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Modelling Industrial Symbiosis of Biogas Production and Industrial Wastewater Treatment Plants - A Review

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Abstract

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The present-day treatment of pulp and paper mill effluents can be significantly improved by incorporating biogas production in the context of industrial symbiosis. In this work a new industrial symbiosis concept is presented, the focus being on modelling it in view of process optimization, design improvement and adoption by the pulp and paper industry. The concept consists of a first stage in which pulp and paper mills effluents are treated by high-rate anaerobic digestion in external circulation sludge bed (ECSB) reactors to produce biogas. In the second stage the removal of organic matter contained in the digestate stream occurs through aerobic activated sludge treatment, aiming to achieve maximum sludge production with minimum aeration requirements. This sludge should in the case study then be co-digested with fish-waste silage to yield methane for energy production, nutrients-rich reject water that can be recycled to the activated sludge treatment for optimum microbial activities and, production of nutrient rich soil amendment. The overall research aim is to develop a mathematical model that describes the relevant process units and the dynamics of the different processes involving organic matter removal, biogas production and nutrients release. The review overall finds that an integrated model is required to simulate this concept and should include recent developments in activated sludge, anaerobic digestion and physico-chemical modelling.

Keywords: biogas, industrial symbiosis, granular sludge bed reactors, anaerobic digestion, high-rate activated sludge system, modelling

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Content

Abstract **1**
Content **2**
Preface **4**
Nomenclature **6**
1 Introduction **7**
1.1 Background 7
1.2 Modelling opportunities 8
1.3 Objectives 9
2 Treatment of pulp and paper mill wastewater **10**
2.1 Pulp and paper mill effluents 10
2.2 Primary mechanical treatment 13
2.3 Aerobic treatment of pulp and paper mill effluents 13
2.3.1 Process description 13
2.3.2 Nutrient requirements for aerobic oxidation 14
2.3.3 High-rate activated sludge process 14
2.3.4 Methane oxidation in activated sludge 15
2.4 Anaerobic digestion of wastewater and sludge 15
2.4.1 Process mechanism description 15
2.4.2 Nutrients release 17
2.4.3 Operating conditions 17
2.4.4 High-rate anaerobic digestion of pulp and paper mill effluent 18
2.4.5 Anaerobic digestion of sludge 19
2.4.6 Anaerobic co-digestion of pulp and paper industry sludge with organic wastes 20
3 Mathematical modelling of pulp and paper mill effluent treatment ... **21**
3.1 Introduction 21
3.2 Activated sludge models 21
3.2.1 General overview 21
3.2.2 Limitation of activated sludge models 22
3.3 Anaerobic digestion models 23
3.3.1 Biochemical models 23
3.3.2 Modelling co-digestion 24
3.3.3 Modelling high-rate anaerobic reactors 24
3.4 Physico-chemical models 25
3.4.1 pH and ion speciation/pairing 25
3.4.2 Mineral precipitation 26

3.4.3 Temperature modelling in ASS ... 27
3.5 Plant-wide models ... 27
3.6 Commercial simulators in the wastewater field ... 28
3.7 Model calibration ... 29
4 Modelling industrial symbiosis for biogas production ... 30
References ... 31

Preface

This report is the first deliverable from the research project Modelling Industrial Symbiosis of Biogas Production and Industrial Wastewater Treatment Plants, running from 2017 to 2020. The project is funded by Sweden's Innovation Agency Vinnova and the leading project partner Scandinavian Biogas Fuels AB. Key partners of the project are Scandinavian Biogas Fuels AB, RISE Research Institutes of Sweden and Lund University (Division of Industrial Electrical Engineering and Automation).

Circular economy is a recent term to express the need for low emission, resource efficient and recycling processes for societies and industries. Industrial symbiosis is a means for achieving circular economy through interconnection of production facilities. Where, one industry benefits from the resources (sometimes considered as waste) form the other and forward (or feedback) resources to a third user. This way recycling loops can be closed, and logistic challenges solved. The highly resource intensive pulp and paper industry (PPI) is one interesting target for industrial symbiosis. One option to obtain a better energy balance for pulp and paper production and other industries generating organic wastes/residues, is to use the residues as substrates for biogas production. The world´s biogas production can be increased and geographically spread by exploring this today largely unused potential, thus contributing to climate change commitments worldwide. By co-locating a biogas production plant with the PPI wastewater treatment plant, waste sludge from the PPI can be co-digested with external substrates for bio-methane production. The nutrient rich digestate supernatant liquid from the biogas plant can be recycled to the PPI where it can replace commercial nutrients otherwise purchased and added. This is an excellent example where both parts (i.e. the biogas producer and the mill) benefits from the co-location and integration through substantial energy savings combined with decreased costs for handling sludge, decreased need for transportation of unrefined digestate and decreased use of commercial nutrients. These measures reduce cost while adding to climate change mitigation. Furthermore, the biosolids can be used as fertilizer by the nearby farms.

The main scope with the present project is to develop a modelling tool, able to assess the interactions of the circular economy concept (a symbiosis connecting an industrial activated sludge system with an anaerobic high rate system for wastewater treatment and a anaerobic co-digestion unit where rejected water from final dewatering serves as nutrient supply to the activated sludge system).

This technical report describes the literature review conducted for the project. The scope of the literature review has the same focus as of the overall project, which involves PPI wastewater treatment and modelling of the specific processes included in the concept. The literature review includes books, academic publications and reports (technical, consultancy, etc.). The literature was retrieved from search in academic databases (Scopus and Web of Knowledge), search on the internet and collected from project partners. Key review articles and reports with extensive reference listings have also been a source for finding relevant literature. The material has been reviewed to collect and compile the present state of knowledge related to the area of the project.

Chapter 1 in this report provides a background to the concept. In Chapters 2 to 4 the literature review is presented with extensive referencing. Chapter 2 is focused on the PPI effluents and wastewater treatment processes, including anaerobic digestion at mills. Chapter 3 is a detailed review of the modelling opportunities of the different subprocesses and in Chapter 4 modelling of the overall concept is laid out.

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Nomenclature

AA	Amino-acids	
AcoD	Co-digestion	
AD	Anaerobic digestion	
ADM1	Anaerobic Digestion Model No. 1	
AnMBRs	anaerobic membrane bioreactors	
ASM	Activated sludge model	
ASM2d	Activated Sludge Model No. 2d	
ASS	Activated sludge system	
BOD_7	7-day biochemical oxygen demand	($gCOD.m^{-3}$)
BSM2	Benchmark Simulation Model No. 2	
$CaCO_3$	Calcium carbonate	
CAS	Conventional activated sludge	
CH_4	Methane	
CO_2	Carbon dioxide	
COD	Chemical oxygen demand	($gCOD.m^{-3}$)
CSTR	Continuous stirred-tank reactor	
CTMP	Chemi-Thermomechanical pulp	
DO	Dissolved oxygen	($g.m^{-3}$)
ECSB	External circulation sludge bed	
EGSB	Expanded granular sludge blanket	
H_2S	Hydrogen sulphide	($mole.\ L^{-1}$)
HRAS	High-rate activated sludge process	
HRT	Hydraulic retention time	(d)
IC	Internal circulation	
LCFA	Long-chain fatty acids	
MLSS	Mixed liquor suspended solids	($gSS.m^{-3}$)
MS	Monosaccharides	
N	Nitrogen	
NSSC	Neutral sulphite semi-chemical	
O_2	Oxygen	
OLR	Organic loading rate	($kgVS/m^3d$)
P	phosphorus	
pH	Hydrogen potential	(standard)
PO_4^{3-}	Phosphate ion	
PO_4-P	Orthophosphate phosphorus	($gP.m^{-3}$)
PPI	Pulp and paper industry	
PPME	Pulp and paper mill effluent	
PS	Primary sludge	
RAS	Returned activated sludge	
SRB	Sulphate reducing bacteria	
SRT	Solid retention time	
TMP	Thermomechanical pulp	
TSS	Total suspended solids	($gSS.m^{-3}$)
UASB	Upflow anaerobic sludge blanket	
VFA	Volatile fatty acids	($gCOD.m^{-3}$)
VFAs	Volatile fatty acids	
VS	Volatile solids	
VSS	Volatile suspended solids	($gSS.m^{-3}$)
WAS	Waste activated sludge	
WWTP	Wastewater treatment pant	

1 Introduction

1.1 Background

The pulp and paper industry (PPI) is a large consumer of water (e.g. debarking, pulp preparation, bleaching water and boiler feed water as well as cooling water system) and generates large quantities of wastewater effluents that contain significant amounts of biodegradable and non-biodegradable organic material. Typical treatment approaches for pulp and paper mill effluents (PPME) involve a sedimentation step to remove suspended solids, and a biological treatment process, usually an activated sludge system (ASS), whereby organic matters are oxidized by the introduction of oxygen (air) into the wastewater (Pokhrel and Viraraghavan 2004). ASS requires large amount of energy for aeration to support the oxidation of organic matter (COD) and is often run at a high solid retention time (SRT), which results in a low waste activated sludge production (WAS) (Eddy et al. 2013). The low organic load and long retention times also mean that large basin volumes are needed for mineralization of organics in the ASS. Furthermore, unlike municipal wastewater, effluents from pulp and paper plants are deficient in vital nutrients like nitrogen (N) and phosphorus (P) that must be added to guarantee an efficient biological wastewater treatment (WWT) (Meyer and Edwards 2014). The WAS produced in the ASS is typically considered as waste and as such handled by incineration, which is becoming restricted due to emissions of greenhouse gases (Veluchamy and Kalamdhad 2017). Hence, the current operation of wastewater treatment in PPI results in a loss of resource (i.e. energy and nutrients) recovery potential.

In this work, a new concept is discussed whereby much of the COD in the wastewater is removed as activated sludge with minimum energy input. The sludge is then used to produce biogas and recover nutrients. This concept is based on three major processes, which have been successfully used at lab and pilot scales (Magnusson et al. 2018). Figure 1 depicts a simplified schematic diagram of the industrial symbiosis concept. Firstly, the pulp and paper mill wastewater, characterized by high concentration of soluble organic materials, is treated using high-rate anaerobic digestion by converting readily biodegradable carbon-rich feedstock into biogas in external circulation sludge bed (ECSB) reactors (Ekstrand et al. 2013). To meet the discharge requirements for final effluent disposal, post-treatment of the produced reject water is carried out in a high-rate activated sludge process (HRAS). Contrary to conventional operations, normally run for degradation of COD while minimizing the WAS production, HRAS systems operate at a higher loading with a shorter residence time and sludge age and more WAS with higher digestion characteristics is thus produced (Ge et al. 2013). Compared to current operations in the pulp and paper mill effluent treatment, the concept is robust and less resource demanding because less energy will be consumed owing to decreased demands for aeration, while at the same time substrate with improved biomethane potential is generated. Furthermore, the integrated treatment concept will result in a lower overall sludge production.

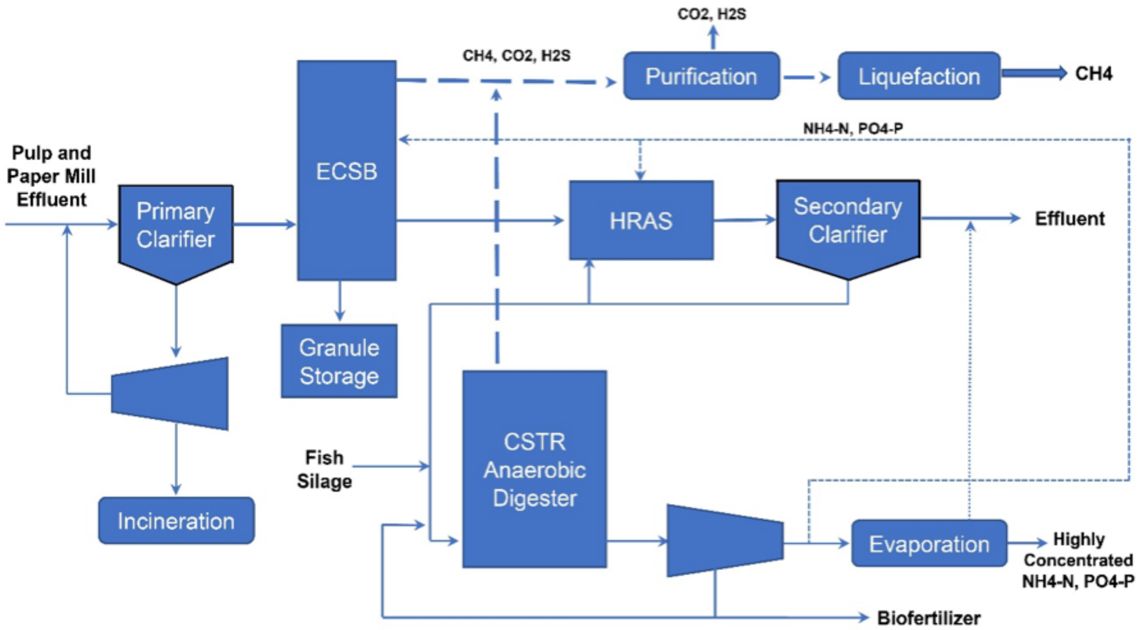

Figure 1. Simplified schematic for the industrial symbiosis between the biogas plant and the pulp and paper mill effluent treatment plant.

To augment the potential of biogas production and profitability of the biogas plant, the WAS stream is co-digested with other substrates, such as fish-waste silage and chicken waste in stirred tank reactors (CSTR). Dewatering of the digestate produces a solid cake, which can be used as bio-fertilizer/soil-improver, whereas a liquid stream rich in nitrogen and phosphorus can be concentrated by evaporation for more efficient transport. In case the biogas plant is co-located with for example a mill, the liquid digestate phase can be recirculated to the ASS thus reducing the need of external N and P (Meyer and Edwards 2014). This concept demonstrates an excellent industrial symbiosis case where all the involved parties benefit from the synergy through substantial energy savings, low costs for sludge handling, no need for untreated digestate transportation over long distances and reduced use of commercial nutrients.

1.2 Modelling opportunities

The above circular economy concept is of great interest to PPI as it gives an opportunity to enhance sustainability, increase productivity and cost-competitiveness owing to large savings in energy and commercial nutrient consumption (Stoica et al. 2009). However, the lack of a general model may hamper the efforts aimed at accelerating knowledge transfer and diffusion of technology to as many mills and wastewater treatment plants as possible.

In this respect, there is a need for the development and application of improved integrated industrial symbiosis modelling tools for the assessment of biogas processes in synergy with pulp and paper mill effluent treatment. A comprehensive mathematical model is a valuable tool for gaining insight into the dynamics in the systems due to variations in operational conditions in the involved processes. Other benefits of modelling the industrial symbiosis are improvements in design, assessment of process configuration, evaluation of operational and control strategies technology development and model-based optimization and design. Process models can be used as an evaluation and decision support tool purposed to deliver cost savings in operational expenditure.

Various industry-standard models have been developed for wastewater treatment. These models include the popular IWA activated sludge model (ASM) series for activated sludge system (Henze 2000) and IWA anaerobic digestion model no1 (ADM1) developed for describing biogas production (Batstone et al. 2002). While the ASM series were primarily developed for evaluating domestic wastewater treatment performance, few studies have used them to simulate activated sludge system treating pulp and paper mill effluents with satisfactory outcomes for the required purposes (Brault et al. 2010, Horan and Chen 1998, Lindblom et al. 2004). In addition, no integrated model exists for an industrial symbiosis centred on biogas production (anaerobic digestion and co-digestion) and high-rate activated sludge system for pulp and paper effluents and some of the processes included in the industrial symbiosis described above are not commonly fully compatible with traditional state-of-the-art process models. This is because the evaluation criteria of conventional treatment technologies are currently focused on treated wastewater quality, whereas, model requirements are considerably more complex with focus on biogas production and resource recovery technologies (Batstone et al. 2015b). The contaminants present in pulp and paper wastewater may also differ from those present in domestic wastewater. This poses significant technical risk, because current models cannot estimate treatment plant performance once emerging technologies have been integrated, and a designed facility may not achieve legislated environmental performance requirements. Modelling pulp and paper mill wastewater treatment and biogas plants driven mainly by resource recovery technologies is an essential development that requires a holistic approach, providing integrated and flexible software tools to transition linear treatment systems into circular biogas resource recovery systems.

1.3 Objectives

To address the above-stated challenges and new opportunities, the main objective of this study is to develop a mathematical process model for systematic evaluation of the proposed industrial symbiosis concept including sustainability assessments through simulations. This would advance the development and application of simulation techniques in biogas processes as well as support and encourage the transition of pulp and paper wastewater treatment towards a higher resource efficiency. The developed model can provide good insights in new projects to show the potentials for saving both electricity and nutrient additions in the WWT and predict discharge values for organic matter (COD), suspended solids (TSS), nutrients (such as N and P) etc. To demonstrate the robustness of the concept, the model can also be used to simulate different scenario analyses under extreme conditions that may cause process disturbances/failure.

2 Treatment of pulp and paper mill wastewater

2.1 Pulp and paper mill effluents

Recent strict environmental regulations in the pulp and paper industry have led to significant reduction in water consumption, particularly for new mills (Kamali et al. 2016). This has been achieved by increasing the recycle of the mill process water (whitewater) at different locations in the papermaking plant. However, the PPI is still highly water-dependent compared to other industries; it generates large quantities of wastewater that requires in-mill treatment to prevent pollution of receiving water bodies (Pokhrel and Viraraghavan 2004).

The effluent loads from PPI are highly heterogeneous depending upon the raw materials and type of the pulping process. The constituents of wood and other plant materials used to make pulp include cellulose (40-45%), hemicellulose (20-30%), lignin (20-30%), and extractives (2-5%) (Rintala and Puhakka 1994). Pulping is aimed at breaking down the bulk structure of the fibre source, such as chips, stems or other plant parts, into the component fibres. The major processes in pulp manufacturing and paper making can be split into five main groups: mechanical, chemical, chemo-mechanical, thermo-mechanical and papermaking, each generating a large volume of wastewater effluent with specific quality (Ekstrand et al. 2013). In general, the pulping and bleaching processes are the primary sources of wastewater production in pulp and paper mill, while a small amount is produced in the paper machine. These effluents contain various substances, extracted from the wood or from the recycled fibres, and some chemicals used in the process that are released from the processes and not retained in the fibres of the manufactured pulp or paper. Table 1 presents the wastewater pollution load from individual pulping and papermaking processes. The pulping and bleaching chemicals used impact significantly the quality of the wastewaters.

The characteristics of the effluents generated from the pulp and paper mills depend on several factors including the type of pulping process, technology applied, internal water recirculation and the amount of water consumed in the process. Table 2 gives the general characteristics of effluents generated in different pulp and paper processes. As typical for an industrial wastewater, the COD content is significantly high in the mill effluent, thus it is the pollutant of concern. Treatment plants are solely designed to remove the wastewater solids and organic matter via mechanical treatment (e.g. primary clarification) and aerobic treatments (e.g. activated sludge process), respectively. These treatment processes in the pulp and paper industries generate large amounts of sludge.

Table 1. Typical wastewater generation and pollution load from pulp and paper industry (Rintala and Puhakka 1994).

Process	Wastewater (m^3/adt pulp and paper)	SS (kg/adt pulp)	COD (kg/adt pulp)
Wet debarking	5-25	not reported	5-20
Groundwood pulping	10-15	not reported	15-32
TMP-unbleached	10-30	10-40	40-60
TMP-bleached	10-30	10-40	50-120
CTMP-unbleached	10-25	20-50	70-120
CTMP-bleached	10-25	20-50	100-180
NSSC	20-80	3-10	30-120
Ca-sulphite (unbleached)	80-100	20-50	not reported
Ca-sulphite (bleached)	150-180	20-60	120-180
Mg-sulphite (unbleached)	40-60	10-40	60-120
Kraft-bleached	40-60	10-20	40-60
Kraft-unbleached	60-90	10-40	100-140
Paper making	10-50	not reported	not reported
Agro-based small paper mill	200-250	50-100	1000-1100

adt – air dry ton, NSSC – neutral sulphite semi-chemicals

Table 2. Characteristics of effluents generated in different pulp and paper processes (Bajpai 2017).

					Parameters					
Process	pH	SS (mg/L)	BOD5 (mg/L)	COD (mg/L)	Carbohydrate (mg/L)	Acetic acid (mg/L)	Methanol (mg/L)	Nitrogen (mg/L)	Phosphorus (mg/L)	Sulphur (mg/L)
TMP	-	383	2800	7210	2700	235	25	12	2.3	72
CTMP	-	500	3000-4000	6000-9000	1000	1500	-	-		167
Kraft bleaching	10.1	37-74	128-184	1124-1738	-	0	40-76	-	-	-
Kraft foul	8.0	16	568	1202	-	-	421	-	-	5.9
Sulphite condensate	2.5	-	2000-4000	4000-8000	-	-	250	-	-	800-850
NSSC pulping										
Spent liquor	-	253	13,300	39,800	6210	3200	90	55	10	868
Chip wash	-	6095	12,000	20,600	3210	820	70	86	36	315
Paper mill	-	800	1600	5020	610	54	9	11	0.6	97

2.2 Primary mechanical treatment

Pulp and paper mill wastewater treatment consists of primary, secondary and tertiary treatment. The primary treatment entails neutralization (equalization), screening and settling. Suspended solids (i.e. fibres, bark particles and organic materials, such as fillers) are removed by mechanical treatments, such as sedimentation, flotation or filtration (Pokhrel and Viraraghavan 2004). Coagulants or flocculants are used to enhance the clarification and separation of the suspended solids, colloids and some dissolved matters. The main constituents of the primary sludge are wood fibres (including cellulose, hemicellulose, lignin and other components) and depending on the process possibly also process chemicals (e.g. fillers, such as kaolin and calcium carbonate), compounds produced during pulping, bark and also sand (Rintala and Puhakka 1994). The amount of fibre and ash in the solid matter of paper mill primary sludge range from 40 to 95% and from 5 to 60%, respectively (Jokela et al. 1997).

2.3 Aerobic treatment of pulp and paper mill effluents

2.3.1 Process description

After primary treatment, a wide-range of processes can be applied to remove the organic materials (BOD). These processes include activated sludge systems (i.e. activated sludge reactors, sequencing batch reactors and aerated lagoons) and biofilm systems, such as MBBR and trickling filters. The activated sludge process is the most widely used in the treatment of PPI waste effluents (Bajpai et al. 1999). In the activated sludge process, dissolved biodegradable and particulate carbonaceous organic matter in the wastewater are transformed in the presence of air by microorganisms to cell tissue (biomass), CO2 and water. Given that the pulp and paper mills effluents are deficient in phosphorus and nitrogen pollutants, the ASS is mainly designed to remove organic compounds (Lindblom et al. 2004).

The performance of activated sludge processes depends on a number of operation and design parameters – one of them is SRT or sludge age. The SRT is defined as the average time the activated solids are in the system (bioreactors and clarifier) and it has an important effect on the selection, survival and growth of microorganisms (Eddy et al. 2013). Generally, the sludge age in ASS lies between 6.5 to 12 days, however, longer sludge age of 15-20 days can be encountered in the pulp and paper industry (Thompson et al. 2001). The SRT plays a key role not only in the ASS but also in downstream processes, such as anaerobic digestion. For instance, decreasing the SRT in the ASS increases both the waste activated sludge amount and methane generation in the anaerobic digestion (Ge et al. 2013, Jimenez et al. 2015). This aspect is further elaborated upon in Section 2.3.3.

After biodegradation, biomass and water are separated, normally using sedimentation tanks. The generated sludge after clarification is referred to as waste activated sludge or bio-sludge, containing high amount of organic matters. Both primary sludge from the primary sedimentation and waste activated sludge from the activated sludge plant are thickened and dewatered, then often mixed with bark and incinerated or landfilled. This conventional waste management is associated with major environmental issues including ground- and surface water contamination, as well as greenhouse gas emissions (Bajpai 2017). However, with increased awareness of the impacts of greenhouse gases and increasing energy costs, the efficient management of waste streams is now of greater concern for the PPI and entities for more sustainable and environmentally friendly approaches.

2.3.2 Nutrient requirements for aerobic oxidation

To support optimum microbial cell synthesis and growth in an activated sludge system, appropriate nutrients, such as nitrogen, phosphorus, calcium, sulphur, copper, magnesium, and potassium, must be available in sufficient amounts. Nitrogen and phosphorus are considered macronutrients because they are required in comparatively large amounts, about 12.2 g of nitrogen and 2.3 g of phosphorus is needed per 100 g of cell biomass (Eddy et al. 2013). However, unlike domestic wastewater containing enough nutrients, effluents from pulp and paper mills are often deficient in these macronutrients, which must be added during biological wastewater treatment to enhance the biomass growth (Slade et al. 2004). Addition of an exogenous source of nutrients will impact the operating expenses of the biological wastewater treatment facility. However, this cost could potentially be offset when anaerobic digestion of sludge is included in the treatment train. Key nutrients can therefore be released and recycled to the biological treatment system (Magnusson et al. 2018).

2.3.3 High-rate activated sludge process

The treatment of pulp and paper mills effluents by the activated sludge process is cost- and energy intensive due to the oxidation of a large fraction of organic matter and subsequent management of sludge. To reduce the cost associated with sludge management, extended aeration approaches have been proposed (Mahmood and Elliott 2006). These approaches are very efficient in lowering the amount of sludge produced, however, they require long SRT and increased need for aeration. Operating an activated sludge system at higher SRT implies large basin volumes, which have cost and space implications. By the way of contrast, high-rate activated sludge (HRAS) treatment, a process modification of activated sludge, was developed, with the intent of reducing electrical energy consumption (Chase and Eddy 1944). HRAS treatment minimizes biological oxidation and takes place by capturing organic carbon in the sludge by adsorption, particulate enmeshment, bio-flocculation, accumulation and assimilation (Jimenez et al. 2005). High-rate aeration systems are highly loaded with organic matters combined with lower suspended solid concentration leading to short SRTs of 1-4 days and hydraulic retention times (HRT) of 2-4 h as well (Ge et al. 2013, Jimenez et al. 2015). As a rule, the sludge loading rate (SLR) of a conventional ASS is roughly 0.25 kgBOD kgVSS-1 d-1, whereas high-rate activated systems operate at SRT ranging from 3 to 13 kgBOD kgVSS-1 d-1 (Ge et al. 2017).

Besides lower energy requirements in comparison to conventional low-rate activated-sludge system, the HRAS systems produce increased volumes of waste activated sludge due to decreasing SRT. Benchtop and pilot-test studies have confirmed that this sludge is of high anaerobic biodegradability with high methane potential and nutrients (i.e. $NH_4^+$and PO_4^{3-}) release (Ge et al. 2016, Ge et al. 2013, Ge et al. 2017). Contrary to HRAS for domestic wastewater, where a two-stage process system may be required to meet effluent discharge regulations, only one step is needed for pulp and paper mill effluent since a nitrification/denitrification step is not required.

The HRAS process approach should be particularly attractive as the focus can no longer only be on oxidizing and removing organic pollutants but also on minimizing the energy input in the ASS while at the same time maximizing recovery of energy and nutrients contained trough anaerobic digestion (Magnusson et al. 2018).

2.3.4 Methane oxidation in activated sludge

Anaerobic sludge digestion liquor may to some extent contain dissolved methane (Hatamoto et al. 2010). For example, the loss of methane in the effluent from an anaerobic system treating domestic wastewater or low-strength wastewater has been estimated in fifteen studies to be between 11 and 100% with degree of supersaturation ranging from 1.34 to 6.9 (Crone et al. 2016). The presence of this dissolved methane represents an environmental hurdle since its emission to the atmosphere contributes to the carbon footprint of a wastewater treatment plant (Daelman et al. 2012). Dissolved methane from the digester may either be stripped or oxidized during downstream processing. It has been shown that approximately 20% of dissolved methane can be stripped in ASS while the remaining is biologically broken down to carbon dioxide and water by methanotrophs (methane-oxidizing bacteria) in activated sludge system treating domestic wastewater (Daelman et al. 2012). However, the fate of dissolved methane that could be lost in the effluent of the proposed industrial symbiosis has not been fully investigated. Due to limited aeration in the HRAS as well as lower levels dissolved oxygen in the high-rate activated sludge system, biological oxidation of dissolved methane may be expected to be prevalent over stripping.

2.4 Anaerobic digestion of wastewater and sludge

2.4.1 Process mechanism description

Anaerobic digestion treatment refers to biological degradation and conversion of carbon-rich feedstock into biogas in the absence of oxygen by a consortium of microorganisms. Digester gas contains about 55-70% methane (CH_4), 30-40% carbon dioxide (CO_2) and small amounts of other gases (N_2, H_2, H_2S, water vapour). The biogas produced can be used directly as fuel in combined heat and power gas engines, or upgraded, after drying and desulphurisation, to natural-gas quality bio-methane for energy production (Eddy et al. 2013). Compared to conventional activated sludge systems, AD offers benefits including the ability to achieve a high degree of organic matter removal at a significantly lower cost while generating a smaller amount of nutrient-rich and stabilized and more dewatered sludge, which in some cases can be used as a bio-fertilizer (Meyer and Edwards 2014).

Anaerobic digestion processes entail four fundamental steps including hydrolysis, acidogenesis (fermentation), acetogenesis and methanogenesis (Figure 2). During hydrolysis, large particulate substrates are firstly broken down to particulate carbohydrates, proteins and lipids, which are further degraded by acidogenic bacteria into monosaccharides (MS), amino acids (AA) and long-chain fatty acids (LCFA), respectively (Batstone et al. 2002). Hydrolysis is immediately followed by acidogenesis or fermentation, which is an acid-forming step. In this step, the organic monomers are used as substrates by acid-forming bacteria, which mainly transform them into higher organic acids or volatile fatty acids (VFAs), such as acetate, propionate acid, butyrate; alcohols like methanol and ethanol; and gases, such as CO2 and H2. Organic substrates serve as both electron donors and acceptors. The propionate and butyrate are subsequently converted by acetogenic microorganisms to H2, CO2 and acetate. Acetate and H2, generated during both acidification and acetogenic reactions, serve as substrates for methanogens. Methanogenesis, the last step of anaerobic digestion, is carried out by two groups of microorganisms collectively known as methanogens (Bajpai 2017). The acetolactic methanogens, split acetate into methane and carbon dioxide. The second group, termed hydrogen utilizing methanogens, use hydrogen as the electron donor and CO2 as the electron acceptor. The free energy change associated with the conversion of propionate and butyrate to acetate and hydrogen requires hydrogen to be at low concentration in the system or the reaction will not proceed (Mosey 1983).

The application of anaerobic digestion technology is increasing worldwide but the number of pulp and paper mills applying AD is still low. One of the leading suppliers of high-rate AD-systems is reporting a total of 278 plants installed, 204 of these at recycled paper mills (Driessen et al. 2007). The reason for the still low degree of implementation is likely resistance to change a working wastewater treatment even if it would mean improved resource efficiency and lower carbon footprints. The success of applying anaerobic digestion processes also depend on the chosen waste streams and on anaerobic digestion process knowhow.

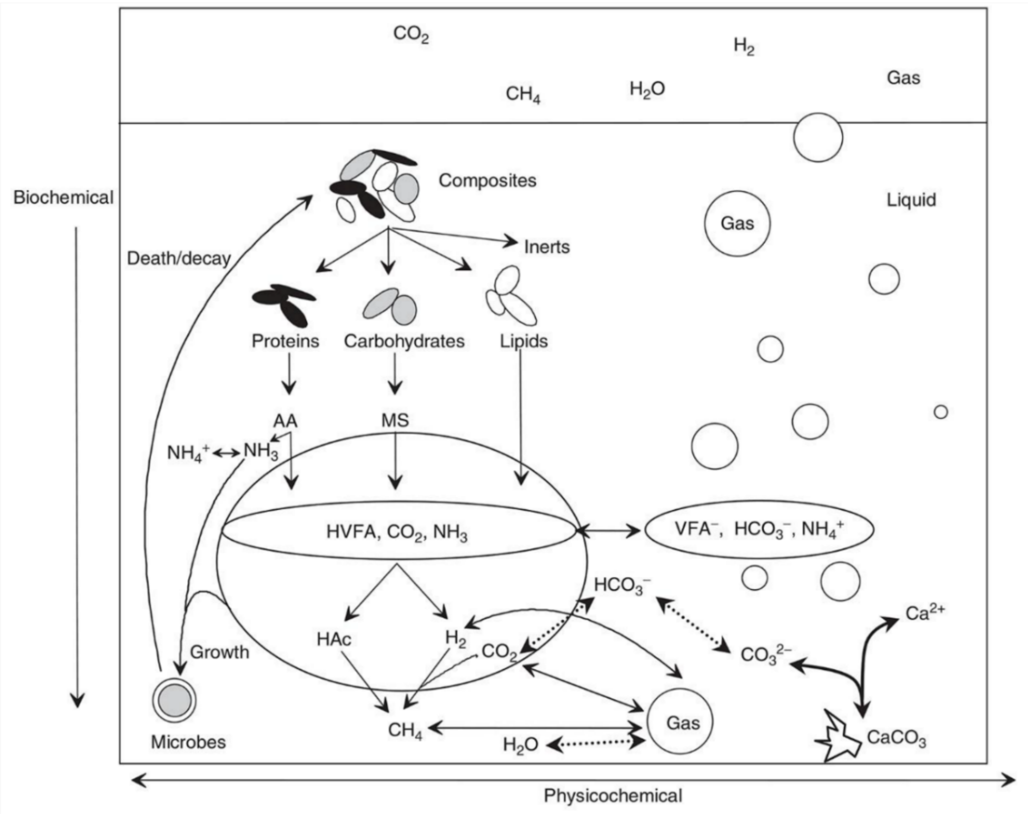

Figure 2. Biochemical (vertical) and physico-chemical processes (horizontal) in an anaerobic digester. AA, amino acids; MS, monosaccharides; HVFA, associated organic acids; VFA-, dissociated organic acids; HAc, acetic acid; Ac-, acetate. Adapted from Batstone and Jensen (2011).

2.4.2 Nutrients release

Besides biogas production, some nutrients of interest, such as nitrogen (N) and phosphorous (P), are released in the digestate, typically as ammonium and phosphate, respectively. The quality of a digestate depends largely on the digestion processes and the characteristics of substrates. After dewatering, the solid digestate contains organics with more recalcitrant structures (i.e. humic substances) and a high proportion of mineral nitrogen, which is available for plant uptake and makes it suitable as biofertilizer (Elliott and Mahmood 2012). On the other hand, the reject water contains dissolved macro- and micro-nutrients (Meyer and Edwards 2014). As indicated above, the recirculation of the reject water to the head of the pulp and paper mill activated sludge system can play a significant role in supplying these nutrients that are essential for growth of heterotrophic microorganisms. This is an added benefit of processing waste activated sludge anaerobically. Therefore, generation of biogas from WAS and other organic substrates through anaerobic digestion creates the opportunity to recirculate nutrients to the AS plant while at the same time humic substances and other recalcitrant structures in the digestate from the anaerobic digestion may be suitable for soil improvement for agricultural use (Hagman and Eklund 2016).

2.4.3 Operating conditions

Anaerobic digestion is affected by a wide range of environmental and operational parameters including temperature, system pH, volatile fatty acid content and conversion, availability of micro and trace nutrient, toxicity, SRT, HRT and volatile solids loading (Bajpai et al. 1999). The pH of an aerobic digester is an important environmental factor for maximum activity of both acidogenenic and methanogenenic microorganisms, which thrive within the pH range of 5.5-6.5 and 7.8-8.2, respectively. However, the operating pH of both cultures is between 6.8 to 7.4 with pH 7 being the optimum (Bajpai 2017). Since the methanogenesis is the rate limiting step for biogas production, it is crucial to hold a pH roughly between 7 and 8 for good methanogenic activity. Low pH is detrimental to the activity of the methanogens causing accumulation of volatiles fatty acid and hydrogen. Introduction of toxic substances can result in process imbalance and accumulation of fermentation products. At higher pH (>8), the metabolisms of methanogenic microbes may be limited owing to the potential formation of minerals from key macro nutrients or trace metals.

The performance of an anaerobic digestion process can also be affected by other important factors including the characteristics of the wastewater. Nutrients, such as nitrogen, phosphorus and sulphur, are required to enhance the bacterial activities (Takashima and Speece 1989). Thus approximatively 10-13 mg/L of nitrogen, 2-2.6 mg/L of phosphorus and 1-2 mg/L of sulphur are desirable by 100 mg of biomass. Furthermore, 50, 10 and 5 mg/L of nitrogen, phosphorus and sulphur, respectively, are required for maximum activity of the methanogenic community (Eddy et al. 2013). Since anaerobic processes releases nitrogen and phosphorus, the addition of macronutrients may or may not be needed depending of the characteristics of the substrates and the SRT of the digesters, and also the needs for trace elements such as Co, Ni and Mo are related to their concentrations in the substrate but also other factors, a main one being the organic loading rate.

On the other hand, higher concentrations of macronutrients, particularly sulphur, may impede methanogenic activities, causing upset or failure of anaerobic reactors (Eddy et al. 2013). In general, effluents from acid sulphite, neutral sulphite semi-chemical, kraft, chemi-mechanical and chemi-thermomechanical pulp mills contain significant amount of oxidized sulphur compounds, such as sulphate, sulphite and thiosulphate (Bajpai et al. 1999). These compounds undergo reduction to sulphide by sulphate reducing bacteria (SRB), which consume organic material in the anaerobic digester and generate hydrogen sulphide (H_2S) and carbon dioxide as end products (Feldman et al. 2017). Competition with SRB for substrate in the form of COD including hydrogen, acetate and methanol, may reduce the quantity of methane generated in anaerobic digesters, while at the same time the presence of sulphide gases degrades the quality of the biogas. Low amount of sulphide (less than 20 mg/L) are needed for optimum methanogenic activity, however, higher concentrations ranging between 50 to 250 mg/L can be toxic and reduce by half or more their activity (Eddy et al. 2013). Besides having an inhibitory effect, hydrogen sulphide can cause operational issues since it is malodorous and corrosive to metals.

2.4.4 High-rate anaerobic digestion of pulp and paper mill effluent

Recent advancements in AD technologies, particularly high-rate anaerobic reactors, have led to an increased application in the pulp and paper mill wastewater treatment (Larsson et al. 2014). The main anaerobic high-rate reactor types include the upflow anaerobic sludge blanket (UASB), anaerobic filters (AFs) and anaerobic membrane bioreactors (AnMBRs) (Bajpai et al. 1999). The first installation of an UASB-reactor at a pulp and paper mill was according to data from Paques done 1983 (Habets and Knelissen 1985). The number of UASB-units has since then increased and are today about a 100 while other types of high rate reactors are today more popular (EGSB from Veolia and IC and ICX from Paques; see below for details). Today a total of about 400 anaerobic reactors treating wastewater from the pulp and paper industry are reported (Driessen et al. 2007). The last years about 30 installations per year have been performed.

Error! Reference source not found. shows an illustration of a UASB reactor, which is the base model from which many of the current high-rate systems have developed. The feed is distributed at the base of the reactor, then percolates through a naturally forming granular microbial sludge blanket, which is partially fluidized and mixed by the upward motion of released gas bubbles (e.g. CH4 and CO2). The separation of the three phases (solid–liquid–gas) takes place in a settler located at the top of the reactor.

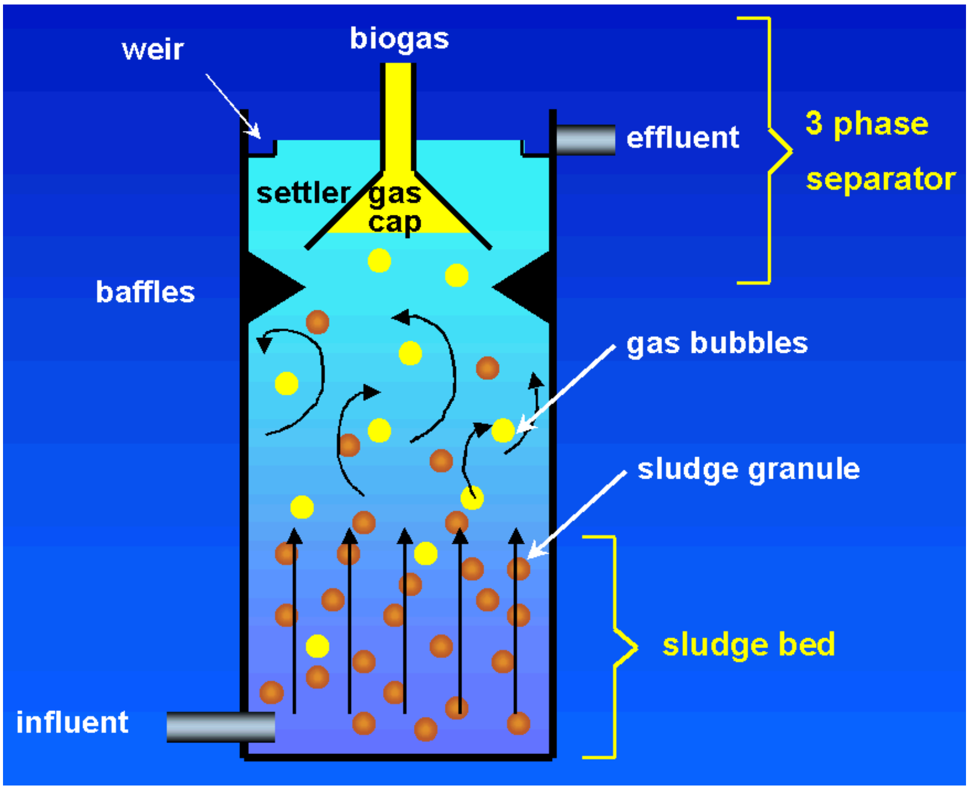

Figure 3. Schematic of an up-flow anaerobic sludge bed (UASB) reactor (http://www.uasb.org).

The variants of UASB technology include expanded granular sludge blanket (EGSB) and, more recently, internal circulation (IC) reactors and external circulation sludge bed (ECSB) reactors as mentioned above. The benefits of the UASB concept and the designs that has evolved from it (including the ICs, EGSB and ECSB) is its phase-separation resulting in two different retention times; one hydraulic (HRT) for the wastewater and one solid (SRT) for the active microflora. This means that the biological process can be run at HRTs as shorts as 2-4 hours without risking a wash-out of the microflora that typically need retention times of 10-15 day as related to their growth rate. A prerequisite for the short HRT is that the COD of the wastewater is easily degradable. To run a high-rate process wastewater with easily digestible, soluble COD concentrations of at least 1500 mg/L at 100% COD-reduction are typically recommended. The low HRT means that large volumes of wastewater can be treated per time unit and thus that the size of the reactors can be reduced. Loading rates in these systems are typically 15-25 kg COD $m^{-3}d^{-1}$ and the methane production will depend on the COD-reduction (350 L/kg of COD reduced). The drawback with the high-rate systems is that they do not tolerated suspended solids levels over 400 mg/L as it will have an undesirable impact on the settleability of the granulated sludge.

2.4.5 Anaerobic digestion of sludge

Anaerobic digestion of sludge is used conventionally for stabilising and reducing the quantity of mixed waste sludge in domestic water treatment and is normally performed in completely stirred tank reactors (CSTR) (Eddy et al. 2013). There is now a growing interest in applying this technology to primary and waste activated sludge from pulp and paper mill wastewater treatment. Both these sludges present various characteristics depending on the raw material used, pulp and paper manufacturing process and downstream wastewater treatment. Primary sludge mainly consists of wood components, papermaking fillers (i.e. kaolin, calcium carbonate ($CaCO_3$)), pitch (wood resin), lignin by-product, inert solids rejected during chemical recovery process and ash (Veluchamy and Kalamdhad 2017). Anaerobic digestion of primary sludge from the pulp and paper industry is very rare and the outcome will depend on the type of pulping process applied: fibres from mechanical pulping is hard to digest due to their high content of lignin while fibres from Kraft and NSSC pulping have been shown more easily digested (Åhrman 2012). The study by Åhrman (2012) showed higher methane potentials for Kraft and NSSC pulp fibres (300-400 m^3/t VS) than for CTMP (70 and 240 m^3/t VS for soft- and hardwood fibres, respectively) and 20 m^3/t VS for TMP fibres (Åhrman 2012). Another study showed methane potentials of Kraft pulp fibre sludge, resulting mainly from degradation of cellulose and hemicellulose, ranging between 190 to 240 m^3 CH_4/t VS, (Bayr and Rintala 2012).

The anaerobic digestion of WAS from pulp and paper mills effluent can result in the turnover of about 21 to 55% of the organic material, while specific methane yields range between 40-200 mL g^{-1} VS (Meyer and Edwards 2014). The biodegradability of the WAS is likely mainly related to the regime of the activated sludge treatment with sludge age as the key parameter (Ge et al. 2013, Magnusson et al. 2018).

Reduced methane potential of WAS is due to its composition. The biodegradable fraction of WAS consists mainly of active bacterial cells in which the biodegradable fraction is below 70%. WAS also contains refractory portions of decayed cells and influent organics. Some of the wood constituents, such as hydrolysed lignin, tannins and resin acids in the activated sludge, also reduce the degradability (Meyer and Edwards 2014). Proper analysis and treatability studies, such as sludge pre-treatment or co-digestion, may be needed to enhance the sludge digestibility and enhance biogas production. The biodegradability of WAS also largely depends on the activated sludge process performance. As indicated above decreasing the sludge age of an ASS can considerably increase sludge-solids production. Such sludge also has higher anaerobic digestibility, which can lead to higher methane production (Ge et al. 2017, Jimenez et al. 2015, Karlsson et al. 2011).

2.4.6 Anaerobic co-digestion of pulp and paper industry sludge with organic wastes

Co-digestion (AcoD) refers to an anaerobic digestion process in which two or more organic substrates are combined and simultaneously digested to enhance biogas production. Given low biodegradability of PS and WAS, AcoD is an attractive means for increasing the methane production and energy available for the WWTP or externally, as for example in gas-powered vehicles. Some studies on co-digestion of PS and WAS from the pulp and paper industry have been performed. Ekstrand et al. (2016) report a methane yield of 230 L/kg VS when co-digesting Kraft fibre sludge with WAS in a CSTR-process with sludge recirculation. The WAS addition was reported to have a stabilising effect on the process. Bayr and Rintala (2012) report as well on co-digestion of Kraft fibres with WAS. In their study methane yields of 150–170 m^3CH_4/tVS was obtained at an organic loading rate (OLR) of 1 kgVS/m^3d. HRTs ranged between 25–31 d.

Mathematical models can be a powerful tool to use for co-substrate selection and determination of optimum loading rates (Xie et al. 2016).

Besides higher methane yields, anaerobic co-digestion offers advantages including a possible improved balance of nutrients (Ekstrand et al. 2016), alleviation of inhibitory effects due to toxic substances via dilution and an enhanced methane production (Hagos et al. 2017, Neshat et al. 2017). Substrate properties and composition are key governing factors for AcoD processes. Difficult-to-digest sludge from pulp and paper mills may be digested together with more easily digestible waste from outside the mill.

3 Mathematical modelling of pulp and paper mill effluent treatment

3.1 Introduction

In recent years, there has been an interest in developing mathematical processes in the field of pulp and paper wastewater treatment. Models have been successfully used as an important engineering tools in research, process design, training, control and optimization of physical, chemical and biological processes. However, to date most models proposed in the literature have been applied on activated sludge systems treating pulp and paper effluent (Horan and Chen 1998, Lindblom et al. 2004). By contrast, modelling work for anaerobic digestion is rare. This is despite that anaerobic digestion is increasingly being applied to treat pulp and paper mill effluents. Therefore, a reliable dynamic and integrative modelling tool is required for designing, simulating and optimizing of symbiotic concepts for supporting effective decision-making as far as biogas production and the function of wastewater (including both the high-rate biogas process and the activated sludge process) is concerned.

The application of plant-wide models is of paramount importance in gaining more insight in the process dynamics of the interactions of the different processes involved in the proposed industrial symbiosis (Section 1). Mathematical modelling platforms can enable in-depth analysis of the main processes, such as high-rate anaerobic digestion, activated sludge treatment at different loads and sludge ages and co-digestion processes.

3.2 Activated sludge models

3.2.1 General overview

Several mathematical models for activated sludge processes have been proposed over the years to describe the conversion and removal of organic carbon, nitrogen (N) and phosphorus (P) during wastewater treatment (Barker and Dold 1997, Gernaey et al. 2004). The activated sludge models (ASM) developed by the International Water Association (IWA) are considered the state-of-the-art for activated sludge processes, and they are implemented in many commercial software packages and have also been the basis for further modelling research on activated sludge treatment (Henze 2000). The series include:

- ASM1 describes the removal of organic carbon (COD) and nitrogen;
- ASM2 describes the removal of organic carbon, nitrogen and phosphorus;
- ASM2d extends ASM2 with chemical phosphorus removal with Al or Fe;
- ASM3 describes the removal of organic carbon and nitrogen, including the storage phenomenon.

The emphasis of the ASM series was on process modelling of activated-sludge biology, which can include the removal of organic carbon compounds, nitrogen and phosphorus, depending on the specific plant design and configuration. The original ASM series only contained limited targeted physico-chemistry (Batstone et al. 2012). For example, the activated sludge model (ASM) series does not model pH or chemical species in the aqueous system, but rather tracks a semi-empirical alkalinity state as an indicator of process resistance to intolerable pH swings (Batstone et al. 2012). One of the limitations of the ASM alkalinity state is that it does not allow modelling of pH effects on minerals precipitation and gas transfer processes.

A few modelling and calibration studies have been carried out to improve understanding of the process or to predict the behaviour of the system for pulp and paper activated sludge treatment plants. Given that the pulp and paper effluents are deficient in nitrogen and phosphorus, nitrification/denitrification and phosphorus removal are not expected to occur, thus simple models are suitable. In this respect, ASM1 has typically been used to describe the activated sludge process for pulp and paper mill effluents and has been used without any modification to study optimization strategies for COD effluent quality and oxygen requirements (Horan and Chen 1998). Quite often ASM1 is slightly modified to describe additional processes, such as nitrogen limitation to heterotrophic growth, hydrolysis of soluble slowly biodegradable compounds and nutrient transformation (Brault et al. 2010, Lindblom et al. 2004). Good predictions were obtained as well for a chemi-thermomechanical pulp effluent treatment using ASM3 (Baranao and Hall 2004). However, none of the models included physico-chemical reactions and could not predict the influence of pH on the activated sludge process. As far calibration is concerned, model parameters that affect biological degradation were adjusted as much as possible to measured steady-state data. Parameters such as heterotrophic yield coefficient, fraction of biomass yielding particulate products, N content in inert particulate matter, P content in inert particulate matter and soluble nitrogen inert fraction in the influent are influential for COD degradation and have been the focus for calibration (Brault et al. 2010, Keskitalo et al. 2010, Lindblom et al. 2004).

3.2.2 Limitation of activated sludge models

The lack of key physicochemical reactions in the original ASM has now been addressed. This development was important because ion activity and ion pairing corrections are needed to enhance plant-wide model predictions (Flores-Alsina et al. 2015, Solon et al. 2015). Thus, a mechanistic model for activated sludge systems treating effluents from pulp and paper mills should necessarily consider all relevant biological processes to mimic the complex physico-chemical and biological transformation interactions that may affect the treatment process performance. While the existing models (e.g. ASM1 or ASM2) have the capability to describe the dynamic microbial interactions of various processes, further model development and upgrade may still be required for application in PPI.

One area that needs further development in the activated sludge model is the inclusion of oxidation of methane by methanotrophs (Arcangeli and Arvin 1999, Daelman et al. 2012). This is particularly important when anaerobic treatment is located upstream of an activated sludge system, thus the residual methane in the AD effluent will either be oxidized and/or stripped depending on the process design and operation. The presence of high levels of dissolved methane in this stream represents a challenge that needs to be thoroughly investigated and modelled in a system-wide context.

Modelling high-rate activated sludge processes is another area that requires attention. This is because the ASM models were developed to simulate the activated sludge treatment with SRT greater than 3 days. Apart from the low SRT high-rate activated sludge systems are characterized by high organic loadings where biological flocculation, adsorption of particulate and colloidal (slowly biodegradable) substrate and storage may become limiting (Haider et al. 2003). The HRAS model of high-rate activated sludge processes includes mechanisms for flocculation of particulate and colloidal substrate, hydrolysis and production of extracellular polymeric substances (EPS) (Nogaj et al. 2013). This model is based on ASM1 and describes the production of storage polymers (using a parallel storage and growth description) and the flocculation of inert biodegradable colloidal substrate into particulate. The HRAS model of Nogaj et al. (2013) is a good candidate for modelling HRAS process in pulp and paper mill effluents. However, inherent complexity of the model of Nogaj et al. (2013) such as a larger number of parameters may limit the application in PPME plant-wide framework.

3.3 Anaerobic digestion models

3.3.1 Biochemical models

A variety of simple steady-state and complex dynamic models have been developed to simulate and assess anaerobic processes (Andrews 1969, Andrews and Graef 1970, Bagley and Brodkorb 1999, Batstone et al. 2002, Mosey 1983). Simple models often fail to predict pH inhibition, product inhibition and VFA accumulation that may lead to failure of the anaerobic digesters (Costello et al. 1991). The anaerobic digestion model proposed by the IWA (ADM1) is probably the most widely accepted and applied for the simulation of anaerobic processes (Batstone et al. 2015a). ADM1 summarises the key biological steps of anaerobic digestion into the grouped reactions of disintegration and hydrolysis, acidogenesis, acetogenesis and methanogenesis, and does represent some physico-chemistry of simple acid-base reactions and liquid-gas transfer (Batstone et al. 2002). The original ADM1 was not able to model the biology nor the physico-chemistry of phosphorus, which is important as its release affects pH and minerals precipitation, that in turn influence the activated sludge system indirectly via the recycling of reject water (Batstone et al. 2006). Recent efforts have been directed at extending biological processes and physicochemical approaches in ADM1 to model more complex systems, with non-ideality corrections for ion pairing and ion activity (Flores-Alsina et al. 2016, Solon et al. 2015). Corrections of activity coefficients and ion pairing appear to be essential for accurate predictions of digester pH in many cases and are required for precipitation processes (Solon et al. 2015).

However, although ADM1 is comprehensively detailed with respect to the biology of anaerobic digestion, it has limitations for application to plant-wide models of wastewater treatment. ADM1 contains different set of state variables compared to the activated sludge model series, thus an interface is often used to translate the variables from one model to another (Nopens et al. 2009).

3.3.2 Modelling co-digestion

The biochemical models described above, particularly ADM1, are often used to describe anaerobic co-digestion in the presence of multiple substrates (Xie et al. 2016). The main modelling approach of anaerobic co-digestion consists of describing operating conditions, such as co-substrates characteristics and ratio, OLRs, HRT, for the key state variables (i.e. methane generation, VS reduction) (Xie et al. 2016). The key feature of AcoD models is the ability to predict maximum energy production and cost efficiency while at the same time avoiding organic overloading by defining the optimal ratio and organic loading of substrates and co-substrates under different process conditions (Aichinger et al. 2015, Zhou et al. 2012). Furthermore, AcoD models can either be used to predict or avoid instability due to e.g. pH changes during operation, particularly when the characterization of the substrates is inadequate. These models are suitable for assessing the impact selection and proportion of co-substrates on the digestion process and feedback effect on the entire treatment (Arnell et al. 2016, Keucken et al. 2018).

While underlying principles of anaerobic co-digestion are well established in ancillary industries, a comprehensive modelling of co-digestion of sludge from pulp and paper mill wastewaters needs to be developed to describe aspects particular to PPI. A model including the competition between sulphate reducing bacteria and methanogens, sulphur cycling and interaction with e.g. Fe and P cycles can be a useful tool in predicting the biogas quality, ammonia nitrogen and economic viability of co-digestion under organic overloading or other inhibitory conditions. Additionally, when the AcoD model is integrated within a plant-wide context, the model should be able to assess not only the impact of co-substrates on the co-digester stability but also the concentration of nutrients in the reject water, which may be beneficial to the activated sludge system given the lack of these nutrients in pulp and paper mill effluent.

3.3.3 Modelling high-rate anaerobic reactors

The possibility to reduce energy input and enhance biogas production from pulp and paper mill wastewater COD is driving much interest in modelling high-rate anaerobic reactors, such as ECSB. Three-phases are present in these reactors: (1) solid phase consisting of bioparticles (granules) composed by active and non-active biomass, (2) liquid phase is composed of the chemical species in aqueous solution (substrates, products, enzymes, ions and water) and (active and non-active) single suspended cells, which are assumed to behave as solutes, (3) gas phase that contains the gaseous products from the anaerobic degradation. Hence, modelling high-rate granular anaerobic processes entails combining the dynamics of these three phases (gas–solid–liquid) present in the reactor, which include biochemical, physico-chemical (i.e. mass transfers between the different compartments and phases) and hydrodynamic processes (Bolle et al. 1986, Fuentes et al. 2011). The flow rate of biomass is often modelled independently of the flow rate of the bulk liquid phase (Costello et al. 1991). Most of these studies have used simplistic models of the biochemical processes while others have included detailed biokinetic conversion models, such as ADM1 (Bolle et al. 1986).

In a recent study, a comprehensive model for an industrial anaerobic granular process was developed using a multi-scale approach (Feldman et al. 2017). In the model a multi-scale modelling approach, which accounts for the reactor hydrodynamics, granule growth and distribution and competition for substrate/space between the SRBs and methanogens taking place inside the biofilm, was used. The high-rate conditions within the reactor were simulated using hydrodynamic model consisting of a series of CSTRs followed by an ideal total suspended solids separation unit (Feldman et al. 2018). This model was validated against data from a full-scale anaerobic granular sludge IC reactor and the simulation demonstrated satisfactory results for organic carbon mineralization, biogas production, nutrients release and precipitation of minerals in the bulk of the bioreactor. While the model of Feldman et al. (2017) solves some of the limitations of previous models, it has some computational requirements due to a large number of hydrodynamic and biokinetic parameters to be calibrated. Hence a reduction in complexity may be required, particularly when the model is integrated within a system-wide model. This model has also numeral issues for dynamic simulations.

3.4 Physico-chemical models

3.4.1 pH and ion speciation/pairing

The pH of a wastewater influences chemical species distribution of weak acid-base systems, such as ammonia, carbonate, phosphate, sulphide, sulphate and volatile fatty acids. These aqueous systems in turn dictate, indirectly via pH or through direct inhibition, the rate of many biological processes such as nitrification/denitrification, uptake/release of phosphorus and anaerobic digestion. Furthermore, the weak acid-base system also impacts chemical precipitation, spontaneous minerals precipitation and stripping of carbon dioxide and ammonia (Batstone et al. 2012). Therefore, pH and ion speciation are crucial for modelling wastewater treatment in a plant-wide context (Batstone et al. 2015a). There has been extensive research in improving the physico-chemical models of wastewater treatment, particularly regarding improved pH prediction. A general aqueous phase chemistry model is essential for describing pH variations and ion speciation/pairing in both ASM and ADM (Solon et al., 2015; Flores-Alsina et al., 2015). The models correct for ionic strength via the Davies' approach to consider chemical activities instead of molar concentrations, performing all the calculations under non-ideal conditions. Ideal behaviour is observed in aqueous solution in which there is no interaction among chemical constituents. This situation can be seen only in very dilute aqueous solutions. As the concentrations of constituents in the water increase in systems such as anaerobic digestion, corrections are required due to deviations from ideality (Solon et al. 2015). The ionic strength determines the content of charged soluble ingredients (anions and cations) in a wastewater and their influence on wastewater chemistry. Ionic strength is calculated as follows.

$$I = \frac{1}{2}\sum_{i=1}^{n} z_i^2 S_i \tag{1}$$

where S_i = concentration of ion species i, n = number of ion species present in the solution and z_i is the charge of the ionic species. The extent of correction for non-ideal behaviour depends on the ionic strength of the wastewater, with increasing corrections required for a higher ionic strength.

Table 3 summarizes typical ionic strengths for various general wastewater types.

The extent to which the chemistry of a wastewater has deviated from ideal infinitely dilute conditions is quantified by using chemical activities instead of molar concentrations for soluble wastewater ingredients. High-strength conditions are expected in the digesters treating co-substrates, such as waste activated sludge and fish silage, thus a robust pH or physico-chemical pH model should consider ion pairing, correction for non-ideal conditions as well as the impact of chemical precipitation.

Table 3. Ionic strength of various types of wastewater (Batstone et al. 2012).

Wastewater type	**Total dissolved salts (mg/L)**	**Ionic strength (*I*, mole/L)**
Drinking water, clean natural fresh water	30-300	<0.001
Weak industrial All domestic	300-800	<0.1
Saline water, anaerobic digesters	5,000-10,000	<1
Strong industrial, landfill leachate, RO brine	10,000-70,000	<5

3.4.2 Mineral precipitation

Due to high content of cations (e.g. Ca^{2+}) and anions (e.g. SO_4^{2-}) in the pulp and paper mill wastewater, release of nutrients (NH_4^+ and PO_4^+) and chemical addition (e.g. $FeCl_3$) in anaerobic digestion, minerals such as calcium carbonate, calcium phosphate and iron sulphide is expected to form and influence the pH of the anaerobic digestion. Therefore, a mechanistic model describing the precipitation and dissolution of multiple minerals is of paramount importance when modelling treatment processes for high-strength wastewater (Flores-Alsina et al. 2015, Solon et al. 2015).

Various models have been proposed for modelling precipitation reactions in wastewater treatment. The key feature in these models is that precipitation equations are described as a reversible process using equilibrium-constraints for aqueous-phase reactions and a kinetic description for precipitation of minerals with dependency on thermodynamic supersaturation as chemical driving force (Kazadi Mbamba et al. 2015, Lizarralde et al. 2015, Musvoto et al. 2000). These models have been used in current industry-process simulation models (such as ASM2d and ADM1) to predict a wide range of precipitation situations including inadvertent scaling, nutrient recovery or sulphide removal via metal precipitation from digesters (Flores-Alsina et al. 2016, Kazadi Mbamba et al. 2016). When chemical phosphorus removal is required in the ASS, the chemistry of phosphate precipitation with calcium, aluminium or iron metal salts could be readily added using a simple modelling approach or step-wise approaches, which involve for example the formation of hydrous ferric oxide, flocculation and adsorption as well as co-precipitation (Kazadi Mbamba et al., 2019).

Chemical precipitation modelling is of great importance to reliably describe major physico-chemical transformations in standard models in plant-wide platforms. Given the high concentrations of divalent and trivalent ions in pulp and paper mill effluents, minerals need to feature in realistic system-wide descriptions when anaerobic digestion is one of the key processes. Chemical precipitation modelling is also important in describing the release of nutrients during anaerobic digestion and their feedback effect on the activated sludge system. In such cases, a combined kinetic-equilibrium precipitation model integrated within a plant-wide system can be a useful modelling tool in developing operational and control strategies for nutrients recycling and biogas production.

3.4.3 Temperature modelling in ASS

An activated sludge system for pulp and paper influent operates at much higher temperature in comparison to domestic wastewater. Higher temperatures are a result of upstream operation in the pulp and paper mill. In general, variations of temperature in a wastewater system have a considerable impact on various physico-chemical and biological processes, owing to changes in equilibrium constants and kinetic parameters. The Arrhenius model is commonly applied to correct kinetic rate parameters in a model.

$$K_T = K_{T_{ref}}\ \theta^{(T - T_{ref})} \tag{2}$$

where K_T, $K_{T_{ref}}$, θ, T and T_{ref} are the substrate utilization coefficient at temperature T, the substrate utilization coefficient at reference temperature T_{ref}, temperature coefficient and the modelled system temperature, respectively.

Existing mathematical models of activated sludge focus primarily on the biological transformations in municipal wastewater and often assume a temperature ranging from 10 to 25 °C (Henze 2000). Hence, the validity of the Arrhenius equation for rate coefficients for modelling treatment of pulp and paper effluents with temperature above 25 °C should be investigated.

3.5 Plant-wide models

In the past, wastewater processes were often modelled as separate unit processes, operated individually (Batstone et al. 2002). Integrated process modelling has received little attention, probably due to computational requirements. However, the focus in modelling of wastewater treatment has recently shifted towards plant-wide systems, where the input/output of one unit is the input/output of another unit downstream or upstream. Plant-wide models connect unit processes and operations, which interact dynamically via different major links of a wastewater treatment plant (Jeppsson et al. 2007, Nopens et al. 2009). Integration of the different processes in the water and sludge lines provides the opportunity to study the effects of process dynamics and transformations for increased process performance. However, a key area of uncertainty of plant-wide applications has been the integration of activated sludge and anaerobic digestion models. The activated sludge model (ASM) series and anaerobic digestion models (ADM1) have not been developed in a plant-wide context and, importantly, have not had a common state variable set. For this reason, the ASM and ADM1 models have not integrated seamlessly in plant-wide models and have typically required state conversion transformers.

In general, two main approaches have been used to develop plant-wide models. In the first approach individual models have been linked via transformers or interfaces that convert the states of one model into the corresponding states of another model (Jeppsson et al. 2006, Nopens et al. 2009). This approach is relatively flexible, allows connection of existing standard models (ASM and ADM1), and reduces computational redundancies compared to supermodels. The BSM2 uses such interfaces (Gernaey et al. 2004). As an alternative to the interfacing approach is using a comprehensive model or a "supermodel" to describe all the state variables and process expressions across an entire wastewater treatment plant (Barat et al. 2013, Seco et al. 2004). The main advantage being to avoid losing information when mapping one model's output to another model's input. Thus, the supermodels are mostly used in the commercial software of the wastewater industry. However, the supermodel approach is less flexible than the alternative (interfaces) and is not easily expanded or contracted when new state variables or components need to be added or removed.

3.6 Commercial simulators in the wastewater field

A wide range of commercially available software packages for modelling and simulation of wastewater treatment plants have been previously developed based on the ASM and ADM1 series (or variants thereof) and are now readily applied in wastewater treatment. Key examples of popular simulators include BIOWIN, WEST, GPS-X, SUMO and SIMBA. These packages are used in almost all sectors, such as consulting, research, utilities and academia. The simulators incorporate most treatment units and technologies as well as a wide range of models i.e. activated sludge, biofilm, anaerobic digestion, clarification and other specialized models, such as aeration and energy use (Melcer 2003). Each treatment unit is described with a reactive or non-reactive mathematical model. A user can configure a process treatment train by connecting different operation/process unit and specifying the operational and loading conditions as to mimic a specific plant performance.

As far as plant-wide modelling is concerned, most of the wastewater simulators do not advocate the use of interfaces that are used to connect ASM series and ADM1. Instead, a consistent set of state variables, stoichiometry and process rate equations is maintained throughout an integrated model.

Computer-based process simulation is a useful tool for modelling wastewater; however, care must be taken when using a simulator. Firstly, the software packages are not open since they have predefined models included and an end-user cannot write or use a specific model if the existing models within the software do not meet his or her modelling requirements. Another drawback is that some of these simulators do not keep pace with academic research developments. For example, reactor configurations such as high-rate anerobic reactors (e.g ECSB) are not included.

Another drwaback is the inclusion of physico-chemical reactions. While the Benchmark Simulation Model no2 (BSM2) has been extensively upgraded with recent modelling developments using a robust physico-chemical framework (Flores-Alsina et al. 2016, Solon et al. 2017), some of the commercial simulators are still using oversimplified approaches, which have failed to predict pH and mineral precipitation under certain conditions (Batstone 2009). The lack of improved physico-chemical models (e.g. activity correction and ion pairing) may restrict the application of these simulators when modelling pH and mineral precipitation for the proposed industrial symbiosis. To avoid overestimation of pH and chemical precipitation, further improvements on physico-chemical aspects, such as ion pairing corrections, are required before using these commercial simulators for cases such as industrial symbiosis between biogas production and pulp and paper wastewater treatment.

3.7 Model calibration

To ensure that the mathematical model of the proposed industrial symbiosis provides reliable outputs as required, it is necessary to adapt or develop systematic model calibration approaches to ensure that models receive reliable input. Wastewater characterization is one of the critical steps in calibrating a model and is essential for predicting performance. Existing characterization methodologies developed for domestic wastewater may require modifications to meet the characteristics of pulp and paper mill effluents.

Whilst plant-wide modelling of wastewater treatment has gained tremendous momentum in recent years (Gernaey et al. 2004), this has not been the case for plant-wide model calibration particularly for pulp and paper mill effluent treatment models. Over the years, several specific calibration methods have been developed for individual activated sludge (Henze 2000) or anaerobic digestion models (Batstone et al. 2002), however, none exists for plant-wide models wherein the sub-models interact in a complex manner. For example, existing calibration methodologies focus on adjusting stoichiometric and kinetic parameters within parts of the whole wastewater treatment plant (Melcer 2003). Such approaches often work well for specific targeted objectives affiliated with specific parts of the whole wastewater treatment plant, e.g. enhancing mainline wastewater treatment to meet minimum effluent quality requirements. Nevertheless, the field of industrial wastewater is rapidly expanding, and the shortcomings of such narrow calibration approaches are becoming increasingly apparent. For instance, in a plant-wide model one part of the wastewater treatment plant (represented by one sub-model) can be very sensitive to changes and calibration of model parameters in another part of the plant (represented by another sub-model). A specific example is the sensitivity of anaerobic digestion model performance to changes in the sludge age in the activated sludge sub-model. A relatively small adjustment of such parameters also very strongly affects the final effluent quality and other plant-wide performance measures via complex model feedbacks. This includes changes to degradability and volume of waste sludge and thus mainline treatment performance and the properties of solids being fed to the anaerobic digesters and thus biogas recovery, and the phosphorus (P) and nitrogen (N) released by anaerobic digestion, and ultimately the nutrient load on the mainline treatment. These effects are not accounted for in existing calibration protocols. They are critical and impossible to capture by considering individual sub-models in isolation. A systematic approach is needed to develop a robust plant-wide model calibration protocol applicable to treatment of pulp and paper mill effluents.

4 Modelling industrial symbiosis for biogas production

This review covered the concept of an industrial symbiosis, which has been proposed as an efficient way to purify pulp and paper mill effluents while at the same time generating products, such as methane, nutrients and biofertilizers, with added values. A reliable description of the movements and cycling of materials within the concept system is very vital both regarding the added values and for guaranteeing the function of the wastewater treatment included in the symbiosis concept. To be able to predict the effect and optimize the process concept regarding energy end resource efficiency comprehensive physico-chemical and biological models are required.

Existing process simulation models for domestic wastewater provide a basis for developing the specific model applicable to this industrial symbiosis. Some improvements are required to achieve such a dynamic model. There is an immense need to improve the activated sludge model with relevant processes, such as dissolved methane oxidation. Since high-rate activated sludge system is part of the treatment, the HRAS model of Nogaj et al. (2013) should be used as the basis for further development to ensure that the storage phenomena, biosorption and bio-flocculation for organic carbon removal are adequately described.

To develop a tractable and robust modelling framework for ESCB reactors, the trade-off between model complexity and performance needs to be analysed. The model proposed by Feldman et al. (2017) is the only published work that accounts for reactor hydrodynamics, microbial interaction/competition with the granules and a comprehensive biochemical model (physico-chemical and biological processes included). Future studies should systematically analyse ways of reducing the layer of complexity to develop an appropriate and simple model structure that provides good general model performance.

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