to anammox-MBR systems (Tao and Hamouda, 2019; Wisniewski et al., 2019; Liu et al., 2019a) did not consider the role of the bioprocesses over membrane fouling issues, which is a very important issue to be addressed in future works.

Regardless of the numerous published data, there needs to be more knowledge concerning SMP kinetics due to their multiple origins and highly complicated nature. The major issue is related to the fact that their kinetics are dependent on many different factors that need to be accounted for in the current modelling approaches. Additionally, the relationship between their nature (e.g., protein or carbohydrates) and the effects over formation, degradation, fouling and many other aspects from a modelling point of view still needs to be improved in the literature. Moreover, depending on the objectives of the model development, changes in model structures are not anodyne: for instance, (Benyahia et al., 2013) it was shown that introducing SMP in simple an-MBR models used for control resulted in significant changes in their mathematical properties (notably in the number and stability of their steady states).

The estimation of EPS has not received much attention in the literature, likely due to the lack of understanding of their formation pathways. According to Scholes et al. (2016), the lack of consensus on the causes of EPS production in the scientific literature is unsurprising given the variation in wastewater influent and microbial populations. The authors also emphasised that each MBR may have its own triggers (SRT, OLR, F/M ratio etc.) for EPS production, which could influence membrane fouling in various ways. For this reason, the establishment of modelling approaches is necessary to encourage new findings and increase knowledge about EPS formation/degradation.

Another serious issue is that most of the data used for modelling SMP and EPS have been obtained from experimental estimation (Scholes

Table 2
Values of the kinetic and stoichiometric parameters in the expressions presented in Table 1.

Symbol	Definition	Unit	Lapidou and Rittmann (2002a, 2002b)	Lu et al. (2001)	Aquino and Sruckey (2008)	Jiang et al. (2008)
UAP						
$k_{f,UAP}$	UAP formation rate constant	$\frac{\text{mg COD}_{UAP}}{\text{mg COD}_{\text{cell}} \cdot \text{d}}$	0.05			
$k_{d,UAP}$	UAP degradation rate constant	$\frac{\text{mg COD}_{UAP}}{\text{mg COD}_{\text{cell}} \cdot \text{d}}$	1.27			
K_{UAP}	UAP affinity constant	$\frac{\text{L}}{\text{mg COD}}$	100			
f_{UAP}	UAP formation yield	$\frac{\text{mg COD}_{UAP}}{\text{mg COD}_{\text{cell}} \cdot \text{d}}$		0.3		
$k_{d,SMP}$	SMP degradation rate constant	$\frac{\text{mg COD}_{SMP}}{\text{mg COD}_{\text{cell}} \cdot \text{d}}$		4.2		
K_{SMP}	SMP affinity constant	$\frac{\text{L}}{\text{mg COD}_{SMP}}$		60		
BAP						
$k_{d,BAP}$	BAP degradation rate constant (Monod equation)	$\frac{\text{mg COD}_{BAP}}{\text{mg COD}_{\text{cell}} \cdot \text{d}}$	0.07			
K_{BAP}	BAP affinity constant	$\frac{\text{L}}{\text{mg COD}_{BAP}}$	85			
$k'_{d,BAP}$	BAP degradation rate constant (First order equation)	$\frac{\text{mg COD}_{SMP}}{\text{L}}$				$7.1 \cdot 10^{-7}$
k_h	BAP formation rate constant from biomass decay	$\frac{\text{mg COD}_{BAP}}{\text{mg COD}_{\text{cell}} \cdot \text{d}}$			0.03	

et al., 2016; Park et al., 2018). Therefore, it is recommended that MBR models be calibrated and validated on the basis of data retrieved from full-scale WRRFs treating real wastewater to consider their real response to dynamic changes in influent composition and operating conditions. Finally, the influence of these components on MBR optimisation can appropriately be validated by correlating them with optimization outputs (e.g., membrane fouling, energy consumption, operating costs, GHG emissions), during model simulations. The successful applications endorse the importance of including conceptual SMP/EPS approaches to model simulations since optimisation of an MBR system could be better assessed by the use of more accurate SMP and EPS estimations.

Given the number of publications that have used the modelling of SMP and EPS formation and degradation to estimate membrane filter performance and energy consumption it seems that these approaches are convincing and, although the models can be complex, they tend to give a monocausal explanation for membrane fouling and MBR behaviour. In practical MBR operations, multiple factors may inflict membrane performance, which can be eventually mistakenly attributed to genuine fouling, but actually may have causes that lie outside the scope of a model (e.g. Hai et al., 2019). Other adverse effects on membrane performance (e.g. coarse fouling, module blocking, filter integrity, uneven flow distribution etc.), which are common at full-scale installations, may lead to an overestimation of the role of EPS and SMPs in a model. Thus, these modelling approaches have to be used with caution and uncertainties at all stages of the model formulation, data collection, set-up, calibration and validation should be taken into account while applying good modelling practices.

In addition to empirical and mathematical models, the application of artificial intelligence (AI) in membrane fouling modelling has been a subject of research for the past two decades (Niu et al., 2022). While these AI models have effectively predicted the increase in TMP resulting from membrane fouling, they have struggled to establish a correlation between permeate quality and TMP (Schmitt et al., 2018; Hamed et al., 2019). This highlights the ongoing significance of mathematical modelling studies focused on understanding the production of SMP, which directly impact the quality of the permeate.

7. Conclusions

The key findings identified from this state-of-the-art review are listed below:

- Accurate estimation of SMP and EPS can contribute to optimising membrane fouling results, which directly influence energy consumption, operating costs, and GHG emissions.
- AI models accurately predict TMP increase from fouling in MBRs but struggle to correlate permeate quality with TMP. This emphasizes the ongoing importance of mathematical modelling to understand SMP production and its impact on permeate quality.
- Although many studies have been published concerning SMPs, there are still gaps in the literature due to their complex nature and multiple origins.
- Only a few studies have focused on the estimation of EPS due to a need for more information on the triggers for their production.
- Most modelling studies have neglected the physicochemical properties of SMP/EPS such as protein and carbohydrate contents or MW.
- Most of the data used for modelling SMP and EPS have been retrieved from experimental estimation, which may limit replicability since such information does not represent the dynamic changes in influent composition and operating conditions.
- The novel conceptual approaches presented in this work primarily focus on biomass-related processes and the role of different bacterial groups in the release of SMP. However, these studies did not consider the direct influence of SMP and EPS on membrane fouling, presenting opportunities for future developments.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Giorgio Mannina reports was provided by University of Palermo. Giorgio Mannina reports a relationship with University of Palermo that includes: employment.

Data availability

No data was used for the research described in the article.

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