



**Final report (public) dated 17.01.2020**

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**ManuMax - Development of a heat integrated steam explosion pretreatment  
process to enhance biogas yields in anaerobic digestion of manure (Phase 1)**

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Bern University  
of Applied Sciences

**Datum:** 17.01.2020

**Ort:** Zollikofen



**Date:** 17.01.2020

**Location:** Bern

**Publisher:**

Swiss Federal Office of Energy SFOE  
Energy Research and Cleantech  
CH-3003 Bern  
[www.bfe.admin.ch](http://www.bfe.admin.ch)

**Subsidy recipients:**

Berner Fachhochschule  
Hochschule für Agrar-, Forst- und Lebensmittelwissenschaften  
Länggasse 85  
3254 Zollikofen  
<https://www.bfh.ch/hafl>

**Authors:**

Michael Studer, BFE, [michael.studer1@bfh.ch](mailto:michael.studer1@bfh.ch)

**SFOE project coordinators:**

Sandra Hermle, [sandra.hermle@bfe.admin.ch](mailto:sandra.hermle@bfe.admin.ch)

**SFOE contract number:** SI/5017 41-01

**The authors bear the entire responsibility for the content of this report and for the conclusions drawn therefrom.**



## Zusammenfassung

Biogas aus Gülle wird durch die Möglichkeit der nachfrageorientierten Stromproduktion in der zukünftigen erneuerbaren Energieversorgung eine wichtige Rolle spielen. Die hohe Rekalzitranz von Gülle gegenüber dem anaeroben Abbau lassen derzeit keine ökonomische Umwandlung zu Biogas in Kleinanlagen zu. Um dies zu ermöglichen, wurde in diesem Projekt eine optimierte, wärmeintegrierte Dampf-Vorbehandlungsmethode entwickelt, welche die Biogas-Ausbeute signifikant steigert. Eine aufwändige Optimierung der Dampfvorbehandlung zeigte, dass eine thermische Vorbehandlung der Flüssigphase der Rindergülle nicht notwendig ist, da die Methan-Ausbeuten dadurch nicht erhöht werden können. Die gewaschenen Feststoffe hingegen profitierten stark von einer Dampfvorbehandlung und die Ausbeuten konnten um bis zu 50% erhöht werden. In einem eigens gebauten Teststand wurde die Pumpbarkeit, die Wärmeübergangskoeffizienten und die Separation in der richtigen Grössenordnung für eine geplante kontinuierlich betriebene Pilotanlage für Vollgülle von 50 Kühen gemessen. Für so kleine Volumenströme ist das Pumpen ohne Verstopfen eine grosse Herausforderung und verlangte nach geometrischen Anpassungen. Ebenfalls ist der Wärmeübergang für die resultierenden tiefen Fliessgeschwindigkeiten sehr niedrig. Die techno-ökonomische Analyse zeigte, dass unter Schweizer Bedingungen, mit einem angenommenen Eigenstromverbrauch und ohne KEV-Beiträge, eine Biogasanlage welche ausschliesslich Rindergülle als Substrat verwendet, erst ab Gülle von mehr als 500 GVE ökonomisch betrieben werden kann. Bei einer durch Vorbehandlung gesteigerten Biogasausbeute verringert sich die Anzahl der notwendigen GVE auf 300, um den break-even zu erreichen. Basierend auf den in diesem Projekt erzielten Ergebnissen, soll in einem nächsten Schritt eine Pilotanlage entwickelt und am Standort der IAG in Grangeneuve gebaut werden.

## Résumé

Le biogaz, issu du lisier, jouera un rôle important dans l'approvisionnement futur en énergie renouvelable grâce à la possibilité de produire de l'électricité en fonction de la demande. La conversion faible du lisier en biogaz lors de la digestion anaérobie, dû à une récalcitrance de la biomasse ne permet pas actuellement une conversion économiquement viable du biogaz dans les petites installations. Pour améliorer la conversion, une méthode de prétraitement à la vapeur optimisée et thermique a été mise au point dans le cadre de ce projet, ce qui augmente considérablement le rendement en biogaz. Une optimisation complexe du prétraitement à la vapeur a montré qu'un prétraitement thermique de la phase liquide du lisier de bovins n'est pas nécessaire, car les rendements en méthane ne peuvent être augmentés. Les solides lavés, au contraire, ont grandement bénéficié du prétraitement à la vapeur et les rendements ont pu être augmentés jusqu'à 50 %. La pompabilité, les coefficients de transfert de chaleur et la séparation ont été mesurés dans le bon ordre de grandeur pour une installation pilote à fonctionnement continu prévue pour le lisier complet de 50 UGB (Unités de Gros Bétails) dans un banc d'essai spécialement construit. Pour des débits volumiques aussi faibles, le pompage sans colmatage est un défi majeur et nécessite des ajustements géométriques. Le transfert de chaleur pour les faibles vitesses d'écoulement qui en résultent est également très faible. L'analyse technico-économique a montré que, dans les conditions suisses, avec une consommation d'électricité supposée propre et sans contribution du KEV, une installation de biogaz qui utilise exclusivement du lisier de bovins comme substrat ne peut être exploitée économiquement qu'avec 500 UGB. Si le rendement en biogaz est augmenté par prétraitement, le nombre d'UGB nécessaires pour atteindre le seuil de rentabilité est réduit à 300. Sur la base des résultats obtenus dans le cadre de ce projet, la prochaine étape consistera à développer une usine pilote et à la construire sur le site de l'IAG à Grangeneuve.



## Summary

Biogas produced in anaerobic digestion plants of manure has the potential to play an important role in future smart and flexible energy system by enabling demand driven electricity production. The high recalcitrance of manure towards anaerobic deconstruction does currently not allow the economic feasible conversion to biogas. To change this, we developed an optimized, heat-integrated steam pretreatment method that significantly enhances the biogas yield. An elaborate pretreatment optimization study showed that a thermal pretreatment of the liquid phase of the manure is not necessary, as the biogas yield cannot be improved. In contrast the washed solids benefitted strongly from steam pretreatment and the yields could be increased up to 50%. In a specifically constructed test rig the pumpability and the heat transfer coefficients were measured in the appropriate dimension of a later continuously operated pretreatment plant operated with manure from 50 dairy LSU (Life Stock Units). The pumping without clogging of such small flow rates appeared to be challenging and required geometrical adjustments. Also, the heat transfer coefficients for the resulting low flow velocities is small. The techno-economic assessment showed, that for a biogas plant using entirely cow manure as substrate, operated under Swiss conditions (*i.e.* no compensatory feed-in remuneration, own consumption of power), can only be economically operated with manure of more than 500 LSU. For pretreatment with an increased biogas yield this number is reduced to 300 LSU to reach break-even. Based on the results obtained during this project, the next step will be the development of a pilot facility at the IAG site in Grangeneuve.

## Main findings



# Contents

|   |           |
|---|-----------|
| <b>Zusammenfassung.....</b>   | <b>3</b>  |
| <b>Résumé.....</b>  | <b>3</b>  |
| <b>Summary .....</b>  | <b>4</b>  |
| <b>Main findings .....</b>  | <b>4</b>  |
| <b>Contents .....</b>   | <b>5</b>  |
| <b>Abbreviations.....</b>   | <b>7</b>  |
| <b>1 Introduction.....</b>  | <b>8</b>  |
| 1.1 Background information and current situation .....  | 8         |
| 1.1.1 Introduction .....  | 8         |
| 1.1.2 Manure as feedstock for anaerobic digestion.....  | 8         |
| 1.1.3 Pretreatment methods to improve the digestibility of manure .....                           | 8         |
| 1.1.4 Steam explosion pretreatment to improve anaerobic digestion performance .....               | 9         |
| 1.1.5 Advantages and challenges associated with steam explosion pretreatment.....                 | 9         |
| 1.2 Purpose of the project .....  | 10        |
| 1.3 Objectives .....  | 10        |
| 1.3.1 Optimization of pretreatment conditions and reasoning for 2-stage pretreatment (Task 1.1).. | 10        |
| 1.3.2 Influence of the explosion step (Task 1.2).....   | 12        |
| 1.3.3 Description of the proposed pretreatment reactor.....                                       | 14        |
| 1.3.4 Option two-stage pretreatment (Task 1.3).....   | 16        |
| 1.3.5 Sizing of the pretreatment pilot plant (Task 2.1).....                                      | 17        |
| 1.3.6 Techno-economic assessment (Task 4.2) .....   | 17        |
| <b>2 Procedures and methodology.....</b>  | <b>18</b> |
| 2.1 Pretreatment optimization.....  | 18        |
| 2.2 Design and construction of a pre-pilot plant.....   | 18        |
| 2.2.1 Reasoning to design and build a test rig .....  | 18        |
| 2.2.2 General Concept .....   | 19        |
| 2.2.3 One-stage pretreatment .....  | 19        |
| 2.2.4 Two-stage pretreatment .....  | 20        |
| 2.2.5 Fundamentals of heat transfer.....  | 20        |
| 2.2.6 Influence of the velocity on heat transfer.....   | 21        |
| 2.2.7 Overall heat transfer coefficient.....  | 21        |
| 2.3 Techno-economic model .....   | 21        |
| <b>3 Results and discussion .....</b>   | <b>22</b> |
| 3.1 Optimization of steam pretreatment of dairy cow manure.....                                   | 22        |
| 3.2 Test stand .....  | 23        |
| 3.2.1 Heat exchanger design.....  | 23        |



|          |  |           |
|----------|--|-----------|
| 3.2.2    | Manure Feed .....  | 27        |
| 3.2.3    | Explosive Manure Outlet .....  | 28        |
| 3.2.4    | Overall heat transfer coefficient for tests with water on both sides of the heat exchanger ..... | 28        |
| 3.2.5    | Pumpability test with liquid manure .....  | 31        |
| 3.2.6    | Conclusion and next steps .....  | 32        |
| 3.3      | Techno-economic model .....  | 33        |
| <b>4</b> | <b>Conclusions .....</b>   | <b>33</b> |
| <b>5</b> | <b>Outlook and next steps .....</b>  | <b>33</b> |
| <b>6</b> | <b>National and international cooperation.....</b>   | <b>34</b> |
| <b>7</b> | <b>Communication .....</b>   | <b>34</b> |
| <b>8</b> | <b>References .....</b>  | <b>34</b> |
| <b>9</b> | <b>Appendix .....</b>  | <b>35</b> |



## Abbreviations

LSU – Live stock units

BMP – Biomethane potential

IAG – Institut Agricole Grangeneuve

CHP – Combine heat and power

THP - Thermal hydrolysis processes



# 1 Introduction

## 1.1 Background information and current situation

### 1.1.1 Introduction

In anaerobic digestion the organic components of waste or agricultural biomass are converted in an oxygen-free environment by a natural microbial consortium to a biogas consisting mainly of CH<sub>4</sub> and CO<sub>2</sub>. Biogas can either be converted to electricity in combined heat and power (CHP) plants or it can be upgraded to natural gas quality methane prior to injection into the natural gas grid. Biogas has the potential to play an important role in smart and flexible energy grids in two ways: i) through demand driven electricity production from biogas temporarily stored on site or from purified biomethane injected into the grid or ii) through a power to methane process at times of surplus supply of electricity (Montgomery and Bochmann 2014). In the latter, H<sub>2</sub>, produced by electrolysis of water and CO<sub>2</sub> from the biogas are converted to biomethane ( $4 \text{ H}_2 + \text{CO}_2 \rightarrow \text{CH}_4 + 2 \text{ H}_2\text{O}$ ) by the action of hydrogenotrophic methanogens in the digester.

### 1.1.2 Manure as feedstock for anaerobic digestion

Animal manure is a promising feedstock for anaerobic digestion and is a largely untapped energy resource in Europe. In Switzerland, currently only 1% of its sustainable energy potential of 21.4 PJ per year is recovered. In Sweden, about 4% of the manure is currently treated by anaerobic digestion, generating approximately 0.17 PJ of biogas mostly in co-digestion plants. Manure contains about 50% lignocellulosic fibres that are recalcitrant to biological deconstruction. Consequently, only modest methane yields (25 to 40%) are achieved in anaerobic digestion of manure. The lowest yields are found for manure of ruminant animals (cattle, cows, sheep etc) with a diet high in lignocellulosic fibers. The low yields render the economic performance of anaerobic digestion plants operating only with manure unattractive for farmers and explain the current low utilization of this huge untapped resource.

### 1.1.3 Pretreatment methods to improve the digestibility of manure

In order to improve the biomethane potential (BMP) of manure, different pretreatment methods have been investigated (Table 1).

Table 1: *Examples of literature results for pretreatment of manure to enhance the BMP.*

| Feedstock                    | Pretreatment  | BMP [mL <sub>CH4</sub> /g <sub>VS</sub> ] | Improvement | Reference                    |
|------------------------------|---|---|-------------|------------------------------|
| Solid fraction of pig manure | Steam explosion, 170°C, 30 min  | 329                                       | +107 %      | (Ferreira et al. 2014)       |
| Solid fraction of pig manure | Aqueous ammonia soaking (10mL of 30% ammonia solution per g <sub>TS</sub> ), 22°C, 3 days | 320                                       | +178 %      | (Jurado et al. 2013)         |
| Cattle manure                | 200°C, 15 min, no explosion   | 331                                       | +21 %       | (Raju et al. 2013)           |
| Swine manure                 | 200°C, 15 min, no explosion   | 344                                       | +29 %       |                              |
| Cattle manure                | Commercial xylanase   | 289                                       | + 9 %       | (Warthmann et al. 2012)      |
| Cattle manure                | Hemicellulose degrading bacterium   | 300                                       | + 30 %      | (Angelidaki and Ahring 2000) |





Based on literature data for pretreatment of the residues of manure-based biogas plants (that are not summarized here due to space reasons) and the one presented in Table 1, it can be concluded that caustic chemicals such as CaO, NH<sub>3</sub> or NaOH are most effective to overcome the recalcitrance towards deconstruction. However, the application of such chemicals has several disadvantages including high costs for the chemicals or their recycling and the generation of undesired salt loads that might prevent the use of the digestate as a fertilizer. Thermal pretreatments of manure that use no chemicals are a promising alternative and have also been shown to be effective, especially if they include an explosive pressure release that disrupts the lignocellulosic structure and reduces the particle size, as we discuss in more detail below.

#### 1.1.4 Steam explosion pretreatment to improve anaerobic digestion performance

To the best of our knowledge, steam explosion is hardly investigated for improving the digestibility of manure. However, it has been shown to be effective with other types of biomass and waste if the conditions are carefully optimized for the feedstock of choice (Table 2).

Table 2: *Optimal steam explosion conditions to improve the BMP of different feedstocks.*

| Feedstock              | Steam explosion conditions | BMP [mL <sub>CH4</sub> /g <sub>VS</sub> ] | Improvement | Reference                |
|------------------------|----------------------------|---|-------------|--------------------------|
| Miscanthus             | 220°C, 10 min              | 374                                       | +345%       | (Menardo et al. 2013)    |
| Hay                    | 175°C, 10 min              | 281                                       | +16%        | (Bauer et al. 2014)      |
| Waste activated sludge | 165°C, 20 min              | 310                                       | +30-40%     | (Oosterhuis et al. 2014) |

For the treatment of waste activated sludge and primary sludge, steam explosion pretreatment has been advanced to commercial-scale application by some companies e.g. CAMBi (NOR), dmt Environmental Technology (NL), teCH4<sup>+</sup> (ESP) or VEOLIA (F). In all these commercially available thermal hydrolysis processes (THP), the sludge is dewatered to increase the solids content, pulped and preheated to 100°C (using recovered heat after steam-ex) and then further heated to the desired pretreatment temperature (e.g. 160°C) by employing high grade (temperature) steam. After the desired residence time, the reactor is decompressed to atmospheric pressure and the released low grade steam is recovered to preheat the solids.

#### 1.1.5 Advantages and challenges associated with steam explosion pretreatment

Steam explosion pretreatment is an effective and cost-efficient method to overcome the recalcitrance of a range of different feedstocks and to reduce their particle size through the explosive pressure release in an energy efficient way. The relevance of the particle size reduction and the explosion was hardly addressed in literature, but the limited data available suggest that it is an essential part of the pretreatment. For instance, Pielhop *et al.* found that an explosive pressure release over a pressure difference of at least 20 bar enhanced the enzymatic digestibility of spruce by 60% compared to a similar pretreatment with a slow pressure release (Pielhop et al. 2016).

However, by the sudden pressure release, high grade heat is dissipated and only low grade heat of 100°C can be recovered. Thus, high pressure steam has to be renewed constantly to heat the biomass to the desired temperature. In order to limit the necessary amount of high grade steam, the amount of water in diluted feedstocks such as manure or waste water sludge is reduced by an initial solid liquid separation step, which however increases the complexity of the pretreatment plant.



Another issue of thermal pretreatments in general is that the two main sugar fractions of the biomass-hemicellulose and cellulose-respond differently to the pretreatment. The high temperatures of 200°C and above required for efficient cellulose hydrolysis lead to thermal degradation of hemicellulosic sugars to HMF and furfural that are known to inhibit the downstream biological processes. A two stage pretreatment process, where the solubilized hemicellulose is discharged after a first moderate temperature stage followed by a high temperature stage can circumvent the sugar loss. Finally, the dominating technology in commercial scale steam explosion plants to continuously feed solids by stuffing screws is complex and prone to fast mechanical wear due to the abrasive nature of the biomass (containing sand; manure contains up to 25 w/w of the DM). Thus, high maintenance costs and prolonged downtime are issues of current pilot scale steam explosion plants.

## 1.2 Purpose of the project

The main aim of the project is to develop a pretreatment method that overcomes the above-mentioned issues of steam explosion and enables economically feasible anaerobic digestion of manure on farm scale to unlock the huge potential of this resource for biogas production. The proposed concept allows for

- recovery of high grade heat,
- particle size reduction through explosive decompression of the biomass,
- the option of two stage pretreatment and
- usage of pumps instead of stuffing screws to feed the biomass slurry into the high pressure reactor.

This sub-project includes also a techno-economic analysis to predict the potential of the process and the consequences of its wider deployment. Furthermore, we aim for in depth understanding of the effects of the operating conditions of steam explosion pretreatment on the anaerobic digestibility of manure.

## 1.3 Objectives

### 1.3.1 Optimization of pretreatment conditions and reasoning for 2-stage pretreatment (Task 1.1)

Steam explosion pretreatment is an effective and cost-efficient method to overcome the recalcitrance of a range of different feedstocks. However, the optimal pretreatment conditions, *i.e.* the pretreatment temperature and duration, depend on the characteristics of the feedstock and must be optimized individually for each feedstock. For example, softwood species require much harsher pretreatment conditions than e.g. herbaceous biomass.

The importance of choosing the right pretreatment conditions is exemplified in Figure 1, where the glucose and xylose yields from beech wood after pretreatment and enzymatic hydrolysis by cellulolytic enzymes are shown as a function of the pretreatment severity. In the severity factor  $\log R_0$ , the pretreatment duration ( $t$  in seconds) and temperature ( $T$  in K) are combined to one value that allows to estimate the harshness of the chosen conditions:

$$\log R_0 = \log(t \cdot e^{(T-100)/14.75}) \quad (1)$$



As shown in Figure 1 and Figure 2, the optimal conditions for maximal xylose and glucose yields do not match at all.

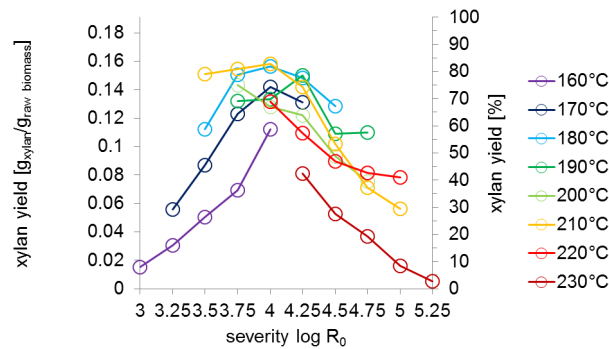


Figure 2: Xylan yield in the liquid phase after steam explosion pretreatment of beech wood at different temperatures and severities. The optimum pretreatment condition for hemicellulose is 180°C, 44Min ( $\log R_0 = 4.0$ )

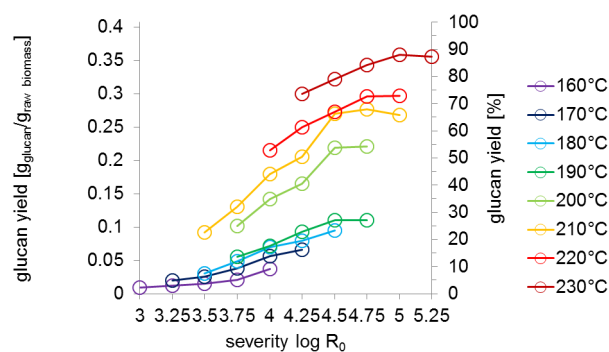
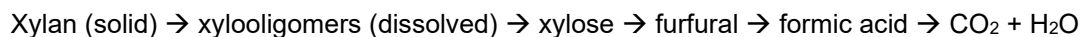


Figure 1: Glucan yield after steam explosion pretreatment of beech wood at different temperatures and severities and enzymatic hydrolysis by Accelerase 1500. The optimum pretreatment condition for cellulose is 230°C, 15Min ( $\log R_0 = 5.0$ )

At the conditions that are optimal for maximal glucose yields, nearly any xylose can be recovered. This can be explained by the heat lability of xylan. At elevated temperatures, a reaction cascade is started that eventually leads to the complete degradation of the substrate to substances that cannot be converted to biomethane:



On the other hand, the conditions that are optimal for maximal xylose yields (180°C,  $\log R_0 = 4.0$ ) are not sufficient to overcome the recalcitrance of the remaining solids. In order to recover both carbohydrate fractions in high yields, a two-stage pretreatment can be performed. The first stage is performed at conditions that are optimal for recovering the maximal of amount of soluble xylan derivatives. The liquid phase containing the dissolved (thermally hydrolyzed) xylan is separated from the pretreatment reactor and only the solid phase is pretreated in the second stage at harsher conditions that are optimal for high glucose release.



Although in the presented example beech wood was used and enzymatic hydrolysis applied the principle considerations are also valid for the pretreatment of manure to enhance the biomethane potential. However, for manure the situation is even more complex, as manure contains besides cellulose and hemicellulose also proteins and fat (and lignin) as components that can be converted to biogas. To achieve high methane yields and to avoid the generation of inhibitors, the degradation of any of these components during pretreatment should be minimized.

### 1.3.2 Influence of the explosion step (Task 1.2)

Steam explosion pretreatment is known to be able to reduce the particle size of the treated biomass in an energy efficient way. However, the relevance of the particle size reduction and the explosive pressure release was hardly addressed in literature, but the limited data available suggest that it is an essential part of the pretreatment. For instance, Pielhop *et al.* found that an explosive pressure release over a pressure difference of at least 20 bar enhanced the enzymatic digestibility of spruce by 60% compared to a similar pretreatment with a slow pressure release (Pielhop *et al.* 2016).

The disadvantage of the explosive pressure release from the pretreatment pressure, e.g. 28bar/230°C, to ambient pressure is, that high grade heat is lost and only low-grade heat of max. 100°C can be recovered. This is due to the fact that for saturated steam *i.e.* steam where simultaneously liquid water and water vapour are present, temperature and pressure are always linked according to the line of saturation vapour pressure. In a pretreatment employing humid biomass this is always the case.

The saturation vapour pressure of water at 100°C is 1bar. Thus, during a very rapid pressure reduction from e.g. 20 to 1bar, the temperature also has to be lowered very rapidly from e.g. 230°C to 100°C (see Figure 3). This temperature reduction is done by dissipation of the high temperature energy through flash evaporation of a part of the liquid water. This sudden phase change from liquid water to water vapour and the associated huge volume increase inside the pores of the biomass causes the biomass to explode.

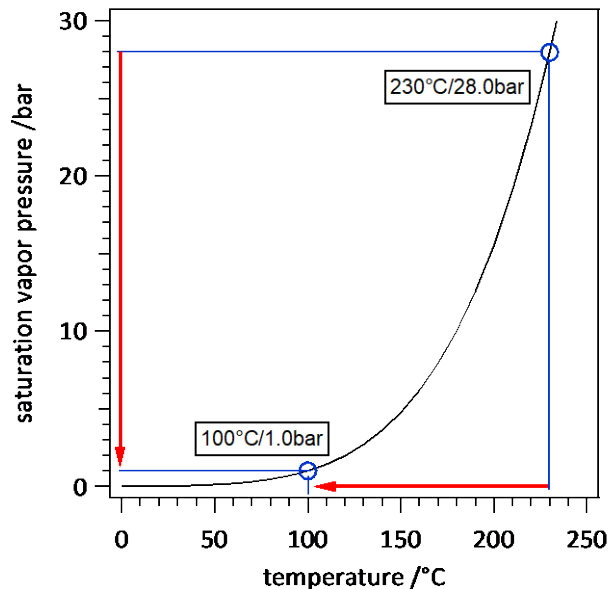


Figure 3: *The saturation vapour pressure of steam as a function of the temperature. During the pressure release from e.g. 28bar, which corresponds to a temperature of the water vapour of 230°C, to ambient pressure (1bar) the temperature must also drop to 100°C. This explosive decompression is the reason that steam-explosion pretreatments only allow for the recovery of low grade heat, i.e. 100°C.*

Due to this temperature reduction during the explosive discharge to ambient pressure only 100°C heat can be recovered and in the following batch or a continuous set-ups high pressure steam has constantly to be renewed to heat the biomass to the desired pretreatment temperature. In order to limit the necessary amount of high grade steam, the amount of water in diluted feedstocks such as manure or waste water sludge is often reduced. (Heat capacity of water: 4.2kJ/(kg K), biomass: ~1.7kJ/(kg K)). However, this separation of the liquid phase i) increases the complexity of the pretreatment plant through an additional unit operation and ii) leaves the liquid fraction of the manure untreated.

The aim of the proposed project is to develop and implement a pretreatment process that allows for the recovery of high grade heat and still offers the beneficial effects of an explosive decompression. Basically, we plan for a counter current double pipe heat exchanger as pretreatment reactor. The manure will be compressed by a pump to a pressure larger than the vapour saturation pressure at the maximum pretreatment temperature (e.g. 30 bar for a pretreatment at 230°C/28 bar) in order i) to prevent evaporation of the manure inside the pretreatment reactor and ii) to allow for an explosive decompression to ambient pressure also at 40°C after recovering - the target temperature of the subsequent anaerobic digestion. To investigate whether this 'cold explosion' will also show a positive effect on the later biochemical conversion, appropriate steam pretreatment pretests were carried out in our steam-gun. Beech wood was pretreated at the previously optimized conditions for glucose release through enzymatic hydrolysis (see Figure 1) but the pressure was slowly released to almost ambient pressure (1.2 bar), thereby preventing flash evaporation of water inside the pores, responsible for the disintegration of the biomass particles in a 'standard' steam explosion pretreatment. After the pressure release bottled nitrogen was fed into the steam-gun pressurizing the reactor to 30 bar when the 'cold' (i.e. ~100°C) pretreated biomass was automatically exploded to ambient pressure (see Figure 4). This pretreated biomass was then enzymatically hydrolysed using commercial cellulolytic enzymes together with the normally pretreated biomass and sugar release after 5 days was measured. Remarkably, the



sugar release from the beech wood samples pretreated at the same conditions but differently exploded showed the same sugar release.

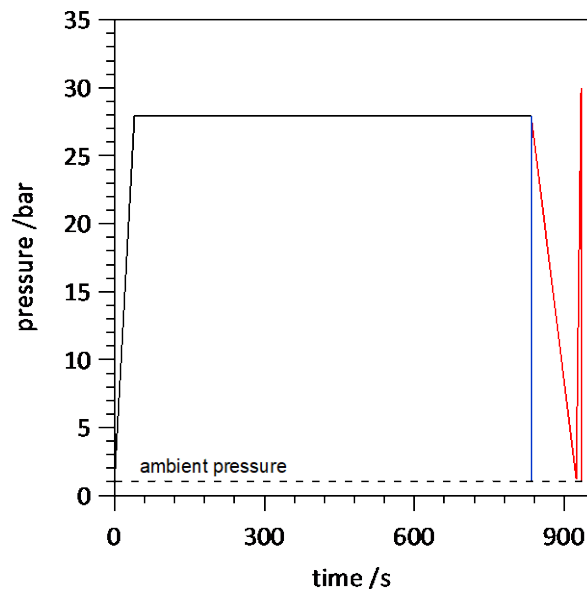


Figure 4: *Pressure course during the steam pretreatment of beech wood at 230°C/28bar. In the graph two explosion strategies are shown. The blue line represents the standard explosive decomposition to ambient pressure applied in a steam-explosion pretreatment. The red line illustrates the 'cold explosion' mimicked in the laboratory. The pressure in the steam-gun is slowly released to ambient pressure which causes the temperature to drop to 100°C. Then the 'cold' pretreated biomass is compressed using bottled nitrogen to 30bar, when the biomass is explosively discharged.*

### 1.3.3 Description of the proposed pretreatment reactor

Besides the above discussed challenges of conventional steam explosion pretreatment processes (dissipation of high grade heat, degradation of hemicellulose), a further issue of large scale pretreatment plants that are operating continuously is the feeding of biomass with a high solids content into the pretreatment reactor. For this purpose, often stuffing screws are employed where biomass is compressed in a special 'screw conveyor' to form a plug, which acts as a seal between the high pressure zone within the reactor and the environment. The stuffing screws are prone to very fast mechanical wear due the abrasive nature of the biomass e.g. due to high sand content in manure solids. Alternatively, we propose in this project to work with manure slurries that can be fed into the pretreatment reactor by conventional slurry pumps.

To this end, we envision a pretreatment reactor where manure and water as heat transfer medium are pumped in counter flow through a series of two double pipe heat exchangers allowing for the recovery of the total thermal energy (see Figure 5).

On the way through the first heat exchanger manure is heated to the desired pretreatment temperature by extracting heat from the water flowing in counter current mode, which is thereby cooled down. Once the target temperature is reached the pretreatment temperature must be kept constant for the previously determined optimum residence time. This will be done in a second well insulated tubular reactor through which the manure is pumped. After this thermal pretreatment, the manure is flowing through a second counter current double pipe heat exchanger, where the heat is



gradually withdrawn from the manure and transferred to the heat transfer medium. These exchanges of heat are dependent on a temperature gradient between manure and water. In the second heat exchanger, this temperature difference is positive, *i.e.*  $T_{\text{manure}} > T_{\text{water}}$ , while the sign of the temperature difference in the first heat exchanger is negative, *i.e.*  $T_{\text{manure}} < T_{\text{water}}$ . Thus, the heated water flowing from the second to the first heat exchanger needs to be further heated in order to raise the temperature of the water to a level above the temperature of the manure, allowing the transfer of heat from the water to the manure. This amount of heat needed to superheat the water is - in an adiabatic system – identical to the heat needed to increase the temperature of the manure from ambient temperature to the fermentation temperature.

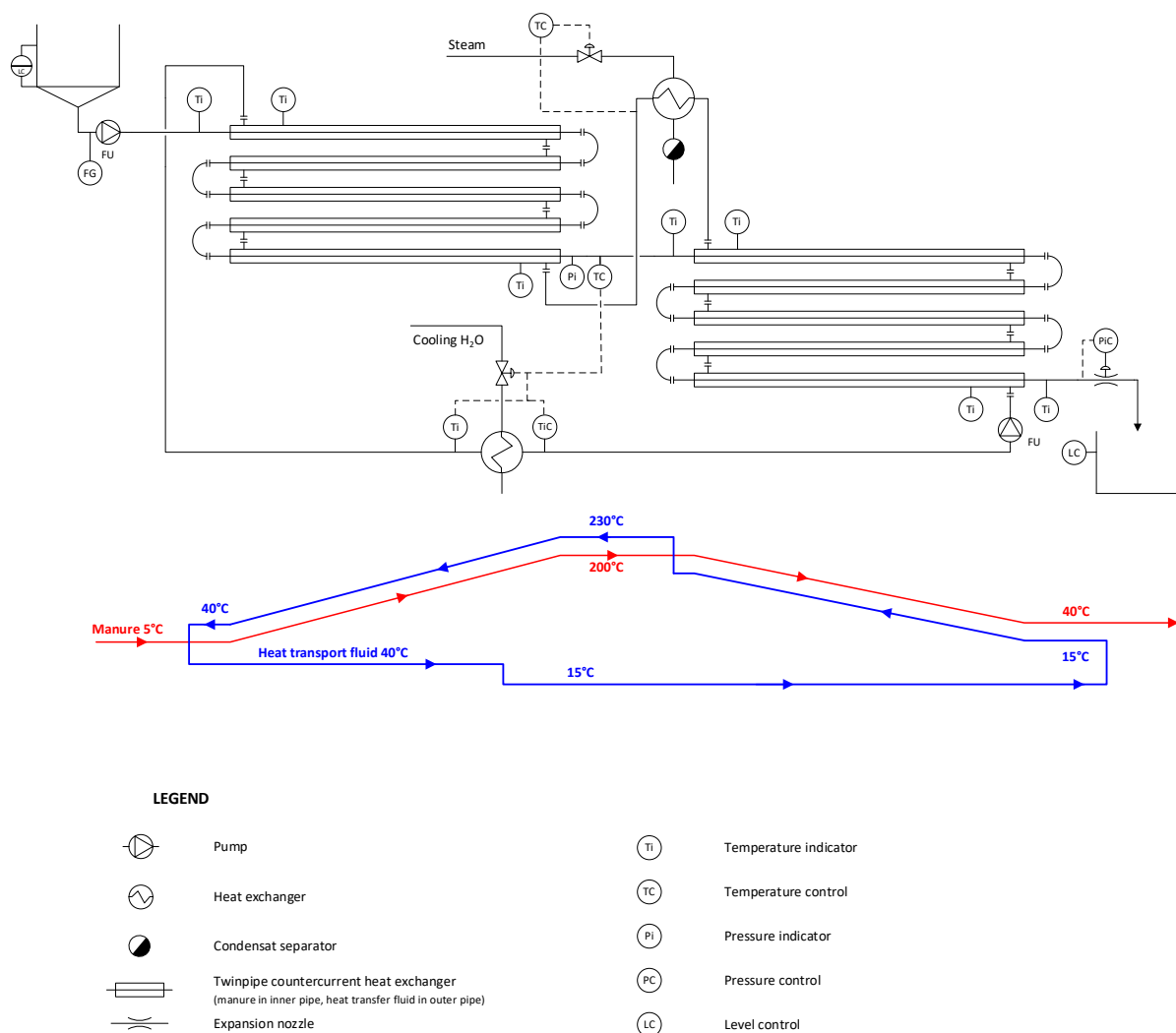


Figure 5: Basic flow sheet of the planned heat-integrated pretreatment reactor. This flow sheet shows the principle of a one-stage pretreatment unit.

Additionally, the manure slurry pumped through this above described pretreatment reactor must be compressed by a pump to a pressure larger than the vapour saturation pressure of water at the maximum pretreatment temperature (e.g. 30bar for a pretreatment at 230°C/28bar). This compression is necessary, in order to prevent boiling of the manure inside the tubular pretreatment reactor. Furthermore, it enables the 'cold explosion', *i.e.* the explosive decompression at the end of the



pretreatment reactor to ambient pressure at 40°C through the expansion of the compressed slurry through a nozzle.

### 1.3.4 Option two-stage pretreatment (Task 1.3)

Based on results from steam or hot-water pretreatment in the cellulosic ethanol industry, it is possible that the pretreatment must be carried in two-stages in order to prevent certain fraction of the manure from degradation and thereby increase biogas yields. If this will be the case our concept for a tubular pretreatment reactor based on heat exchangers in series would have to be modified (see Figure 6). One modification would have to enable the separation of the liquid from the whole slurry after reaching a certain lower temperature (e.g. 180°C). In this case a screw press could be installed after the first heat exchanger, which would allow for the solid/liquid separation under pressure. The remaining solids then need to be heated to the second pretreatment temperature. This will be done in an additional double-walled heat exchanger in which heat from the water is transferred to the manure

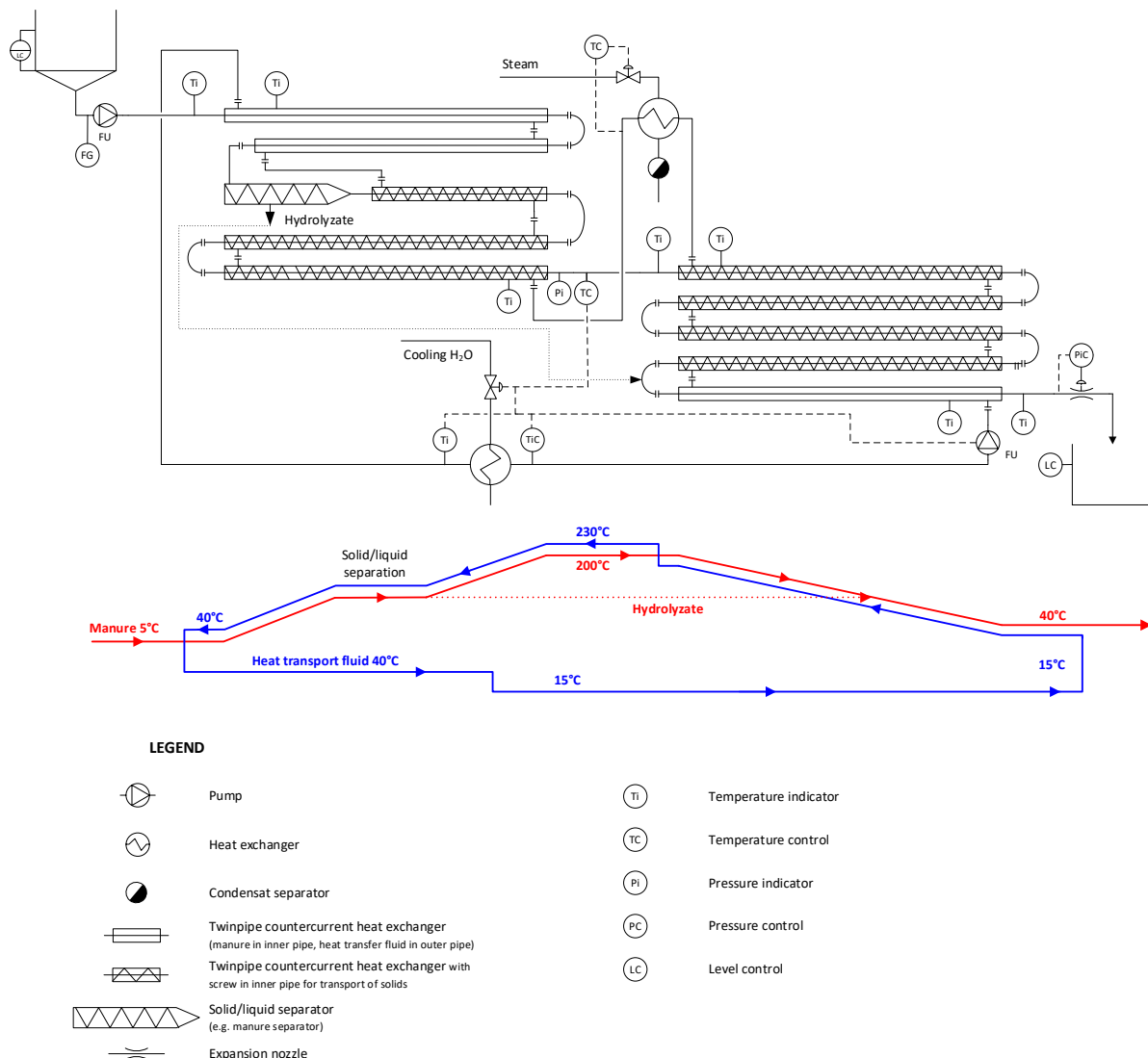


Figure 6: *Basic flow sheet of the planned heat-integrated pretreatment reactor with solid/liquid separation for two-stage pretreatment. The separated hydrolysate is mixed back to the more severe pretreated solids for high grade heat recovery.*





solids. In this heat exchanger, likely a screw conveyor would have to be installed, thus a similar reactor as the high-pressure steam pretreatment reactors in the cellulosic ethanol industry. The secondary heat exchanger, which extracts the heat from the pretreated manure would have to be identically constructed.

### 1.3.5 Sizing of the pretreatment pilot plant (Task 2.1)

In this project we aim at developing the technology to unlock the energetic potential of manure. As in the two participating countries the average dairy farm holds less than 100 cows (CH: 25, A: 30, SE: 70)) it is necessary to develop technologies that are applicable on farms with 50 to 100 dairy cows. For this proposal, we have an agricultural school at hand (IAG Grangeneuve, Switzerland) which is in the process of building a dairy stable for 50 milk cows and which would be willing to operate a biogas plant entirely using their dairy manure by applying our pretreatment technology to be developed in this proposed ERA-NET project. A dairy cow with a milk yield of 6'000kg/a produces approximately 22m<sup>3</sup>/a of manure slurry. For IAG's 50 cows stable this calculates to a volume of 1'100m<sup>3</sup>/a or a volume flow rate of roughly 2L/Min for a continuously run pretreatment facility. In order to be able to estimate the size of such a pretreatment reactor for this given flow rate of manure, the diameter of the inner tube (through which the manure slurry is flowing) of the double heat exchanger must be estimated.

However, the diameter is influenced by minimum flow velocity, the heat transfer coefficient and with that length of the tube, the solids content in the manure slurry as well as the resulting pressure drop. Estimations showed that for a tube with 20 mm diameter the mean velocity would be 0.11 m/s, which is higher than the published necessary minimum velocity to prevent sedimentation for a manure slurry with 9% dry matter. Furthermore, with a typical heat transfer coefficient for double pipe heat exchangers of 1'000W/(m<sup>2</sup> K) (Verein deutscher Ingenieure 2013) the length of the tube can be calculated to be ca. 20m (G. Hörnig 1982). For two heat exchangers in series, *i.e.* 40 m of 20 mm pipe, the resulting pressure drop is in the range of only 3 bar. Thus, if the heat exchange tube is split into pieces of e.g. 2 m length connected with 180° pipe elbows the whole installation including storage tanks, feed pump and control unit would easily fit in e.g. a 12ft container.

The goal of this task is to investigate and experimentally test the required key components in the appropriate size. These include for example pipe diameters to determine the pumpability and the pressure drop, or the heat transfer coefficients in order to determine the required length of the pretreatment equipment. Furthermore, the back pressure and explosive discharge valve must be designed, constructed and tested, as well as an eventual solids/liquid separation under pressure.

### 1.3.6 Techno-economic assessment (Task 4.2)

An assessment of the economic and environmental soundness of the process concept is in the centre of WP 4. Leader of this task is Chalmers Industriteknik (CIT). The assessment is conducted in two stages, an initial basic assessment in order to identify potential improvements to the process scheme and a detailed assessment prior to construction of a pilot plant.

A techno-economic assessment will be performed to estimate biogas production costs for representative production cases, including contributions from feedstock supply, electricity, capital costs and operational costs. A well-to-gate approach will be used to make the results relevant for commercial implementation. The overall economics are evaluated under a wide range of financial parameters in terms of euros (€) per gigajoule (GJ), based on an underlying component-level capital cost estimate.

For this second analysis an important share of the analysis and modelling work will be carried out in the Swiss sub-group.



## 2 Procedures and methodology

The Swiss part of the Era-Net project ManuMax contained three work packages:

1. *Pretreatment optimization* to find the optimal condition for each biomass fraction to maximize methane production. These results from the optimization yield the answer whether a multi-stage pretreatment must be considered. And finally, the effect of a 'cold explosion' must be investigated.
2. *Design and construction of a pre-pilot plant* test rig to test important parameters at the later scale of a pilot-plant
3. *Techno-economic model* for the Swiss case

### 2.1 Pretreatment optimization

The goal of this study was to optimize the pretreatment of manure using steam explosion and to understand i) the effect of the pretreatment on each component and ii) the influence on methane production. To this end, the manure was separated into washed solids and liquid fraction, and each fraction pretreated by steam explosion and analyzed separately. This solid-liquid separation together with the washing of the solids was necessary because the dry matter content of pressed does not exceed 30% and with that an accurate compositional analysis of the solids would otherwise not be possible.

The manure used for the experimentation was sampled on the 07/01/2019 from a dairy farm in Kernenried, Switzerland. The manure contained short (<40 mm) wheat and rape seed straw used as bedding material in the resting pens. The total dry matter content of the whole manure (*i.e.* total solids, TS) was 8.75% w/w and the organic matter (*i.e.* volatile solids, VS) was 6.26% w/w.

The solid and the liquid fraction of manure were pretreated separately between 130 and 210°C and between 5 to 40 min using a steam gun system and saturated steam at 30 bars. Hemicellulose, cellulose, lignin, ammonia and protein were analysed before and after pretreatment for each fraction. The methane potential of each fraction was also measured using a custom-made methane potential test system at 37°C during 16 days for the liquid and 21 days for the solid fraction in triplicate, using a inoculum from a biogas plant using cow manure as substrate (Vögtli, Hochwald, Switzerland). The inoculum was filtered through a cheese cloth to remove larger plant biomass particles.

The whole manure was also pretreated, with remixing the solid and the liquid fraction of manure before the steam explosion pretreatment. The manure was pretreated at 150 and 170°C from 10 to 40 min.

### 2.2 Design and construction of a pre-pilot plant

#### 2.2.1 Reasoning to design and build a test rig

The dimensions of the novel pretreatment reactor components are expected to be small compared to standard biogas plant installations as only manure from 50 livestock units (LSU) will be available at Grangeneuve. These small dimensions pose difficulties in pumping the manure through the heat exchanger pipes. Furthermore, manure is a challenging fluid due to its physical properties such as high viscosity, two phase flow and high sand load. Hence the design, construction and testing of a pre-pilot plant was deemed necessary to observe and measure critical parameters and the behavior of



manure in the pretreatment reactor as well as to test individual process units such as pumping and heating manure. The main challenge was to prevent the heat exchanger pipes from clogging by straw and other particulate matter. The planned test stand should give insights into the practical application of the pretreatment and allow for in-depth analysis of the different design options, such as one and two-stage pretreatment (section 2.2.2). Moreover, the test stand was built based on dimensions potentially later applied in the pilot facility. With a pre-pilot plant unexpected challenges and design problems can be detected and solved early in the design stage.

The following parameters must to be analyzed and their influence on the system had to be understood:

- Pressure drop in the heat exchanger
- Heat transfer as function of the tube geometry, *i.e.* tube diameter, surface geometry
- Pumpability of manure
- Heat-carrier fluid
- Explosive discharge
- Solid/liquid separation under pressure

The pipe diameter influences the pressure drop of manure in the heat exchanger and the flow velocity which are the critical parameters for the pumpability of manure. Heat transfer and especially the heat transfer coefficient for different fluid and geometrical configurations was necessary to calculate the final heat exchanger area for the pilot plant as the heat exchanger area determines the size and thus the investment cost of the heat exchanger. The explosive discharge unit is a major component of the system whose functionality must be tested, understood and optimized. Two general design concepts are presented, one concerning pretreatment of whole manure and one showing solid/liquid separation to pretreat both fractions separately.

### 2.2.2 General Concept

Manure is first compressed to reaction pressure to prevent boiling at pretreatment temperatures above 100°C. The reaction pressure corresponds to a value slightly above the vapor pressure of water at reaction temperature. Compressed manure is then brought to reaction temperature in a double pipe heat exchanger operated in counter-current flow configuration (see Figure 9). After reaction temperature is reached and held for a specified period hot manure is cooled in the second zone of the heat-exchanger to 40°C, corresponding to the fermentation temperature. In the heat exchanger high grade energy used to heat the manure is recovered during cooling of the manure and used in the heating section. After thermal pretreatment the pressurized manure at 40°C is explosively discharged to atmospheric pressure to mechanically disrupt the lignocellulosic biomass structure. In the following subsections the different options for the heat exchanger and pretreatment configurations are described.

### 2.2.3 One-stage pretreatment

In the one-stage pretreatment, the whole manure is heated to reaction temperature and subsequently cooled to fermentation temperature in a counter-current heat exchanger. In the test rig water was used



as the heat transfer fluid. The planned temperature profile of water (blue) and manure (red) are shown in Figure 9. The only external energy input is at the most elevated manure temperature where the sign of the temperature difference  $\Delta T$  in the heat exchanger must be changed, i.e. the water from the manure-cooling-section of the heat exchanger must be superheated against the manure temperature.

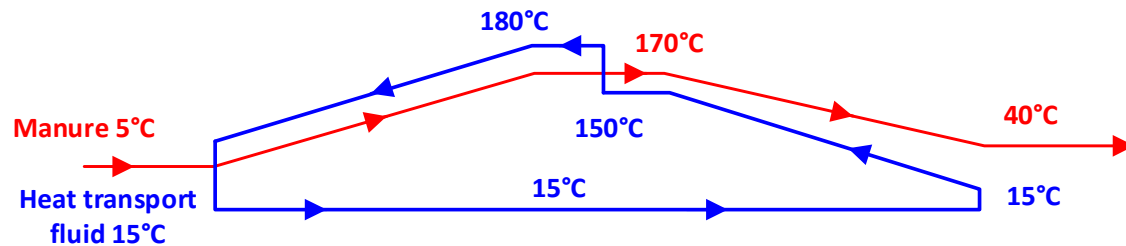


Figure 7: Temperature profile for manure one-stage pretreatment at 170°C. Manure shown in red is heated and cooled with the water cycle depicted in blue.

#### 2.2.4 Two-stage pretreatment

A two-stage pretreatment must be applied if certain fractions from manure are thermally unstable. In that case the goal is to pretreat first at a lower temperature, solubilize these thermally unstable substances and recover them with the liquid phase. The remaining solids can then be pretreated at a higher temperature to prepare them for the subsequent biochemical conversion in the biogas fermenter. Thus, the goal of the experiments in the test rig was to test a solids/liquid separation.

#### 2.2.5 Fundamentals of heat transfer

The theory of heat transfer presented in this section is a short summary, more information can be found in the VDI heat atlas (Verein deutscher Ingenieure 2013). In heat transfer, three modes are distinguished:

- Conduction
- Convection
- Radiation

Conduction is the transfer of energy due to the random motion of the atoms between neighboring molecules. It occurs in solids, liquids and gaseous media. The heat flux in one dimension is given by equation (2) with  $\lambda$  called thermal conductivity.

$$\dot{q} = -\lambda \frac{\partial T}{\partial x} \quad (2)$$

Convective heat transfer refers to the heat transport in flowing media. The simplest form to calculate the heat flux is shown by equation (3) with  $\alpha$ , heat transfer coefficient and  $T_W - T_F$ , the temperature difference between a wall and the bulk fluid.

$$\dot{q} = \alpha \cdot (T_W - T_F) \quad (3)$$

Thermal radiation is the energy emitted by any matter to its surroundings. Radiative heat transfer comes does not play an important role for the temperature range in this work for liquid heat exchanger and is therefore not further discussed.



Heat flow  $\dot{Q}$  between two temperature levels can be calculated in two different ways. Either using enthalpies or with the heat capacity. In general, the heat flow is given by equation (4).

$$\dot{Q} = \dot{m} \cdot (h_2 - h_1) \quad (4)$$

For small temperature differences the enthalpy can be approximated as heat capacity times temperature (equation(5)). The obtained value diverges from the actual value calculated with enthalpies when the temperature difference is increasing because the heat capacity  $c_p$  is temperature dependent.

$$\dot{Q} = \dot{m} \cdot c_p \cdot (T_2 - T_1) \quad (5)$$

### 2.2.6 Influence of the velocity on heat transfer

Heat transfer in pipe flow is heavily dependent on the flow regime. The dimensionless number describing the flow regime is the Reynolds number  $Re$ . It is defined by equation (8) with the velocity  $w$ , the pipe diameter  $d_i$  and  $\nu$  the kinematic viscosity (Verein deutscher Ingenieure 2013).

$$Re = \frac{wd_i}{\nu} \quad (6)$$

If the Reynolds number is less than  $Re = 2300$  pipe flow is laminar. In the transition region  $2300 < Re < 10^4$  the type of flow is influenced by the form of the pipe inlet and the nature of the inlet stream. At  $Re > 10^4$  the flow is turbulent. In the turbulent flow regime heat is transferred very rapidly due to the agitation of the fluid. In laminar flow a boundary layer is formed restricting heat transfer. The transition regime is between these two heat transfer ranges but still shows inferior heat transfer properties (Verein deutscher Ingenieure 2013).

### 2.2.7 Overall heat transfer coefficient

Heat transmission through a plane wall is the common heat transfer mechanism for heat exchangers. Heat is transferred from a bulk fluid to the heat exchanger wall by convection, conducted through the wall and transferred to the second fluid again by heat convection. The overall heat transfer coefficient can be calculated using each individual heat resistance added together. Thus, the overall heat resistance is given as follows:

$$R = \frac{1}{\alpha_i A} + \frac{\delta}{\lambda A} + \frac{1}{\alpha_o A} \quad (7)$$

Where  $\delta$  is the wall thickness and  $A$  signifies the heat transfer area.  $\alpha_i$  and  $\alpha_o$  are the convective heat transfer coefficient at the pipe wall on inner and outer side, respectively.  $\lambda$  signifies the heat conduction coefficient of the pipe wall material.

## 2.3 Techno-economic model

A techno-economic model was developed to calculate the net present value (NPV) for the envisioned manure-based biogas plant at IAG in Grangeneuve employing a thermal pretreatment. All biogas was assumed to be converted to electricity in a combined heat and power plant (CHP). The techno-economic model was programed in Matlab. Cost functions for the investments of i) fermenter, ii) the pretreatment equipment, iv) the gas storage and v) for the CHP were developed. The operating costs



were estimated employing a sixth cost function and the calculated revenue was divided into an electricity and a heat part. A sensitivity analysis gave insight into the most influential parameters on the NPV.

## 3 Results and discussion

### 3.1 Optimization of steam pretreatment of dairy cow manure

*Detailed information about the optimization of steam pretreatment of manure will be provided in this report as soon as the data were published in a peer-reviewed journal.*

The amount of glucose/cellulose, xylose/hemicellulose and protein was very low ( $< 1$  g) in the liquid fraction of manure, and with that substances were highly diluted and close to the detection limit. The pretreatment of this fraction at conditions of 130 to 190°C caused high losses of carbohydrates of 20 to 60% w/w and of proteins from 40 to 60%. Moreover, the pretreatment did not have a positive effect on methane yield of the liquid phase for all tested conditions. If the BMP is based on the mass VS after pretreatment the methane yield is approximately the same as for the raw manure, but if the BMP is based on the mass VS in the raw manure the yield is lower than for the raw manure.

However, for all conditions tested – including the raw manure liquid – the fermentations were terminated within 10 days.

*In conclusion, the liquid fraction of manure does not benefit from a steam pretreatment. Neither the methane yield nor the methane production rate was increased.*

The manure solids were pretreated separately from the manure liquid. Saturated steam was injected into the washed wet manure solids to heat them up to the target pretreatment temperature. Thus, during pretreatment of wet manure solids a slurry is produced. The condensate together with dissolved substances from the solids formed the hydrolysate, which was separated from the remaining pretreated solids. These solids were washed in a filter bag and pressed to increase the dry matter content. The hydrolysates were centrifuged to remove any particulate matter. The precipitates solids were (washed and) added to the solids obtained by filtration.

A mass balance shows, that during steam explosion, degradation and solubilisation of the solids remained approximately constant for all tested pretreatment conditions up to 170°C and only increased for higher temperatures (190 and 210°C). The maximum fraction which was solubilized was 24% of the organic matter at the pretreatment condition of 210°C and 10 Min.

Cellulose was neither solubilised nor degraded at the chosen pretreatment conditions. This is likely due to the not sufficiently harsh pretreatment condition, but also due to the high pH and the high buffer capacity of manure, which prevented auto-hydrolysis reactions of cellulose.

Analogous to the cellulose the recovery of the hemicellulose remained constant for all pretreatment conditions at temperatures up to 170°C and solubilization was low. However, for pretreatment severities above 170°C/10 min the degradation as well as the solubilization increased up to a maximum of 40% for the highest tested condition of 210°C/10 min. Furthermore, this increased solubilization was accompanied with an increased degradation of the solids of approximately 40% w/w, which corresponds to a loss of ca. 25% of the standard biogas yield from raw manure.



Proteins were the substance that was most negatively affected of all non-water-soluble substances. A very high loss of 35% w/w was measured already for the pretreatment condition with the lowest tested severity, 130°C/10 min. The protein loss during the pretreatment increased with increasing pretreatment temperature and time and reached a maximum of 60% w/w for the highest tested temperature of 210°C. In the hydrolysate no dissolved proteins were detected. The amount of ammonia increased with increasing pretreatment severity, thus we speculate that proteins were degraded to form ammonia.

The initial methane production rate of the pretreated solids increased with steam pretreatment for the tested conditions, likely due to the thermochemically solubilized fractions. 70 to 117% of the BMP for pretreated solid was already produced at 10 days (compared to the 65% for the raw solid). The maximum biogas yield (in [mL<sub>CH4</sub>/g<sub>VS, raw manure</sub>]) was only increased for certain conditions compared to reference case of raw manure solids. Generally, only the pretreatment conditions at temperatures of 130°C and 170°C increased the biogas yield (at 25 days) by ca. 30%. This percentage increase may seem small, however the total BMP (including the liquid phase) was 290 Nm<sup>3</sup>/t<sub>VS</sub>, which is 70% higher than the often-used default value of 170 Nm<sup>3</sup>/t<sub>VS</sub> for methane potential from dairy cow manure.

*In conclusion, i) BMP of manure solids can be increased by steam pretreatment for certain conditions and ii) the fermentation time was reduced to < 15 days.*

## 3.2 Test stand

### 3.2.1 Heat exchanger design

#### Pipe diameter

The most critical parameter for the heat exchanger design is the pipe diameter. With smaller pipe diameters the flow velocity in the pipe increases along with the pressure drop. However, high pressure loss leads to an increased energy requirement for manure pumping through the long heat exchanger pipes (varying between 10-100 m depending on the heat-transfer coefficients). Furthermore, at very low flow velocities it must be assumed that sand and other small particulate matter settles in the pipe to build up a plug, which completely clogs the pipe. Hörnig published a minimum flow velocity of 0.18 m/s for cow manure in a pipe with a diameter of 80 mm in order to prevent clogging (G. Hörnig 1982). Natural Resources Conservation Service of America reported a minimum flow velocity of even 0.5 m/s independent of the pipe diameter. As due to the number of cows a constant volume flow rate of manure of 4.4 L/Min is given, the flow velocity and the following pressure drop must be calculated.

The equation for the pipe velocity is as follows:

$$v = \frac{4\dot{V}}{\pi d^2} \quad (8)$$

And the pressure loss can be estimated employing the Hagen Poiseuille equation, which is actually only valid for homogeneous and Newtonian fluids, which is both not given for manure.

$$\Delta p = \frac{8\dot{V}\eta l}{\pi r^4} \quad (9)$$



Table 3: Influence of pipe diameter on pressure loss and flow velocity

| Pipe diameter | Pressure loss | Flow velocity |
|---------------|---------------|---------------|
| DN 10         | 0.85 bar/m    | 0.93 m/s      |
| DN 15         | 0.34 bar/m    | 0.41 m/s      |
| DN 20         | 0.11 bar/m    | 0.23 m/s      |
| DN 25         | 0.05 bar/m    | 0.15 m/s      |

Based on these results, a pipe diameter of 25 mm was chosen. The heat exchanger in the test-stand had a length of 24 m corresponding for a DN25 pipe to a total pressure loss of 1.3 bar, which was assumed to be acceptable. Furthermore, the flow velocity is with 0.15 m/s similar to the published 0.18 m/s.

### Energy balance for one-stage pretreatment

With the heat exchanger design and assuming adiabatic conditions, all heat needed to reach reaction temperature of e.g. 170°C can be recovered when cooling the heated manure. However, a certain additional heat input remains to change the heat exchanger configuration from heating to cooling manure. Therefore, to change the sign of the  $\Delta T$  in the heat exchanger an electric heater was installed to increase the temperature of the recirculating water. The needed power input is given by the energy balance. A reaction temperature of 170°C was assumed for the energy balance based on preliminary results of the pretreatment optimization. The following parameters are used for the energy balance:

Table 4: Parameters used for the energy balance of the one-stage manure pretreatment

| Parameter   | Value                    | Source                                 |
|---|--------------------------|--|
| Manure flow rate, $\dot{V}_m$   | 4.4 l/min                | Given by IAG for 50 LSU                |
| Manure density at 20°C, $\rho_m$  | 1034.1 kg/m <sup>3</sup> | (Achkari-Begdouri and Goodrich 1992)   |
| Heat capacity of wheat straw, $c_{p,ws}$  | 2.1 kJ/kgK               |  |
| Wheat straw mass fraction, $x_{ws}$   | 0.04                     | Given by the IAG                       |
| Manure inlet temperature, $T_{in}$  | 5°C                      | Assumption in winter                   |
| Manure reaction temperature, $T_{react}$  | 170°C                    | Given by the pretreatment optimization |
| Enthalpy of liquid water at reaction temperature and 25 bar, $h_{w,react}(170^\circ\text{C})$ | 720.61 kJ/kg             | (Moran and Shapiro 1995)               |
| Enthalpy of liquid water at manure inlet temperature and 25 bar, $h_{w,in}(5^\circ\text{C})$  | 23.69 kJ/kg              | (Moran and Shapiro 1995)               |





|  |              |                          |
|--|--------------|--------------------------|
| Enthalpy of liquid water at water inlet temperature and 25 bar, $h_{w,h,in}(180^{\circ}\text{C})$  | 763.97 kJ/kg | (Moran and Shapiro 1995) |
| Enthalpy of liquid water at water outlet temperature and 25 bar, $h_{w,h,out}(15^{\circ}\text{C})$ | 65.43 kJ/kg  | (Moran and Shapiro 1995) |

First, the energy to heat manure from inlet temperature,  $T_{in} = 5^{\circ}\text{C}$  to reaction temperature,  $T_{react} = 170^{\circ}\text{C}$  is calculated with equation (10).

$$\dot{Q}_{heat} = \dot{m}_{manure} \cdot x_{ws} \cdot c_{p,ws}(T_{react} - T_{in}) + \dot{m}_{manure} \cdot (1 - x_{ws}) \cdot (h_{w,react}(170^{\circ}\text{C}) - h_{w,in}(5^{\circ}\text{C})) \quad (10)$$

The heat calculation is split into two parts, one for the solid fraction and one for the liquid fraction of manure. For the liquid part the enthalpy of water is used. For the solid fraction the heat capacity with the given temperature difference was inserted since the heat capacity can be assumed to stay constant over the temperature for solid material. For the solid fraction the heat capacity of wheat straw was applied.

The heat needed to reach manure reaction temperature was provided by a counter current water flow. Assuming no heat losses to the environment for the heat exchanger, the mass flow of water can be calculated according to equation (11). The temperature difference between both liquids over the length of the heat exchanger was assumed to be a constant value of  $10^{\circ}\text{C}$ .

$$\dot{m}_w = \frac{\dot{Q}_{heat}}{h_{w,h,in}(180^{\circ}\text{C}) - h_{w,h,out}(15^{\circ}\text{C})} \quad (11)$$

For the heat recovered from cooling manure to fermenter temperature equation (12) is applied:

$$\dot{Q}_{cool} = \dot{m}_{manure} \cdot x_{ws} \cdot c_{p,ws}(T_{react} - T_{ferment}) + \dot{m}_{manure} \cdot (1 - x_{ws}) \cdot (h_{w,react}(170^{\circ}\text{C}) - h_{w,ferment}(40^{\circ}\text{C})) \quad (12)$$

Again, assuming no heat losses over the heat exchanger, the obtained final water temperature can be calculated. The equation for the output enthalpy of the water stream at the hot side is given by (13).

$$h_{w,c,out} = \frac{\dot{Q}_{cool}}{\dot{m}_w} + h_{w,c,in}(15^{\circ}\text{C}) \quad (14)$$

The corresponding temperature can be found in the enthalpy tables (Moran and Shapiro 1995).

Finally, the heat to change the  $\Delta T$  of the water in the heat exchanger can be calculated, which is the actual heat input to the system provided by the electrical heater.

$$\dot{Q}_{overheat} = \dot{m}_w \cdot (h_{w,h,in}(180^{\circ}\text{C}) - h_{w,c,out}) \quad (15)$$



Table 5: Results of the energy balance for the one-stage pretreatment

| Parameter            | Value        |
|----------------------|--------------|
| $\dot{Q}_{heat}$     | 51.79 kW     |
| $\dot{m}_w$          | 0.074 kg/s   |
| $\dot{Q}_{cool}$     | 40.93 kW     |
| $h_{w,c,out}$        | 617.52 kJ/kg |
| $T_{w,c,out}$        | 150°C        |
| $\dot{Q}_{overheat}$ | 10.86 kW     |

The heat input needed to superheat the water is the same as the difference  $\dot{Q}_{heat} - \dot{Q}_{cool}$  which is the energy to heat manure from the input temperature  $T_{in} = 5^\circ\text{C}$  to fermentation temperature  $T_{ferment} = 40^\circ\text{C}$ . Analyzing the energy balance over the whole system, for an adiabatic system the only energy input is given by the temperature difference of manure inlet to manure outlet being  $\dot{Q}_{heat} - \dot{Q}_{cool} = 10.86 \text{ kW}$ .

#### Energy balance for two-stage pretreatment

For the two-stage pretreatment, the solid phase is separated from whole manure. A solid fraction of 30 % w/w ( $x_{solid,sep}$ ) is assumed for the separated solids, which is a typical value for commercial manure separators. Using the total manure flow rate with a solid fraction of  $x_{ws} = 0.04$  the final mass flow rate of the solid phase is calculated (equation (16)).

$$\dot{m}_{solid\ phase} = \dot{V}_m \cdot \rho_m \cdot \frac{x_{ws}}{x_{solid,sep}} \quad (16)$$

Using the same temperature levels and equations as for the one stage pretreatment, the total energy necessary to heat wet manure solids is  $\dot{Q}_{solid\ phase} = 5.9 \text{ kW}$ . Thus, if we assume that the manure liquid must not be pretreated, the heat input to pretreat the solids is even lower than the heat necessary for the one stage pretreatment. Or in other words, the energy necessary to pretreat the separated solids alone at  $170^\circ\text{C}$ , is not sufficient to heat the remixed whole manure (i.e. non-pretreated liquid plus pretreated solids) to fermentation temperature. Additional energy is necessary to heat the whole manure to fermentation temperature. Thus, in either process configuration (whole manure in counter-current heat exchanger or only pretreating the solids (and remixing them with non-pretreated liquid) no additional energy is consumed by the supplementary pretreatment compared to a conventional biogas plant. The only difference for heat provision is the higher temperature level (i.e.  $170^\circ\text{C}$  instead of ca.  $80^\circ\text{C}$ ).

Table 6: Results of the energy balance for the two-stage pretreatment

| Parameter                | Value       |
|--------------------------|-------------|
| $\dot{m}_{solid\ phase}$ | 0.0101 kg/s |
| $\dot{Q}_{solid\ phase}$ | 5.9 kW      |



### Heat exchanger length

The length of the heat exchanger in the test rig was set to 12 m for each heating and cooling of the manure giving a total length of 24 m. The length was a first estimate because the goal of the test stand was primarily to measure the heat transfer coefficient and not to reach the final pretreatment temperature.

### 3.2.2 Manure Feed

Manure feed was achieved by pressurizing the storage vessels equipped with a dip tube using compressed air. To convey manure through the heat exchanger, first two parallel tanks of 150 L volume are filled with fresh manure (Figure 27). Two valves on top of both tanks (V5, V6) allow air inside the tank to escape. A radar level sensor (FMP51, Endress&Hauser, Reinach, Switzerland) indicates the tank level. Both tanks are connected to a feed pipe by two valves located on the bottom and top of the tank, the bottom valve allows liquid manure to enter the feed pipe. Through the top valve (V1), air can enter the tank while feeding manure. During one cycle, one tank is connected to the feed pipe while the valves to the other tank remain closed. The feed pipe is equipped with an immersion pipe, when air enters the feed pipe the pressure raises and manure is fed through the heat

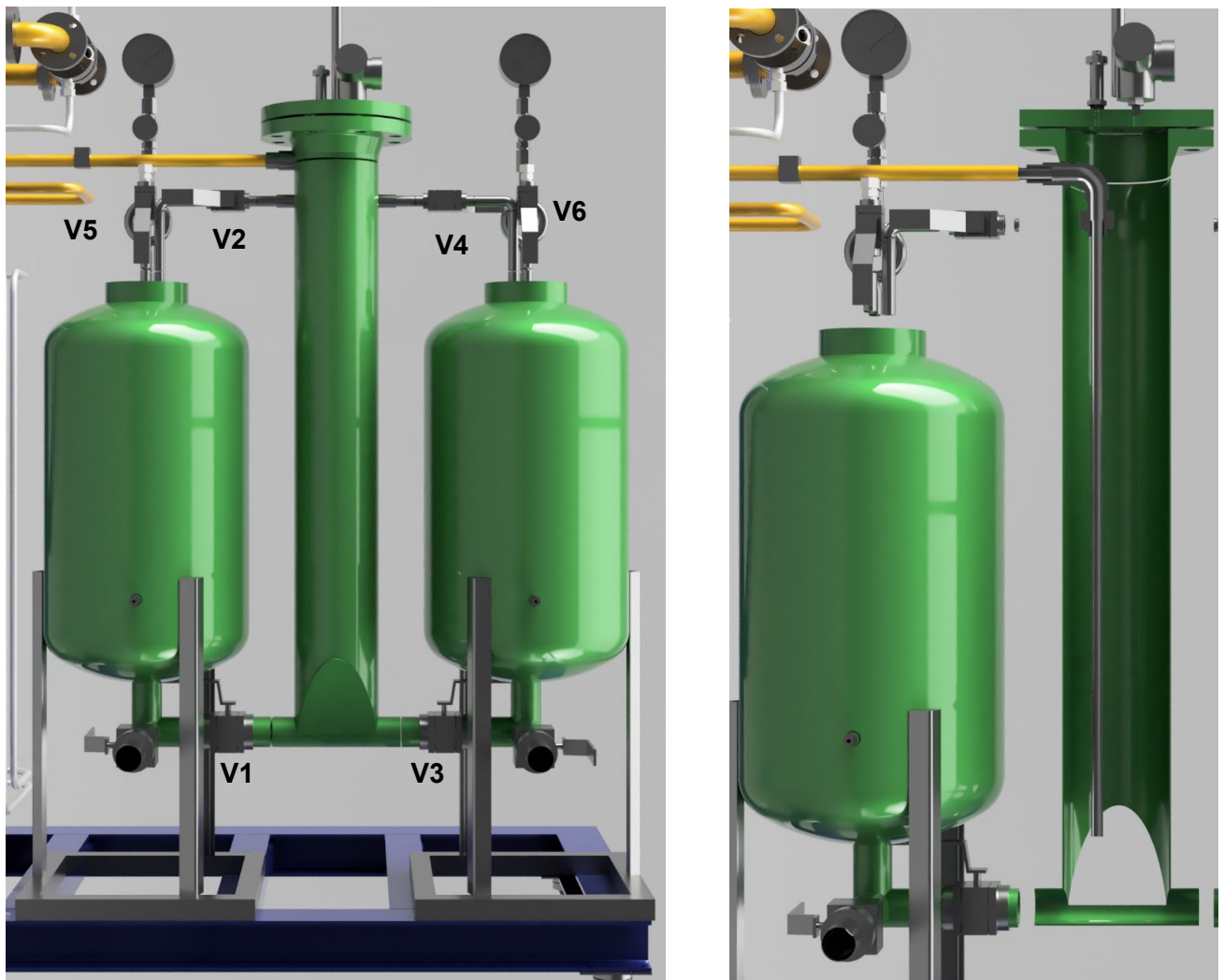


Figure 8: Left is the manure feeding device in green. On the right the immersion pipe is shown into the feed tank.



exchanger. After one tank is emptied, the two valves connecting it to the feed pipe (V1, V2) are closed and both valves (V3, V4) at the second tank are opened. Since the second tank is filled with manure and does not contain any air, it is directly pressurized when both valves are opened.

### 3.2.3 Explosive Manure Outlet

After manure was heated and cooled in the heat exchanger, it is explosively discharged through a self-designed valve (Figure 28). A conical piston is spring loaded pressed against the valve seat to maintain the high pressure (higher than the saturation pressure of the manure at the hottest position in the heat exchanger). The adjustable spring force sets the threshold pressure to release a constant manure flow.

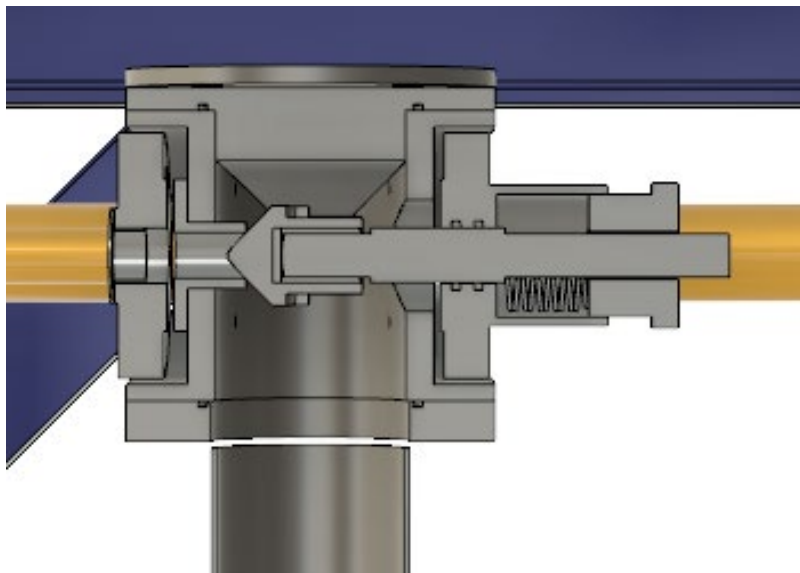


Figure 9: Manure outlet valve. A spring loaded conical piston is pressed against the valve seat.

Figure 29 shows the complete test stand. The total length of the stand is 8 m and the height is 2.3 m. The pilot plant is mounted on a supporting structure shown in blue. To move the PrePilot plant wheels are fixed to the support structure.

### 3.2.4 Overall heat transfer coefficient for tests with water on both sides of the heat exchanger

In a first test series, all systems were tested with water and the overall heat transfer coefficient for water was measured.

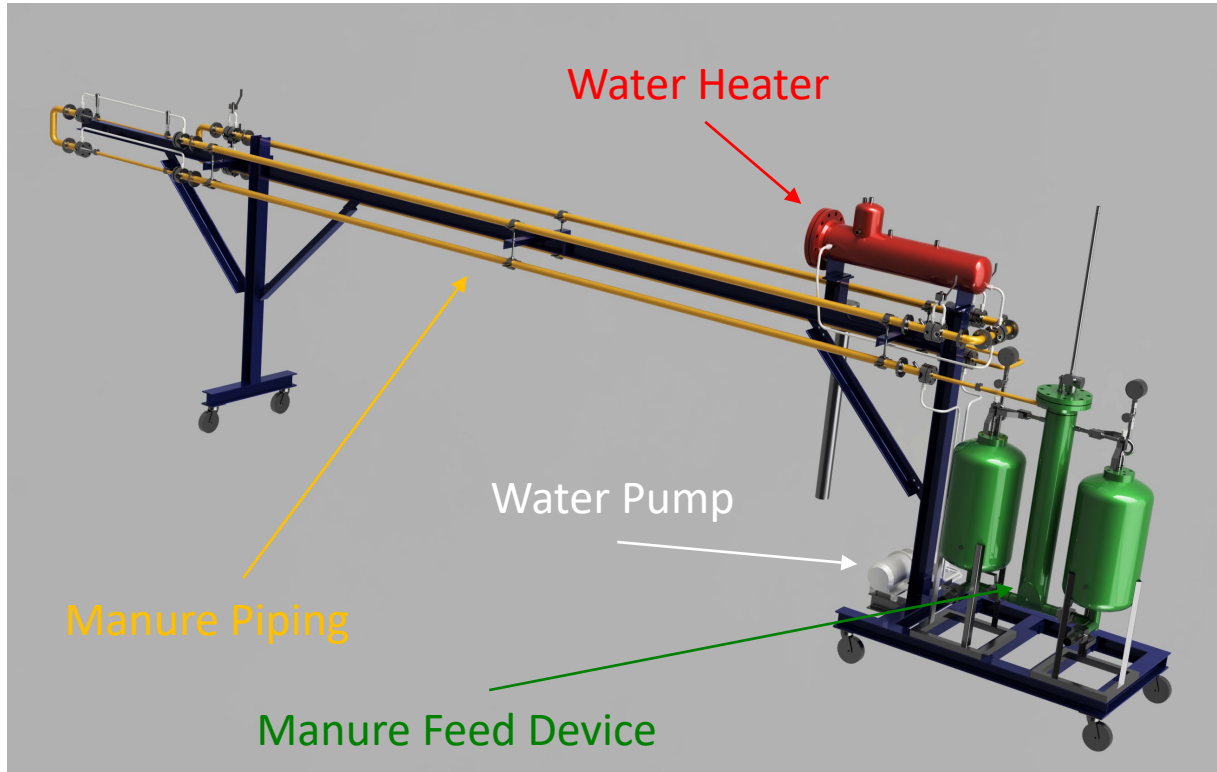


Figure 10: Test stand to analyze the feasibility of pretreating dairy cow manure in a counter-current heat-exchanger. Manure is first filled into the green feeding system. Then it is pumped through the yellow heat exchanger pipes and released at the end through the explosion valve. Water is circulated by the water pump shown in white in counter-current direction through the heat exchanger and heated with the immersion heater shown in red.

### Experimental procedure

To calculate the overall heat transfer coefficient, the temperature at the beginning and the end of each heat exchanger pipe (every 6 m) is measured and logged every 5 min, together with the volume flow rate for both liquids. Using the densities and the heat capacities, the overall heat transfer coefficient for the heat exchanger can be calculated for each 6 m heat exchanger tube.

In a first step the heat transferred over the heat exchanger is calculated (equation (18)(17)).

$$\dot{Q}_{12} = \dot{V} \cdot \rho \cdot (T_2 - T_1) \quad (17)$$

The heat exchanger area is given by:

$$A_{12} = l_{12} \cdot \pi \cdot \frac{d^2}{4} \quad (18)$$

The log mean temperature difference for the heat transfer coefficient is defined with equation (19)

$$\Delta T_{lmtd} = \frac{(T_{hot,in} - T_{cold,out}) - (T_{hot,out} - T_{cold,in})}{\log \left( \frac{T_{hot,in} - T_{cold,out}}{T_{hot,out} - T_{cold,in}} \right)} \quad (19)$$



The overall heat transfer coefficient  $k$  can be calculated according to equation (20), if all these values are known for one 6 m heat exchanger tube,

$$k_{12} = \frac{\dot{Q}_{12}}{A_{12} \cdot \Delta T_{lmtd}} \quad (20)$$

For the experiments, the flow rate of the primary liquid was set to a constant value by adjusting the spring force at the manure outlet valve.

Also, the water mass flow rate was adjusted so that the temperature of the water must increase by 20°C over the heater (71.0 – 51.5°C in Figure 30). This was calculated based on the energy required to heat the manure from ambient temperature to 40°C and the manure flow rate.

### Results for the mass transfer coefficients

The following  $k$ -value were calculated at two different water flow rates on the primary side (manure side)

Table 7: Experimental results for the overall heat transfer coefficient of the manure heat exchanger at a flow rate of 2 and 4 l/min.

| Water flow rate (manure side) | Water flow rate (water side) | Energy input heater | $k$ -value                                    |
|-------------------------------|------------------------------|---------------------|---|
| 2 l/min                       | 2.338 l/min                  | 3.19 kW             | 213 W/m <sup>2</sup> K ± 17W/m <sup>2</sup> K |
| 4 l/min                       | 4.68 l/min                   | 6.39 kW             | 307 W/m <sup>2</sup> K ± 14W/m <sup>2</sup> K |

Figure 30 and Figure 31 show the temperature profile for the experiment with 2 l/min and 4 l/min water flow rate on the manure side. In both experiments the ‘manure’ could be heated in the first heat exchanger tube and cooled in the second to reach the targeted 40°C at the outlet. However, the temperature curves are not parallel as expected. The reason for this is the different heat amount that must be transferred to and from the manure in the heating and cooling heat exchangers, respectively, but their identical length. For a constant  $k$ -value the second heat exchanger would have to be shorter, as the manure must be heated from ambient temperature to pretreatment temperature and cooled down to 40°C.

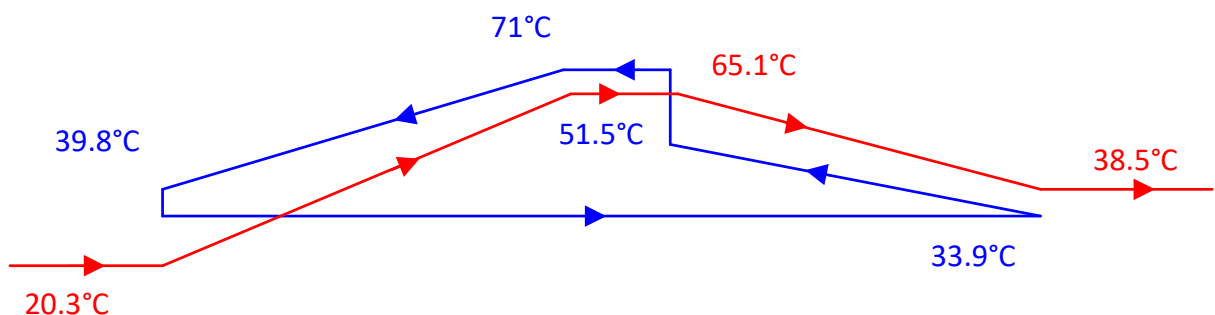


Figure 11: Temperature profile for water test with a water flow rate at the manure side of 2 l/min

The VDI heat atlas states  $k$ -values of 300 – 1'400W/m<sup>2</sup>K for liquid/liquid heat transfer for double pipe heat exchangers (Verein deutscher Ingenieure 2013). Thus, the here measured heat transfer



coefficients are very small. The reason for such small values is the very low liquid volume flow rate. For the given diameters the primary side is in the transition regime and the water side in the laminar region. Typical heat exchangers operate with both liquids in the turbulent flow regime.

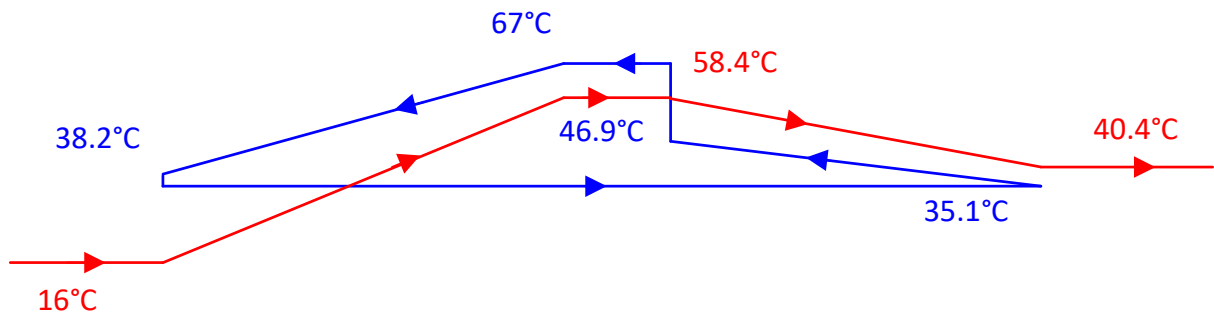


Figure 12: Temperature profile for water test with a water flow rate at the manure side of 2 l/min

### 3.2.5 Pumpability test with liquid manure

As a first test the pumpability of manure without heating was tested through the whole heat exchanger system.

#### Experimental procedure

The procedure is as described in paragraph 3.2.2 Manure Feed. The water was not circulated as only the pumpability was tested in a first run. The flow rate was set to 4 l/min by adjusting the backpressure through the explosion valve for a given pressure in the feed tank.

#### Results for the pumpability of manure

When feeding whole manure to the heat exchanger pipes, the pipes were instantly blocked. In each test run a plug of manure solids (basically chopped straw, ca. 40 mm length) was formed in the heat exchanger tube. These plugs could not be removed even by increasing the pressure in the feed tanks up to 20 bar. There are different several reasons leading to plug formation within the tubes: i) a too low flow velocity, ii) sharp edges at the connection of two heat exchanger pipes or at the temperature sensor inlets, iii) too small diameters of the inner tubes, and iv) the backpressure valve does not allow solids to pass. Figure 32 left, shows a manure plug at the connection flanges of two heat exchanger pipes. The straw in the plug is well visible. In a first attempt the flow rate of manure was increased by removing the backpressure valve from 10 to ca. 45 l/min. With such high flow velocities, whole manure could be pumped without problems through the tubes without clogging (Figure 32, right). However, such high flow rates are 10-fold too high for a pilot plant at the IAG site.



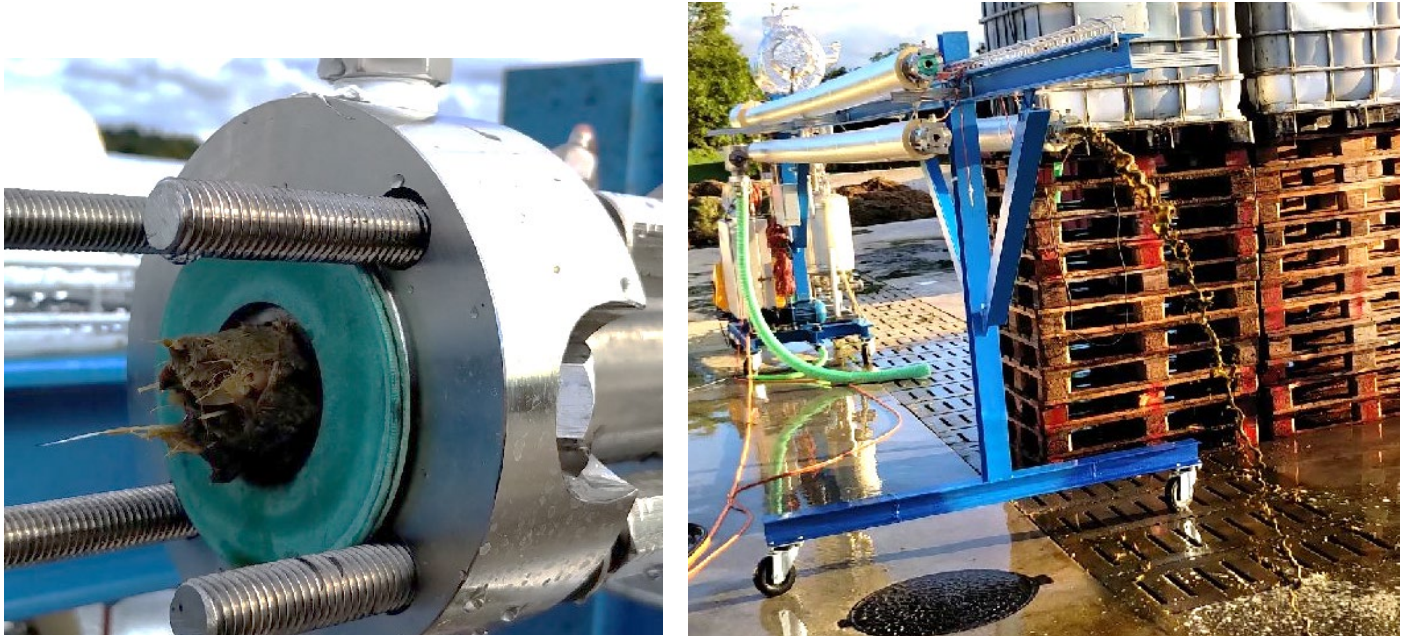


Figure 13: Left: Manure plug at the connection of two heat exchanger pipes. Right: Manure flowing out of one heat exchanger pipe at a flow rate of 45 l/min

### 3.2.6 Conclusion and next steps

To prevent abrasion of pump devices when pumping manure, the feeding system is performed pneumatically. The implementation and execution of that feeding device worked well and is a suitable solution for small flow rates of manure of 4 l/min at Swiss farm scale. The energy input to produce compressed air for the compression and feeding is very low with 0.4 kW, compared to the in total produced energy of 16 kW.

In the current heat exchanger design, manure solids are likely to clog the heat exchanger tubes due to sharp edges and a too low flow velocity. The sharp edges will be removed by changing the design of the heat exchanger connections (adjusting edge, sealing by O-rings) and by replacing the sensors sticking into the manure flow by plugs which will be smoothed on the inside of the tubes. Increased flow rates showed no clogging and could therefore be a solution by adding e.g. compressed air to the manure to form a three-phase flow. However, the increased flow rate would consume large amounts of compressed air and would thus be an energy intensive design change. Moreover, increasing the gas fraction of the manure stream would reduce the heat transfer even further. With these design changes the pumpability of manure will be tested again.

The heat transfer coefficients of the designed heat exchanger are for the low water flow rates of 2 and 4 l/min very small compared to literature values. The reason is the low flow velocity and the resulting laminar flow regime in the pipe and the transition flow regime for the water in the annular section. Such low heat transfer coefficients lead to very large necessary heat exchanger surfaces. Furthermore, the heat transfer coefficients for manure is expected to be even lower than for water due to the high viscosity. For the given DN25 tubes, this would result in very long heat exchangers in the range of 100-200 m. Such lengths would not only result in higher investment costs and higher pressure drops but would also increase the risk of clogging.





Summarizing, it must be emphasized that pumping whole dairy cow manure (*i.e.* containing short straw) continuously at such small volume flow rates (*i.e.* manure from 50 LSU) does not seem feasible without a high risk of clogging. Furthermore, due to the small heat transfer coefficients measured for the given situation, a potential double pipe heat exchanger would have to be built very long and would with that be quite expensive to acquire and operate, due to the high pressure drop.

### 3.3 Techno-economic model

*Detailed information about the techno-economic model will be provided in this report as soon as the data were published in a peer-reviewed journal.*

The evaluation of the techno-economic model yields a break-even for the number of cows for the net present value (NPV) for Switzerland at 221 cows and for Sweden at 331 cows. The difference is due to different values for both countries for the labour cost and electricity and heat selling prices.

The sensitivity analysis for both countries showed that the total investment cost have the largest impact on the NPV followed by the methane yield.

## 4 Conclusions

With an elaborate pretreatment optimization study, we found out that the liquid fraction of dairy cow manure does not need to be pretreated. Organic matter in the liquid is lost during steam pretreatment and with that the biomethane potential is hampered. The manure solids however benefit from steam pretreatment and under the given experimental conditions (manure solids/liquid separation, washing of solids) an increase in biomethane of up to 50 % could be shown. Nevertheless, the option not to use the liquid phase for biogas production but only the pretreated solids is not feasible as for all tested pretreatment conditions always a minimum of 30 % of the total biogas is produced from the raw liquid. Another important finding about the steam pretreatment is the much-reduced fermentation time of the pretreated solids. The fermentation is thus completed for the raw liquid and the pretreated solids in less than 10 days.

A techno-economic model showed that a biogas plant producing electricity for self-consumption at a price of 20 Rp/kWh would need manure from more than 300 and 500 LSU with and without a pretreatment step, respectively. Thus, often cited 'smallest-scale' biogas plants on Swiss farms do not seem feasible. A sensitivity analysis also revealed that total investment cost, followed by the methane yield and personnel expenses are the main cost drivers. Thus, a pretreatment step might indeed contribute to a profitable biogas production from manure in several ways. First, the biogas yield is increased leading to a higher revenue. And second, thanks to pretreatment also the residence time in the fermenter is sharply reduced, which offers potential cost savings by smaller fermenters. The fermenters must potentially also be designed differently as 10 days residence time in standard stirred tank reactors might lead to wash out of certain groups of microorganisms

## 5 Outlook and next steps



The results from the pretreatment optimization at laboratory scale and from the techno-economic analysis seem promising to increase the use of manure as a substrate for biogas production. However, it is unclear how such a process can be operated continuously and for an extended period of time, as well as by how much the biogas yields can be increased under real conditions. We are therefore confident to bring the project ManuMax to the pilot phase. We plan to design, construct and commission a pretreatment unit. In order to be able to analyse the fermentation of the pretreated material we will also need a small biogas fermenter, in which only the pretreated manure will be digested. We plan to pilot the process on a scale using manure of only 50 LSU, knowing that manure of more than 300 LSU would be necessary for an economic operation. But even if the size is smaller, it allows the technology to be tested on a real scale - so that it can then be implemented on farms – but features the advantages of lower investments and the installation at an agricultural school where only fewer cows but additional manpower are available.

## 6 National and international cooperation

'ManuMax' was a joint-project between Swiss, Swedish and Austrian partners granted by the Era-Net Bioenergy under the 11th Joint Call for Research and Development Proposals with the topic 'Bioenergy as part of a smart and flexible energy system'.

## 7 Communication

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Cazier, E:A. , Bühler, P., Brethauer, S., Studer, M. (2019) Steam explosion pretreatment improves the conversion efficiency of cattle manure to methane, **6th International Conference on Renewable Energy Gas Technology**

Cazier, E:A. , Brethauer, S., Studer, M. (2019). Optimization of methane production of cattle manure using steam explosion. **16th IWA World Conference on Anaerobic digestion**

Studer, M. et al. (2020). ManuMax – Development of a heat integrated steam explosion pretreatment process to enhance biogas yield in anaerobic digestion of manure → phase I. **6<sup>th</sup> Central European Biomass Conference**, Graz, Austria

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## 9 Appendix

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