



A review of modeling bioprocesses in membrane bioreactors (MBR) with emphasis on membrane fouling predictions

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ABSTRACT

Membrane bioreactor technology is an attractive approach for combined wastewater treatment and water reclamation. Although considerable practical experience and experimental data are available, regarding the operation of MBR, mathematical models that would be valuable for improved design and optimization of MBR systems are still at an unsatisfactory state. This paper presents a critical review of existing activated sludge mathematical models and variations thereof, with emphasis on the special requirements which arise from the strong interaction of the concurrently occurring biological process and membrane filtration in MBR systems. The desirable attributes of an activated sludge model, focused on MBR technology, emerge by assessing the influence of this interrelation on both processes. Special attention is paid to identifying key variables that can help establish a direct link between bioprocess modeling and membrane fouling models. Various activated sludge models, originally developed for, and commonly used in the conventional activated sludge systems, as well as some modified versions employed in MBR systems, are reviewed. It is pointed out that these models, by design, have inherent deficiencies for MBR simulation mainly because they do not provide sufficient data to comprehensively simulate the fate of different microbial products (like EPS and SMP) that play an important role in membrane fouling. The necessity of an alternative bioprocess modeling framework, *de novo* focused on the specific needs of the MBR technology, as well as guidelines concerning the development of such a model are suggested in the paper.

Keywords: Membrane bioreactor (MBR); Membrane fouling models; Wastewater treatment; Activated sludge modeling; EPS; SMP

1. Introduction

Membrane Bioreactors (MBR) have emerged as an attractive technology for advanced municipal and industrial wastewater treatment and reclamation, combining the biological degradation process of activated

sludge with a direct solid-liquid separation through membrane filtration [1]. The MBR process can no longer be considered as a novel process but rather as a reliable alternative to *Conventional Activated Sludge Process (CASP)* [2], since the commercial significance of this technology is already considerable, with applications in municipal and industrial wastewater treatment becoming increasingly widespread [3]. The

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advantages of MBR are well known, including small footprint and reactor volume requirements, high and consistent effluent quality, effective rejection of pathogenic bacteria and viruses, stability of operation in high or shock loadings, independent control of *Solids* and *Hydraulic Retention Time* (*SRT* and *HRT* respectively), higher volumetric loading and reduced sludge production, compared to CASP [1,3–5]. However, membrane fouling, i.e. the deposition and/or adsorption of particulates, colloidal and soluble species onto and into the membrane, resulting from interactions between the mixed liquor and the membrane [5], remains the main, albeit unavoidable, handicap of the system, significantly contributing to operating cost and inhibiting the widespread application of MBR technology [2].

Extensive research on membrane fouling has been carried out since the early days of MBR technology and numerous papers have been published on membrane fouling and the effect that system parameters (i.e. operating conditions, hydrodynamic conditions, membrane properties, feed and biomass characteristics) have on filtration performance [6]. However, integrated modeling studies linking the above system parameters to system performance indicators, and in particular to membrane fouling propensity are at a relatively rudimentary state. This unsatisfactory situation may be attributed to the intrinsic complexity and uncertainties that characterize the MBR process. Indeed, there are many parameters involved in the design and operation of MBR systems, reviewed in various papers [2,3,6–8], that have a direct or indirect effect on MBR fouling [2]. The complex interactions between these parameters may have hindered the efforts to link the two main operations of MBR process, i.e. *biological treatment* and *membrane separation*, and to establish an integrated modeling framework that could provide a significant thrust towards the broader use of MBR technology for wastewater treatment.

The scope of this paper is to present a critical review of existing activated sludge mathematical models and variations thereof with emphasis on the special requirements which arise from the strong interaction of the concurrently occurring biological treatment and membrane filtration in MBR systems. The requisite features of an activated sludge model, focused on MBR technology, emerge through an evaluation of the influence of the biological process on membrane filtration and *vice-versa*. Particular aspects of, and concepts involved in, the bioprocess modeling, that need to be revisited and implemented in an integrated framework, are briefly reviewed. Subsequently, an assessment is made of various activated sludge models, originally developed for, and commonly used to simulate CASP, as well as some

modified versions thereof employed in MBR systems. Special care is paid to clarify the extent to which existing mathematical models can provide suitable key parameters that could link biological treatment and membrane filtration processes. Along these lines, the paper indicates some guidelines concerning the development of appropriate bioprocess models for MBR systems.

2. The interrelationship between biological treatment and membrane separation

In MBR technology, the biological treatment and the membrane separation cannot be considered as independent sequential unit operations [9], since they are closely interrelated, resulting in an entirely new process with its own particular characteristics. This *de facto* interaction which is, after all, the typical feature of the MBR technology [9] is outlined in the subsequent sections.

2.1. Influence of biological processes on membrane separation

Since the membrane, in MBR systems, is in direct contact with an active biological suspension (called mixed liquor), the physicochemical characteristics of the latter inevitably affect the membrane filtration process. Mixed liquor may be subdivided into three idealized components, i.e. *suspended solids*, *colloids* and *solutes*. This approach has often been employed to account for the relative contribution of each biomass fraction on MBR fouling, though it neglects any coupling or synergistic effects, which may occur between these components, and there is no standard classification methodology, which would allow the interpretation of the results from different studies [2]. Although the contribution to fouling of each component is still unclear, the contribution of the mixed liquor supernatant (i.e. soluble matter and colloids) seems to be relatively more important than that of the suspended solids [2,3]; indeed, in terms of fouling mechanisms, the biomass supernatant is considered to be responsible mainly for the irreversible and difficult to tackle narrowing and blockage of the membrane pores, while suspended solids tend to form a mostly reversible cake layer [2,3,6].

2.1.1. The role of the mixed liquor supernatant

The composition and concentration of organic matter present in the mixed liquor supernatant (referred to as *Dissolved Organic Matter* or *DOM*) appear to play a significant role in the membrane filtration process. The abbreviation *DOM* is a general term which is used to

describe the organic matter present in the bulk of a liquid, and, although there is no standard definition, it usually refers to organic compounds with size smaller than 0.45 μm , thus comprising both solutes and colloids/macromolecules. In MBR systems, a large portion of DOM consists of soluble organic compounds of microbial origin that are frequently referred to as *soluble Extracellular Polymeric Substances (sEPS)* or *Soluble Microbial Products (SMP)*. These substances result from substrate intermediates and/or end products as well as from hydrolysis, lysis and biomass decay [10]. Nonetheless, it should be noted that another portion of DOM consists of the originally present non-biodegradable organic matter and the potentially residual biodegradable organic matter of the influent; thus, the term sEPS/SMP, although it is used so by many authors, does not represent the total organic matter present in the mixed liquor supernatant.

During filtration, DOM compounds are absorbed onto and/or into the membrane, block membrane pores and form a partly irreversible gel structure on the membrane surface and into the membranes pores; moreover, they provide an excellent base layer for the attachment of bacteria and they also serve as a possible nutrient source for attached bacteria growth and biofilm formation. All these mechanisms are considered responsible for increased hydraulic resistance to permeate flow [11]. Le-Clech et al. [2] and Meng et al. [6] have reviewed, among other issues, the relationship between membrane fouling and DOM concentration and have concluded that, in general, higher concentration of DOM results in greater loss of hydraulic performance, although its composition seems to be a more important factor. In particular, a direct relationship between the carbohydrate level in DOM with various parameters indicating fouling propensity (i.e. fouling rate, filtration index, critical flux etc.) has been identified [2,6].

The above observations have been also taken into account in various mathematical modeling efforts. Ishiguro et al. [12] proposed a simple mathematical expression to describe membrane flux, being proportional to the difference of DOM concentration between the mixed liquor and the permeate. Fan et al. [13], investigating the effect of sludge characteristics on critical flux, concluded that the latter, measured by the stepwise flux method, was almost solely related to the colloidal *Total Organic Carbon (TOC)* concentration and recommended an empirical mathematical expression. Liang et al. [14] developed a mathematical model for membrane fouling in which both reversible and irreversible fouling were quantified; it was also speculated that DOM was the key contributor to irreversible fouling and the analytical expressions that were obtained both for permeate flux and *Transmembrane Pressure*

(TMP) evolution used the DOM concentration as model input variable. Busch et al. [15] presented a detailed model for submerged outside-in hollow fiber filtration which considered various fouling mechanisms like pore blocking, cake layer formation and biofilm formation. The mathematical expression of pore blocking and cake layer formation was based on the work of Broeckmann et al. [16], in which the pore blocking resistance is proportional to the concentration of the species in the bulk phase that can penetrate the membrane pores, i.e. to the concentration of DOM compounds. Moreover, the concentration of DOM was used as an input in the biofilm formation resistance model representing the organic matter that can either be attached to the biofilm or serve as substrate for bacteria growth in the biofilm [15]. Despite the fact that different model variables are used in this work [15], all of them literally stand for the DOM concentration. Finally, Guglielmi et al. [17] presented a subcritical flux fouling model that could predict the time at which a sharp change in the TMP-time profile (TMP jump) occurred. In this model the time when the TMP jump occurred was inversely proportional to the concentration of the DOM.

2.1.2. The role of the mixed liquor suspended solids

Suspended solids seem to be responsible for the formation of a cake layer and/or biofilm onto the membrane surface [2,6]. The *Mixed Liquor Suspended Solids (MLSS)* concentration or the *Mixed Liquor Volatile Suspended Solids (MLVSS)* concentration are the usually employed parameters to define the concentration of suspended biomass in wastewater treatment plants since they are directly measurable and monitored on a daily basis [18]. Meng et al. [6] reported various, mainly empirical, mathematical expressions describing membrane flux or membrane fouling rate which included MLSS/MLVSS concentration. Furthermore, Liang et al. [14], in their mathematical model, considered MLSS as the major component of the reversible fouling and concluded that the resistance of the reversible cake layer was proportional to the MLSS concentration. However, it appears that the MLSS/MLVSS concentration has a rather complex relation to MBR fouling, and contradictory findings about the effect of this parameter on membrane filtration have also been reported [2,6]; thus, the MLSS or MLVSS concentration alone seems to be a poor indicator of biomass fouling propensity [2]. This unclear picture should not be unexpected since the MLSS/MLVSS concentration is a lump parameter, which represents a large number of different kinds of suspended organic matter with possibly different fouling propensity.

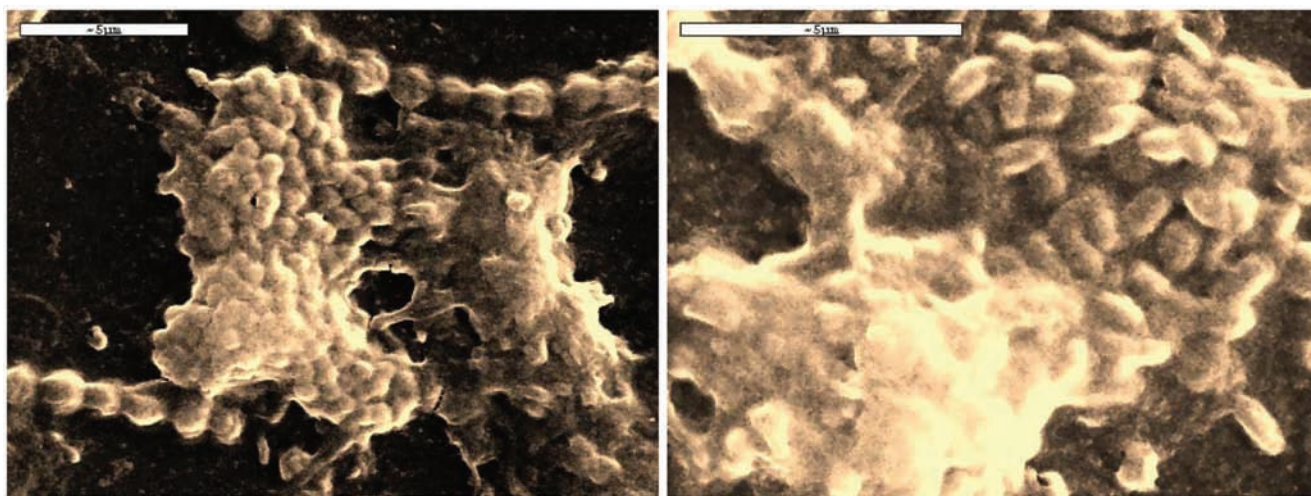


Fig. 1. SEM images of suspended biomass aggregates [20].

A detailed analysis of the biofloc structure reveals the great importance of a highly hydrated gel polymeric matrix, formed by *Extracellular Polymeric Substances (EPS)*, in which the bacteria are embedded and more or less immobilized. By definition, EPS are bound at or outside the cell surface regardless of their origin [19]; hence, the term *bound EPS (bEPS)*, which is used in many papers to differentiate sEPS/SMP from EPS content of bioflocs, may be considered superfluous. EPS refers to various classes of organic macromolecules such as polysaccharides, proteins, nucleic acids, phospholipids, humic substances and other polymeric compounds, which have been found to fill the intercellular spaces of microbial aggregates such as biofilms, activated sludge flocs and anaerobic sludge granules. Furthermore, their composition may be due to various processes, i.e. active secretion, shedding of cell surface material, cell lysis and adsorption from the environment [19]. Fig. 1, obtained in this Laboratory [20], shows Scanning Electron Micrographs (SEM) of suspended biomass aggregates. Although the dewatering procedure required for SEM examination produces artifacts, it is evident that bacteria are embedded in a slime matrix of EPS.

Despite the fact that EPS do not have such a clear and/or direct correlation with membrane fouling as DOM, their effect on MBR filtration has received considerable attention in recent years [2,6]. Empirical mathematical expressions were presented in some studies in attempts to quantify the impact of bioflocs EPS content on membrane fouling. Meng et al. [21] developed an empirical expression for fouling resistance under constant pressure filtration which included the EPS content of the mixed liquor. Guglielmi et al. [17] also included EPS concentration in their subcritical flux

fouling model outlined above. However, the EPS impact on the fouling process is rather complicated [6], since EPS are found to influence considerably activated sludge structural characteristics (mean floc size and floc size distribution, floc shape, porosity and water content) as well as physico-chemical properties like zeta potential, surface charge and hydrophobicity [2,6,22]. The EPS content apparently also affects various properties of the resulting fouling cake layer such as the specific cake resistance, the cake compressibility as well as the stickiness or adhesion coefficient [2,6,23]. Busch et al. [15] suggested that the removal rate of a biofilm from the membrane surface depends on the degree of cross-linking within the biofilm which was considered to be proportional to the mass ratio of EPS to microorganisms. They also suggested a modeling approach, where if there is no EPS the cross-linking degree approaches zero, while high EPS concentrations lead to a cross-linking degree of unity [15]. Le-Clech et al. [2] proposed that there may be a near optimum EPS level at which a stable biofloc structure could be maintained without exhibiting high fouling propensity

Furthermore, the concentration of EPS seems to play an important role in the regulation of DOM concentration, since these two organic fractions are closely interrelated. Rosenberger et al. [24] compared the LC-OCD chromatograms of extracted EPS and the organics in the sludge supernatant (i.e. DOM) and found that the two chromatograms exhibited the same pattern. Thus, it can be concluded that DOM and EPS are of the same nature and their relative concentrations are under a dynamic equilibrium which can be easily shifted by changing conditions in the mixed liquor environment [24]. Indeed, various processes and/or

conditions may result either in the biosorption of DOM by the bioflocs and the formation of EPS [25] or in the dissolution/hydrolysis of EPS and the release of DOM in the bulk liquid [26]; for example, the concentration of divalent cations (Ca^{+2} , Mg^{+2}) [23, 26], the addition of coagulants [6], pH or temperature variations, enzymatic hydrolysis [27], fluid shear stresses and oxygen concentration [28], have been reported to affect the EPS - DOM equilibrium. In fact, fouling mitigation via addition of coagulants, such as the so-called *Membrane Fouling Reducers (MFR)* or *Membrane Fouling Enhancers (MFE)* [2, 6], is apparently based on the aforementioned interrelationship between EPS and DOM; indeed, addition of coagulants directly to the bioreactor was found to reduce the DOM concentration, leading to a proportional increase in the concentration of EPS [6].

2.2. Influence of membrane separation on biological processes

Membrane separation has a significant influence on the biological processes in MBR wastewater treatment plants, which can be attributed to two different mechanisms, acting directly and indirectly. Membrane filtration *directly affects* the biological treatment through the complete retention of all the mixed liquor components that are larger than the membrane pores. This is in contrast to CASP where biomass components with poor settling characteristics are washed out with the effluent from the sedimentation tank. Ng and Hermanowicz [29] studied the performance and the biomass characteristics of a CASP system and a MBR, operating under the same HRT and SRT and treating the same synthetic wastewater. It was clearly observed that for all operating conditions studied, the amount of non-flocculating microorganisms in the MBR was much higher than in the CASP. Moreover, the composition and structure of biomass flocs in MBR was different than that in CASP despite identical inoculums, wastewater influent characteristics and similar operating conditions. This difference was attributed to the fact that in CASP, non-flocculating microorganisms are not retained in the system while in a MBR both flocc-forming and dispersed microorganisms are retained. Masse et al. [30] also compared biomass structure of a MBR and CASP using various techniques, which showed significant differences in sludge morphology. At similar SRT, the number of non-flocculating bacteria was clearly higher in the MBR. Moreover, it was concluded that in MBR systems a significant proportion of the DOM is rejected by the membranes and, thus, retained in the bioreactor, unlike the CASP where it is washed out [30]. It is noted here that most of the DOM consists of SMP which are considered by some

researchers to act as inhibitors for some bacteria species (mainly nitrifying) [31]. Regardless of being toxic or not, the elevated concentration of DOM in MBR supernatant tends to influence the overall bioprocess. Finally, the complete retention of suspended biomass in MBR favors, under appropriate operating conditions (i.e. high SRT), the growth of a widely diversified bio-community; the latter includes specialized bacteria that enhance breakdown and eventual assimilation/mineralization of organic components, like organic micropollutants, that are otherwise difficult to biodegrade.

Membrane filtration can also have an *indirect influence* on biological processes and biomass kinetics. Since the separation of biomass from the treated wastewater in MBR plants is independent of the suspended solids settling characteristics, it is possible to independently control the HRT and the SRT in order to achieve an optimum biological treatment with no constraints resulting from sludge settling characteristics. Currently, MBR tend to be operated with longer SRT (10–30 days) than CASP, thus maintaining higher biomass concentration, reducing solids production and minimizing reactor volume [1,5]. Under such operating conditions, the biomass is kept as close as possible to a food-limited environment in order to encourage endogenous decay. This uncoupling of metabolism, so that catabolism of substrate continues unhindered while anabolism of biomass is restricted, leads to a reduction in the observed biomass yield and hence to reduced sludge production [9]. At such low growth rates or long-term limitations, which are typical in MBR operation, microorganisms undergo changes in metabolism [32] that are not common in the CASP and should be taken under consideration by models of activated sludge describing the MBR process [33]. In addition, the aeration rate in MBR is greater than in CASP in order to promote shear stresses on the membrane surface and reduce fouling [2]. These hydrodynamic stresses in MBR tend also to influence the biofloc morphology by generally reducing floc size compared to the CASP and promoting dissolution of EPS [28,30]; indeed, a comparison of the aggregate size distribution of CASP and MBR mixed liquor reveals a distinct difference in terms of mean particle sizes and particle size distribution [2,9].

2.3. The requisite features of an activated sludge model for MBR technology

The fundamental concepts of the bioprocesses applicable to MBR systems are essentially the same as those employed to describe CASP; thus, it is understandable that literature studies of bioactivity modeling in MBR systems are based on existing or slightly

modified activated sludge models for the CASP [34]. However, the review in the preceding section suggests that these mathematical models as well as the bioprocess kinetics and parameters used to describe CASP should not be carelessly adopted for bioprocess modeling in MBR systems, and that a careful evaluation of existing models should be carried out, especially addressing the prevailing different operating conditions in MBR. For example, a bioprocess mathematical model for MBR systems should account for, and successfully describe, the elevated concentration of DOM in MBR and the higher MLSS concentration. Moreover, the partial inhibition of the nitrifying activity and/or the enhanced biodegradability of activated sludge in MBR should be considered. *Therefore, regarding an activated sludge model for MBR, one should take into account the substantially different properties of the activated sludge, compared to the CAS process, and the likelihood that a quite different set of model parameters and/or various model modifications and adaptations may be needed.*

Furthermore, since the physicochemical characteristics of the biological suspension inevitably affect the filtration performance, which is of crucial importance for system reliability and process economics, models of biomass activity in MBR should have a dual perspective. *They should be capable of both adequately describing the complex biological processes that take place in the bioreactor as well as accounting for the biomass characteristics that greatly affect membrane filtration performance.* In particular, a basic model of biomass kinetics in MBR should at least be capable of providing estimates of the EPS concentration in the activated sludge flocs and the concentration of DOM in the bioreactor supernatant taking into account the existence of SMP. Knowledge of the variation of these foulants, in response to changes of operating parameter values (i.e. SRT, HRT, aeration rate, etc), is of paramount importance for adjusting the design and operation of MBR with the objective of minimizing fouling.

3. Review of activated sludge models

Mathematical models have gained wide acceptance and are used extensively for designing and simulating the CASP. Despite the fact that activated sludge modeling has been thoroughly dealt with and reviewed elsewhere [35], a short review of the main CASP models is provided. The scope of this review is not to provide an extensive and in depth analysis of every variation or version of CASP models but rather to outline the features and capabilities of these models for describing the specific biological processes and operational conditions in MBR systems and to point out their limitations. Additionally, certain modified CASP models, which

have been developed for MBR systems, are assessed regarding the extent to which they fulfill the aforementioned requirements for MBR applications.

3.1. Activated Sludge Model no 1 (ASM1)

This model is the outcome of work by a task group formed in 1983 by the International Association on Water Quality (now called International Water Association – IWA). The final report was presented in 1987 [36] and the proposed model, called Activated Sludge Model No1 (ASM1), soon gained general acceptance. Although ASM1 has been upgraded since that time, it is still the most widely used model for conventional municipal wastewater treatment processes, serving as a reference in any subsequent modeling effort [37,38]. Many of its concepts, like bisubstrate hypothesis, death-regeneration process, the Monod type kinetics, the use of *Chemical Oxygen Demand* (COD) as the suitable parameter for defining the organic matter, the COD fractionation and the hydrolysis process, are widely employed by most of the CASP models, and it is still considered as “state of the art” model if biological phosphorus removal is not taken into account [35]. Sperandio and Espinosa [33] evaluated the performance of ASM1 for modeling the biological processes in an aerobic submerged MBR for a large range of SRT. Their study was mainly focused on predicting suspended solids, excess sludge production, and nitrification kinetics. ASM1 provided satisfactory predictions although adjustments in some model parameters as well as some modifications in model structure were proposed, especially at prolonged SRT.

However, in ASM1 there is no provision for simulating the fate of, and providing data for, some organic fractions that play an important role in MBR operation. In particular, the ASM1 is unstructured in regard to bioflocs which are simply considered as a uniform aggregate of biomass. Indeed, there is no reference to the extracellular polymeric matrix that surrounds the bacteria population and is responsible for the structural and functional integrity of bioflocs, despite the fact that it is proposed that both biodegradable and non-biodegradable particulate organic matter is enmeshed in, or entrapped by, the bioflocs (i.e. the extracellular polymeric matrix). Moreover, there is no provision to describe the production of EPS by the bacteria population and thus it is impossible to simulate and predict their concentration. Furthermore, ASM1 does not provide the necessary information to accurately determine the DOM concentration of the supernatant in MBR systems. Despite the fact that the bisubstrate hypothesis improves the model prediction of the activated sludge process, an ambiguity exists

since it is not clear whether the slowly biodegradable COD should be treated as a soluble or particulate fraction. Also, ASM1 does not account for SMP despite the fact that their existence in activated sludge supernatant is undeniable [10]. Instead, it is stated that the organic matter present in the activated sludge supernatant is of the same origin as that in the influent and either it is left in the supernatant due to process limitations (on the rate of removal of the biodegradable organic matter originally present in the influent stream) or it is the non-biodegradable soluble organic matter present in the wastewater influent which leaves the system at the same concentration as that at the entry. In other words, ASM1 does not account for the influence that the operating conditions have on DOM concentration, through SMP production, but it is assumed that DOM concentration depends entirely on the influent characteristics [39].

3.2. Activated Sludge Model no 2/2d (ASM2/2d)

Activated Sludge Model No2 (ASM2), was presented in 1995 [40] and extended the capabilities of ASM1 by including biological phosphorus removal. The model incorporated a new group of bacteria, which originally consisted of heterotrophs and autotrophs. The new group, called *phosphorus-accumulating organisms (PAO)*, encompasses the different types of microorganisms capable of accumulating, under aerobic conditions, phosphorus and storing it in the form of cell internal structures. The activated sludge model No2d (ASM2d) [41], incorporated the observation that PAO can use internal cell organic storage products for denitrification and can grow under anoxic conditions, through the addition of two new rate processes. All other details of ASM2 were carried over to ASM2d [34]. Since these two models were extensions based on ASM1 the aforementioned deficiencies of the latter apply to them as well.

3.3. Activated Sludge Model no 3 (ASM3)

The Activated Sludge Model No3 (ASM3) was presented [18] in an attempt to correct some of the deficiencies of the ASM1 that have become apparent during its 10 years of application. In ASM3 the importance of internal storage polymers in the heterotrophic biomass is recognized, especially in systems subjected to feast and famine conditions [42]. Despite the fact that ASM3 has not been tested as extensively in full-scale wastewater treatment plants as ASM1 [18], it is generally considered equivalent to ASM1, in terms of describing the dynamic behavior of municipal wastewater treatment plants, after being properly calibrated [43]. However, it has been found that the ASM3 fails to model some experimental observations of

storage phenomena and in particular the fact that storage and growth occur simultaneously during feast periods [42]. This observation has led to the development of various modified models of simultaneous storage and growth that have attempted to better represent these experimental data [42,44].

In their study, Sperandio and Espinosa [33], apart from ASM1, evaluated the performance of ASM3 for describing the bioprocesses in an aerobic MBR under various SRT; despite the fact that in some cases ASM3 performed better than ASM1, adjustments in model parameters as well as some modifications in model structure were required. Wintgens et al. [45] used ASM3 to simulate the steady state operation in a full-scale MBR plant and reported that the simulated values corresponded well with the data. In that paper, an attempt was made to combine filtration with bioprocess modeling; however, there was no coupling or interaction between these two modeling compartments since the semi-empirical filtration model did not employ any data input from the ASM3. The above findings suggest that in ASM3 little progress has been made in terms of the requisite features of an activated sludge model catering to the special needs of MBR technology, as previously outlined. As in the case of ASM1, ASM3 does not provide information on the biofloc structure and thus it is impossible to make any predictions regarding EPS concentration. Although, Wintgens et al. [45] recognized that the amount of EPS may be crucial for the filtration process, its concentration could not be quantified due to the lack of an appropriate model framework. Saroj et al. [46] used a modified version of ASM3 [42] and coupled it with a very simple EPS model in order to overcome the aforementioned problem; they assumed a linear dependence of EPS concentration on the biomass concentration through an EPS factor which was taken to be a function of several environmental and operating conditions. Despite being a step towards the introduction of more detail, by considering organic fractions that play an important role in fouling phenomena, the proposed model lacks the basic underpinning that could provide a comprehensive treatment of the formation and degradation rate of EPS organic fraction. Finally, it is pointed out that in ASM3 the production of SMP is not included in the mathematical model, though their existence is rather loosely implied; thus, in ASM3 (as in ASM1) the concentration of DOM in the supernatant is regarded to be practically independent of the operating conditions.

3.4. Soluble Microbial Products (SMP) models

The incorporation of SMP into the modeling of wastewater treatment was first attempted by Daigger and

Grady [47], but it was Namkung and Rittmann [48] who investigated SMP formation kinetics in a biofilm reactor and successfully [10] developed an extended steady state biofilm model based on the classification of SMP into two categories; i.e. the growth associated microbial products or *Utilization Associated Products (UAP)* that are produced at a rate proportional to the rate of substrate metabolism, and the non-growth associated microbial products or *Biomass Associated Products (BAP)* formed at a rate proportional to the concentration of the biomass. Furumai and Rittmann [49], based on a study of Rittmann et al. [50] for the kinetics of UAP and BAP formation, presented an activated sludge model that could describe carbon oxidation and nitrification by a mixed population of both heterotrophs and autotrophs. Both kinds of bacteria would produce SMP but only heterotrophs could degrade and use them for cell synthesis with multiple-substrate degradation kinetics. Eleven mass balance equations for each model component were provided. However, it should be noted that the model did not consider anoxic conditions (denitrification) and did not adopt the bisubstrate hypothesis; thus, the hydrolysis process was ignored. De Silva et al. [51] used a modified version of the model of Furumai and Rittmann [49] to quantify the relationships between heterotrophic bacteria, autotrophic bacteria and key chemical constituents in a MBR operated under aerobic-anoxic cycles. The model, which was evaluated against experimental data from a pilot MBR system, including MLSS concentration and effluent quality (COD, total nitrogen, NH_4^+ and NO_3^-), seemed to capture the trends for soluble COD and nitrogen species.

In an attempt to take advantage of the benefits of both the ASM family and the SMP models, various researchers have tried to incorporate the formation and degradation of SMP in the well-known models of ASM family. Orhon et al. [52] introduced a mathematical model which combined ASM1 with SMP production. They examined the theory of Rittmann et al. [50] and considered that only BAP contributed significantly to soluble COD of the mixed liquor. The model was calibrated for a set of experimental data derived from a sequencing batch CASP reactor. However, the model evaluation was only based on the soluble COD and MLVSS concentration. This model [52] was further developed to include UAP [53] in order to account for the effect of the initial substrate concentration on the performance of CASP. Model simulations of a number of practically significant scenarios were performed; nonetheless, the resulting model was rather simple and poor experimental support was provided.

Lu and coworkers [39,54] modified ASM1 and ASM3 by incorporating mechanisms that described the

fate of SMP; they also recognized the existence of both UAP and BAP despite the fact that in the model they were grouped together in a single state variable defined as SMP. Moreover, hydrolysis products of particulate biodegradable organic matter arising from biomass decay were also classified as BAP because their biodegradable characteristics were considered the same as BAP. The models obtained were quite complicated since eight new SMP-related parameters were introduced. The ASM1-SMP model was evaluated under steady-state conditions for an intermittent aerobic MBR system and the simulation results were generally in good agreement with experimental data, despite the fact that a significant underestimation of MLVSS concentration was noticed [39]. The ASM3-SMP model was not evaluated against experimental data; their study [54] provided the simulated predictions of the performance of an intermittent aerobic MBR system under various HRT and SRT conditions. Moreover, some processes of ASM3 (i.e. the storage of internal biopolymers) were not considered in their ASM3-SMP model and they were replaced by concepts originally found in ASM1.

Lee et al. [55] based on the study of Lu et al. [39] proposed a slightly different model which also incorporated SMP formation kinetics into ASM1. Their study was mainly focused on the effect of SMP on membrane performance and the developed activated sludge model, which was inadequately described, was coupled with a resistance-in-series filtration model. However, the resistance of the fouling layer was considered to be dependent only on the MLVSS concentration and the effect of SMP on fouling was neglected. Moreover, their model was not evaluated against experimental data and only numerical case studies on the effects of various operating conditions (mainly SRT) on flux decline rate or MLSS accumulation rate were provided [55]. Di Bella et al. [56] adopted the modified version of ASM1, proposed by Lu et al. [39], in an attempt to present an integrated model for MBR process which could also estimate the cake layer effect on the permeate COD concentration. The model was evaluated against the MLSS concentration and the permeate COD, NH_4^+ and NO_3^- concentrations of a submerged MBR pilot plant and successfully simulated the measured data, after being calibrated. The model results confirmed the important role of SMP in the biological processes in MBR and quantified the significance of cake deposition in the filtration process. Recently, Gonzalez et al. [57] developed an integrated model that coupled biomass transformation processes, membrane fouling and the effects of filtration cycles with intermittent coarse bubble aeration. The sub-model describing the biological transformations was

also based on the study of Lu et al. [39] and was coupled with the total filtration resistance sub-model through two basic variables; i.e. MLSS and SMP concentration. The model was evaluated against the TMP profile of a pilot MBR system operated under various conditions and the simulation results were reported to be in good agreement with the experimental data. This modeling procedure [57] constituted a step forward since it linked a fouling model with parameters that were directly influenced by the bioprocesses.

Oliveira-Esquerre et al. [58] proposed a modification of ASM3 that could take into account the process of production and consumption of microbial products in a submerged MBR. They considered that SMP were mainly produced by biomass decay (BAP) and the simulation results of the proposed modified ASM3 were compared with steady-state experimental results of a pilot MBR as well as with the results of the modified ASM1 proposed by Lu et al. [39]. Evaluation of the results showed that the carbonaceous materials were more accurately estimated by the modified ASM3, while the model of Lu et al. [39] performed slightly better in the estimation of nitrate. Jiang et al. [59] extended the existing ASM2d to ASM2dSMP by introducing kinetics for formation and degradation of SMP. Dynamic batch experimental results were used for the estimation of the additional SMP-related parameters and the model was validated using independent experimental results of a lab-scale MBR monitored under steady state conditions. The simulated sludge and effluent concentrations using the ASM2dSMP showed good agreement with the measurements and they were generally better than the simulation results using the original ASM2d. Additionally, the model was used to evaluate, through simulation, the impact of operational parameters on the SMP concentration and concluded that SRT is the key operational parameter controlling SMP concentration [59].

It is evident that most of the aforementioned MBR modeling studies [39, 54–59] stress the importance of SMP in MBR operation. Undoubtedly, the incorporation of production and degradation of SMP in the conventional activated sludge models constitutes progress in terms of bioprocess modeling for MBR. These models allow for a better prediction of the DOM concentration in the MBR supernatant, suggesting trends of operating conditions that may lead to control and reduction of SMP production. However, most of these hybrid models (ASM+SMP) are generally too complicated and over-parameterized; furthermore, various process variables are introduced that are impossible to determine experimentally in full-scale MBR systems (e.g. UAP and BAP) and, thus, serious identifiability issues are raised. Moreover, various ambiguities of the

ASM family models are not resolved; e.g. whether or not the products of hydrolysis, together with the produced SMP are released back to the bulk liquid before being assimilated by the bacteria. Finally, the major drawback of these models is that they do not account for the EPS of the bioflocs. Despite the fact that Lu et al. [39] imply the existence of the EPS, by attributing the difference between the measured and the calculated concentration of MLVSS to some large molecular weight organic matter that is absorbed around the biomass, there is neither a specific variable nor any expression describing the EPS production. Thus, it is impossible to simulate and predict their concentration in the activated sludge biofloc. For the same reason (i.e. the inability to simulate EPS content), some model parameters, which are employed in the filtration model equations and stand for important physicochemical properties (i.e. the stickiness of the biomass, the compression coefficient of the cake layer) in the Di Bella et al. [56] model, are considered independent of the EPS activated sludge floc content, though they can be significantly influenced by it. In the work of Gonzalez et al. [57] this kind of inconsistency also exists; specifically, in their total filtration resistance sub-model, they take into consideration the effect of the EPS concentration on the specific resistance of the biomass cake. However, their biological sub-model does not include a variable that refers to the EPS concentration; thus, they employ a questionable assumption that the EPS content of the bioflocs is equal to the SMP concentration in the mixed liquor supernatant.

3.5. The Laspidou and Rittmann model

In 2002 Laspidou and Rittmann [60,61], reviewing the relationship between EPS and SMP, recognized the existence in the literature of two different approaches that have treated these groups of compounds separately and has resulted in using different terminology to describe the same organic fractions. A unified theory was proposed that coupled these different approaches and reconciled apparent contradictions. This theory took into account the following groups of organic matter: original soluble substrate, active and inert biomass, EPS and SMP. They also proposed a mathematical model that quantified the relationships among these organic fractions and an electron acceptor such as oxygen. The model, comprised of seven mass balance equations, was evaluated against experimental data obtained from Hsieh et al. [62] and good agreement was observed for both steady state and transient conditions. It is noted that these data [62] were obtained in an attached growth wastewater treatment process but the theory is also considered applicable to suspended

biomass systems [60]. The unified model of Laspidou and Rittmann seems to properly capture the interrelation between bacteria, EPS and SMP. Moreover, it clearly defines the physical state (soluble or particulate) of all the variables involved. Finally, in contrast to the traditional view, which considers that the assimilated substrate is either converted to new biomass or catabolized to produce energy, Laspidou and Rittmann provide strong evidence that part of the substrate is shunted to EPS (mainly) and SMP production, and therefore the available substrate for biomass formation is reduced.

Despite the fact that the Laspidou and Rittmann modeling effort is the first one that successfully incorporates EPS and SMP formation in bacteria growth models, it still cannot be used as is for bioprocess modeling in MBR, since it is too simple to describe the complicated biological phenomena therein. More specifically, the Laspidou and Rittmann model was developed and evaluated for a biosystem with rather simple substrate input that is considered merely soluble and readily biodegradable. These conditions are very different from those in a real wastewater treatment plant, where the organic matter in influent wastewater is very complex and consists of both soluble and particulate fractions with different biodegradability rates. The Laspidou and Rittmann model does not adopt either the bisubstrate hypothesis or the hydrolysis process that is very important in real systems [35]. Moreover, the model does not account for autotrophic bacteria species, does not include nitrification and cannot provide any information about nitrogen removal processes. Finally, since oxygen is the only electron acceptor, the model applies only to aerobic conditions and it cannot be used under anoxic ones (denitrification).

Jang et al. [63] underlined the effect of operating conditions, such as SRT and HRT, on the characteristics and concentration of EPS and SMP, and the need to further investigate their effect on membrane fouling in relation to biological kinetics. Thus, in their study [63] the unified approach of Laspidou and Rittmann [60, 61] was coupled with a semi-empirical fouling model in an attempted to relate the modified fouling index with the *Food to Microorganism (F/M)* ratio and the concentration of DOM in the MBR supernatant. The reported simulation [63] was carried out to predict trends of the fouling potential, in relation to operational parameters of the MBR. Experimental data were used only to apply the semi-empirical filtration model parameters and not to validate the model performance to a different range of operating conditions. Moreover, the parameters used are those of Laspidou and Rittmann [61] obtained under completely different

conditions (pure culture and single substrate experiments) from those in a real MBR system; thus, although these results are interesting and reproduce correctly some general trends observed experimentally, further research is clearly required to obtain a comprehensive modeling framework. Ahn et al. [64], attempted to resolve the deficiencies of Laspidou and Rittmann [60, 61] model and integrated their unified theory into the ASM1 in order to predict the fate of SMP and EPS under various SRT conditions. Basic model parameters of ASM1 were determined experimentally by a respirometric method with samples from three pilot MBR systems operated under different SRT. However, the proposed model is very complicated and the parameters related to EPS and SMP formation seem almost impossible to be determined. Moreover, despite the fact that the analytical measurements of SMP and EPS concentration [64] were generally in good agreement with the modeling results, the measured SMP concentration was very low and inconsistent with literature data [10, 11].

3.6. An overview of activated sludge models

To facilitate a comparison and provide an overall view of the various activated sludge models, that were assessed in this review, Table 1 is presented summarizing key characteristics. In this overview, the following significant features of the mathematical models are considered:

1. *Bioprocesses simulation*: This criterion considers which of the following basic bioprocesses are dealt with in the mathematical model: carbon oxidation, nitrification, denitrification, hydrolysis process and phosphorus removal.
2. *Provision of key variables for fouling predictions*: The models are judged on whether they can describe the fate of different microbial products, like SMP and EPS, which play an important role in the membrane fouling process.
3. *Application to MBR*: The criterion considers if the models have been applied to simulate the bioprocesses in MBR systems.
4. *Model evaluation*: It is examined whether the models have been evaluated on the basis of experimental data.
5. *Model ease-of-use*: The number of the bioprocesses and the state variables that are defined in the models are provided as an indicator of the model complexity.

Upon inspection of Table 1, it is observed that the ASM family models include all the basic bioprocesses taking place in wastewater treatment plants and their validity has been proven in numerous studies.

Table 1
Overview of activated sludge models assessed in this paper

Model	Bioprocesses simulation				Provision of key variables for fouling predictions		Application to MBR	Model evaluation	Model ease-of-use	
	Carbon oxidation	Nitrification	Denitrification	Hydrolysis	Phosphorus removal	SMP				EPS
ASM1 [36]	✓	✓	✓	✓	×	×	×	✓	8	13
ASM2/2d [40, 41]	✓	✓	✓	✓	✓	×	×	✓	19/21	19/20
ASM3 [18]	✓	✓	✓	✓	×	×	×	✓	12	13
Orhon et al. [52]	✓	×	×	✓	×	×	×	×	3	8
Artan et al. [53]	✓	×	×	✓	×	×	×	×	3	5
Furumai and Rittmann [49]	✓	✓	×	×	×	✓	×	×	?	11
De Silva et al. [51]	✓	✓	✓	×	×	✓	×	✓	?	10
Lu et al. [39]	✓	✓	✓	✓	×	✓	×	✓	10	12
Lu et al. [54]	✓	✓	✓	✓	×	✓	×	×	17	12
Lee et al. [55]	✓	✓	✓	✓	×	✓	×	✓	12	13
Lapidou and Rittmann [61]	✓	×	×	×	×	✓	✓	×	11	7
Ahn et al. [64]	✓	✓	✓	✓	×	✓	✓	×	15	16
Oliveira-Esquerre et al. [58]	✓	✓	✓	✓	×	✓	×	✓	14	14
Jiang et al. [59]	✓	✓	✓	✓	✓	✓	×	✓	27	22

✓: Yes, ×: No, ?: Incomplete reported information

However, they cannot provide key variables for fouling predictions and for this reason, several modified versions, that combine ASM family models with the SMP theory of Furumai and Rittmann [49], are proposed. Some of these models (i.e., Orhon et al. [52], Artan et al. [53] and De Silva et al. [51]) are very simple and cannot be used for full scale MBR systems while others (i.e., Lu et al. [54] and Lee et al. [55]) are inadequately evaluated and quite complicated. On the other hand, the models presented by Lu et al. [39] and Jiang et al. [59] can be considered as more integrated approaches concerning modeling of bioprocesses in MBR, but they do not include the EPS fraction. The Laspidou and Rittmann [61] model, despite being too simple, is the first one that successfully incorporates EPS and SMP formation in bacteria growth models. Ahn et al. [64] combine their theory with the ASM1 model in an attempt to simulate the concentration of both SMP and EPS organic fractions in MBR systems; however, the proposed model is quite complicated, with fifteen processes and sixteen state variables, and poorly evaluated.

4. Concluding remarks

This review suggests that key issues related to membrane fouling in MBR systems, such as reliable estimation of the effect of concentration of various organic fractions of mixed liquor, on reversible and irreversible fouling rates, have not been settled yet. Basic problems hindering progress in resolving these issues are the lack of a broadly accepted and clear definition of the organic fractions, accompanied by easy to implement experimental techniques for their determination, and the inability to relate, in a reliable manner, the concentration of these organic fractions to externally controlled MBR system parameters, through an effective modeling framework of the bioprocesses that take place in the bioreactor. Unless both these problems are effectively tackled, progress in improved fouling predictions and overall system optimization should not be expected. It is, therefore, evident that reliable mathematical modeling of the bioprocesses in MBR is of paramount importance for linking membrane fouling to controlled system parameters, leading to overall progress in this field.

However, the bioprocesses taking place in wastewater treatment plants are characterized by great complexity and consequently by incomplete understanding of the phenomena involved; therefore, regarding their simulation, a comprehensive approach is preferable, largely based on phenomenological, macroscopic type modeling, with some theoretical underpinning. By necessity, such models are usually developed for a specific task and their range of validity

largely depends on the objectives set in their formulation. This situation also holds for the case of MBR technology which is characterized by an additional intrinsic complexity resulting from the interaction between the con-currently occurring biological processes and membrane filtration. Regarding the mathematical modeling of bioprocesses in MBR, this inevitable, rather strong, interaction necessitates both the introduction of additional features - i.e. simulating organic fractions that are considered to be important fouling parameters - as well as appropriate model modifications and adjustments to cope with the substantially different operating conditions of MBR compared to CASP. It is argued in this review that existing conventional activated sludge models, by design, do not provide a level of detail sufficient to describe in an integrated manner microbial components (i.e. EPS and SMP) that play a critical role in MBR processes; moreover, for some cases specific to MBR operating conditions, they seem to be unsuitable for describing the biomass activity and kinetics. Additionally, modified versions of CASP models, that have been developed by quite a few researchers, are characterized by excessive complexity and insufficient demonstration of the validity of the model structure and of the recommended parameter values.

In view of the above considerations, this paper underlines the need for the development of an alternative bioprocess modeling framework which should be *de novo* focused on the specific needs of the MBR technology. Such a model should both adequately describe the complex biological processes, taking place in the bioreactor, and produce reliable and representative mixed liquor parameters needed to model and predict the membrane filtration operation. The unified theory of Laspidou and Rittmann [59, 60], despite the fact that it is at a rather rudimentary level, as well as the accumulated knowledge and expertise available in the ASM family models, could serve as a basis for the development of a comprehensive model, appropriate for the MBR process. Such a model could provide a significant thrust towards the broader use of MBR technology for wastewater treatment.

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