INTRODUCTION

1. INTRODUCTION

Wet sanitation systems connected to sewage treatment facilities are currently considered the gold standard for urban sanitation in high-income countries. However, approximately 670 million people are estimated to still engage in open defecation, and around 4.2 billion lack access to secure sanitation systems (Tsalis et al., 2020). On average, a standard flush toilet consumes about 5 L of water per flush, and it is estimated that each person in high-income countries consumes around 15,000 L of water annually to flush away 500 L of urine and 35 kg of faeces (Lamichhane, 2007). Significant energy and chemicals are required to remove nutrients present in the human waste that is flushed away at municipal wastewater treatment facilities (Maurer et al., 2006).

Despite the type of sanitation system, most of the nutrients, energy and water in human waste are not recovered. In light of the paradigm shift towards a circular economy in the water sector, innovative strategies for recycling nutrients from household wastewater with a specific focus on source-separated human urine are necessary. Source-separated sanitation systems, which divide urine and faeces, can effectively recycle valuable nutrients from human waste (Malisie et al., 2007). Source-separated sanitation systems can effectively recycle valuable nutrients, recovering up to 91% N, 83% P and 59% K (Vinnerås & Jönsson, 2002). The recovered nutrients can be used as fertilizers, reducing the need for chemical fertilizers and promoting sustainable agriculture (Xu et al., 2017). Moreover, source-separated sanitation systems have been found to possess a lower carbon footprint and higher nutrient recovery compared to traditional systems (Kjerstadius et al., 2017). Dehydrating treated urine in a circular system can reduce energy demand and some heat and water can be recovered. Urine source separation involves the separation of urine from other waste streams at the point of origin, which allows for the efficient capture and recovery of valuable nutrients. This approach is gaining recognition as a transformative strategy for sustainable waste management and environmental conservation. Urine source separation offers a range of benefits, including the reduction of nutrient pollution, enhancement of agricultural productivity, and fostering of economic growth (Wilsenach & Van Loosdrecht, 2004). However, problems associated with source-separating sanitation systems need to be solved for these technologies to be mainstream.

1.1. THE NEED FOR SOURCE SEPARATION OF URINE

The primary environmental advantage of urine source separation is the substantial reduction in nutrient pollution. Urine contains a significant portion of the nitrogen, phosphorus, and potassium (NPK) found in human waste, which are essential for plant growth (Jönsson et al., 1997). In conventional sewage systems, these nutrients often end up in water bodies, causing eutrophication, a detrimental process characterized by excessive nutrient levels that lead to rapid algae growth, depleting oxygen, and severely impacting aquatic ecosystems (Martin et al., 2022). By separating urine at the source, these nutrients can be captured and repurposed as fertilizers, thus minimizing nutrient runoff and reducing the need for synthetic fertilizers. This practice not only helps protect water bodies and aquatic life but also lowers the carbon footprint associated with the production of synthetic fertilizers, which are both energy-intensive and a significant source of greenhouse gas emissions (Martin et al., 2022). Urine source separation also enhances the efficiency of wastewater treatment systems. Traditional treatment processes are often energy-intensive and struggle to remove nutrients effectively. By separating urine at the source and treating it separately from faeces, the volume of wastewater requiring treatment is significantly reduced, leading to a more efficient and cost-effective treatment process (Wilsenach & Van Loosdrecht, 2004).

Additionally, the nutrients recovered from urine can be used to enhance the nutrient value of the soil, promoting healthier plant growth and reducing the need for synthetic fertilizers (Kishor et al., 2020). This approach also has the potential to improve the overall sustainability of agriculture by reducing the environmental impact of fertilizer production and use. Urine-derived fertilizers are rich in essential nutrients required for plant growth, providing a sustainable and eco-friendly alternative to synthetic fertilizers (Kishor et al., 2020). Synthetic fertilizers not only demand substantial energy but also depend on finite resources such as phosphate rock. Utilizing urine as a fertilizer can enhance soil fertility while conserving these non-renewable resources. This approach supports the concept of a circular nutrient economy, where nutrients are recycled back into the agricultural system rather than being lost as waste (Karak & Bhattacharyya, 2011). The implementation of urine source separation has the potential to significantly benefit urban and rural areas (Karak & Bhattacharyya, 2011). In densely populated regions, this practice can alleviate the strain on sewage treatment facilities, enhancing their operational efficiency and sustainability. By reducing the volume of waste that needs to be treated, urine separation contributes to the development of a more resilient and adaptable sanitation infrastructure capable of accommodating the demands of growing populations and addressing the challenges posed by climate change.

Economically, urine separation can lead to significant cost savings for farmers by reducing their reliance on expensive synthetic fertilizers (Sohn et al., 2023). Additionally, the decreased burden on sewage treatment facilities can lower operational costs for municipalities (Wilsenach & Van Loosdrecht, 2004). The potential to develop commercial products from treated urine, such as nutrient concentrates, can open new revenue streams and stimulate economic growth in both urban and rural areas. These economic benefits underscore the viability of urine source separation as a financially sustainable practice that can support long-term agricultural productivity and rural development.

Health improvements are another critical aspect of source-separated sanitation (Feachem et al., 1983). Traditional sewage systems can be vectors for disease transmission, especially in regions with inadequate infrastructure. Urine which is typically sterile is a low-risk fertilizer due to low pathogen content.(Fuhrmeister et al., 2020). Effective urine source separation and treatment are crucial for preventing pharmaceuticals and other pollutants from entering water bodies and potentially impacting human health through the water supply. This is particularly important in areas where water resources are scarce and the quality of drinking water is a significant public health concern.

Advancements in sanitation technology are necessary to successfully implement urine source separation. Innovative toilet designs, such as urine-diverting dry toilets (UDDTs) and vacuum toilets, are vital in facilitating the separation process (Deka et al., 2022). UDDTs, for example, separate urine from faeces at the source, enabling efficient collection and treatment. These toilets are particularly advantageous in areas with limited water supply, as they require minimal to no water for flushing. Vacuum toilets, commonly used in aircraft and trains, can also be adapted for urine separation in buildings. They employ a vacuum system to transport urine to a separate storage tank, reducing water usage and simplifying the collection and storage of urine for further processing (Mulec et al., 2016).

One of the major challenges in implementing urine source separation is gaining social acceptance (Barton et al., 2021). Cultural taboos and misconceptions about using human waste in agriculture can impede the adoption of this practice. Overcoming these barriers requires extensive public education and awareness campaigns emphasising urine-derived fertilisers' safety and benefits. Demonstrating the efficacy of these fertilizers through pilot projects and community involvement can aid in altering public perception and fostering trust. Equally important for the successful implementation of urine source separation is behavioural change. To achieve this, individuals need to be willing to employ source-separating toilets and participate in the separation process. Enhancing the user-friendliness and aesthetic appeal of toilets can facilitate acceptance and usage (Aliahmad et al., 2023). Additionally, engaging communities in the planning and implementation phases can create a sense of ownership and commitment to the system's success. Community engagement and education are essential to ensure that individuals comprehend the significance of urine separation and are motivated to participate in the process.

Robust policy and regulatory frameworks are necessary to support the adoption of urine source separation (Aliahmad et al., 2023). Governments can incentivize the use of source-separating sanitation systems through subsidies, tax breaks, and grants for research and development. Developing standards for the treatment and application of urine-derived fertilizers can ensure safety and quality, fostering confidence among farmers and consumers. Policy measures should also focus on integrating urine separation into broader waste management and environmental protection strategies. Furthermore, aligning urine separation with objectives related to sustainable agriculture, water conservation, and climate change mitigation can amplify its impact. For instance, incorporating urine source separation into national and regional sustainability plans can create synergies that enhance overall environmental sustainability. Governments and policymakers play a crucial role in creating an enabling environment for the adoption of urine source separation by providing the necessary support and resources for its implementation.

However, the use of urine in agriculture raises several nutritional concerns. One of the main issues is the inconsistency in nutrient concentration (Mie et al., 2017). The nutrient content of human urine can vary significantly based on diet, health, and hydration levels, making it challenging to apply it consistently as a fertilizer. To address the issue of excess or insufficient nutrient application, researchers are investigating techniques for standardizing the nutrient content of urine-based fertilizers. Methods such as dilution and nutrient extraction are being explored to achieve this goal (Bischel et al., 2015). However, there is also concern about the presence of contaminants in urine, such as pharmaceuticals, hormones, and pathogens (when it comes in contact with feces), which could pose risks to human health if crops absorb them (Bischel et al., 2015). While treatment methods like composting and advanced filtration can help mitigate these risks, they add complexity and cost to the process, potentially offsetting some of the environmental benefits. It is crucial to develop effective and affordable treatment technologies to ensure the safe use of urine-derived fertilizers.

1.2. ALKALINE URINE DEHYDRATION FOR NUTRIENT RECOVERY AND CHALLENGES

Source-separated urine can be either applied directly to agricultural fields (Pradhan et al., 2009) or can be further treated so that the nutrients can be recycled more efficiently in the form of fertilizers or similar products (Winker et al., 2009). One way to recycle nutrients is by removing the water from urine to urine-based fertilisers that are concentrated liquids, slurries, or dry powders. Such technologies include passive evaporation (Pahore et al., 2010), nitrification-distillation (Udert & Wächter, 2012), membrane distillation (Tun et al., 2016) and forward osmosis (Volpin et al., 2018). Another strategy is to selectively extract nutrients present in urine via processes such as precipitation (Le et al., 2020), adsorption (Pillai et al., 2014), stripping (Başakçilardan-Kabakci et al., 2007) and ion exchange (Tarpeh et al., 2017). These treatments also concentrate nutrients present in urine but produce wastewater that either needs to be managed or requires further treatment before it can be discharged into the environment (Harder et al., 2019).

Alkaline urine dehydration presents a promising approach to nutrient recovery from human urine. It involves the addition of alkaline earth chemicals, such as calcium hydroxide ($Ca(OH)_2$), to fresh urine to prevent the enzymatic degradation of urea into ammonia (Simha et al., 2020). The incorporation of alkaline substances in fresh urine inhibits the activity of urease, an enzyme that catalyzes the hydrolysis of urea into ammonia and carbonate. By increasing the pH to above 10, urease activity is suppressed, preventing the conversion of urea into ammonia, which would otherwise volatilize and lead to nitrogen loss (Simha et al., 2020). This high pH environment also helps to inactivate pathogens, making the urine safer for handling and further processing (Senecal et al., 2018). $Ca(OH)_2$ or $Mg(OH)_2$ are commonly used for this purpose due to their availability, effectiveness, and additional benefits such as facilitating the precipitation of phosphates, which are crucial for nutrient recovery. The resulting product is a nutrient-rich solid fertilizer that can be used in agriculture, reducing the need for synthetic fertilizers and closing the nutrient loop. However, maintaining a stable pH throughout the dehydration process is a significant challenge due to the carbonation process, which leads to a drop in pH and can undermine the benefits of the initial alkalization (Simha et al., 2020). The process can be summarized by the following reactions:

CO_2 (gas) + H_2O (liquid) ↔ H_2CO_3 (aqueous)

H_2CO_3 (aqueous) ↔ H^+ (aqueous) + HCO^{3-} (aqueous) (Simha et al., 2018)

During evaporation, the alkalized urine absorbs carbon dioxide (CO_2) from the air, leading to the formation of carbonic acid (H_2CO_3). The formation of carbonic acid and its subsequent dissociation into hydrogen ions (H^+) and bicarbonate ions (HCO_3^-) leads to a decrease in pH. As the pH drops, urease activity can potentially reactivate, leading to the hydrolysis of urea and the release of ammonia, thereby resulting in nitrogen loss, odour and reduced efficacy of the dehydration process. This is one of the major drawbacks associated with alkaline urine dehydration. Addressing this drawback would be a notable step in the right direction for this technology to be scaled up and accepted by the masses.

1.3. STRATEGIES TO BUFFER THE pH OF URINE DURING ALKALINE URINE DEHYDRATION

Providing users with a comparable level of comfort to conventional water-flushed toilets poses a challenge in resource-oriented sanitation systems that recycle urine. Traditional water-flushed toilets are perceived as more convenient and comfortable, creating a barrier to the adoption of urine recycling systems. Expecting users to continuously add alkaline chemicals, such as calcium hydroxide, to negate the pH reduction due to carbonation is impractical and restricts users from making the switch to resource-oriented sanitation systems. To address this issue, this thesis explores two innovative and user-friendly solutions to buffer the pH of dehydrating alkaline urine.

The first proposed solution is fabricating a passive chemical dosing system in the form of biodegradable polymer pouches loaded with alkaline chemicals. A passive chemical dosing system delivers chemicals into a process stream or environment that does not rely on active mechanical components like pumps or motors. Instead, this system utilizes natural processes such as diffusion, osmosis, or gravity to control the release of chemicals (Kreutzer et al., 2022). The design and operation of passive chemical dosing systems have both advantages and disadvantages that make them suitable for various applications. One key advantage of passive systems is their simplicity, which results in fewer moving parts and lower maintenance requirements and operational costs. This makes them ideal for use in remote or challenging environments, as well as for projects with limited budgets. Additionally, because passive systems do not rely on external power sources, they are highly energy-efficient, making them suitable for remote or off-grid applications where energy resources are scarce or non-existent (Smith et al., 2015).

However, passive systems may lack the precision and control of active systems, as the rate of chemical release is often influenced by environmental conditions, leading to variability in dosing. This can be problematic in processes requiring tight control over chemical concentrations. Furthermore, passive systems are generally designed for low to moderate dosing requirements and may not be suitable for applications that require large volumes of chemicals to be delivered quickly or in precise amounts (Smith et al., 2015). Additionally, passive systems may have slower response times to changes in the required dosing rate, which can be a limitation in dynamic processes where rapid adjustments are necessary to maintain optimal conditions. Factors such as temperature, pressure, and humidity can adversely affect the performance of passive systems and lead to inconsistencies in chemical delivery (Kreutzer et al., 2022). The passive nature of these systems makes them susceptible to either overdosing or underdosing, which may compromise process efficiency and effectiveness. This can also pose safety hazards in certain applications (Kreutzer et al., 2022).

The pouches would be designed in a way that they can buffer the pH of the dehydrating urine for a longer duration. These pouches would be placed in a urine evaporating cassette, where they would dissolve at controlled rates, gradually releasing alkaline chemicals to maintain optimal pH levels, as illustrated in the accompanying figure (Fig. 1). This steady release would counter the pH drop due to carbonation during convective evaporation. The degradation rate of these polymers in urine would depend on their properties, such as thickness, type of enantiomer and water solubility, as well as the properties of the urine, such as pH, temperature, and ionic strength (concentration factor). In the long term, these biopolymer pouches could also be used to administer various additives, such as peroxides, to break down organic micropollutants and/or pathogens in urine.

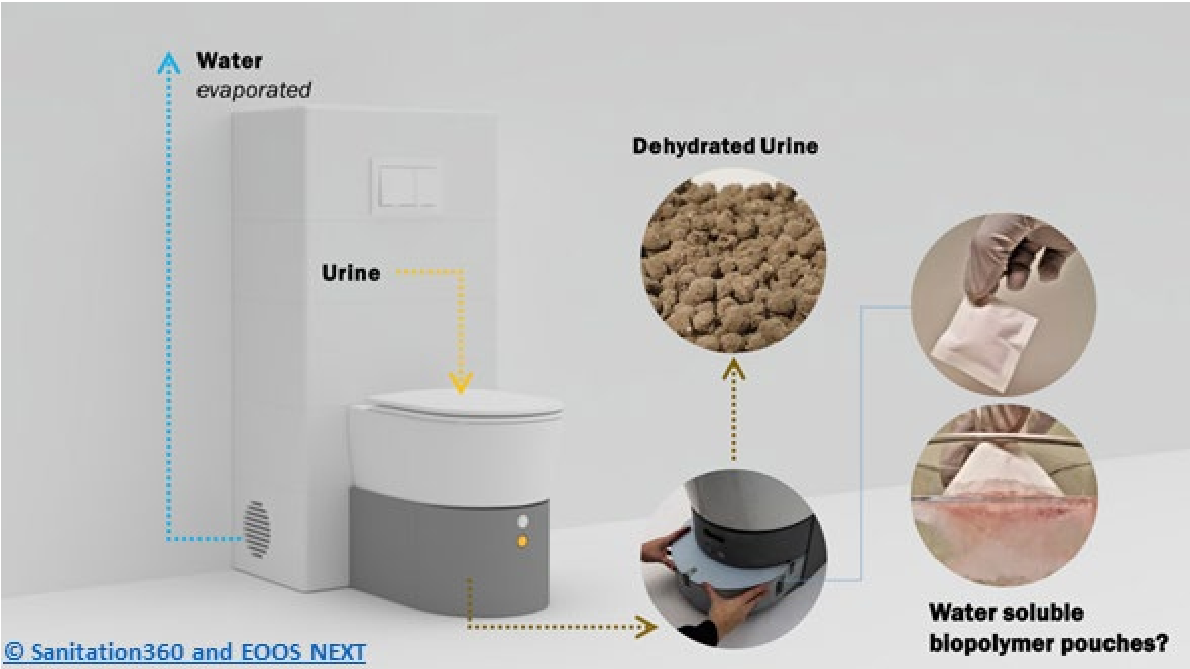

Figure 1: *A novel toilet that separately collects and dehydrates human urine by evaporation. This toilet has been installed at the offices of VA Syd in Malmo and in use for >2 years. This picture depicts the proposed passive chemical dosage mechanism to buffer the pH of urine during alkaline urine dehydration.*

The second proposed solution (Fig. 2) is to develop a closed-loop circular urine dehydrating system that would prevent the entry of CO_2. This solution would employ super absorbent polymers (SAPs) to dehumidify the humid air generated during urine evaporation, enabling its subsequent recycling. The working principle of the circular system would be similar to a heat pump, reusing energy to maintain a constant temperature and reducing overall energy consumption.

One of the key advantages of circular urine dehydrators is their ability to recover water from urine. By utilizing SAPs, water can be reclaimed and potentially reused, which is particularly beneficial in water-scarce environments. Additionally, circular conditions can help to neutralize the odour associated with urine, making the dehydrator system more pleasant to use and reducing the risk of unpleasant smells in the vicinity. However, the setup and installation of a circular loop alkaline urine dehydrator can be expensive and technically complex, requiring significant upfront investment and specialized knowledge to implement effectively. While the system itself may not have many moving parts, maintaining the appropriate conditions and ensuring the efficiency of the dehydration process requires regular monitoring and maintenance. Some aspects of the dehydration process, such as heating or circulating air, may require energy input, which can be a disadvantage in off-grid or energy-limited environments. The efficiency of the dehydration process can be influenced by environmental factors such as temperature and humidity. Inconsistent conditions can lead to variability in system performance.

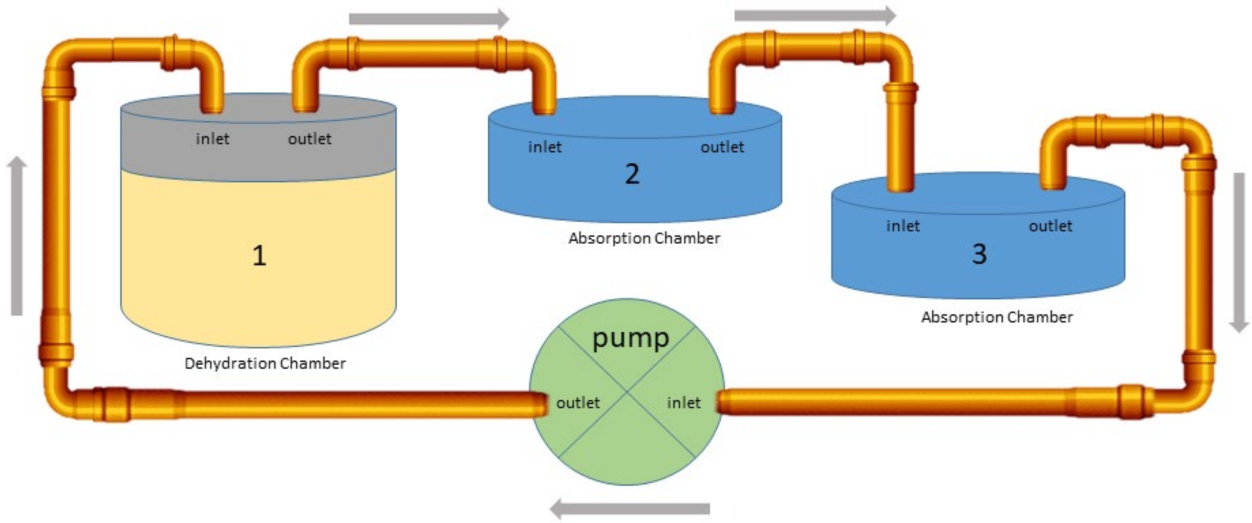

Figure 2: *The circular alkaline dehydration setup. Chamber marked 1 is the dehydration chamber where urine is loaded to dehydrated while chambers marked 2 and 3 are the absorption chambers which contain super absorbent polymers to dehumidify the humid air generated during the dehydration of urine. Each chamber had an inlet and outlet for recycling the sir via an aquarium pump. Fins were attached to the inside of the lids of the absorption chambers (2 and 3) to break the flow of the recycled air and increase turbulence and interaction between the super absorbent polymers and moist air*

1.4. REDUCED ENERGY DEMAND

Implementing source-separating sanitation systems along with advanced technologies can significantly reduce the energy footprint of the wastewater treatment plants and synthetic fertilizer industry. In modern wastewater treatment facilities that treat mixed domestic wastewater, 0.5-2 kWh of energy is utilized to treat each cubic meter of wastewater (Hamawand, 2023). Domestic wastewater comprises more than 99% water and the remaining 1% contains suspended and dissolved organic and inorganic matter, as well as microorganisms (Lara-Martín et al., 2020). The average nitrogen, phosphorus, and potassium content in municipal wastewater from Indian cities is 30 mg/L, 7.50 mg/L, and 25 mg/L, respectively (CPHEEO, Govt of India, 2013). Although urine accounts for less than 1% of the volume of domestic wastewater, it contributes approximately 80% of the N, 56% of the P, and 63% of the K found in domestic wastewater (Randall & Naidoo, 2018). Thus, source-separating urine can reduce the energy needed to remove N, P and K introduced via urine.

The energy-intensive Haber-Bosch process, which is used for ammonia production, requires approximately 8-12 MJ of energy per kilogram of ammonia produced (Summaries, 2021). In 2021, around 600 Exajoules (10^{18} J) of energy were utilized to produce 150,000 tonnes of ammonia using the Haber-Bosch process (Agency, 2021). The Haber-Bosch process consumes about 2% of the world's total energy supply, and recovering nutrients from urine has the potential to reduce this demand by up to 40% (Iddya et al., 2020). Phosphorous is usually mined as apatite rock or sedimentary phosphate rock and requires around 10 kWh of energy to mine 1 kg P (von Bahr, 2016). Recycling the nutrients from urine and using them in agricultural fields can not only reduce the energy used to produce synthetic fertilizers but also reduce environmental pollution.

1.5. OBJECTIVES AND STRUCTURE OF THE THESIS

The primary objective of this thesis was to explore the use of polymers for buffering the pH of urine during the alkaline dehydration of urine and to enhance the overall value of this process. The specific objectives were to evaluate:

- the degradation of the polymers in concentrated and unconcentrated urine to find the best polymer for the fabrication of the passive chemical dosing system.
- the degradation of Poly-L-lactic acid as a function of layer thickness and crystallinity of the polymer films and pH and temperature of urine.

- the efficacy of the pouches in regulating the pH of the dehydrating urine and accessing the release of chemicals as a function of the number of layers of the pouches and as a function of the pH and concentration factor (CF) of the urine.
- the efficacy of the circular- urine dehydrating setup in buffering the dehydrating urine's pH and removing organic metabolites in the recycled water.

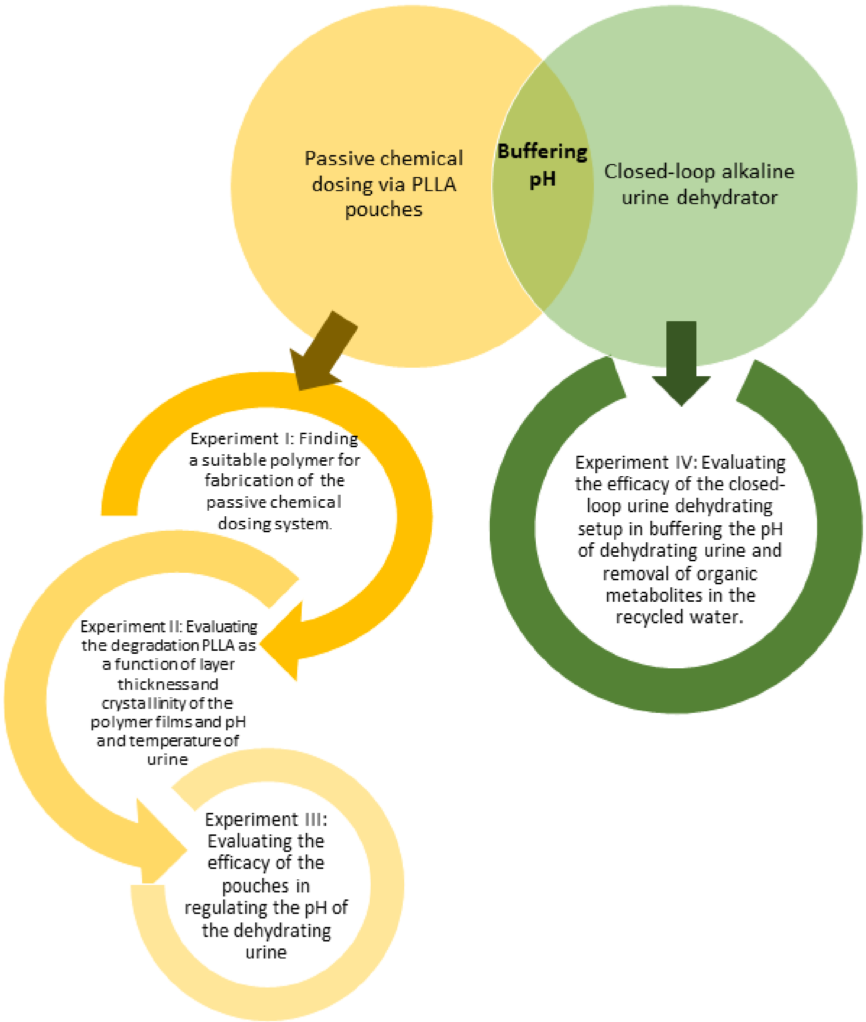

Figure 3: *Structure of the thesis and relation between each experiment and the overall aims and objectives of the thesis.*

The first two investigations were conducted to understand the various aspects of polymer degradation in urine. The data from these experiments were extremely important to design an effective passive chemical dosing system to buffer the pH of urine during alkaline urine dehydration as seen in the third investigation. The fourth investigation was conducted to evaluate the efficacy of using Super Absorbent Polymers (SAPs) in a novel circular alkaline urine dehydration setup in buffering the pH and recycling reusable water.

This first investigation (Experiment I) evaluated the potential of four different polymers, polypropylene (PP), polylactic acid (PLA), polycaprolactone (PCL), and polyvinyl alcohol (PVOH), as candidates for a passive chemical dosing system designed to buffer the pH of source-separated urine during alkaline dehydration. Each polymer possesses distinct properties that make it suitable for specific applications in the context of urine treatment. Polymer films with a diameter of 2 cm and a thickness of 2 mm were fabricated and stored in alkaline urine for sixteen days. The films were destructively sampled on days two, four, eight, and sixteen and evaluated for degradation.

The results of the first investigation revealed that PLA was the polymer that degraded the most and formed non-toxic by-products. In the second investigation (Experiment II), the effect of urine pH, temperature, and polymer film thickness on polymers' degradation rate and degradation mechanism was evaluated. Poly-L-lactic acid (PLLA) films were fabricated into different thicknesses (0.05 mm, 0.1 mm and 0.25 mm) and stored in Mili-Q water (pH 7) and alkaline urine (pH 12) at 20 °C and 45 °C for eight days. Destructive sampling was done on days two, four, and eight, and the degradation of the PLLA films was evaluated.

The third investigation (Experiment III) assessed the effectiveness of the PLLA-based passive chemical dosing system in maintaining a safe pH (>10) during alkaline urine dehydration. Pouches with various layer configurations were fabricated, each containing a KOH pellet and stored in alkaline urine (CF 1 and CF 10, pH 11 and pH 14) for thirty-two days. The pouches were destructively sampled on days two, four, eight, sixteen, twenty-four, and thirty-two. The change in pH and K concentration of the urine and degradation of the PLLA pouches were examined.

The fourth study (Experiment IV) devised a novel circular-alkaline urine dehydration setup to evaluate its capacity to maintain a safe pH during dehydration. SAPs were used to absorb moisture generated during urine dehydration and were then extracted as water. The extracted water was also tested for the removal of organic metabolites (OMs) to determine its suitability for non-potable domestic applications such as gardening. The thermodynamics and energy demand of the circular setup were also assessed.