

Evaluation and techno-economic analysis of packed bed scrubber for ammonia recovery from drying fumes produced during the thermal drying of sewage sludge

Ali Saud^{1*}, Jouni Havukainen¹, Mika Mänttari² and Mika Horttanainen¹

¹School of Energy Systems, LUT University, Lappeenranta, Finland

²School of Engineering Science, LUT University, Lappeenranta, Finland

Abstract. This study investigates the recovery of ammonia from drying fumes during thermal drying of sewage sludge with packed bed acid scrubbers to recover ammonia and to produce ammonium sulfate. The process is modelled for two concentrations, 75 and 100 ppm, and 1000m³/h inlet flowrate of drying fumes containing air and ammonia gas. It results in finding optimal parameters for scale-up of drying fumes during thermal drying of 7700t/a sewage sludge of Lappeenranta city. It is found that a single scrubber, with a 24000 m³/h of inlet gas and an ammonia concentration of 75 ppm, liquid to gas ratio of 1.5, temperature and pH of liquid acid as 100°C and 3 respectively, gives the efficiency of more than 99%, and reduces ammonia concentration in the outlet stream to 0.2 ppm. The capital cost is 290 k€, operating cost is 113k€/a, removal cost with and without revenue of ammonium sulfate is 20 €/t and 18 €/t of sludge. The packed bed acid scrubber would be suitable to remove ammonia in the drying fumes to recover ammonia from the drying fumes, but the initial economic analysis highlights that the production of commercial grade ammonium sulfate fertilizer would be an expensive option.

1. Introduction

The population of the world is increasing and is predicted to reach 8-10 billion in 2050. This increase in population results in a high demand for food production [1]. Given this, the demand for fertilizers is also increasing by 4% annually to support food needs for an additional 2.3 billion people in 2050. Thus, the production of sustainable fertilizers is the need of the hour [2].

Nitrogen and phosphorus are the two unique elements of agriculture. Annually, 120 million tons of atmospheric nitrogen is converted into reactive nitrogen for the production of fertilizers by the Haber Bosch process, but the high temperature and pressure requirements for reactions consume a large amount of energy: it has been estimated that 1kg of liquid ammonia requires 42MJ of energy, and emits 1.9 kg of CO₂ [3]. Sludge is, however, a global growing waste problem and, at the same time, a potential source of recoverable nutrients. Therefore, in many countries, it is incinerated [4], which is an effective disposal method but easily destroys the nutrients. Sewage sludge contains nutrients such as phosphorus and nitrogen along with harmful substances. Due to this fact, sewage sludge utilization poses the risk of soil contamination directly or after treatment [5,6].

During the thermal drying of sewage sludge, a large amount of ammonia combusted to N₂ and NO_x which can be recovered. Previous research [7], proposed ammonia recovery from the drying fumes resulting from the thermal drying of mechanically dewatered sewage sludge. The ammonia exiting as fumes during thermal drying can be absorbed with sulfuric acid or nitric acid to produce ammonium sulfate and ammonium nitrate, which are commercially used fertilizers.

Wet acid scrubbers have shown remarkably efficient ammonia recovery, between 91 and 99%, from exhaust air in animal facilities [8]. The application of ammonia scrubbing has been studied in different animal facilities, including poultry and swine farms, to neutralize gas emissions. Scrubbers have been installed in animal facilities in which the ammonia in exhaust air reacts with dilute sulfuric acid to produce ammonium sulfate [9]. The design of scrubbers have been widely studied and optimized in many studies, details of which are given in [8,10,11]. On the other hand, the absorption of ammonia to produce a commercial product such as ammonium sulfate fertilizer from the drying fumes produced during the thermal drying of sewage sludge has been little studied.

This study is part of the ongoing effort to find ways to recover nitrogen from mechanically dewatered sewage sludge during its thermal drying phase. The aim of this

*Corresponding author: ali.saud@lut.fi

study is evaluating the possibilities and feasibility of acid scrubbing for the recovery of ammonia from sludge drying fumes for fertilizing purposes by changing different parameters such as pH, liquid flow rate and temperature and by conducting initial economic analysis.

2. Methods

2.1 Process Description

A process flow diagram of scrubbing system with inlet and outlet streams is shown in Fig. 1. The air containing ammonia enters from the bottom of the scrubber at atmospheric pressure as stream GASIN, and sulfuric acid enters as stream H2SO4 and water enters as stream LIQIN from the top of scrubber. From the top of the scrubber, the EXHAUST stream vents out air to ambient environment after treatment, and the LIQOUT stream, comprised of water and ammonium sulfate, leaves the scrubber from the bottom. The LIQOUT stream further goes into splitter where 50% of liquid goes in RECYCLE stream to circulate again with the diluted acid and remaining goes to PRODUCT stream. A MIXER combines streams before entering scrubber to stream LIQMAI and liquid ammonium sulfate comes out in stream AMMLIQ. Equilibrium reactions are considered, and the required equilibrium data is taken from the literature and the ASPEN databank. Acid scrubbing of ammonia gas is modelled in the ASPEN Plus simulator. ASPEN Plus also supports estimations of process behaviour by applying engineering knowledge. The Radfrac column with rate-based calculation method is selected as a suitable option to design scrubber for vapor liquid streams, equilibrium and rate based reactions. [11]. The electrolyte NRTL (ENRTL) method is selected for thermophysical property analysis to determine the thermodynamic properties of the liquid phase, including electrolytes [12-15].

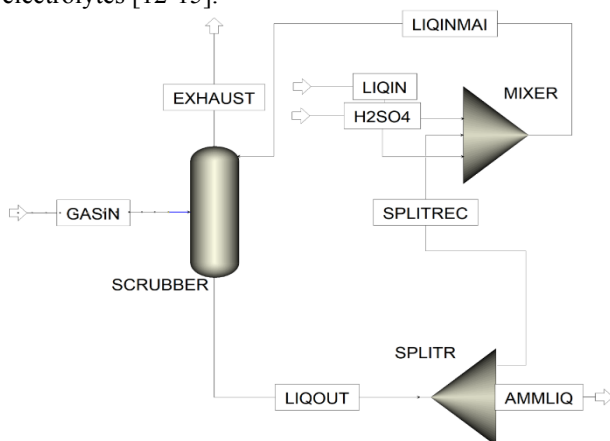


Fig. 1. A simplified representation of a scrubber in ASPEN Plus V9.

2.2 Reactions

In the modeling of scrubber, the reactions shown in Table 1 are considered. For the calculation of

equilibrium constant K for reactions, the following rate equation was used for calculation [17]:

$$\ln K = A + \frac{B}{T} + C \ln T + DT \quad (1)$$

Table 1. Equilibrium reactions included in the model.

Reaction	Type of Reaction	Chemical Equation
R1	Equilibrium	$H_2O + HSO_4^- \leftrightarrow H_3O^+ + SO_4^{2-}$
R2	Equilibrium	$H_2SO_4 + H_2O \leftrightarrow H_3O^+ + HSO_4^-$
R3	Equilibrium	$NH_3 + H_2O \leftrightarrow OH^- + NH_4^+$
R4	Equilibrium	$2H_2O \leftrightarrow OH^- + H_3O^+$
R5	Salt	$(NH_4)_2SO_4 \leftrightarrow NH_4^+ + SO_4^{2-}$

where, T is the absolute temperature and A, B, C, and D are equilibrium parameters. Their values are available in the ASPEN databank, as shown in Table 2.

Table 2. Reaction parameters for the equilibrium constant K.

	A	B	C	D
R1	-5.393	1.73×10^3	0	0
R2	-3.898	3.47×10^3	0	0
R3	-1.257	-3335	1.497	-0.037
R4	132.9	-1.34×10^4	-22.477	0
R5	-216.6	4.26×10^3	37.518	-0.0799

2.3 Data for modelling

Two studies were selected as a reference for the preliminary design of the scrubber Melse and Ognik et al. [8] and Khakharia et al. [16]: The first one was the work of Melse and Ognik et al. [8], in which acid packed bed scrubbers were developed for the removal of ammonia (NH₃) from pig and poultry facilities. It was concluded in the study that NH₃ removal was in the range of 40% to 100% with an average value of 96% and the second study was work of Khakharia et al. [16] which describes an acid scrubber to treat ammonia emissions from a post-combustion CO₂ capture plant and in this study, the ammonia inlet concentration of 150 mg/m³, and the ammonia outlet concentration decreased to 5 mg/m³. These two studies are used to estimate required parameters as listed in Table 3, to evaluate the model and configuration, cost estimation, and scale-up of ammonia scrubber for the drying fumes produced by sewage sludge. The selected data for validation is mentioned in Table 3.

Table 3. Selected process parameters for modelling.

Parameter	Values
Flow Rate of Gas	1000 m ³ /h
Inlet concentration of Ammonia	75 and 100 ppm
H ₂ SO ₄ pH	1,2,3,4,5
Liquid to Gas Ratio (L/m ³)	4.3
Liquid Flow Rate	4025 kg/h
Inlet Temperature	100 °C
Pressure	1 atm
Superficial Velocity	1.4 m/s

The proposed scrubber is validated with the data from Khakharia [16] and Melse [8]. In first validation, liquid flow rate and inlet concentration of ammonia is varied to analyze the outlet concentration of ammonia. pH, gas flow rate and the temperature is kept constant while in second validation, pH is varied to analyze the ammonia removal efficiency of scrubber. Liquid flow rate, gas flow rate and temperature are kept constant. The selected packing material is Mellapakplus 252Y packing and liquid to gas (L/G, L/m³) ratio is 4.3 [8,16].

2.4 Scale-up and Cost Estimation

Scale up is needed to investigate the initial feasibility of acid scrubbing for ammonia removal. The scale up includes the calculation of scrubber dimensions. The height of the column, packing height, and diameter of the scrubber were calculated as follows:

The height of the scrubber and packing is calculated with Eq. 2 and Eq. 3 [10]:

$$H_{column} = 1.4H_{pack} + 1.02D + 2.81 \quad (2)$$

where, H_{column} is the height of the scrubber, H_{pack} is the height of the packing, and D is the diameter of the scrubber.

$$H_{pack} = N \times HETP \quad (3)$$

where, N is the number of equilibrium stages, and $HETP$ is the height equivalent to theoretical plate. The superficial velocity, u_s is estimated as 1.4 m/s [8], and diameter, D_c was calculated from:

$$D_c = \frac{\sqrt{4A}}{\pi} \quad (4)$$

where, A is the area of the scrubber calculated from Eq. 5, and the value of π was taken as 3.14. The area is calculated from Eq. 5:

$$A = \frac{Q}{u_s \times 3600} \quad (5)$$

where, Q is the flow rate of gas in m³/h.

2.5 Cost Estimation

In cost estimation, the method cost curves and equations for preliminary estimation is considered due to unavailability of empirical data. The method of cost calculation for equipment cost, and different percentages of variations have been taken from literature [18,19]. Eq. 6 is used to calculate the mass of the scrubber and cost of equipment purchased:

$$C_e = a + bS^n \quad (6)$$

where, C_e = Cost of equipment purchased, €
 a, b = cost constants and values, taken as 11600 and 34 respectively [18]
 S = size parameter (Shell mass, kg)
 n = exponent for equipment, value 0.85 [18]

The shell mass of the scrubber, S , is the size parameter to calculate purchase cost. It is calculated from Eq. 7:

$$S = \pi D_c H_c t_w \rho \quad (7)$$

where, D_c = vessel diameter, 2.4 m

H_c = vessel height, 12 m

t_w = wall thickness, 0.009 m

ρ = metal density, 8000 kg/m³

The values of wall thickness, t_w and metal density, ρ are taken from literature [18].

Other investment includes costs for installation, instrumentation, piping, electrics, and costs related to engineering, construction, and services, are also calculated from factors given in literature [18] and multiplying with cost of purchased equipment, C_e . The operational cost includes the cost of sulfuric acid and water, these being 0.4 €/kg and 1 €/m³ respectively.

3. Results and Discussion

a. Model Validation

Model validation is required to verify the designed model consistency and to analyze behaviour of system. The process of model validation in this study is based on the data from literature.

In Fig.2(a), the designed model behaved in the similar way as in the referenced model. In the work of Khakharia [16] and in this current study, the inlet concentration of ammonia was varied from 152-155 mg/m³ (218-223 ppm) and outlet concentration was measured below 2 mg/m³ (3 ppm).

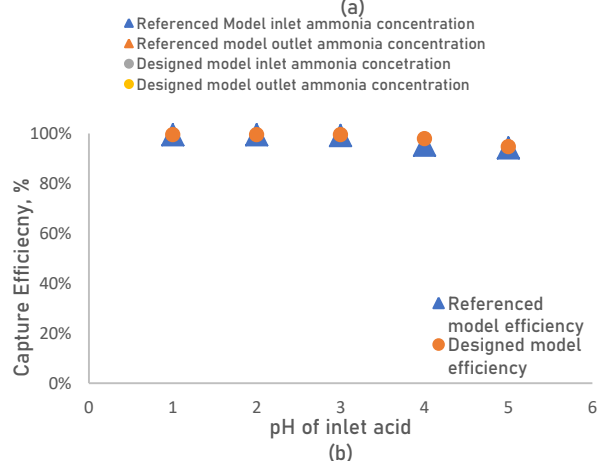
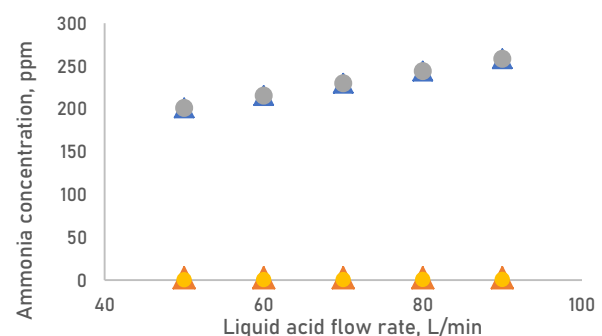


Fig. 2. a) Validation off referenced and modelled scrubber with changing liquid flow rate and ammonia concentration **b)** effect of changing pH on efficiency of scrubbers.

In the second case of validation, the efficiency of both referenced and modelled scrubber were above 99%. In Fig.2(b), it can be seen clearly that with the lower pH, the efficiency of both scrubbers is above than 99% and as pH increases, the efficiency started to decrease due to lower concentration of acid.

b. Effect of pH

pH is very substantial in the absorption of ammonia, and according to literature should be kept in the range of 1-5 for acid scrubbers [8,16,20]. The lower water flow rate assists in ammonia capture. Hence, the pH of acid is varied from 1-5 to analyze changes in the capture of ammonia and the efficiency of the scrubber. The high solubility of ammonia in water and establishing reaction equilibrium between gas and liquid phase in absorption led to the use of sulfuric acid to keep the ammonia in an ionized form [16].

The water flow rate, temperature, and gas flow rate were kept constant at 70 L/min, 100°C, and 1000 m³/h, and only the pH of acid varied from 1 to 5 to estimate the effect of pH. Two inlet concentrations of NH₃ in the inlet of scrubber, 75 ppm and 100 ppm, were studied. Fig. 3 shows that the pH had no effect on efficiency of scrubber from pH 1-3, but that efficiency started reducing at pH 4 and pH 5. The ammonia gas outlet concentration was declined to 0.2 ppm and 0.3 ppm at pH 1 to 3 for both inlet concentration of 75 and 100 ppm respectively.

On the other hand, at pH 4 and 5, the ammonia concentration reduced to 0.9 ppm and 8.6 ppm from 75 ppm respectively, while on 100 ppm it was reduced to 2.3 and 12.5 ppm respectively as shown in Fig. 3. The observed efficiencies are higher than 99% but for pH 5, this reduced to 88%. The same trend was observed in the study of Khakharia et al.[16] and Melse and Ognik [8] have also suggested a pH range from 1-4.

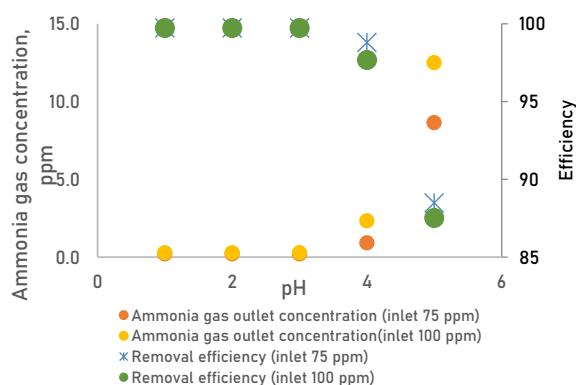


Fig. 3. Outlet gas ammonia concentration and removal efficiency with varying pH of inlet sulfuric acid.

c. Effect of Flowrate

The appropriate L/G ratio is always required for the efficient flow of liquid through selected packing. The L/G ratio promotes mass transfer between gas and liquid phases [8,21].

L/G ratio is the reason that the flowrate of liquid is varied between 70 and 150 L/min (L/G ratio of 4.3-8) to evaluate the effect of flowrate on ammonia removal and scrubber efficiencies. The gas flow rate is fixed at 1000m³/h, pH of acid at 3, temperature at 100°C, and only the liquid flow rate was varied in the simulation.

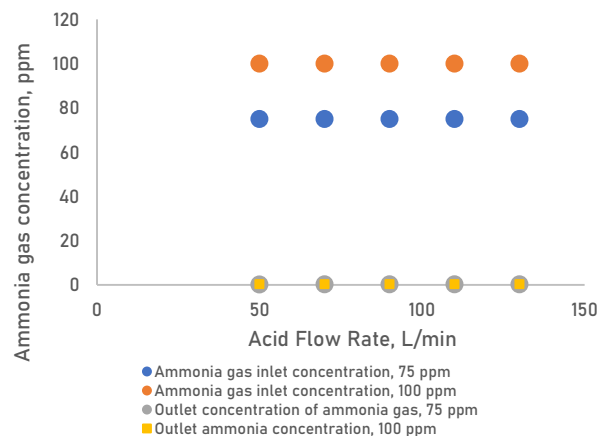


Fig. 4. Effect of inlet acid flow rate on the outlet concentration of ammonia for both 75 and 100 ppm ammonia concentration

In Fig. 4, the variation in flow rate has had little effect on the removal of ammonia because, at the lowest flow rate, 70 L/min, the efficiency of ammonia removal is greater than 99%. It is also noticeable that if the concentration of ammonia is increased, the liquid flow is sufficient to remove the additional ammonia to the desired level.

d. Presence of ammonium sulfate

Ammonium sulfate is present in a very diluted form in the liquid outstream of the scrubber. The presence of ammonium ions (NH₄⁺) and sulfate ions (SO₄²⁻) in the liquid outlet stream confirms the existence of ammonium sulfate. The production (or concentration) of ammonium ions varies with the concentration of inlet ammonia gas, and the concentration of sulfate ions varies with the pH of the liquid. Higher the inlet concentration of ammonia gas, the more ammonium ions will be produced. In Figs. 5(a) and 5(b), it shows that the selection of pH 3 is very favorable for a higher amount of ammonium ions as the mass flow rate of NH₄⁺ ions is about 0.06 kg/h, whereas SO₄²⁻ ions is 0.43 kg/h respectively. In Fig. 5(b), the dissociation of acid decreases with increasing pH, resulting in a decreased amount of SO₄²⁻. Similarly, NH₄⁺ also shows a low dissociation behaviour at pH 4 and 5.

e. Scale-Up and Cost Estimation

The scale-up of acid scrubber is done on the sewage sludge production rate of Toikansuo wastewater treatment plant, situated in the small Finnish town of Lappeenranta, treats wastewater with a capacity of 16000

m³/day for 72,000 habitants. The total sludge 7700 t/a with 20% total solids and a total nitrogen content of 5.3% of TS, and, on average 12% of the total nitrogen ended up in drying fumes as ammonia [7]. It summarizes that one scrubber with a flow rate of 24000 m³/h and 8610 kg-Nitrogen/a of ammonia with concentration of 75 ppm will be treated.

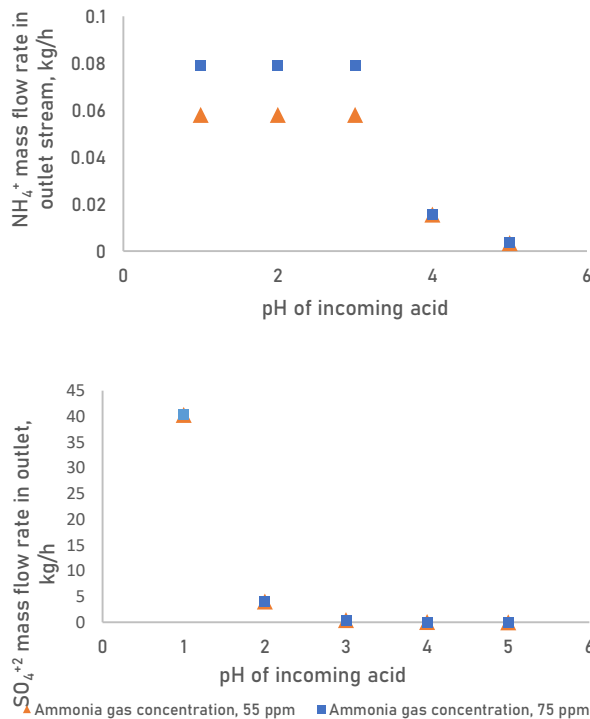


Fig. 5. Mass flow rate at different pH levels of (a) NH₄⁺; and (b) SO₄⁻² ions in liquid ammonium sulfate outlet stream.

Table 4 summarizes the cost of a scrubber, packing, and selected material whereas Table 5 summarizes process specifications, the number of scrubbers required, and the dimensions of a scrubber.

Table 4. Dimensions of scrubber.

Specifications	Values
Diameter of scrubber	2.4 m
Height of scrubber	12 m
Packing height	6 m
Required number of scrubbers	1

Table 5. Cost calculated for scrubber.

Equipment	Material	Cost (k€)
Scrubber	Stainless steel	78
Packing (Sulzer Mellapakplus 252Y)	SS 304L	21
Erection and commissioning cost		165
Start-up cost		26
Total		290

The total capital cost for scrubber with the capacity of handling 24000 m³/h of gas and 575 L/min of liquid is

290 k€. In operational cost, sulfuric acid and water are the main contributor with cost of 14 k€/a and 99 k€/a. We assume, that the scrubber can be operated by the personnel of the sludge treatment plant and so it does not increase the personnel costs.

In Fig.6, a graph between liquid to gas ratio (L/G) and recycling rate is made to predict the best economical cost of water. Firstly, liquid to gas ratio is changed from 1.5 to 4.3 and recycling rate kept constant at 50% and then L/G ratio kept constant at 1.5 and recycling rate was varied from 50-70 %.

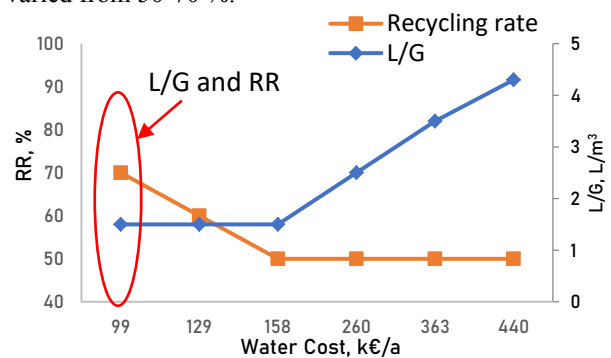


Fig. 6. Relation of water cost to recycling rate (RR) and liquid-to-gas ratio (L/G).

When recycling rate is varied above 70%, simulation started giving errors due to flooding limit of 80%. The encircled values show that the 70% recycling rate and 1.5 liquid to gas ratio gives optimal results with lower cost of water. It helps in calculation of operational cost of sulfuric acid and water which is 14 k€/a and 99 k€/a respectively with total of 113 k€/a.

Table 6 gives further details of the per-ton cost of treated sludge and possible income generated from recovered nitrogen by using it in fertilizer production. The value of mineral fertilizers in terms of nitrogen is 1.6 €/kg-Nitrogen [22]. The annual nitrogen production from recovered ammonia (based on 99% recovery) is calculated as 8610 kg-Nitrogen/a. The expected production of ammonium sulfate is 9 t/a and the cost of ammonia recovery per ton of sewage sludge treated would be 18 €/ton. The total annual cost is calculated as 155 k€ taking into account an interest rate of 10% and a 10-year lifetime for scrubbers.

Table 6. Annual cost and expected income generation

Recovery	Cost
Income from recovered nitrogen	14 k€/a
Total annual cost	155 k€/a
Additional cost for sludge treatment	20 €/t of sludge
Cost of ammonia recovery	18 €/t of ammonium sulfate
Cost of ammonia recovery	18 €/kg of nitrogen

4. Conclusions

A preliminary study of acid scrubber to recover ammonia from the drying fumes produced during the thermal

drying of sludge was conducted including modelling and cost estimation. The ASPEN Plus simulations were conducted for ammonia concentrations of 75 and 100 ppm. It was observed that low pH values between 1-3 are suitable for ammonia recovery, but with higher pH such as 4-5, efficiency would decrease from 99% to 88%. The L/G ratio was changed by changing the liquid acid flow rate and value of 1.5 with recycling rate of 70% was found to be a feasible L/G ratio.

The total capital investment of a scrubber for a sludge treatment plant with capacity of 7 700 ton/a, was calculated as 290 k€ with an operational cost of 113 k€/a, including the cost of sulfuric acid and water. The cost of ammonia removal is 20 €/t of sludge and the cost of ammonium sulfate produced is 18 €/t of sludge which is practical treatment cost, but product is very diluted as compared to commercial grade and cost is high for optional source of ammonia fertilizer production and treatment.

The result is disproving the previous proposals that this kind of scrubbing could be feasible not only for ammonia emission reduction but also for production of ammonia fertilizer. If the costs will be covered by sludge treatment costs, it can be possible to utilize the end product in some cases. But the additional costs for refining the product to fertilizer seem to be too high to compete with other nitrogen fertilizers and more cost-effective methods are needed. Further research by the authors will focus on finding more suitable and feasible methods to recover ammonia from the fumes produced by the thermal drying of sewage sludge.

References

1. J. van der Hoek, R. Duijff, and O. Reijnders, *Sustainability* **10**, 4605 (2018)
2. M. Xie, H. K. Shon, S. R. Gray, and M. Elimelech, *Water Res.* **89**, 210 (2016)
3. L. F. Razon, *Am. Inst. Chem. Eng.* **33**, (2014)
4. Y. Cao and A. Pawłowski, *Renew. Sustain. Energy Rev.* **16**, 1657 (2012)
5. D. Fytili and A. Zabaniotou, *Renew. Sustain. Energy Rev.* **12**, 116 (2008)
6. A. M. Mahon, B. O. Connell, M. G. Healy, I. O. Connor, R. O. R. Nash, and L. Morrison, (2017)
7. M. Horttanainen, I. Deviatkin, and J. Havukainen, *J. Clean. Prod.* **142**, 1819 (2017)
8. R. W. Melse and N. W. M. Ogink, *Trans. ASAE* **48**, 2303 (2005)
9. L. J. Hadlocon and L. Zhao, *Agric. Eng. Int. CIGR J.* **2015**, 41 (2015)
10. P. R. Bhoi, R. L. Huhnke, A. Kumar, K. N. Patil, and J. R. Whiteley, *Fuel Process. Technol.* **134**, 243 (2015)
11. C. Van der Heyden, P. Demeyer, and E. I. P. Volcke, *Biosyst. Eng.* **134**, 74 (2015)
12. J. Liu and D. S. Chen, *Computer-Aided Design for Energy Saving in an Ammonia-Based Post-Combustion CO₂ Capture Process* (Elsevier Masson SAS, 2018)
13. Y. Tang, Y. Gao, D. Liu, F. Zhang, S. Qu, Z. Hao, X. Zhang, and Z. T. Liu, *RSC Adv.* **7**, 23591 (2017)
14. D. Bravo, F. J. Álvarez-Hornos, J. M. Peña-roja, P. San-Valero, and C. Gabaldón, *J. Environ. Manage.* **213**, 530 (2018)
15. D. Bolzonella, F. Fatone, M. Gottardo, and N. Frison, *J. Environ. Manage.* **216**, 111 (2018)
16. P. Khakharia, A. Huizinga, C. Jurado Lopez, C. Sanchez Sanchez, F. De Miguel Mercader, T. J. H. Vlugt, and E. Goetheer, *Ind. Eng. Chem. Res.* **53**, 13195 (2014)
17. Aspen Technology Inc., (2013)
18. G. Towler and R. Sinnott, *Chemical Engineering Design : Principles, Practice, and Economics of Plant and Process Design*, 2nd ed. (Butterworth-Heinemann, 2012)
19. M. S. Peters, K. D. Timmerhaus, and R. E. West, *Plant Design and Economics for Chemical Engineers*, 5th ed. (McGraw-Hill Education, 2003)
20. L. S. Hadlocon, L. Zhao, R. B. Manuzon, and I. E. Elbatawi, *Trans. ASABE* **57**, 949 (2014)
21. L. S. Hadlocon, R. Manuzon, and L. W. Zhao, (2014)
22. J. Havukainen, **13**, 1050 (2018)