CLOSING THE LOOP BY COMBINING UASB REACTOR AND REACTIVE BED FILTER TECHNOLOGIES FOR WASTEWATER TREATMENT: MODELLING AND PRACTICAL APPROACHES

Raúl Antonio Rodríguez Gómez

March 2016
Dedication

A mis padres e hijos

“Pues se fue la niña bella,
bajo el cielo y sobre el mar,
a cortar la blanca estrella
que la hacía suspirar”

Rubén Darío
Wastewater treatment is mainly based on chemical and biological processes. Biological treatment is subdivided into aerobic and anaerobic processes. The former requires an inert support medium and aeration to create good conditions for the development of microorganisms. In the latter, microorganisms degrade organic material in the absence of oxygen and instead use another electron acceptor such as sulphate, ferric iron, nitrate, carbon dioxide or organic compounds.

Among the anaerobic systems, the upflow anaerobic sludge blanket (UASB) reactor has been widely used due to its capacity for forming granules without the need for an inert support medium for the development of microorganisms. The UASB reactor and similar reactors are based on internal circulation and an expanded granular sludge bed can be used to remove organic material from wastewater. The main advantages of the UASB reactor are that it produces biogas that can be used as an energy source and it uses a simple technology which is cheap compared with aerobic processes. The main disadvantages are that the removal of nutrients is poor and that the effluent needs post-process treatment before it can be discharged into water bodies.

Nutrients, mainly phosphorus and nitrogen, are essential for life and for the survival of species, including humans, whereas nutrients in excess concentrations in seas, lakes and rivers are regarded as contaminants. Gaseous nitrogen is the most abundant element in the air; plants and some animals can absorb it and convert it into nitrogen-based compounds in food. Phosphorus is the second most abundant mineral in the human body. It is predicted that global phosphorus resources will be exhausted in a couple of hundred years. Thus, these nutrients will be viewed in future as valuable resources rather than as contaminants. Nowadays, in most countries the nutrients in wastewater are wasted because they are not recovered. However, researchers are working on developing methods to capture and recycle these nutrients.

Natural mineral-based sorbents are an attractive solution for recovery of nutrients from wastewater. Various sorbents have been studied recently, mainly for their potential for removing phosphorus. The Royal Institute of Technology in Stockholm (KTH) has developed two sorbents (Sorbulite® and Polonite®) based on calcium-silicate materials for use in tertiary treatment of wastewater. The former has the ability to remove organic material and phosphorus, but the removal of pathogens is low. The latter is very efficient in removing pathogens and phosphorus, but its removal performance may be affected if the incoming wastewater contains high concentration of organic materials. It has been also reported that both remove nitrogen to a minor extent compared with phosphorus.

The Swedish Environmental Protection Agency recommends percentage removal of organic material, phosphorus and nitrogen of 90%, 90%, and 50%, respectively, for residential wastewater before its discharge into water bodies.

Taking into account Swedish regulations, a system composed of a UASB reactor and a packed bed reactor (PBR) filled with Sorbulite and Polonite was devised. The hypothesis was that the UASB reactor would remove most of the organic material, Sorbulite would protect the Polonite from the solids and organic material present in the effluent of the UASB reactor and Polonite would remove pathogens and phosphorus from the wastewater.
Models were developed to simulate the performance of the UASB reactor and the PBR with filter media. The models included dispersion, advection and reaction terms. The reaction term in the UASB reactor was described by two expressions for the granules which have analytical and numerical solutions. They involve the substrate bioconversion rate, the mass transfer coefficient in the film, the intra-particle diffusivity and the volumetric fraction of biomass in the reactor. The reaction term in the PBR was assumed to follow the Langmuir isotherm.

As a first attempt, the models developed for the UASB reactor were validated using data from the literature on a UASB reactor used to treat sugarcane mill wastewater and another used to treat slaughterhouse wastewater. The results showed good agreement between simulated and observed values.

Next, a laboratory-scale UASB reactor followed by a PBR was built to treat residential wastewater. The results revealed that the proposed system effectively treated the residential wastewater and that the effluent complied with the Swedish regulations on wastewater discharge. The system also removed >99% of *Escherichia coli* and *Enterococcus faecalis*.

The models developed successfully predicted changes in the concentration of substrate, biomass and granule size in the UASB reactor. As regards percentage removal of organic matter (chemical oxygen demand, COD), the simulation predicted a slightly higher value (91%) than observed in practical experiments (89%). As regards the concentration of volatile suspended solids, the model underestimated the final concentration, giving a value of 40458.35 mg L⁻¹ compared with 45803.36 mg L⁻¹ observed in practical experiments. The simulation predicted 6% expansion of the sludge bed, which was lower than the 20% observed in practical experiments. This difference can be attributed to channelling and bubbles observed in the practical experiment, which contributed to greater expansion of the sludge bed.

There was also a difference in the size of the granules. In the simulations a final granule radius of 0.12 mm was obtained, while in the practical experiment the value ranged from fine particles to 0.2 mm.

The PBR removed a large percentage of phosphorus even at the end of the experiment. In the lower part of the reactor, where the Sorbulite was located, it was observed that solids coming from the effluent of the UASB reactor were retained.

The model developed for the PBR successfully predicted changes in the concentration of phosphorus throughout the reactor. The predicted percentage removal at the end of the experiment was 98.31%, which was slightly higher than the 97.83% measured in practical experiments.
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LIST OF APPENDED PAPERS

This thesis is based on the following papers, which are referred to in the text by their corresponding Roman numeral.


III. Rodríguez-Gómez R., Renman G. 2016. Sequential UASB and dual media packed-bed reactors for domestic wastewater treatment – Experiment and simulation. Accepted for publication in the journal Water Science and Technology.


Reprints are published with the kind permission of the journals concerned.

The contribution of the authors to each of the papers was as follows:

- In Paper I, Raúl Rodríguez-Gómez developed the model, wrote the corresponding numerical code to solve the model under the supervision of Luis Moreno, and wrote the manuscript, which was reviewed by the co-authors.

- In Paper II, Raúl Rodríguez-Gómez developed the model under the supervision of Luis Moreno; Longcheng Liu participated in discussions about how to improve the model. Raúl Rodriguez-Gómez and Gunno Renman revised several practical cases until they found a study case adequate to test the model developed. Paper II was written by Raúl Rodríguez-Gómez and reviewed and modified by the co-author.

- In Paper III, Raúl Rodríguez-Gómez developed the model, carried out the practical experiment with the guidance of Gunno Renman and wrote the manuscript, which was reviewed and modified by the co-author.

- In Paper IV, Raúl Rodríguez-Gómez developed the model, carried out the kinetics experiments in the laboratory under the guidance of Gunno Renman and wrote the paper, which was reviewed and modified by the co-author.

Raúl Rodríguez-Gómez was responsible for the results and all diagrams presented in Papers I-IV.
Relevant publications not included in the thesis:

Papers

Rodríguez-Gómez R., Renman G. 2013. Triggered phosphorus recovery from wastewater using UASB and fixed-bed reactor technology. Poster presented to NORDIWA-13th Nordic Wastewater Conference, October 8-10, Malmö, Sweden


Thesis


Books


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<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tbody>
<tr>
<td>AAC</td>
<td>Aerated autoclaved concrete</td>
</tr>
<tr>
<td>ADM</td>
<td>Axial dispersion model</td>
</tr>
<tr>
<td>Al</td>
<td>Aluminium</td>
</tr>
<tr>
<td>ATCP</td>
<td>Amorphous tricalcium phosphate</td>
</tr>
<tr>
<td>B</td>
<td>Contois constant</td>
</tr>
<tr>
<td>b</td>
<td>Langmuir coefficient</td>
</tr>
<tr>
<td>BOD</td>
<td>Biochemical oxygen demand</td>
</tr>
<tr>
<td>bioden</td>
<td>Density of biomass in the sludge bed</td>
</tr>
<tr>
<td>Ca</td>
<td>Calcium</td>
</tr>
<tr>
<td>CaO</td>
<td>Calcium oxide</td>
</tr>
<tr>
<td>CaCO$_3$</td>
<td>Calcium carbonate</td>
</tr>
<tr>
<td>Ca$_5$(PO$_4$)$_3$OH$_2$</td>
<td>Hydroxyapatite</td>
</tr>
<tr>
<td>CaHPO$_4$</td>
<td>Dibasic calcium phosphate</td>
</tr>
<tr>
<td>CH$_4$</td>
<td>Methane</td>
</tr>
<tr>
<td>COD</td>
<td>Chemical oxygen demand</td>
</tr>
<tr>
<td>CÔD$_{UASB}$</td>
<td>Mass of substrate that stays in the UASB reactor</td>
</tr>
<tr>
<td>CÔD$_0$</td>
<td>Mass of substrate in the influent</td>
</tr>
<tr>
<td>CÔD$_e$</td>
<td>Mass of COD in the effluent of the UASB reactor</td>
</tr>
<tr>
<td>CO$_2$</td>
<td>Carbon dioxide</td>
</tr>
<tr>
<td>CSTR</td>
<td>Continuous stirred tank reactor</td>
</tr>
<tr>
<td>CSH</td>
<td>Calcium silicate hydrates group</td>
</tr>
<tr>
<td>D$_A$</td>
<td>Diffusion coefficient within the granule</td>
</tr>
<tr>
<td>D</td>
<td>Diffusion coefficient</td>
</tr>
<tr>
<td>DCP</td>
<td>Dibasic calcium phosphate</td>
</tr>
<tr>
<td>E</td>
<td>Concentration of non-active biomass</td>
</tr>
<tr>
<td>ECP</td>
<td>Extracellular polymer</td>
</tr>
<tr>
<td>f</td>
<td>Fraction</td>
</tr>
<tr>
<td>Fe</td>
<td>Iron</td>
</tr>
<tr>
<td>GLSS</td>
<td>Gas-liquid-solid separator</td>
</tr>
<tr>
<td>HAp</td>
<td>Hydroxyapatite</td>
</tr>
<tr>
<td>HRT</td>
<td>Hydraulic retention time</td>
</tr>
<tr>
<td>HPO$_4^{2-}$</td>
<td>Hydrogen phosphate</td>
</tr>
<tr>
<td>H$_2$PO$_4$</td>
<td>Dihydrogen phosphate</td>
</tr>
<tr>
<td>H$_3$PO$_4$</td>
<td>Phosphoric acid</td>
</tr>
<tr>
<td>KH$_2$PO$_4$</td>
<td>Potassium dihydrogen phosphate</td>
</tr>
</tbody>
</table>
i  CSTR under analysis, type of mineral
K  Kinetic rate
k  Volumetric conversion rate
KD  Decay of active biomass
km  Mass transfer coefficient
KS  Substrate concentration at $\frac{1}{2}\mu_{max}$
L  Height of the reactor
Mg  Magnesium
MgCl2  Magnesium chloride
Mn  Manganese
n  Number of CSTRs
N  Nitrogen
Na  Sodium
Np  Number of granules per volume of reactor
NaCl  Sodium chloride
NaOH  Sodium hydroxide
NH4  Ammonium
NH4Cl  Ammonium chloride
OLR  Organic loading rate
OSWWTP  On-site wastewater treatment plant
P  Phosphorus
PBR  Packed bed reactor
Pe  Péclet number
PO4−  Phosphate
Q  Flow rate
q  Flux
qmax  Maximum adsorption capacity
R  Radius of the granule
r  Radial distance from the center of the granule
R  Reaction rate
S  Substrate concentration
Si  Silicon
SiO2  Silica
Sp  Substrate within the granule
Spgrad  Gradient of substrate inside the granule
TISM  Tanks-in-series model
TiO2  Titanium dioxide
TN  Total nitrogen
TSS  Total suspended solids
UASB  Upflow anaerobic sludge blanket
V  Volume of the reactor
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$v$</td>
<td>Upflow velocity</td>
</tr>
<tr>
<td>VSS</td>
<td>Volatile suspended solids</td>
</tr>
<tr>
<td>$V_{CH_4}$</td>
<td>Volume of methane</td>
</tr>
<tr>
<td>$V_g$</td>
<td>Volume of granule</td>
</tr>
<tr>
<td>$X$</td>
<td>Biomass concentration</td>
</tr>
<tr>
<td>$x$</td>
<td>Fraction</td>
</tr>
<tr>
<td>$X_p$</td>
<td>Biomass concentration in the granule</td>
</tr>
<tr>
<td>$Y$</td>
<td>Yield of biomass</td>
</tr>
<tr>
<td>$Y_{CH_4}$</td>
<td>Yield of methane</td>
</tr>
<tr>
<td>$z$</td>
<td>Direction of flow</td>
</tr>
<tr>
<td>$\phi_p$</td>
<td>Volumetric fraction of biomass</td>
</tr>
<tr>
<td>$\phi_{p_{max}}$</td>
<td>Maximum fraction of biomass</td>
</tr>
<tr>
<td>$\phi$</td>
<td>Thiele modulus</td>
</tr>
<tr>
<td>$\tau$</td>
<td>Contact time in every mesh solution</td>
</tr>
<tr>
<td>$\mu_{max}$</td>
<td>Maximum utilisation constant</td>
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</table>
Abstract

A laboratory-scale upflow anaerobic sludge blanket (UASB) reactor followed by a packed bed reactor (PBR) filled with Sorbulite® in the lower part and Polonite® in the upper part was used to treat household wastewater in a 50-week experiment. A model was developed to describe the performance of the UASB reactor, including mass transfer through the film around anaerobic granules, intra-particle diffusion and bioconversion of the substrate. In a second model, a numerical expression describing the kinetics occurring in the granules was developed. It includes the resistances through which the substrate passes before biotransformation. These expressions were then linked to governing equations for the UASB reactor in order to describe degradation of the substrate, biomass growth (active and inactive), and variation in granule size over time. A third model was developed to describe the profile of the phosphorus (P) concentration throughout the PBR. In a first attempt, the analytical and numerical model was applied to data taken from previous studies in which UASB reactors were used to treat sugarcane mill wastewater and slaughterhouse wastewater. The results showed good agreement between observed and simulated results. Sensitivity analysis showed that diffusion coefficient and yield were important parameters in the UASB reactor model.

The laboratory bench-scale experiment revealed that the combined UASB-PBR system efficiently treated the residential wastewater. Phosphorus, BOD, and pathogenic bacteria all showed average removal of 99%, while total nitrogen showed a moderate reduction in the system (40%). Application of the numerical solution model to the experimental UASB reactor used resulted in good agreement between simulated and experimental values. Regarding the PBR, the model developed successfully predicted P removal. For both models, the capability and sensitivity analyses identified important parameters. A treatment system aiming to close the loop is suggested based on sequential UASB and PBR with biogas collection, nutrient recycling via sludge and filter media and elimination of pathogenic organisms.

Key words: Bench-scale experiment, Modelling, Polonite, Sorbulite, Wastewater.

1. Introduction

Wastewater treatment is mainly based on chemical and biological processes. Chemical processes usually work on the principle of coagulation-flocculation and have the following disadvantages: handling of dangerous substances, risk of intoxication, high cost, poor efficiency and production of high concentrations of sludge (Zeman, 2012). Biological treatment is mainly subdivided into aerobic and anaerobic processes. The former requires an inert support medium and aeration to create good conditions for the development of microorganisms. In the latter, microorganisms degrade organic material in the absence of oxygen and instead use another electron acceptor such as sulphate, ferric iron, nitrate, carbon dioxide or organic compounds (Rodríguez-Gómez, 2011).

Among the anaerobic systems, the upflow anaerobic sludge blanket (UASB) reactor has been widely used due to its capacity...
for forming granules without the need for an inert support medium for the development of microorganisms. While the UASB reactor provides benefits such as removal of organic material and production of biogas, it is poor at removing nutrients and pathogens. Discharge of nutrients to water bodies such as the Baltic Sea (Fleming-Lehtinen et al., 2015) causes eutrophication. It also causes the spread of pathogens that may result in epidemics like cholera or foodborne illness, such as that which occurred during spring 2011 in Germany (Flieger et al., 2015). It was recently reported that phosphorus (P) can be transported in high concentrations through groundwater to lakes (Meinikmann et al., 2015), which can have implications for the use of wastewater infiltration systems.

Natural mineral-based sorbents are an attractive solution for recovery of nutrients from wastewater. Various sorbents have been studied recently, mainly for their potential for removing P. Sorbulite and Polonite are engineered filter minerals for removing organic material, pathogens and nutrients present in wastewater. Sorbulite is an autoclaved aerated concrete (AAC) like-material. Polonite is a calcium-silicate material produced after a thermal and grinding treatment procedure. These two materials complement each other. Sorbulite has the capacity to remove organic material and P, while Polonite has the ability to remove pathogens and P. In addition, both minerals may remove some nitrogen (N) from wastewater. Due to the alkalinity of Polonite, the treated water has a high pH, but this tends to decrease with time. An effluent with high pH can help to tackle the problem of acidification in water bodies. If neutral pH is desired in the effluent, bark can be used to decrease the pH. Bark can also be a useful material since it has been reported that it can reduce total N by up to 39% (Dalahmeh et al., 2014).

The P adsorption capacity of a filter material must be calculated carefully, since its value can vary widely depending on the method (e.g. Langmuir, Freundlich, batch, column etc.) and the conditions (i.e. particle size, temperature, source of water etc.) used for calculation (Cucarella & Renman, 2009). If the influent wastewater undergoes good pre-treatment, with efficient removal of organic material, many of the sorbents provide an acceptable level of P removal. However, in real treatment conditions the influent wastewater can exhaust the filter material, which affects its lifetime and requires frequent replacement to maintain high removal performance.

By combining the UASB reactor and the Sorbulite-and Polonite technology in order to treat wastewater, a loop of benefits that in optimal conditions ends with minor waste volumes is generated. These benefits are:

- Removal of organic material
- Removal of phosphorus
- Removal of nitrogen
• Removal of pathogens
• Removal of solids from wastewater
• Production of biogas
• Production of sludge and minerals rich in nutrients that can be used as fertiliser.

For that reason, in this thesis a system composed of an UASB reactor and a reactive bed filter (a packed bed reactor, PBR) filled with Sorbulite and Polonite was devised to treat wastewater. The system was tested for treatment of household wastewater, but it could be scaled up suit to industrial and municipal wastewater treatment plants.

In addition to practical experiments, a series of models to describe the performance of the UASB reactor and the PBR was developed. These models are mathematical expressions obtained from a mass balance of substrate in the granules of the UASB reactor and in both reactors (i.e. UASB and PBR).

This thesis is divided into seven chapters. Chapter 1 provided an introduction to the work, while in Chapter 2 the research problems are described. Chapter 3 presents the objectives and Chapter 4 the background to the UASB reactor, Sorbulite and Polonite. Materials and methods are discussed in Chapter 5, while Chapter 6 contains the results and discussion. Chapter 7 presents the conclusions.

2. Research Problem

A number of UASB reactors have been built since their development in the late 1970s (Lettinga, 2001). Most of these works in an acceptable way, but there are still many reactors that need optimisation. Common problems associated with use of a UASB reactor are: low biogas production, poor organic material removal, no development of granules, bad smell generation and low effluent quality (Korsak, 2011). All these problems are linked each other. To overcome these issues, it is necessary to understand the processes and mechanisms involved in the UASB reactor. There are basically two ways of studying these processes: in practical experiments and using computer programmes to simulate the performance of the reactor.

Computer simulation involves the development of models capable of predicting the behaviour of the reactor. The model programme is created in a computer and it is executed to obtain a prediction of the performance of the reactor. Simulation results can be obtained in a very short time compared with in practical experiments, a lot of money is saved and risks of accidents due to manipulation of equipment and objects are avoided. However, the models produced have limitations and work under assumptions.

Simulations and practical experiments both have their advantages in work to understand the mechanisms occurring in UASB reactors.

However, the UASB reactor itself cannot be used as a single system to treat wastewater (Lim & Kim, 2014). It must be coupled
with other systems to remove nutrients and/or to meet national water discharge regulations.

Most researchers working with Sorbulite or Polonite have carried out experiments at laboratory scale, but few at pilot or real scale. However, Polonite has been widely used by companies working on wastewater treatment as a tertiary treatment in on-site wastewater treatment plants, with excellent results.

By combining these USAB and PBR systems, a loop of utilisation of the sub-products may be generated. For instance, in a house provided with a septic tank and located in the countryside where no municipal treatment plant and sewer system exists, the proposed system can be very useful since the sub-products generated are not wasted, but are exploited, as shown in Figure 1. Loops that have to be broken, however, are those of pathogens (Vinnerås, 2007) and micropollutants (Luo et al., 2014).

Fig. 1. Loops generated with the proposed upflow anaerobic sludge blanket (UASB) and packed bed reactor (PBR) system.

3. Objectives

The overall aim of this thesis was to develop models able to predict the performance of a UASB reactor and a PBR filled with two commercially produced materials; Sorbulite and Polonite. Moreover, practical experiments were performed to demonstrate and validate the models. Specific objectives of the work were to:

- Develop an analytical and numerical expression to describe the conversion of substrate in the UASB reactor
- Develop a model to describe the profile of the P concentration along the height of a PBR filled with Sorbulite and Polonite
- Study the performance of a UASB reactor fed with residential wastewater and inoculated with non-granulated anaerobic biomass
- Study the performance of a PBR filled with the filter materials Sorbulite and Polonite and receiving pre-treated
wastewater from a UASB reactor, with particular regard to the P removal efficiency

- Validate the models with data from the literature and results obtained from the UASB and PBR experiments.

4. BACKGROUND

This chapter reviews current knowledge of the UASB technology and of the two filter materials used in the experiments (Sorbulte and Polonite).

4.1. The UASB reactor technology

The UASB reactor is basically a tank in which wastewater rich in organic materials enters from the bottom and flows upward. Inside the tank, an anaerobic sludge blanket containing microorganisms acts as a filter for small particles present in wastewater and as a bioreactor to consume the organic material. Biogas is produced as a by-product of the microbial digestion. The rising bubbles of the biogas result in agitation of the sludge blanket and over time dense granules are formed. In the upper part of the UASB reactor, a gas-liquid-solid separator (GLSS) is installed to prevent washout of particles reaching that point. Granules can reach the GLSS due to their buoyancy, which is induced by the biogas trapped inside the granules and bubbles attached to their surfaces. After a while, the bubbles of biogas are released and the density of the granules increases, causing them to move in a downward direction until they reach the sludge blanket.

The main goal of the UASB reactor is degradation of organic material, which ends in the production of biogas. The biogas is mainly composed of carbon dioxide ($CO_2$) and methane ($CH_4$) and it is generated from complex polymers through a series of bio-reactions in which processes of hydrolysis, acidogenesis, acetogenesis and methanogenesis take place. Figure 2 shows these processes and the typical percentage of product for each reaction.

In addition to biogas, which can be used as an energy source, the UASB reactor provides other benefits such as: low growth rate of biomass, reduction of pathogens under thermophilic conditions, simple technology that can be used in developing countries, and no need for an inert support medium for the development of microorganisms, as dense granules are formed. The main disadvantages of the UASB reactor are that: it requires a long time to start up, it is susceptible to toxic substances and it achieves poor removal of nutrients.

It has been reported that the UASB reactor can work with a variety of wastewater types, including: brewery, beverage, distillery, fermentation, food leachate, pulp, household, textile, paper industry, municipal and petroleum refinery wastewater (Rodríguez-Gómez, 2011). However, a post-treatment step is needed in order to meet the standards for wastewater discharge (Lim & Kim, 2014). Malovanyy et al. (2015) used a moving bed biofilm reactor to treat municipal wastewater pre-treated by a UASB reactor. However, the scope of their study was limited to nitrogen removal.
4.1.1. Granulation in the UASB reactor

The processes involved in the creation of granules are not well understood, but there are some theories which can be classified as relating to physical, microbial and thermodynamic processes. Table 1 presents a brief description of these theories (Rodríguez-Gómez, 2011).

**Table 1. Theories on the origin of granulation**

<table>
<thead>
<tr>
<th>Classification</th>
<th>Name</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Physical</td>
<td>Growth of colonised suspended solids</td>
<td>Suspended fine particles attach themselves and may colonise suspended solids coming from influent, forming granules. A fraction of fine particles may be dragged to the effluent of the UASB reactor.</td>
</tr>
<tr>
<td></td>
<td>Selection pressure</td>
<td>Biogas causes pressure in the biomass. High pressure removes lightweight and dispersed biomass. Heavier particles remain in the reactor. Initially the biomass consists of voluminous aggregates, but with time they become denser due to bacterial growth.</td>
</tr>
<tr>
<td>Microbial</td>
<td>Extracellular polymer (ECP) bonding model</td>
<td>ECP is a bio-glue secreted by cells. When two cells collide, they attach themselves and over time they reproduce and consequently a granule is formed.</td>
</tr>
<tr>
<td></td>
<td>The Capetown model</td>
<td>Amino acids are over-secreted from the microbial population; this encourages Methanobacterium to produce ECP, which in turn induces formation of granules.</td>
</tr>
<tr>
<td></td>
<td>Spaghetti theory</td>
<td>The filamentous Methanoseta entangle each other, forming a structure that resembles a spaghetti ball. Microorganisms grow in this ball and consequently the size and density of the ball increase and thus a granule is created.</td>
</tr>
<tr>
<td>Thermo-dynamic</td>
<td>The four-step model for granulation</td>
<td>Granulation follows four steps: i) Transport of cells to the substratum; ii) reversible adsorption of the cell to the substratum by physicochemical forces; iii) irreversible adhesion of the cell to the substratum by microbial or polymer attaching; and iv) multiplication of the cell and growth of the granules.</td>
</tr>
<tr>
<td></td>
<td>The proton translocation-dehydration model</td>
<td>This theory consists of four stages: i) Dehydration of bacterial surfaces; ii) embryonic granule formation; iii) granule maturation; and iv) post-maturation.</td>
</tr>
</tbody>
</table>
Nowadays, researchers try to develop granules able to remove not only organic material, but also nutrients from wastewater. For instance, Zhang et al. (2015) reported that denitrifying bacteria can colonise the outer layer of the granule if the wastewater is enriched with nitrate. If the aim is to remove P from wastewater, Hao et al. (2015) stated this can be achieved by adding iron to the anaerobic digestion process. Moreover, Tervahauta et al. (2014) reported that P can be captured in the form of calcium phosphate by anaerobic granules. However, their study was limited to blackwater and the percentage of captured P was only 2% of the concentration in the inlet wastewater. Thus, a post-treatment step is recommended to recover the P leaving the UASB reactor.

4.1.2. **Biogas produced from the UASB reactor**

Biogas is mainly composed of CH$_4$ and CO$_2$ with traces of hydrogen sulphide (H$_2$S), N$_2$ and H$_2$ (Salam et al., 2015). The presence of H$_2$S in biogas causes corrosion and odour problems, which may be an issue for equipment that comes into contact with the biogas. However, various methods have been developed to remove H$_2$S, including: adsorption, precipitation, reaction with alkaline substances, biochemical oxidation of sulphide to sulphate, thiosulphate or elemental sulphur, and microaeration to reduce sulphide to elemental sulphur (Krayzelova et al., 2014). Removal of CO$_2$ from the biogas in order to upgrade it is also of interest and can be easily achieved using the well-known Mariotte flask technique, where the CO$_2$ present in the biogas is absorbed by a solution of NaOH. This method not only purifies the biogas, but also measures the volume of CH$_4$, since displacement of the NaOH solution occurs (Priya & Narayani, 2012). Other techniques such as degassing membrane (Angelidaki et al., 2014), injection of H$_2$ into the reactor (Hao et al., 2015), fixation of CO$_2$ using Ca(OH)$_2$, activated carbon (Karim et al., 2016), zeolite and TiO$_2$ have also been developed to refine the biogas (Liu et al., 2015). The generation of biogas is directly linked to the degradation of organic material. It has been reported that between 31 and 63% of the COD present in the influent is converted into methane (Tawfik et al., 2013). However, the volume of biogas produced is influenced by several factors, for instance type of wastewater.

**Table 2. Yield of biogas from UASB reactors treating different types of wastewater**

<table>
<thead>
<tr>
<th>Type of wastewater</th>
<th>Biogas yield, m$^3$ (kg COD$_{rem.}$)$^{-1}$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Brewery</td>
<td>136.5</td>
<td>Kumar, 2012</td>
</tr>
<tr>
<td>Cane-molasses</td>
<td>0.168</td>
<td></td>
</tr>
<tr>
<td>Cheese whey</td>
<td>0.28-0.55</td>
<td></td>
</tr>
<tr>
<td>Slaughterhouse</td>
<td>0.41-0.45</td>
<td></td>
</tr>
<tr>
<td>Vinasses</td>
<td>0.46</td>
<td></td>
</tr>
<tr>
<td>Cane vinasses</td>
<td>0.42</td>
<td></td>
</tr>
<tr>
<td>Pulping coffee</td>
<td>0.37</td>
<td></td>
</tr>
<tr>
<td>Olive mill</td>
<td>0.328</td>
<td>Siciliano et al., 2016</td>
</tr>
<tr>
<td>Municipal</td>
<td>0.18-0.22</td>
<td>Khan et al., 2015</td>
</tr>
</tbody>
</table>
Table 2 summarises biogas yield from different sources of wastewater.

Existing models to describe the production of biogas are complex since they involve several mechanisms, including: dispersion, biogas transfer rate, advection, settling velocity of granules etc. For instance, the model developed by Kalyuzhnyi et al. (2006) involves many empirical equations and a total of 35 variables, which decreases the reliability of the model.

4.1.3. Factors influencing the operation of the UASB reactor

pH
Biodegradation of organic material is very effective within the pH range 6.5-7.5. Outside this range, the methanogenic microorganisms are affected. The reactor normally has the ability to self-control the pH changes and addition of buffer solutions is not necessary (Jain et al., 2015).

Temperature
The UASB reactor can work under psychrophilic, mesophilic and thermophilic temperatures. In psychrophilic conditions, the activity of the microorganisms decreases. Mesophilic temperatures have the disadvantage of poor start-up of the reactor. The issue with working in thermophilic conditions is that a lot of energy is needed to heat the reactor. The preferred temperature is 35°C, since the removal of organic material and the production of biogas are efficiently balanced (Rodríguez-Gómez, 2011; Jain et al., 2015).

Organic loading rate (OLR)
OLR is an important factor that in some cases must be adjusted by varying the concentration of COD in the incoming wastewater, especially during the start-up step. Low OLR (i.e. <1.5 kgCOD m$^{-3}$d$^{-1}$) causes starvation of microorganisms and granule disintegration. Accumulation of volatile fatty acids occurs if the reactor is fed with high OLR and consequently the pH decreases, which disturbs the performance of the UASB reactor. The optimal OLR range is 2-4.5 kgCOD m$^{-3}$d$^{-1}$ (Habeeb et al., 2011).

Hydraulic retention time (HRT) and upflow velocity
The HRT is directly linked to the upflow velocity. Increased HRT means a reduction in upflow velocity. This action improves the removal of suspended solids and COD from the substrate, since the microorganisms have more time to degrade the organic material. If the opposite occurs (decreased HRT) not only is the COD affected but there is also a risk of granule break-up, resulting in washout of biomass (Abbas et al., 2015).

Type of sludge
In principle, any sludge containing anaerobic microorganisms can be used to inoculate a UASB reactor. The sludge can be obtained from environments such as swamps, lakes, oceans, paddy fields, hot springs and manure (Parawira, 2004). However, the best microbial cocktail to inoculate a UASB reactor is one taken from an existing UASB reactor, either granular or slurry.
Every time a UASB reactor is inoculated and fed with a new substrate, the microorganisms living in the sludge bed experience a condition known as lag phase or aclimatisation phase, where they become habituated to the new substrate. The log or exponential phase then takes place; at this point the multiplication of the microorganisms is fast until it reaches a steady state. When the stationary state is reached, the reactor is in equilibrium; the dying cells equal the cells that are born. If the harmony in the reactor is strongly disturbed, for instance by toxic compounds, a decay phase can occur which can create a chaotic environment and destabilise the system (Rodríguez-Gómez, 2011).

4.1.4. Modelling the UASB reactor

There are two different types of models describing the performance of UASB reactors. The first type are models in which statistical correlations are obtained for an existing reactor and the results are used to optimise the reactor. The second type are phenomenological models based on the mechanisms taking place in the reactor. The latter are more interesting, since they may be applied to any kind of UASB reactor without prior physical construction. The most frequently used models are the axial dispersion model (ADM) and the tanks-in-series model (TISM). The latter is also known as the multi-CSTR model. The main difference is that in the ADM, the UASB reactor is seen as a single column, while the TISM hypothetically divides it into several small reactors which behave as continuous stirred tank reactors (CSTR). Both models include terms of dispersion, advection and reaction. However, in the ADM, the dispersion term is directly linked with the mathematical expression, while in the TISM the term is intrinsically included in the number of small CSTRs into which the UASB reactor is divided.

According to Sreekrishnan and Saravanan (2006), models for UASB reactors can also be classified into fluid flow models and reactor models. The former study the fluid dynamics in the UASB reactor, while the latter, besides studying the hydrodynamics, also simulate the possible reactions occurring in the UASB reactor.

Existing fluid flow models

A series of conceptual models for flow distribution in UASB reactors is presented in Ojha and Singh (2002). They studied those different configurations to study the flow distribution in the reactor. They concluded that if the flow resistance increases, disturbance of the flow occurs. They also reported that there is no generalised model to describe the hydrodynamics in UASB reactors.

Wu and Hickey (1997) hypothetically divided the UASB reactor in two sections, the first corresponding to the lower part of the reactor, where the flow is assumed to be completely mixed, and the other corresponding to the upper part, where plug flow is assumed. This leads to a model that behaves as a non-ideal CSTR. Those authors assumed that a fraction of the substrate passes
directly from the influent to the effluent, in order to take into account dead volume present in the sludge bed.

Using a non-reactive trace, Gomes et al. (1998) studied the flow dynamics of a UASB reactor. They divided the UASB reactor in two zones, the first representing the sludge bed and the second corresponding to the blanket zone. Similarly to Wu and Hickey (1997), they also assumed that there is a dead zone, but assumed that it is located in the sludge bed. They used the axial dispersion model to get a flow profile of the tracer in the reactor. Their experimental and simulated results showed good correlation.

Wu et al. (2005) applied the model proposed in Gomes et al. (1998) using a rhodamine tracer instead of lithium to study the hydrodynamics of a UASB reactor. They concluded that the dispersion term played an important role in describing the flow of the rhodamine. In their simulation, they assumed that the dispersion coefficient was linearly dependent on the upflow velocity.

Existing reactor models

Gonzalez-Gil et al. (2000) used the TISM to describe the substrate consumption in a UASB reactor. The simulation included the hydrodynamics and the reaction terms. They assumed that the conversion of the substrate followed the Monod model. Based on a mass balance, they developed an expression for the degradation of substrate in the granule. However, the granule size was assumed to be constant. By manually varying the diameter of the granule and running the simulation several times, they showed that the particle size affects the removal of the substrate and that the internal mass transport is the limiting parameter in anaerobic granules. On the other hand, they concluded that the external mass transfer can be neglected, since it did not affect the kinetic term.

A model to describe the bioconversion of the substrate taking into account dispersion, advection and reaction was proposed by Korsak et al. (2008). The governing equation followed the axial dispersion model. The dispersive term was described as a function of the flow regime (i.e. Reynolds number) and the Péclet number. The reaction term was able to predict the mass flow of the substrate and its corresponding kinetics within the granule, which included the ratio of convective to diffusive mass transport in the stagnant film (i.e. Sherwood number).

Rodríguez and Moreno (2009) applied the TISM to predict the change in substrate and biomass in a laboratory UASB reactor fed with glucose as the carbon source. The reaction terms for both the substrate and biomass were assumed to only follow the Monod model. The model included substrate degradation, biomass growth and death, and washout of microorganisms. A year later, the same authors improved the model by introducing mass balance of substrate in the granules. This led to an expression able to describe not only the substrate degradation and the biomass profile, but
also the size of the granules and their location along the reactor height (Rodriguez & Moreno, 2010).

In 2002, Batstone et al. proposed an extensive anaerobic digestion model (ADM1) to describe processes occurring in anaerobic reactors. However, this model has been criticised since it is not adequate to describe start-up of UASB reactors and it involves at least 26 dynamic variables and many parameters (López & Borzacconi, 2009; Zhao et al., 2010). To describe the start-up of UASB reactors, Zhao et al. (2010) linked the linear phenomenological equation with the ADM1. Consequently, the hydrolysis rate and the specific uptake rate coefficients varied over time during the start-up. Their results demonstrated that its model was able to describe the performance of UASB reactors during the lag phase (i.e. acclimatisation of anaerobic biomass), but for wastewater containing toxic compounds the model was not suitable.

The axial dispersion model was used by Zeng et al. (2015) to describe the performance of a laboratory-scale UASB reactor fed with synthetic wastewater. The reaction term in the model was assumed to follow the kinetics described in the ADM1. The model showed good correlation between experimental and simulated data. A weakness of that model was that it did not take into account the variation in granule growth. However, the model was able to describe the fluid dynamics of a non-reactive trace injected into the UASB reactor.

The art of modelling lies in the development of mathematical expressions able to represent mechanisms occurring in the system. For UASB reactors, most existing models are too complex or too simple and only a few models consider the mechanisms occurring in the reactor. Despite researchers agreeing that the granule is an essential issue in the UASB reactor, existing models do not include the variation in granule size. Overcoming this particular shortcoming is one of the contributions described later in this thesis.

The main benefits of the UASB reactor are the significant removal of organic material and the production of biogas. However, it is necessary to treat the effluent of the UASB reactor before its discharge to water bodies, since it still contains nutrients, pathogens and, in some cases, organic material. Post-treatment options are not completely developed, but systems such as stabilisation ponds, sequence batch reactors, activated sludge, rotating biological contactors and biofilters have been tested (Lim & Kim, 2014). In the present thesis, this issue was addressed using calcium silicate-based filter materials to treat the effluent of the UASB reactor.

4.2. The filter media

4.2.1. Sorbulite

Sorbulite was developed based on the production of AAC, which is a lightweight porous building material composed of a mixture of cement, lime, silica, water and aluminium (Renman & Renman, 2012). Table 3 shows the properties of Sorbulite.
Renman and Renman (2012) reported an adsorption capacity for P of 39.6 mg g\(^{-1}\) and high efficiency of BOD removal. Other studies, e.g. that performed by Lima et al. (2015), indicate that Sorbulite may also be used to remove organic material. The trademarked Sorbulite is manufactured and distributed by the company Ecofiltration AB (formerly Bioptech AB), Sweden.

Few studies have been carried out using Sorbulite as a medium to treat wastewater. However, the results that exist are promising, as they demonstrate that Sorbulite can reduce the concentrations of P and bacteria in wastewater taken from the effluent of a septic tank. Sorbulite is also efficient in removing organic material and N from the same source of wastewater (Nilsson et al., 2013a). In addition, it has been reported that the P captured by Sorbulite can be recovered by a desorption process using HNO\(_3\) (Kassa, 2013).

The mechanisms for N removal are not well studied, but one explanation could be that ammonium is converted to ammonia when it is contact with Sorbulite, due to its high pH. With time, the pH in the Sorbulite decreases and it is possible that microorganisms consuming N are formed at the surface of the Sorbulite particles.

In this study, Sorbulite was mainly used as a filter medium to stop the transport of particulate and dissolved organic material coming from the effluent of a UASB reactor. This protects the Polonite in the PBR and its efficiency and lifespan may be prolonged, since it has been demonstrated that presence of organic material in the Polonite bed can affect the removal of P (Nilsson et al., 2013a).

### 4.2.2. Polonite

Polonite is a calcium-silicate mineral resulting from a thermal process applied to a particular type of the bedrock Opoka. The trademarked Polonite is manufactured and distributed by the company Ecofiltration AB (formerly Bioptech AB), Sweden.

Interest in using Polonite in small to medium-scale wastewater treatment plants is growing, as it has been successfully used to remove P from wastewater and as a soil amendment for crops. The chemical and physical properties of Polonite are presented in Table 4 (Nilsson et al., 2013a). In terms of chemical compounds, Polonite mainly contains silica (SiO\(_2\)) and calcium oxide (CaO), at percentages of 40.2% and 42.6%, respectively.
While the mechanism involved in the removal of P by Polonite is not fully understood, it is almost certainly the case that it involves an adsorption process followed by a reaction between CaO and P present in the wastewater.

The common forms of P in wastewater are orthophosphate, polyphosphate and organic phosphate. Of these, the most relevant in domestic wastewater is orthophosphate because polyphosphate in aqueous solution forms orthophosphate, while the concentration of organic phosphate can be neglected for domestic wastewater (Tchobanoglous et al., 2003). Forms of orthophosphate can include phosphate ($\text{PO}_4^{3-}$), hydrogen phosphate ($\text{HPO}_4^{2-}$), dihydrogen phosphate ($\text{H}_2\text{PO}_4^{-}$) and phosphoric acid ($\text{H}_3\text{PO}_4$). A possible stoichiometric reaction can be:

$$2\text{H}_3\text{PO}_4 + 3\text{CaO} \leftrightarrow \text{Ca}_3(\text{PO}_4)_2 + 3\text{H}_2\text{O}$$ (1)

The product of this reaction is known as amorphous tricalcium phosphate (ATCP). Gustafsson et al. (2008) reported that this compound is formed when Polonite is used to remove P from wastewater. However, they used synthetic wastewater [prepared by mixing ($\text{KH}_2\text{PO}_4$) and ($\text{NH}_4\text{Cl}$) with tap water] to carry out batch experiments. Gustafsson et al. (2008) stated that other compounds that may be formed include hydroxyapatite ($\text{HAp}, \text{Ca}_5(\text{PO}_4)_3\text{OH}_{(s)}$) and dibasic calcium phosphate ($\text{DCP}, \text{CaHPO}_4$), through the stoichiometric equations:

$$5\text{Ca}^{2+} + 3\text{PO}_4^{3-} + \text{H}_2\text{O} \leftrightarrow \text{Ca}_5(\text{PO}_4)_3\text{OH}_{(s)} + \text{H}^+$$ (2)

$$\text{Ca}^{2+} + \text{PO}_4^{2-} + \text{H}^+ \leftrightarrow \text{CaHPO}_4$$ (3)
Most previous research focuses on the behaviour of orthophosphate, since it is the most abundant form of P in domestic wastewater. However, the regulations on discharge of residential wastewater for on-site wastewater treatment plants (OSWTP) in Sweden (Naturvårdsverket, 2006) state that total P must be measured and reported in order to prove that the wastewater treatment plant is efficient. The required percentage removal of total P depends on the zone or municipality in which the treated wastewater will be discharged. The required values can be classified into high level and normal level, which require a removal rate of total P of 90% and 70%, respectively. Another acceptable way of evaluating OSWTP performance is achieving a total P concentration in the effluent of up to 1 mg L\(^{-1}\) and 3 mg L\(^{-1}\) for high and normal level, respectively.

The key factor in complying with regulations on discharge of wastewater is the capacity of the Polonite to remove P. This has been studied applying Langmuir and Freundlich isotherms (Cucarella, 2007). It has been reported that Polonite has very high adsorption capacity, up to 119 mg PO\(_4\)-P g\(^{-1}\) (Brogowski & Renman, 2004). Cucarella (2007) reported a value of 60 mg P g\(^{-1}\), which is much higher than that reported for other minerals such as bauxite (0.35-0.61 mg g\(^{-1}\)), zeolite (0.215 mg g\(^{-1}\)), limestone (0.68 mg g\(^{-1}\)), shellsand (8-17 mg g\(^{-1}\)), Filtralite (2.5-4.5 mg g\(^{-1}\)) and red mud (27 mg g\(^{-1}\)) (Nelin, 2008).

Once the efficiency of Polonite decreases, the captured P in the Polonite bed could be recovered using extraction methods. Few experiments have been carried out to study this issue. According to Kassa (2013), P can be extracted with nitric acid and a less effective extraction can be obtained with an ammonium lactate solution. Those authors also tested sodium bicarbonate and water, but the extraction rate was practically zero. Renman and Renman (2010) reported that the breakthrough in Polonite takes a long time. They performed an experiment using municipal wastewater and after 67 weeks they did not find evidence of a breakpoint for P, since at that time the P concentration in the effluent was 0.2 mg L\(^{-1}\). Cucarella (2007) demonstrated that used Polonite can be directly recycled in the form of soil amendment to crops, so it is not necessary to extract the P in the Polonite bed before it is used in agriculture. An interesting property of used Polonite is that it can be regenerated if a thermal process is applied. However, this topic needs more investigation (Khan, 2007).

It has been also proven that Polonite not only removes P but also pathogens. Nilsson et al. (2013b) reported maximum removal of enterococci of 100%. Likewise, in a previous study Renman et al. (2004) reported that Polonite removes >99% of coliform bacteria. This high removal is attributable to the high pH in the Polonite.

5. MATERIALS AND METHODS

This chapter is divided into two parts, modelling and experimentation. Models were developed to describe the
behaviour of degradation of the substrate, growth of biomass and granule size in the UASB reactor. The main contribution of this chapter is the development of an expression able to predict granule size over time, which was solved in both analytical and numerical ways. In a first attempt to validate the models, data from the literature were used. Then once the practical experiment had been completed, the model was applied to the experimental results.

A model to predict the behaviour of the concentration of P in a PBR filled with the two minerals was also developed. The expression assumes that the source term is described by an adsorption equation.

Both models (UASB and PBR) include terms of dispersion, advection and reaction.

In the experimental part, a system composed of two reactors, an UASB reactor followed by a PBR filled with the two calcium silicate-based minerals, was studied during 350 days. Based on the Swedish regulations on discharge of wastewater from OSWTP, it was decided to study biochemical oxygen demand (BOD$_7$), total N and total P. In addition, analyses of chemical oxygen demand (COD), solids (volatile suspended solids (VSS) and total suspended solids (TSS)) and pathogens (Escherichia coli and Enterococcus faecalis) were performed.

5.1. UASB reactor models

In the following section, the conceptual model for the UASB reactor is explained. The development of an expression to describe the reaction term in the granule is then presented and the model describing the PBR is described. Finally, the geometry and conditions for the practical experiment are described.

5.1.1. Conceptual model

The UASB reactor is viewed as comprising of several CSTRs, all with the same volume, connected in series (Figure 3).

**Fig. 3. Conceptual model of the UASB reactor.**
Each CSTR has the capacity to store a certain amount of biomass ($\varphi_{P_{\text{max}}}$), since in practical experiments the sludge bed is not only composed of biomass, but also of water. $\varphi_{P_{\text{max}}}$ takes into account active and inactive biomass.

Initially, the biomass is distributed in a certain number of CSTRs with a volume fraction of $\varphi_{P_{\text{max}}}$ and one reactor on the top of the sludge bed with the remaining biomass (if any). The other CSTRs have no biomass. During operation of the UASB reactor, the sludge bed expands as the amount of biomass increases. However, the volume fraction ($\varphi_p$) of each CSTR can only reach $\varphi_{P_{\text{max}}}$ at most, with the surplus being passed onto the next CSTR.

The main assumptions used in the model are:

- In the sludge bed, the distribution of granule size is the same at any height in the bed
- No transition zone is considered in the sludge distribution
- The substrate consists of only one biodegradable organic material
- Substrate degradation rate is controlled by a single process (the rate-limiting process)
- The model is one-dimensional and transient; only variations along the height of the reactor are considered
- Granules are spherical in shape.

The governing equations for the UASB reactor to describe the concentration of substrate, active biomass and inactive biomass in each CSTR can be written as:

\[
\frac{dS_i}{dt} = \frac{Q}{V_i} \left( S_{i-1} - S_i \right) - K_i * S_i \tag{4}
\]

\[
\frac{dX_i}{dt} = K_i * S_i * Y - K_d * X_i \tag{5}
\]

\[
\frac{dE_i}{dt} = K_d * X_i \tag{6}
\]

where $S$ is the substrate concentration in the reactor, $t$ is the operating time, $Q$ is the flow rate, $V$ is the volume of the reactor, $K$ is kinetic rate, $X$ is the active biomass concentration, $Y$ is the yield, which is defined as the mass of biomass produced per mass of substrate consumed, $K_d$ represents the decay of active biomass, and $E$ is the concentration of non-active biomass. The subscript $i$ represents the CSTR under analysis. These governing equations apply for the simulation of the UASB reactor in Papers I-III, but the expression describing the kinetic term varies between papers. In Paper I the kinetic expression is solved in an analytical way, while in Papers II and III it is solved in a numerical way.
In equations (4), (5) and (6), the term on the left-hand side corresponds to the accumulation term. The first term in the right-hand side of equation (4) is the advective term, and the last one represents the reaction term. In equation (5), the second term on the right-hand side represents the decay term that describes the inactivation of biomass. The advective terms is not taken into account in equations (5) and (6), since it is assumed that the biomass does not leave the reactor. The dispersive term is accounted in the number of CSTRs \((n)\) into which the UASB reactor is divided, which in turn is a function of Péclet number as shown in equation (7).

\[
n = \frac{Pe}{2} + 1
\]  

(7)

This expression indicates that a system composed of few CSTRs has a low Péclet number, i.e. large dispersion. On the other hand, a reactor divided into several CSTRs has a high Péclet number \((Pe \rightarrow \infty)\), i.e. low dispersion.

Since the concentration of the substrate (i.e. COD) at the effluent of the UASB reactor can be calculated from equation (4), the mass of substrate that remains inside the UASB reactor \((\dot{C}OD_{UASB})\) may be determined by equation (8).

\[
\dot{C}OD_{UASB} = \dot{C}OD_o - \dot{C}OD_e
\]

(8)

where \(\dot{C}OD_{UASB}\) represents the mass of COD removed by the UASB reactor, \(\dot{C}OD_o\) is the mass of the influent concentration of COD and \(\dot{C}OD_e\) represents the mass of COD in the effluent of the UASB reactor. Hence, the volume of methane produced \((V_{CH_4})\) can be predicted, since it corresponds to a fraction of the mass of COD retained in the UASB reactor, as shown in Figure 4.

![Biogas diagram](image)

**Fig. 4.** Representation of the mass balance for COD in the UASB reactor.
From experimental work, the volume of methane produced per mass of COD removed (i.e. $Y_{\text{CH}_4}$) is known. Thus, the volume of methane produced in the UASB reactor can be projected by applying equation (9).

$$V_{\text{CH}_4} = Y_{\text{CH}_4} \text{COD}_{\text{UASB}}$$  \hspace{1cm} (9)

where $V_{\text{CH}_4}$ is the volume of methane produced and $Y_{\text{CH}_4}$ is the yield of methane.

### 5.1.2. Kinetic term-analytical solution

The kinetic rate ($K$) occurring in a granule can be described by equation (10), which is the result of a mass balance of substrate in a granule (Paper I). Figure 5 shows a schematic view of the granule in which the mass balance was applied.

$$K = 3k_m \frac{\varphi_P D_A (\phi \cosh \phi - \sinh \phi)}{R D_A (\phi \cosh \phi - \sinh \phi) + R k_m \sinh \phi}$$  \hspace{1cm} (10)

where $k_m$ is the mass transfer coefficient, $\varphi_P$ represents the volumetric fraction occupied by the granules in the reactor, $R$ is the radius of the granule, $D_A$ is the diffusion coefficient within the granule and $\phi$ is the Thiele modulus, which describes the relationship between reaction rate and the diffusion rate in the granule.

---

**Fig. 5. Representation of a granule.**
Since $\phi_p$ represents the volume occupied by particles relative to the total bed volume, the number of granules per volume of reactor ($N_p$) can be determined as:

$$N_p = \frac{\phi_p}{\frac{4}{3}\pi R^3}$$  \hfill (11)

$K$ is then introduced in the governing equations of the UASB reactor [equations (4) and (5)]. Hence, the value of $K$ is calculated for every CSTR and constantly recalculated as the substrate and biomass change over time. The biomass is redistributed in the CSTR according to $\phi_{p_{\text{max}}}$.

In the governing equations [equations (4), (5) and (6)] the parameters are linked and they are dependent on each other. Thus, the change in granule radius can be determined if the density of the biomass in the sludge bed (bioden) is known. Examples of bioden values are shown in Figure 2 in Paper I. The values can be determined experimentally or using Stokes equation.

With the initial conditions of the UASB reactor, a new value of $X$ can be determined. Therefore, a new value of $\phi_p$ can be found by dividing $X$ per bioden. The volume of the new granule ($V_g$) can then be determined by dividing $\phi_p$ per $N_p$. Because it is assumed that the shape of the granule is spherical and knowing $V_g$, the new radius of the granule can be calculated using equation (12).

$$R^3 = \frac{V_g}{\frac{4}{3}\pi}$$  \hfill (12)

This new $R$ is introduced into $K$, which modifies $S$, $X$ and $E$ in the governing equations. Consequently, the behaviour of the concentration of substrate, concentration of biomass (active and inactive) and change in granule size in the UASB reactor can be predicted.

5.1.3. Kinetic term-numerical solution

By performing a mass balance of the substrate in a granule (Figure 5) and neglecting the external mass transfer coefficient, the reaction term is significantly simplified and can be written as:

$$\mathcal{R} = q \cdot 4 \pi r^2 N_p$$  \hfill (13)

where $q$ is the flux. The number of granules ($N_p$) is a function of the biomass (active and inactive), density and volume of the granule. $q$ in equation (13) is a function of $D_A$ and the gradient of the substrate inside the granule ($S_{p_{\text{grad}}}$), which involves the equation of the mass balance applied to the granule, which in turn is related to the Monod equation. This interaction is shown in a schematic way in Figure 6. It was the focus in Paper II.
Once $K$ is determined, it is included in the governing equation of the UASB reactor [equations (4) and (5)] and then a new iteration is run, which depends on time. In Paper III, a similar model was developed, but it differed in that the Contois model was used instead of Monod. In addition, the model was applied to our own experimental data, while in Papers I and II experimental data from the literature were used to validate the model. A sensitivity analysis was also performed in Papers I and II.

### 5.2. Packed bed reactor model

Modelling of the PBR filled with the two minerals Sorbulite and Polonite (Figure 7) is described in Paper IV. The parameter under study was the concentration of total P.

\[
X_P = f(S_P)
\]

\[
D_A \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial S_P}{\partial r} \right) - \frac{\mu_{\text{max}}}{Y} \frac{X_P}{K_S + S_P} S_P = 0
\]

\[
N_P = f(X_P)
\]

\[
K = q \text{ Area } N_P
\]

**Fig. 6. Interaction of the variables involved in bioconversion of substrate in the granule.**

**Fig. 7. Representation of the packed bed reactor.**

$L_S$: Height of the Sorbulite layer, $L_P$: Height of the Polonite layer.
The ADM was used and it was assumed that the reaction term follows the Langmuir isotherm. So, the governing equation controlling the behaviour of the reactor was:

\[
\frac{\partial P_i}{\partial t_i} = D_i \frac{\partial^2 P_i}{\partial z_i^2} - \nu_i \frac{\partial P_i}{\partial z_i} - \left( q_{\text{max}_i} b_{1i} P_i + b_{2i} P_i \right) P_i \tau_i
\]  

(14)

where \( P \) represents the concentration of total phosphorus, \( D \) is the diffusion coefficient, which is \( f(\text{Pe}, \nu, L) \), \( \nu \) is the upflow velocity, \( z \) represents the direction of the flow, \( q_{\text{max}} \) is the maximum adsorption capacity, \( b \) is the Langmuir coefficient, \( \tau \) is the contact time between the mineral and the water in every mesh solution, and the subscript \( i \) denotes the type of mineral (i.e. Sorbulite or Polonite). The conditions at the end of the Sorbulite layer become initial conditions for the Polonite layer. To perform this simulation, it was necessary to study the kinetics of Sorbulite and Polonite to obtain the kinetics parameters for the Langmuir isotherm.

The simulation was divided into three cases:

1- Simulation of the experiment: The simulation followed the practical experiment. Working conditions during the simulation were the same as in the experimental PBR.

2- Hypothetical cases:
   - The simulation was run under the assumption that the PBR was filled with 20% Sorbulite and 80% Polonite (as in the experiment)
   - The packed bed reactor was assumed to be filled with only Sorbulite
   - The packed bed reactor was assumed to be filled with only Polonite

In the hypothetical cases, the contact time between the mineral and the water was set at 10 min. This was done to obtain transient results and show how the P profile varied along the height of the reactor.

3-Sensitivity analysis: The value of one parameter at a time was varied while keeping the others constant. The simulation was then run and the decrease in the concentration of P was studied. Hence, the importance of the parameter for the model was assessed.

### 5.2.1. UASB and PBR-Practical experiment

A description of the practical experiment concerning the UASB reactor and the PBR is presented in this section.

**UASB reactor**

The practical experiment on the UASB reactor is described in Paper III. The reactor consisted of a transparent cylinder of polymethyl methacrylate with a base of PVC, which contained a series of rings that worked as wastewater distributors across the cross-sectional base area of the reactor (Figure 8).
The height of the reactor was 110 cm and the diameter 10.5 cm. In the upper part of the reactor, a GLSS made of PVC was installed (Figure 9), and the outlet for biogas was located at the top. A pulse water pump was used to introduce wastewater into the UASB reactor with an upflow velocity of 0.11 m h$^{-1}$ in the lag phase. During the experiment (i.e. after acclimatization phase), however, the upflow velocity was increased to 0.18 m h$^{-1}$, since at that point waste sludge already was washed out from the UASB reactor. Wastewater generated by the daily needs of a family composed of four people was first processed in a septic tank, where most of the solids were removed. Wastewater was collected at the outlet of the septic tank and taken to the site where the experiment was performed (KTH laboratory). A timer was used to control the flow of wastewater. It operated for three periods per day (i.e. 8-hour per day), with the aim of imitating wastewater flow in single house conditions.

The hose connecting the water pump and the UASB reactor was fitted with a one-way valve to avoid backflow due to the pressure exerted by the sludge bed and the water in the UASB reactor. The biogas produced was collected in a Mariotte-like cylinder (Figure 10) filled with 10% NaOH solution to purify the biogas.
The biogas inlet was located at the bottom of the Mariotte-like cylinder and while the biogas passed through the liquid, CO₂ was removed, leaving only CH₄. At the same time a slight pressure was applied in the cylinder and the NaOH solution was intentionally leaked through a fine syringe installed in the bottom of the cylinder. In this way, it was possible to measure the volume of biogas produced.

The anaerobic sludge (not granular) used to inoculate the reactor was taken from the anaerobic digester of a large-scale wastewater treatment plant (Henriksdals Reningsverket, Sweden). The TSS concentration was 51263.12 mg L⁻¹, giving a VSS/TSS ratio of 0.82. The height of the sludge bed before the start-up of the reactor was 60% of the height of the reactor.

**Packed bed reactor**

The PBR studied in Papers III and IV was made of the same material as the UASB reactor, with a height of 55 cm and internal diameter of 10.5 cm. Water entered by the bottom, where a water distributor was installed (Figure 11). At a height of 16 cm, a sampling point with diameter 5 cm was installed.

The materials forming the bed were Sorbulite and Polonite with particle size range 2-6 mm. The height of the Sorbulite was 20% of the height of the packed bed, and the remaining 80% was filled.
with Polonite. Figure 12 shows the system composed of the UASB reactor (a) and the PBR (b) used in the practical experiment.

Analytical procedures

Samples from the influent to the UASB reactor (wastewater from the septic tank), the effluent from the UASB reactor (influent to the PBR) and the effluent from the PBR were collected in 100-mL acid-washed plastic bottles once a week and analysed immediately, except for solids and bacteria, which were analysed every second week. The chemical oxygen demand (COD) concentration was determined according to methods CSN ISO 6060 and CSN ISO 15705. The concentrations of BOD$_7$, TSS, VSS and total N were determined according to method CSN EN 1899-1/-2, CSN 757350, CSN EN 872 and CSN EN 12260, respectively. Total P
was determined using the continuous flow analysis technique (Seal Analytical-Auto Analyzer detector). The concentrations of *Escherichia coli* and *Enterococcus faecalis* were calculated according to methods described in SS 028167-2:1996 and EN ISO 7899-2:2000, respectively. A portable meter (Hach Lange HQ40D multi-meter) was used for measurement of pH and temperature.

6. RESULTS AND DISCUSSION

6.1. Kinetic term-analytical solution

The model that involved the analytical expression (Paper I) was validated with data reported by Nacheva et al. (2009), who studied an UASB reactor treating wastewater from a sugarcane mill. Every 60 days, the upflow velocity in the UASB reactor was increased by a factor of two and the removal of the substrate (i.e. COD) was measured. A convenient volumetric substrate conversion rate (k) (accounted for by Thiele modulus) was adjusted to get the same value of substrate removal in the first time interval. However, for the following intervals, the adjusted parameter was kept constant. Comparison of the simulated results with observed values reported by Nacheva et al. (2009) showed good agreement (Figure 13) (Paper I). The highest removal of substrate occurred at the lowest upflow velocity for both the practical experiment and the simulations. This can be attributed to high HRT when the upflow velocity was lower than in other stages. The tendency for removal of substrate in the practical experiment and in the simulation results was similar. In stages 2 and 3, however, the simulation underestimated the practical results, while in stage 4 it slightly overestimated the observed values. In the last stage, the removal of substrate greatly decreased, since the HRT was shorter and washout of biomass occurred when the upflow velocity increased. Consequently, a shorter contact time for substrate degradation and fewer anaerobic microorganisms characterised the UASB reactor, affecting its performance.

In the simulation, the operating time was then increased to two years and the UASB reactor was divided into 16 CSTRs. This was done to show the capability of the model. The operating time was divided into four intervals to show the profile of COD, biomass and granule size over time (Figure 14).

The number of CSTRs into which the reactor was divided is simply the height of the UASB reactor. Initially the degradation of the substrate took place in the first two CSTRs (lower part of the reactor) and as time passed the biomass grew and was rearranged in the CSTRs that were directly above. With this rearrangement, a characteristic curve proposed by Narnoli and Mehrotra (1997) for location of biomass in the UASB reactor was produced. Hence, the degradation of the substrate occurred in the CSTRs filled with biomass, which were located in the lower part of the UASB reactor. The percentage of substrate removal reached 96% in the last interval (Figure 14a).
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![Graph](image1.png)

**Fig. 13.** Comparison between the experimental and model response data.

![Graph](image2.png)

**Fig. 14.** a) Profile of substrate along the height of the reactor, b) active biomass in the sludge bed, c) changes in active, inactive and total biomass over time, and d) variation in granule size over time.
Figure 14b shows how the biomass was propagated with time. It also shows the profile of the concentration of active biomass for the four intervals studied and the increase in the sludge bed over time. Figure 14c shows the profile of the active, inactive and total biomass in the four intervals. The active biomass practically remained constant and the inactive biomass and total biomass grew over time. A higher increase in inactive biomass compared with active biomass has also been reported by Soto et al. (1998), who studied the biomass profile of a laboratory-scale UASB reactor. This behaviour occurred due to decay rate and washout of the biomass. In this hypothetical case, the increase in inactive biomass and a constant active biomass over time meant that the UASB reactor did not work in stationary state; the values of decay constant and washout affected the production of biomass responsible for the degradation of the organic material. However, the active biomass did not decrease and the substrate removal slightly increased over time. Figure 14d shows the change in granule size over time. According to the simulations, the radius of the granule increased from 0.25 mm to 0.38 mm after 2 years (Paper I). This granule growth rate (0.17 µm d\(^{-1}\)) was lower than the range reported by Habeeb et al. (2011), which was 7-31 µm d\(^{-1}\) for different types of granules. However, there was a similar tendency for granule size to increase over time.

6.2. Kinetic term-numerical solution

The model that involved the numerical expression (Paper II) was validated with data reported by Nacheva et al. (2011), who studied an UASB reactor treating slaughterhouse wastewater. The validation process was similar to that applied in the case of the analytical solution (Paper I). The operating condition was divided into four stages, each with different upflow velocity.

In this case, the equation describing the mechanisms occurring in the granules was a function of the Monod model. The yield was adjusted in the first stage in order to get equal removal of substrate and was kept constant for the following stages. A comparison between simulated and experimental results is shown in Figure 15. The simulation results showed good agreement with the experimental results, with the simulated values falling within the range of maximum and minimum values measured in the experimental part. The percentage of substrate removal tended to increase from one stage to another. This was attributed to the increase in the upflow velocity from one stage to other, since it improved the interaction area between substrate and microorganisms. This was mathematically demonstrated by Korsak (2011), who studied the effect of upflow velocity on removal of substrate in anaerobic granules of UASB reactors. As in the analytical model, the numerical model was able to predict the profile of the substrate along the UASB reactor, the expansion of the sludge bed, the concentration of biomass over time and the change in size of the granules over time.
A sensitivity analysis was performed for both the analytical and the numerical model. The percentage removal of substrate was studied as one parameter at a time was varied while the others were held constant. The number of CSTRs into which the UASB reactor was hypothetically divided \((n)\), the diffusion coefficient \((D_A)\), the maximum percentage of solids that a CSTR could contain \((\phi_{P_{max}})\), the mass transfer coefficient \((k_m)\) and the kinetic constant \((k)\) were the parameters analysed for the analytical model (Paper I). For the numerical model, however, \(n\), \(D_A\), \(\phi_{P_{max}}\) and \(Yield\) were analysed (Paper II).

It was found that \(n\) had the same effect in both models, with the removal of substrate increasing with the number of CSTRs used to describe the performance of the UASB reactor. Large dispersion caused a reduction in percentage substrate removal. However, in the analytical model the effect was slightly higher, since it went from 88% to 97% for a number of CSTRs equal to 8 and 40, respectively (Paper I). The numerical expression ranged from 93 to 98% for the same values of \(n\) (Paper II).

High values of \(D_A\) did not affect removal of the substrate, but it was strongly affected by low values in both the analytical and numerical expression (Papers I and II). For both models there was no major difference in percentage substrate removal when \(\phi_{P_{max}}\) was varied. This was because the biomass is always the same in the UASB reactor under study, and \(\phi_{P_{max}}\) affects only the height of the sludge bed. For values of \(\phi_{P_{max}}\) in the range 15-40%, removal of substrate was 96-90% for the analytical solution. For the numerical solution, the range was 97-94% for \(\phi_{P_{max}}\) of 18-40%. This indicates that the analytical expression had slightly
higher sensitivity than the numerical expression when $\phi_{\text{P}_{\text{max}}}$ was the parameter varied.

$k_m$ and $k$ had the same effect on percentage removal of substrate for the analytical expression. At low values, the efficiency of the UASB reactor decreased, while at higher values the efficiency increased.

The effect of Yield in the numerical expression played an important role due to the high sensitivity of the model response. Low values of this parameter indicated low activity of microorganisms, and consequently removal of the substrate was low. On the other hand, when Yield took higher values, the removal of the substrate increased significantly. For instance, with a yield value of 0.025 and 0.1, the removal of substrate reached 30% and 98%, respectively.

Despite the fact that the studies carried out by Nacheva et al. (2009) and Nacheva et al. (2011) were similar, they obtained opposing results. In both studies, the performance of a UASB reactor where the upflow velocity increased from stage to stage was evaluated. In the first study the percentage removal of the substrate decreased from one stage to the other (Nacheva et al., 2009; cf. Paper I). In their second study, however, it increased (Nacheva et al., 2011; cf. Paper II). This probably occurred since in the first study the inoculated biomass was non-granulated, while in the second study the UASB reactor was inoculated with granular biomass. Hence, the effect of upflow velocity in the first study was negative, since it induced washout of biomass from stage to stage. In the second study, the effect of increasing upflow velocity was positive, since it improved the mass transfer of substrate from the liquid bulb to the granule (Korsak, 2011). Moreover, due to the fact that dense granules were already formed, no significant washout occurred.

### 6.3. Practical experiment and its simulation

This section summarises the results presented in Papers III and IV. The results are divided into two sections. The first concerns the experimental part, which studied the performance of the processes in the system (i.e. UASB reactor and PBR). The results focus on: BOD$_7$, COD, pH, total N, total P, TSS, VSS, *Escherichia coli*, *Enterococcus faecalis* and granule size.

The second part corresponds to the application of the model to simulate the behaviour of the UASB reactor (Paper III) and the PBR (Paper VI). The parameters under study were: COD, solids and granule size for the UASB reactor, and the concentration of total P for the PBR. Comparisons between experimental and simulated results and sensitivity analysis were also performed.

#### 6.3.1. Experimental results

Table 5 shows the average values obtained in the experiment and their corresponding standard deviation (Paper III). On average, the efficiency of BOD$_7$ removal for the UASB reactor and the PBR was 88% and 89%, respectively. The whole system achieved a removal rate for BOD$_7$ of 99%.
The profile of BOD$_7$, over time for both reactors and the whole system is shown in Figure 16a. Similar behaviour was obtained for the COD, with an observed average removal rate of 89% and 86% for the UASB and PBR, respectively. This corresponds to a removal rate of 98% for the whole system. The removal of organic material in the PBR was mainly attributed to the action of Sorbulite (Paper III). The mechanisms behind organic matter removal in Sorbulite remain to be investigated. However, Chen et al. (2015) reported that the removal of organic material when autoclaved aerated concrete is used could be explained by the high porosity of this material. Likewise, those authors found microbial activity in the pores, which may consume not only organic material but also nutrients.

The pH did not show significant changes in the UASB reactor. In the PBR, however, the pH went from neutral to basic (Figure 16b). This increase in pH was due to the high calcium content in both Sorbulite and Polonite. The high pH in the effluent can work as a buffer to tackle problems of acidification in water reservoirs (Paper III).

The removal of total N was low in both the UASB (18%) and the PBR (27%), reaching a removal rate of 40% for the whole system. An explanation for the consumption of N in the UASB reactor could be that anaerobic microorganisms use it for their reproduction and respiration (Rodríguez-Gómez, 2011). In the PBR, the N removal could be explained by development of bacteria consuming N in the Sorbulite bed, as reported by Nilsson et al. (2013b).

### Table 5. Experimental results and standard deviation*

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Inlet</th>
<th>Outlet UASB</th>
<th>Outlet (SP)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOD$_7$, mg L$^{-1}$</td>
<td>269.08 (40.94)</td>
<td>34.22 (17.30)</td>
<td>3.28 (1.07)</td>
</tr>
<tr>
<td>COD, mg L$^{-1}$</td>
<td>475.66 (71.53)</td>
<td>52.53 (25.46)</td>
<td>7.11 (1.70)</td>
</tr>
<tr>
<td>pH</td>
<td>7.42 (0.25)</td>
<td>7.52 (0.22)</td>
<td>11.34 (0.41)</td>
</tr>
<tr>
<td>Total N, mg L$^{-1}$</td>
<td>62.28 (4.81)</td>
<td>51.11 (4.31)</td>
<td>37.21 (3.31)</td>
</tr>
<tr>
<td>Total P, mg L$^{-1}$</td>
<td>12.77 (1.49)</td>
<td>10.00 (1.09)</td>
<td>0.17 (0.07)</td>
</tr>
<tr>
<td>TSS, mg L$^{-1}$</td>
<td>53.63 (5.10)</td>
<td>22.75 (2.86)</td>
<td>3.02 (0.59)</td>
</tr>
<tr>
<td>VSS, mg L$^{-1}$</td>
<td>14.00 (1.96)</td>
<td>7.79 (0.94)</td>
<td>0.89 (0.15)</td>
</tr>
<tr>
<td><em>Escherichia coli</em>, CFU (100 mL)$^{-1}$</td>
<td>160960.00 (21597.61)</td>
<td>150120.00 (17159.35)</td>
<td>62.94 (31.48)</td>
</tr>
<tr>
<td><em>Enterococcus faecalis</em>, CFU (100 mL)$^{-1}$</td>
<td>62880.00 (11395.61)</td>
<td>52320.00 (9136.56)</td>
<td>19.51 (9.46)</td>
</tr>
</tbody>
</table>

*Numbers in brackets refer to standard deviation.
On the other hand, the removal of total P was very high in the system, reaching 99%. This was due to the action of Sorbulite and Polonite; the removal of total P in the PBR was 98%, whereas it was only 21% in the UASB reactor. These results are shown in Figures 16c and 16d, respectively (Paper III). The mechanism responsible for P removal in the UASB is biological assimilation (Ping et al., 2011). The mechanisms behind P removal in Polonite and Sorbulite are well documented as calcium phosphate precipitation and crystallisation (e.g. Gustafsson et al., 2008).

The TSS concentration in the incoming wastewater was 54 mg L\(^{-1}\), of which 58% was removed in the UASB reactor and 87% in the PBR. Therefore a total removal rate of 94% was achieved in the system (Figure 17a).

The removal of VSS presented similar behaviour (Figure 17b); 44% and 89% in the UASB reactor and PBR, respectively. The total removal of VSS in the system was 94%. The VSS/TSS ratio in the sludge bed of the UASB reactor was stable during the experiment, with an average value of 0.74 and a minimum and maximum value of 0.71 and 0.81, respectively.

No clogging problems were observed during the experiment, but solids were accumulated in the Sorbulite bed, as shown in Figure 18 (Paper III).
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Fig. 17. Experimental results: a) Total suspended solids (TSS), b) volatile suspended solids (VSS), c) Escherichia coli and d) Enterococcus faecalis.

Fig. 18. Accumulation of solids in the bottom of the Sorbulite bed.
The concentrations of *Escherichia coli* and *Enterococcus faecalis* both showed a removal rate of 99%. According to Geeraerd et al. (2014), this occurs due to the fact that these pathogens cannot live under high pH. Changes in the concentrations of bacteria are shown in Figures 17c and 17d (Paper III).

Previous research by Nilsson et al. (2013a, 2013b) also demonstrated the capacity of Polonite and Sorbulite to reduce bacteria during wastewater filtration. However, their experiments were performed under high organic loading or extreme wastewater discharge, which resulted in a much lower bacteria reduction. The design flow used in our system was able to control the pathogenic bacteria concentration at below 100 CFU 100 mL⁻¹, which is much lower than the EU limit for surface water of bathing water quality (Directive 2006/7/EC of the European Parliament and of the Council of 15 February 2006).

Concerning the granules, biomass was not granulated at the beginning of the process. As time passed, the biomass attached to itself, forming irregularly shaped aggregates. These aggregates ranged in size from fine particles to 0.4 mm in diameter. Photographs of the granulation process are presented in Figure 19, which shows (a) the sludge used to inoculate the UASB reactor, (b) irregularly shaped and fine particles above the sludge bed after 1 month of operation, (c) drag effects in the biomass induced by the biogas, and (d) granules at the end of the experiment; these particles were washed and selectively chosen. The water pump used in this study played an important role for the granulation. It sent pulses of water, which induced agitation of particles in the sludge bed (Paper III).

**Fig. 19.** Granulation processes during the experiment. a) Seed sludge, b) after 1 month, c) drag of biomass due to biogas, and d) at the end of the experiment.
As a consequence of the paths created by biogas, channelling and stagnant biogas were observed in the sludge bed (Figure 20a). It has been reported that channelling is a problem since the HRT is reduced due to preferential flow (Rodríguez-Gómez, 2011). However, it was also observed that with time the channelling disappeared due to the effect of collisions between particles caused by the upflow velocity and the buoyancy force of the biogas. Similarly, stagnant biogas was constantly released due to fine bubbles of biogas on their way upwards colliding and releasing the stagnant biogas bubbles.

Due to the collision of biogas bubbles, they merged with each other and their buoyancy force increased. This allowed breaking of the barrier formed by the particles in the sludge bed and the bubbles made their way to the top of the UASB reactor. From time to time, samples of biogas were taken and burned to ensure it was flammable. The biogas was probably methane (Figure 20b).

The Swedish regulations on discharge of water from household recommend removal of BOD$_7$, total P and total N at a rate of 90%, 90% and 50%, respectively, or to a concentration at the outlet of 30 mg BOD$_7$ L$^{-1}$, 1 mg total P L$^{-1}$ and 40 mg total N L$^{-1}$, respectively (Naturvårdsverket, 2006). Based on the results of our experiment, the regulations on BOD$_7$ and total P removal were fulfilled. For total N, the removal in the experiment was 10% less than the recommended rate (in percentage terms). However, the concentration of total N in the effluent of the experimental system was less than that recommended by the regulations and hence the regulatory requirements also were fulfilled for N. Figure 21 shows a schematic representation of the experimental system and the concentration of parameters measured in each reactor.
6.3.2. Simulation

UASB reactor

The model that involved the numerical solution (Paper II) was used to simulate the practical experiment. However, the kinetic was assumed to follow the Contois model (Paper III), since it has been reported that this is a more accurate representation for residential wastewater (Donoso-Bravo et al., 2013). This model was also chosen because it requires fewer variables than the analytical solution. The operating time of the UASB reactor was 50 weeks. Results were plotted every 10 weeks, except for the granule size, for which data were presented every 2.5 weeks. The parameter studied in the simulation was the organic material (measured as COD). Batch culture experiments were performed to study the kinetics of the sludge. Contois kinetic parameters were determined as: \( \mu_{\text{max}} = 0.109 \text{ d}^{-1} \), \( B = 0.486 \), \( Y = 0.13 \), and \( K_d = 0.0068 \text{ d}^{-1} \).

The parameters used to run the simulation were taken from the practical part, except for the initial radius of the granule. It was assumed to be 1.00E-4 m, since the biomass inoculated in the reactor was in the form of sludge (i.e. non-granulated). Likewise, the diffusion coefficient was assumed to be 3.00E-7 m\(^2\)h\(^{-1}\), which corresponds to the value used by Rodríguez-Gómez et al. (2013) in a simulation of a UASB reactor. Table 6 shows the parameters used during the simulation.

The concentration of COD at the outlet of the UASB reactor decreased with time, while the same behaviour was observed in the experimental part. During the first time interval, the removal of COD was lower in the simulation (63%) than in the experiment (76%). Gradually, the removal of COD in the simulation increased until it reached 91% in the last interval; this value was slightly higher than that obtained in the experiment (i.e. 89%). Figure 22a
Table 6. Parameters used for simulation of the UASB reactor

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Height of the UASB reactor, m</td>
<td>1.100</td>
</tr>
<tr>
<td>Diameter of the UASB reactor, m</td>
<td>0.105</td>
</tr>
<tr>
<td>Initial COD, mg L(^{-1})</td>
<td>476</td>
</tr>
<tr>
<td>Operation time, d</td>
<td>350</td>
</tr>
<tr>
<td>Upflow velocity, m d(^{-1})</td>
<td>1.44</td>
</tr>
<tr>
<td>Flow rate, m(^{3}) d(^{-1})</td>
<td>0.0125</td>
</tr>
<tr>
<td>Initial radius of the granules, m</td>
<td>1.00E-04</td>
</tr>
<tr>
<td>Diffusion coefficient, m(^{2}) h(^{-1})</td>
<td>3.00E-07</td>
</tr>
<tr>
<td>Initial VSS in sludge bed, mg L(^{-1})</td>
<td>33934.709</td>
</tr>
<tr>
<td>Density of the biomass, mg L(^{-1})</td>
<td>1026000</td>
</tr>
</tbody>
</table>

Figure 22b shows a comparison of the predicted concentration of VSS in the sludge bed based on the simulation and the values obtained during the experiment. The concentration of VSS was the object of study because it was assumed that the VSS represents the microorganisms responsible for degradation of the substrate. The concentration of VSS in the sludge bed was underestimated by the simulation. However, a similar increase in the concentration of VSS in every interval was observed for both the simulation and the experiment. The TSS can easily be determined because of the assumption \( \frac{\text{VSS}}{\text{TSS}} = 0.74 \), which was the average ratio value at the end of the experiment (Paper III).

The simulation revealed the profile of expansion of the sludge bed in the reactor. In this case the value of \( \varphi \) was 10\% and was determined experimentally. Figure 22c shows the profile of the sludge bed with time. At the beginning, eight CSTRs contained biomass. The height of the sludge bed expanded as reproduction of microorganisms occurred, reaching 8.5 CSTRs with biomass during the simulation. At the end of the experiment, however, the height of the sludge bed was 66 cm, which corresponds to 9.6 CSTRs with biomass.

Despite the fact that there were no granules when the experiment started, an initial granule radius of 0.1 mm was assumed in the simulation. The growth rate of the granules in the simulation was lower than that in the experiment. Figure 22d shows the variation in granule radius with time. The final radius of the granule in the simulation was 0.12 mm, whereas granules of radius up to 0.2 mm were observed in the experiment (Paper III). To predict the biogas (measured as methane) production in the UASB reactor, equation (9) was applied. In the simulation, the \( \text{COD}_0 \) was obtained from the average value of the influent concentration of COD.
Concerning the $Y_{\text{CH}_4}$, the average value determined during the experiment was 0.20 (Paper III). This value is in the typical range for municipal wastewater (i.e. 0.18-0.22) (Khan et al., 2015). Likewise, the $Y_{\text{CH}_4}$ was slightly lower than that reported by Borggren (2007), who studied biogas production from the anaerobic digester located in the wastewater treatment plant from where the anaerobic sludge was taken in this study.

Figure 23a shows a comparison of the volume of cumulative methane produced and simulated in the UASB reactor at 10-week intervals (Paper III). It was observed that during the experiment the production of methane increased during weeks 10, 20 and 30, but it decreased in week 40 and then remained almost constant. This can be attributed to the change in temperature experienced by the reactor, which was within the range 18-27°C during weeks 10, 20 and 30 and 16-22°C during weeks 40 and 50. However, the removal of COD was not affected by the change in temperature, as shown in Figure 22a.

The same behaviour has been reported by Lew et al. (2004), who observed high removal of COD but low production of methane when the temperature decreased in an UASB reactor. The low production of methane occurs due to inactivation of methane-producing bacteria when the temperature decreases and high removal of COD due to retention of solids in the sludge bed (Lew et al., 2004). Besides decreasing the activity of methanogenic bacteria, it has also been reported that methane production is affected by resistances that the substrate experiences during its transport to the core of the granule (Korsak, 2011). Hence, the production of methane could be affected because methanogenic bacteria were located in the core of the granule (MacLeod et al.,...
1990) for the last two intervals studied (weeks 40 and 50). In weeks 10, 20 and 30, the methanogenic bacteria were dispersed in the sludge bed, since granules had not yet formed.

The simulated production of biogas showed an increasing tendency from one interval to the next. However, the simulated values underestimated the actual values in the first 30 weeks, while for the last 20 weeks the values were overestimated. This occurred because in the simulation the change in temperature was not accounted for, indicating that this parameter is important in predicting the volume of production of methane. Moreover, in the simulation the influent concentration of COD and $Y_{CH_4}$ were assumed to be constant, while in the experiment their values varied.

The profile of the volume of methane produced weekly is shown in Figure 23b (Paper III). The $Y_{CH_4}$ in the experiment ranged from 0.11-0.32 m³CH₄ (kgCODrem)⁻¹

Figure 23a also shows the trends in biogas production taken as $Y_{CH_4}$, the average value obtained from the experiment. Moreover, it shows the temperature profile, from which it is apparent that
methane production is temperature-dependent. This is in agreement with Zhao (2011), who studied the production of biogas at different temperatures in an anaerobic digester.

**Packed bed reactor**

The expression used to simulate the PBR is presented in equation (14). The behaviour of the concentration of P was studied and the simulation was divided into three cases (Paper IV):

1- Simulation of the experiment: The simulation followed the practical experiment. Working conditions during the simulation were the same as in the PBR.

2- Hypothetical case: The simulation was run under the assumption that the PBR was filled with: 20% Sorbulite and 80% Polonite (as in the experiment, but HRT=10 min), only Sorbulite, and only Polonite. This provided information about how the minerals and their combination affected the profile of P along the reactor height.

3- Sensitivity analysis: The value of each parameter was varied while keeping the others constant. The simulation was then run and the removal of P was studied. Hence, the importance of different parameters in the model was identified.

**Comparison between simulation and practical experiment**

To validate the model, a simulation was run using the same operating conditions as in the practical experiment (Table 7). In addition, the operating time was divided into four intervals to show the variation in P over time.

<table>
<thead>
<tr>
<th>Description</th>
<th>Sorbulite</th>
<th>Polonite</th>
</tr>
</thead>
<tbody>
<tr>
<td>Height of the reactor, m</td>
<td>0.110</td>
<td>0.440</td>
</tr>
<tr>
<td>Reactor radius, m</td>
<td>0.053</td>
<td>0.053</td>
</tr>
<tr>
<td>Operation time, d</td>
<td>350</td>
<td>350</td>
</tr>
<tr>
<td>Flow rate, m$^3$ d$^{-1}$</td>
<td>0.0125</td>
<td>0.0125</td>
</tr>
<tr>
<td>Upflow velocity, m d$^{-1}$</td>
<td>1.44</td>
<td>1.44</td>
</tr>
<tr>
<td>Péclet number</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>Initial P conc., mg L$^{-1}$</td>
<td>10</td>
<td>P at $z = 0.11t$</td>
</tr>
<tr>
<td>$q_{max}$ , mg g$^{-1}$</td>
<td>1.67</td>
<td>2.18</td>
</tr>
<tr>
<td>$b$ , L g$^{-1}$</td>
<td>1.49</td>
<td>25.43</td>
</tr>
</tbody>
</table>

Due to the fact that the effectiveness of Polonite varies depending on its use, a kinetic experiment for the Polonite was performed during every interval. To do this, a sample of Polonite was taken 5 cm above Sorbulite level and a Langmuir experiment was performed to determine the adsorption capacity using HRT = 10 min for the Polonite in that interval. The values of adsorption capacity used were 2.18 mg g$^{-1}$, 1.45 mg g$^{-1}$, 0.99 mg g$^{-1}$ and 0.64 mg g$^{-1}$ and the intervals were 88 d, 175 d, 263 d and 350 d,
respectively. Because the amount of Sorbulite in the PBR was limited, the kinetic experiment was not performed for the different intervals. Hence, in the simulation it was assumed that the kinetics kept their initial condition.

Figure 24 shows the percentage P removal for both the simulated and experimental cases during the different intervals. The simulation overestimated the average values of the experimental results for the first three intervals. For the last interval, however, the simulation underestimated the experimental results.

Nevertheless, the simulated results fitted within the maximum and minimum range of the experimental results. As time passed, percentage P removal gradually decreased, giving average values of 98.74%, 98.65%, 98.38% and 98.31% for the four intervals. During the whole experiment, the maximum and minimum percentage removal of P was 99.35% and 96.81%, respectively.

![Fig. 24. Comparison of simulated and experimental results.](image)

**Hypothetical cases**

The simulation was run using parameters as in the practical experiment with the exception that HRT was set at 10 min. This was done to show the transient state of the concentration of P along the height of the reactor.

Figure 25 shows the profile of the P concentration along the reactor height. The removal of P was 55% when it was assumed that the reactor was filled with only Sorbulite. On the other hand, when it was assumed that it was filled with only Polonite, the removal was 96%. In the case where the reactor was filled with Sorbulite and Polonite, the curve showed poor removal in the Sorbulite phase (i.e. 9%) and most of the P was removed due to Polonite, which achieved removal of 95%. However, this does not mean that Sorbulite cannot be used to remove P, but a higher HRT is needed to get significant percentage removal of P. The
purpose of Sorbulite in this experiment was not to remove P, but to remove organic material and solids from the incoming wastewater.

Sensitivity analysis for the PBR

A sensitivity analysis was performed on HRT, D, adsorption capacity and height of the packed bed in the PBR. While these values were varied one at a time, the other parameters in the simulation were held constant. The percentage P removal was the key parameter in the sensitivity analysis. During the analysis, it was assumed that the PBR was filled with only Polonite. The exception was when the height of the packed bed was tested, in which case the total height of the reactor was 0.55 m, but it was assumed that the reactor was filled with Sorbulite and Polonite at different levels.

Figure 26 shows the simulation response to the sensitivity analysis. Figure 26a shows the effect of HRT on removal of P, i.e. the longer the HRT the greater the removal of P. At low values of HRT the variation in the removal of P was significant. On the other hand, when HRT took high values, the percentage removal of P did not change significantly. Figure 26b shows the variation in percentage removal of P at different values of dispersion coefficient. At relatively high values, the percentage removal of P decreased and at low values it tended to remain constant. The value of the dispersion coefficient of particles in a PBR is normally in the order of 1E-9 m²s⁻¹. This means that high values of dispersion coefficient are unlikely. Figure 26c shows the variation in percentage removal of P at different values of maximum adsorption capacity. This parameter was important when it took low values, but at high values the removal of P became constant.
Figure 26c also shows the effect of the Langmuir adsorption constant; low values decreased the effectiveness of P removal, but as the constant increased the effectiveness of P removal also increased until a steady profile was reached. Figure 26d shows the profile of percentage removal of P at different heights of the packed bed (i.e. Sorbulite and Polonite). It was assumed that Sorbulite was placed under Polonite. Thus, when the height of Sorbulite was 0.15 m, the height of Polonite was 0.4 m. The percentage removal of P was 85% for Sorbulite and 98% for Polonite with that specific arrangement. On the other hand, when the height of Sorbulite was 0.4 m and the height of Polonite was 0.15 m, the percentage removal of P was 94% for Sorbulite and 89% for Polonite. Figure 26d also provides a comparison between Sorbulite and Polonite at the same height, which shows that Polonite was more efficient in removing P than Sorbulite. This was explained by the higher adsorption capacity of Polonite compared with Sorbulite. In both cases, however, the height of the packed bed was more sensitive when it had low values. With high values, the percentage removal of P tended to be constant, with values close to 100%.

6.4. Technology outlook

In this thesis, a novel system composed of a UASB reactor and a PBR to treat residential wastewater was successfully tested in bench-scale experiments. However, other researchers have proposed the use of anaerobic reactors coupled with other processes to treat wastewater. Verstraete et al. (2013) applied a microbial electrolysis cell (MEC) to produce biogas in a septic tank.
where blackwater was collected. The system was able to remove 85% of COD, 90% of suspended solids and 68% of total phosphorus. For nitrogen, however, it did not achieve removal since the concentration in the influent and effluent were of the same order of magnitude. The same technology has been tested in UASB reactors (e.g. UASB-MEC) (Verstraete et al., 2013). The use of this system improved biogas production, but electricity was needed to induce the generation of hydrogen and/or methane.

The activated sludge process has been used to treat effluent of UASB reactors (Tawfik et al., 2008). The results show very good removal of organic material and solids, but poor removal of pathogens. A comparison of the results from this work and the system studied by Tawfik et al. (2008) shows that the use of a PBR filled with Sorbulite and Polonite to treat effluent of UASB reactors has advantages over the activated sludge process. For example, the PBR does not consume energy since aeration is not needed, and the PBR is able to remove pathogens from wastewater due to the high pH, usually ranging between 9 to 12 in Polonite. Vinnerås (2007) used 3% ammonia to increase pH above 9 in faeces and manure and achieved a good reduction in the indicator organisms for bacteria.

Other technologies which use the UASB reactor as a primary treatment and another system for post-treatment are summarised in Chong et al. (2012). Examples of systems combined with UASB reactors include: UASB-sequencing batch reactors, UASB-trickling filters, UASB-downflow hanging sponges, UASB-stabilising ponds, UASB-rotating biological contactors, UASB-constructed wetlands, etc.

7. Conclusions

Three models were developed and validated in this work. Two of these models were able to predict the behaviour of the concentration of substrate and biomass and the change in granule size in UASB reactors. The third model was able to predict the concentration of phosphorus along the height of a packed bed reactor filled with Sorbulite and Polonite. In addition, a practical experiment using a system composed of an UASB reactor followed by a PBR was carried out. The conclusions drawn from the work presented in this thesis are as follows:

- The analytical model developed successfully described the performance of an UASB reactor treating sugarcane mill wastewater. The analytical expression described the reaction within the granule, which takes into account mass transfer through the liquid film around the granule, intra-diffusion and specific reactions within the granule.

- The numerical expression developed described the performance of the UASB reactor focusing on degradation of the substrate, biomass concentration, height of sludge bed and granule size. The simulated and experimental results showed very good agreement. The numerical expression can be solved...
Closing the loop by combining UASB reactor and reactive bed filter technologies for wastewater treatment: modelling and practical approaches

using a Monod or Contois model to describe the bioconversion of substrate within the granule.

- Sensitivity analysis revealed that the diffusion coefficient within the granule and the yield parameter play an important role in the performance of the UASB reactor. It also revealed that the height of the PBR and its filter media and the HRT are important for achieving high P removal.
- A bench-scale system composed of a UASB reactor followed by a PBR filled with Sorbulite and Polonite was operated successfully, achieving high BOD₇, TSS and total P removal, as well as a reduction in pathogenic bacteria to bathwater quality. Moderate total N removal was accompanied by a single benefit of the UASB, the formation of combustible gas.
- The combination of UASB reactor and reactive filter media in the PBR should be run in full-scale operation to test the possibility of closing the loop.

8. Future research

This thesis demonstrated that an UASB reactor followed by a PBR filled with Sorbulite and Polonite can be used to treat residential wastewater. Future research should concentrate on:

- Studying the proposed system in pilot, industrial and on-site, single-house scale
- Studying the system under different operating conditions
- Studying the production of biogas and how it can be utilised
- Analysing economic aspects of the system

Since mechanisms for P removal in the PBR are not fully understood, it is also necessary to perform more studies on this topic. Moreover, analyses to reveal the mechanisms involved in the removal of organic material, pathogens and N in Polonite and Sorbulite must be addressed. Chemical modelling applying e.g. COMSOL Multiphysics could assist in further technical development of commercial solutions.

The current models for the UASB reactor can be improved by studying the growth rate of granules of different sizes. Similarly, a model to predict the production of biogas can be developed.

9. References


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