REMOVALS OF ORGANIC AND NITROGEN IN BIOGAS EFFLUENT BY AAO SYSTEM

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ARTICLE INFO		ABSTRACT
Received:	14/4/2021	Livestock wastewater effluents containing high content of organic and
Revised:	06/5/2021	nutrients are discharged to the receiving water environment. Although wastewater is treated by biogas digester, the effluent concentrations still
Published:	20/5/2021	remain high in COD, ammonium (NH_4^+) and phosphate (PO_4^{-3-}) . This
		research used a three-chambers system anaerobic-anoxic-aerobic
KEYWORDS		(AAO) for simultaneous removal of organic, nitrogen pollutants via
		continuous experiments. The biogas effluents with COD (2000-5000
Livestock wastewater		mg/L), NH ₄ ⁺ (155-188 mg/L), PO_4^{3-} (13.00-21.95 mg/L) were used as
Wastewater treatment		influences for the continuous experiments. The results show that
Organic pollutants		removal efficiency of COD and NH_4^+ were achieved at 82% and 70%,
Nutriant removal		respectively. The concentration of COD was obtained at the safe level
Nutrient removal		regulated by the QCVN 62-MT:2016/BTNMT. To optimize the AAO
AAO		system for higher input concentrations of pollutants, a further study on operation conditions such as hydraulic retention time and oxygen supply should be conducted.

XỬ LÝ CHẤT HỮU CƠ VÀ NITƠ TRONG NƯỚC THẢI SAU BIOGAS BẰNG CÔNG NGHỆ AAO

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I BÁO	ΤΟΜ ΤΑΤ
14/4/2021 06/5/2021 20/5/2021	Nước thải chăn nuôi có hàm lượng chất hữu cơ và dinh dưỡng cao được thải ra môi trường tiếp nhận. Mặc dù phần lớn nước thải từ chăn nuôi lợn được xử lý bằng hệ thống biogas nhưng nồng độ COD, amoni (NH_4^+) và phốt phát (PO ₄ ³⁻) vẫn rất cao. Nghiên cứu này sử dụng hệ
	thông ba ngăn kỵ khí-thiêu khí-hiêu khí (AAO) để loại bỏ đông thời các chất ô nhiễm hữu cơ, nitơ và phốt pho thông qua các thí nghiệm liên tục ở quy mô phòng thí nghiệm. Nước thải đầu ra hệ thống biogas
	có COD từ 2000-5000 mg/L, NH ₄ ⁺ từ 155-188 mg/L, và PO ₄ ³⁻ từ 13,0- 21 95 mg/L được sử dụng làm đầu vào của hệ thống xử lý. Kết quả cho
	thấy khả năng xử lý chất hữu cơ (COD) đạt 82%, amoni đạt 70%, nước sau xử lý dưới ngưỡng cho phép theo QCVN 62-MT:2016/BTNMT. Để tối ưu hóa hệ thống AAO xử lý nước thải có nồng độ chất ô nhiễm cao, cần có những nghiên cứu sâu hơn về điều kiện vận hành như thời gian lưu và lượng oxy cung cấp.
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1. Introduction

The livestock sector has shifted from household farming to large scale farming in recent years. Our country had 28.15 million pigs in 2018, with porkers accounting for 24.1 million of those. The Red River Delta is home to 7.1 million of pigs [1]. Emissions from livestock production are estimated to be 84.5 million tons of solid waste per year, with 50 million m³ of liquid waste. Only about 20% of livestock waste is reused effectively (biogas, composting, worm farming, fish feeding, etc.), while other 80% of waste is discharged into environment causing pollution [2], [3].

The development of biogas technology has helped to solve the problem of organic pollution in wastewater, as well as to reduce greenhouse gas emissions and investment costs. Data analysis of input and output wastewater samples from 9 biogas systems revealed that using biogas systems to treat pig wastewater significantly reduced pollutant concentrations. COD parameters decreased by 84.7%, BOD₅ decreased by 76.3%, TSS decreased by 86.1%, VSS parameters decreased by 85.4%, total nitrogen decreased by 11.8%, TP parameters decreased by 7.0%, and fecal coliform decreased by 51.2% [4]. Furthermore, the use of biogas technology generates methane gas, which can be used as a new environmentally friendly energy source. However, the concentration of pollutants in the effluent remains quite high, exceeding the Ministry of Natural Resources and Environment's regulated standards [5]. The content of organic matter, total nitrogen, and total phosphorus, in particular, remains extremely high, potentially causing eutrophication when discharged into bodies of water. Therefore, a more appropriate solution is required to effectively and thoroughly treat the polluted nutrients in wastewater produced by biogas.

Many types of wastewater are treated using AAO technology, including domestic wastewater, medical wastewater, and livestock wastewater [6]. AAO stands for anaerobic-anoxic-oxic. AAO technology employs a continuous biological treatment process that employs a wide range of microorganisms for wastewater treatment, including anaerobic, anaerobic, and anaerobic microbiota [7]. Currently, AAO or AO technologies are widely used in high-efficiency concentrated wastewater treatment systems in industrial zones; however, the process of operating A2O systems in wastewater treatment livestock has received little attention. The purpose of this research is to assess the ability to treat organic matter and nutrients in wastewater after biogas treatment.

2. Research method

2.1. Material

Wastewater for this experiment was collected from biogas effluent in a pig farms in Van Giang district, Hung Yen province during 2020. The farms are raising 80 pigs including 8 sows, 50 fatten pigs and 22 piglets. The average amount of water used for pigs is 10-20 liters/head. This amount of water varies depending on the season and method of bathing (summer, winter), but a standard of 10 liters/head/day is recommended. Before feeding into the system, wastewater was filtered by a clean cloth. The daily samples were regularly taken and analyzed to observe the influent concentration of the AAO system. The COD, amoni (NH₄⁺) and pH are in the range of 2000-5000 mg/L, 155-187 mg/L and 6.8-9.5, respectively. These values are higher than the National technical regulation on the effluent of livestock (QCVN 62-MT:2016/BTNMT) (Table 1).

Parameter	Unit	Concentration	QCVN 62
COD	mg/L	2000-5000	300
${ m NH_4}^+$	mg/L	155-187	150
pH	-	6.8-9.5	5.5-9

Table 1. Characteristics of wastewater used in the experiment

2.2. System design

AAO system was designed at the laboratory scale (Figure 1). It was made up of three consecutive anaerobic, anoxic, and oxic compartments with the following dimensions: $45 \times 25 \times 30$ cm (*length* × *width* × *height*). The working volume is 33 liters. The compartment structure includes: 1- anaerobic compartment with dimensions of $10 \times 25 \times 30$ cm (*length* × *width* × *height*), working capacity is 7 liters; 2- anoxic has a size of $15 \times 25 \times 30$ cm (*length* × *width* × *height*) with working capacity is 11 liters; 3- oxic has a size of $20 \times 25 \times 30$ cm (*length* × *width* × *height*), working capacity is 15 liters; 4- Settled tank $28 \times 25 \times 15$ cm (length × width × height). The continuous water flow was set at 11 L/h and 6.6 L/h to obtain hydraulic retention time (HRT) at 3 hours and 5 hours.



Figure 1. Experimental model of AAO technology

Note: 1- Anaerobic compartment; 2- Anoxic; 3- Oxic; 4- settled compartment; 5- Output point after processing; 6- sewage inlet.

To improve the efficiency of the adhesion process, Kaldnes PE05 filter particles were added to create a contact environment between water and microorganisms. The Kaldnes PE05 grain is 25x10 mm in size and has a working temperature of 5-45 0 C. The material is made of HDPE plastic and has a specific surface area of 650-750 m²/m³. Oxygen supplied for the aeration chamber (chamber 4) with a gas flow rate of 3.5 liters per minute. Emina (10^{8} CFU/mL) probiotic was added to the feeding wastewater before entering the system.

2.3. Operation and data collection

The AAO wastewater treatment system was set up in a continuous flow with different COD concentrations, an initial COD concentration of 2000 mg/L with a microorganism addition of 1.0% (v/v). Water inlet samples were collected at the start of system operation (C_0), water samples after treatment were collected at the outlet point after anaerobic treatment (chamber 2), and water samples after treatment were collected at the outlet point after anaerobic treatment (chamber 5). Sampling was taken every 2 days after the operation (days 1, 3, 5, 7... 23 after operation). Samples were taken in accordance with TCVN 6663-1:2011 (ISO 5667-1:2006) and TCVN 6663-3:2008 (ISO 5667-3:2003).

The pH was measured in accordance with TCVN 6492:2011 using the portable equipment. COD was analyzed according by titration (TCVN 6491:1999) using the oxidizing chemical

 $K_2Cr_2O_7$. Total nitrogen was analyzed based on TCVN 6638:2000. NH_4^+ analysis using UV/Vis. The COD concentrations were analyzed in 3 replicates.

3. Results and discussion

3.1. Removal of organic matter

Although pig breeding wastewater was treated by biogas system before discharged into the environment, the organic matter content remains high due to the large amount of water used to push of manure into the biogas system. Because of the short retention time, the COD treatment efficiency of biogas systems was low. The effluents from biogas at the targeted farm was observed around 2000 mg/L, which was much higher than the regulated value [5]. The wastewater from biogas effluents ranging from 1800-2000 mg/L. Figure 2 shows the COD treatment efficiency at different functions chambers. COD concentration decreased significantly anoxic - oxic chambers. Treatment efficiency in the anaerobic following anaerobic compartment reaches 60% after 4 days, anoxic prevention reaches 70%, and oxic compartment reaches 82% after 14 days of operation. It was clear that the ability to handle COD in the anaerobic compartment has significantly reduced the amount of organic matter. The results showed that treated water was lower than 300 mg/L which meet the regulation at column B, QCVN 62-MT:2016/BTNMT for discharge into environment. When the COD increased up to 5000 mg/L, the treatment efficiency was estimated to be 40-60% within 10 days. However, treatment efficiency increased to 77% during 23 days of operation. This result indicates a limitation of the current AAO system. Therefore, a larger capacity of AAO system should be evaluated.



Figure 2. Removal of COD at different compartments

Compartment 1: Anaerobic, Compartment 2: Anoxic, Compartment 3: Oxic

AAO technology is not a new concept in wastewater treatment but it attracts great attention from scientist for improvement and optimization for wastewater treatment. Donkin and Rossell [8] used the AAO activated sludge configuration for the treatment of a milk powder/butter wastewater with HRT of 7 days and a nominal sludge age of 20 days. COD removal efficiency remained excellent throughout the trial at over 90% removal while overall nitrogen removal remained unchanged at 66%. Recently, number of researches has been focused on the addition of catalysts for removal efficiency enhancement. Zulkifli et al. [9] tried the AAO coated with carbon nanotubes and gold nanoparticles and used as the supports for the catalytic. The Au-CNTs-AAO exhibited superior catalytic activity, with a reaction rate (k) of 0.678 min⁻¹. In this research, we proposed the coupling catalysts of plastic Kaldnes to enhance specific surface area within the aerobic chamber. The result shows high removal efficiency with short hydraulic retention time at 3 to 5 hours while most of the research obtained similar removal efficiency at 8 hours.

3.2. Nitrogen treatment efficiency

Nitrogen ammonia (NH₄⁺) is an important criterion in determining the pollution content of pig wastewater. High concentrations of ammonium released into the environment would cause eutrophication and be harmful to humans and aquatic organisms. Figure 3 shows that removal efficiency of NH₄⁺ was achieved in the range of 20-70%. Since the influence of NH₄⁺ was not stable against time course change, a normalized concentration was calculated for its concentration at measured time. The removal efficiency of NH₄⁺ increases gradually from anaerobic compartment to anaerobic compartment. Because there was no existing nitrate bacteria in anaerobic conditions, NH₄⁺ treatment was obtained lowest at anaerobic counter from 5.5-15.7%. As a result, NH₄⁺ treatment was not significantly occurred here.



Figure 3. Nitrogen treatment efficiency



Figure 4. Ammonium removal after 14 days

Removal of nitrogen normally takes places in two processes, nitrification and denitrification. The treatment efficiency of NH_4^+ was in the range from 4.8% to 26.2%. However, in order to remove nitrate, denitrification process takes precedence during the operation. *Nitrosomonas* and *Nitrobacter* bacteria grow and develop in the aerobic compartment, this results in treatment efficiency of NH_4^+ in this compartment ranges from 17.4% to 72%. Nitrosomonas and Nitrobacter were two bacteria added to the aerobic compartment to take oxygen and convert

 NH_4^+ to NO_3^- while growing. The treatment efficiency of NH_4^+ gradually increases from the anaerobic to the aerobic compartment (Figure 4). Zhang et al. [10] evaluates the enhancement of and phosphorus removal and sludge reduction in an nitrogen AAO-CFSSDR (Anaerobic/Anoxic/Oxic combined with CFS-Sludge disintegration reactor) process suggested that the removal efficiencies of TN, NH₄⁺-N and TP increased from 71.15, 79.23 and 85.52% to 85.05, 87.70 and 90.06%, respectively; and the sludge was reduced by 34.79%. This research shows an application of cheap plastic materials as a medium for microorganism so that the removal efficiency of nitrogen ammonium could enhance up to 70%. Therefore, further research on enhancing removal performance against life cycles of material should be conducted.

4. Conclusion

The study results show that the AAO system coupling with Kaldnes plastic material for treatment of pig wastewater achieved high removal efficiency. The removal of organic matter (COD) reaches higher than 82%, and results in the treated concentration below the threshold set by QCVN 62-MT: 2016/BTNMT. Removal of nitrogen (primarily ammonium- NH_4^+) accounts for 70%. It is recommended to continue studying the operating conditions and optimizing the system based on operating condition such as retention times and types of wastewater.

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