Strategies For Sustainable Wastewater Treatment Based On Energy Recovery And Emerging Compounds Control

Wu G.[1,*], He K.[2], Miao J.[1], Yin Q.[1], Zhao Y.[1]

[1] Key Laboratory of Microorganism Application and Risk Control (MARC) of Shenzhen, Graduate School at Shenzhen, Tsinghua University, Shenzhen 518055, Guangdong, China

[2] Department of Environmental Engineering, Graduate School of Engineering, Kyoto University, Nishikyo, Kyoto 615-8540, Japan

*corresponding author:

e-mail: wu.guangxue@sz.tsinghua.edu.cn

Abstract

Wastewater treatment contributes significantly to protect human health and ecological safety. With sustainable development of wastewater treatment, recovery of resources and energy from wastewater has been the focus for conventional contaminants, while removal and control of ecological toxicity is the focus for emerging compounds. Under anaerobic conditions, especially with enhancement of direct interspecies electron transfer, methanogenesis and/or nitrous oxide production could be achieved separately or simultaneously, and removal of some types of emerging compounds could be enhanced. During nitritation, both nitrogen and emerging compounds removal could be achieved simultaneously. Therefore, a new wastewater treatment concept based on recovery of energy and control of emerging compounds was proposed, including anaerobic treatment technology by incorporating directly electron transfer enhancement and nitritation-based nitrogen removal. The proposed strategy provided a new technology for advancing the sustainable development of wastewater treatment through recovery of energy and control of emerging compounds.

Keywords: Wastewater treatment; Energy recovery; Emerging compounds; Anaerobic treatment; Nitritation

1. Introduction

Wastewater treatment contributes significantly to the control of disease spreading, and the protection the human health and ecological safety. Therefore, it has been considered as one of the most important technologies in the previous century. Pollutants including organic carbon, nitrogen and phosphorus removal have been historically considered during development of wastewater treatment, corresponding the wastewater treatment stages could be divided into biological wastewater treatment, biological nutrient removal, enhanced biological nutrients removal and limit of technology (LOT). Currently, enhanced biological nutrients removal is the main focus in many countries, while in American and some other developed countries, consideration and practice of LOT have been carried out (Bott *et al*., 2012). Since 2014, the year of one hundred years of anniversary of activated sludge, experts from all over the world have begun to think about future development of wastewater treatment. The basic consensus has been brought forward that wastewater should be considered as a resource rather than waste. Recovery of energy and resources for conventional contaminants in wastewater, such as organic matters, nitrogen, phosphorus and so on, should be focused. In addition, ecological risk and toxicity control of emerging compounds should be the near future focus.

2. Recovery of energy/resources from and control of emerging contaminants in wastewater

With the sustainable development of wastewater treatment, concepts are gradually changing from conventional pollutant removal to the recovery of energy and resources from wastewater. Various concepts for the recovery of energy and resources from wastewater have been proposed (i.e, McCarty *et al*., 2011; Sheik *et al*., 2014). For energy recovery, the main approach includes converting organic matters in wastewater directly into energy through anaerobic treatment. Otherwise, the other pathway for energy recovery is to convert liquid organic carbon to the solid organic matter firstly and then recovery energy by anaerobic digestion of sludge. For resource recovery, nitrogen and phosphorus are the core focus, such as recycled as fertilizer through struvite precipitation and so on. Among all the proposed recovery of energy and resources from wastewater, one typical example is the wastewater treatment Roadmap 2030 proposed by STOWA (Acronym of Foundation for Applied Water Research). The Roadmap included "water factory", "energy factory" and "nutrient factory", and the corresponding feasible technologies were also proposed for each "factory". McCarty *et al*. (2011) analyzed the feasibility of anaerobic wastewater treatment process for generating energy, and considered anaerobic treatment for energy generation is a core technology. Sheik *et al*. (2014) proposed the concept of "wastewater biorefinery column" for wastewater treatment, which achieved the recovery of energy and resources based on biological regulation of the activated sludge process. Emerging compounds and their toxicity are key future issues during wastewater treatment, especially for the protection of environmental and ecological health. Systematic researches on the effect and mechanism of emerging compounds removal such as pharmaceutical and personal care products (PPCPs) in wastewater treatment (i.e., Review from Luo *et al*., 2014) have been carried out. Carballa *et al*. (2004) examined the removal of PPCPs and endocrine disrupting chemicals (EDCs), and found that biodegradation of activated sludge contributed a lot for PPCPs and EDCs removal. As biological removal of emerging compounds is complex, the biodegradation pathway and ecological toxicity of emerging compounds and their byproducts have been gradually received intensive attention (Evgenidou *et al*., 2015). Therefore, in future, effective control technology for emerging compounds should be developed. Sustainable development concept based on economy, society and environment has been gradually introduced to wastewater treatment. Griggs *et al*. (2013) proposed a new concept of sustainable development, including the sustainable development of core economy, mesosphere of society and outer loop of ecology, and especially, sustainable water safety is a significant component. Therefore, maintaining the healthy and safe development of ecosystem become the final target from the perspective of sustainable development. As shown in Fig. 1 (Li *et al*., 2016), a new concept for sustainable wastewater treatment so as to achieve an ecosystem based wastewater treatment is proposed. The new concept includes three levels of energy, material and ecosystem. The internal support is energy, which includes the electricity for systematic operation and so on. The mesosphere is technology which is mainly considered from material flow such as wastewater, nitrogen and phosphorus. The outer sphere is ecology and the target of wastewater treatment is to maintain the sustainable development of ecosystem. Ecosystem not only includes water ecosystem, but also air ecology and the existing environment for humans, animals and plants. Thus wastewater treatment process includes not only nutrients removal, but also ecosystem sustainable development. Therefore, for conventional pollutants management, energy and resources recovery should be achieved in future wastewater treatment. Meanwhile, the effective control technology for emerging compounds should be developed to promote ecological safety and sustainable development of wastewater treatment. With these new concepts, strategies for new wastewater treatment should be brought forward to tackle challenges we are facing with.

3. Anaerobic energy recovery and emerging compounds control through enhancing electron transfer

Due to the low organic concentration in municipal wastewater, there are some key problems when conventional anaerobic treatment is applied for energy recovery directly: (1) a low methane recovery efficiency, especially due to the high solubility of methane, and (2) a low methane production rate due to the low organic carbon concentration. When direct anaerobic treatment of municipal wastewater is applied, enrichment of methanogen and/or development of high-rate anaerobic reactors are bottlenecks, especially for the low-temperature anaerobic treatment technology (Chang, 2014). To date, anaerobic treatment technology based on anaerobic membrane bioreactor is intensively developed and investigated. For the anaerobic membrane bioreactor, to reduce the operating cost, mechanism of membrane fouling and its control strategy are still key research aspects (Martin-Garcia *et al*., 2011; Skouteris *et al*., 2012). For anaerobic treatment of wastewater, efficient recovery of methane and maintaining a low effluent concentration of methane are important to avoid the release of greenhouse gases, which may probably cause secondary pollution (Bandara *et al*., 2011). Anaerobic treatment includes several complicate stages, such as hydrolysis, acidification and methane production, which requires different types of microorganisms to participate together. Therefore, facilitating the syntrophism among these microorganisms is capable of improving anaerobic treatment performance. Recently, some species like Geobacter were found to be able to transfer electron directly to methanogens via their functional organs such as pili. This electron transfer mechanism is especially common in granular sludge, which can enhance the efficiency of anaerobic treatment (Rotaru *et al*., 2014). For some methanogens such as Methanosaeta and Methanosarcina, direct interspecies electron transfer (DIET) has been proved to be another syntrophic methane production mechanism, except interspecies electron transfer through H_2 and formate (Rotaru *et al*., 2014). Dosing conductive materials such as activated carbon, conductive magnetite and activated carbon fiber, accelerated DIET and therefore improved the efficiency of anaerobic processes, including anaerobic treatment of wastewater and anaerobic digestion of sludge (i.e., Liu *et al*., 2012). Conductive materials mainly shortened the lag phase of methane production and increased the methane production rate (Liu *et al*., 2012). Microbial and metabolic mechanism of anaerobic treatment still requires further investigation, and clarification of the mechanism of DIET will be valuable for reactor optimization and development of new technologies for energy recovery from wastewater. The structure of emerging compounds is complex and the metabolic pathways of these pollutants are also diverse. According to degradation conditions, aerobic degradation and anaerobic degradation are commonly applied. Degradation mechanisms of emerging compounds during wastewater treatment mainly consisted of biological adsorption and biodegradation in activated sludge systems, especially aerobic biodegradation. Although anaerobic biodegradation attracts less attention, some emerging compounds which cannot be degraded by aerobic processes, can be degraded easily under anaerobic conditions, such as sulfamethoxazole (Fig. 1). Meanwhile, dosing redox mediator or conductive materials further improved the anaerobic treatment efficiency of emerging compounds (Fig. 1). Toxicity of emerging compounds is mainly related to their concentration. In addition, their biodegradation products are usually so complicated that their ecological effects cannot be ignored. Recently, Völker *et al*. (2016) found that by prolonging nutrient-limited anaerobic conditions, or with the anaerobic pretreatment of iron reduction, removal of endocrine and dioxin-like activities could be increased by 40-75%, and therefore improving the water ecological safety. Thus, anaerobic treatment of emerging compounds and their ecological toxicity worth further investigation.

4. Nitritation-based technologies for energy recovery and emerging compounds control

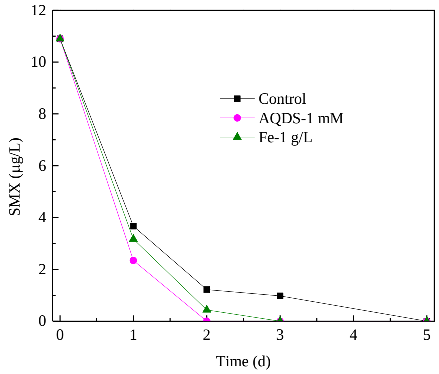

Figure 1. Example of effect of dosage of anthraquinone-2,6-disulfonate (AQDS) or ferroferic oxide (Fe) on anaerobic degradation of sulfamethoxazole (SMX)

Effective management of nitrogen is very important for energy and resource recovery from wastewater. To date, nitrite-based partial nitrification/denitrification and shortcut nitrification/anammox technologies are being extensively investigated. The key point is to achieve nitrite accumulation or couple it with other nitrogen removal processes. Nitrification comprises of ammonia oxidation and nitrite oxidation steps, with ammonia-oxidizing bacteria (AOB) being the functional bacteria in the first step, and nitrite-oxidizing bacteria (NOB) in the second step. Advantages of nitrite-based processes are as follows: (1) reduction in energy consumption; (2) reduction of electron donor demand; (3) low sludge production; and (4) high reaction rate and efficiency. Since AOB and NOB are of different physiological properties, nitrite accumulation can be effectively achieved by enhancing the activity of AOB while inhibiting NOB. Low dissolved oxygen concentrations, proper pH and multiple anoxic/aerobic alternative operation mode are efficient methods to achieve nitrite accumulation (Suthersan and Ganczarczyk, 1986; Yoo *et al*, 1999; Kornaros *et al*., 2010). The ammonia monooxygenase (AMO) gene in AOB makes it possible to simultaneously co-metabolize certain types of emerging compounds. For example, the biological removal of 17α-ethinylestradiol and bisphenol A, which belong to the EDCs category, is related to nitrification, specifically to the AMO activity, which was verified by testing the AMO enzyme activity (Yi and Harpper, 2007; Sun *et al*., 2012). Fernandez-Fontaina *et al*. (2012) also concluded that under high ammonia nitrogen load rates, emerging compounds could be efficiently removed via biological processes, with the AMO-based cometabolism being the possibly dominating metabolic pathway. Batt *et al*. (2006) found that the removal efficiency of iopromide in nitrifying activated sludge was 61%, while almost no removal was discovered in common activated sludge, and for trimethoprim, the removal efficiency in nitrifying activated sludge was 50%, while it was only 1% in common activated sludge. Biodegradation of triclosan, bisphenol A and ibuprofen by *Nitrosomonas europaea* was investigated by Rho *et al*. (2009), they found that triclosan and bisphenol A could be degraded by N. *Europaea*, while ibuprofen could not. In the process of biodegradation of triclosan and bisphenol A mixture, activity of N. *Europaea* would be inhibited by the degradation products or the toxicity of triclosan (Rho *et al*., 2009). Tran *et al*. (2009) studied removal of several PPCPs by enriched nitrifiers, and found that the removal efficiency of clofibric acid, diclofenac, carbamazepine, and propyphenazone all increased with increasing ammonium nitrogen concentrations. In addition, the existence of heterotrophs can promote certain PPCPs removal (Tran *et al*., 2009). In particular, acclimation of heterotrophs that can degrade emerging compounds through cometabolism or directly deserves further investigation, especially under oligotrophic nitrifying conditions. Under anoxic conditions, denitrifiers can successively reduce nitrate into nitrite, nitrogen monoxide and nitrous oxide (N_2O), with dinitrogen being the ultimate product. Therefore, N_2O is an obligatory intermediate during denitrification, which can be generated when N_2O reductase is inhibited. N_2O can be used as burning agent or rocket oxidizer. In particular, the energy generation rate can be promoted during the co-combustion of methane and N_2O mixture. Therefore, N_2O production by denitrification is an effective way for energy generation. Separated dosage of organic carbon and nitrite during denitrification could achieve effective nitrogen removal as well as a high N_2O generation. Carbon source is added firstly under anaerobic condition so as to be accumulated as internal carbon source, followed by the addition of nitrite, leading to the generation of N_2O during denitrification. However, the reaction rate of this process is relatively low, which usually takes more than 20 hours (Scherson *et al*., 2013). Otherwise, a high N_2O production rate can be achieved by specific denitrifiers acclimated with protein or carbohydrate, with nitrate or nitrite being the electron acceptor (Fig. 2). Therefore, coupling denitrification for N_2O production is not only a technology to effectively control nitrogen, but an ideal way to achieve energy generation during wastewater treatment.

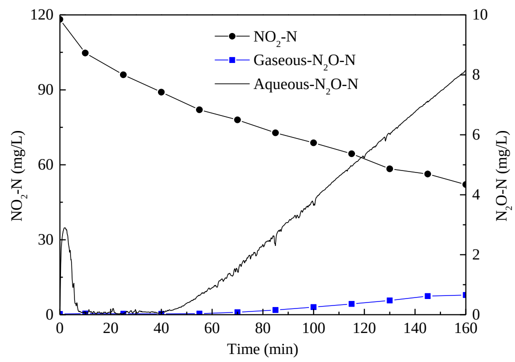

Figure 2. Example of nitrous oxide (N_2O) production in a typical denitrifying SBR reaction cycle with starch as the organic carbon and nitrite as the electron acceptor.

5. Strategy for energy recovery and emerging compounds control during wastewater treatment

Based on the research results above, a new wastewater treatment concept coupling energy recovery and emerging compound control is proposed (Fig. 3). This new technology mainly includes anaerobic treatment and high-efficient nitrogen removal. The two processes are carrying out under the optimal condition separately, which would improve the system performance. Anaerobic treatment may include methanogenesis, denitrification for N_2O production and enhanced emerging compounds removal. Under anaerobic conditions, the methanogenesis process can be enhanced by dosing conductive materials. If necessary, effluent from the nitritation reactor could be recycled to specific places of the anaerobic reactor to enhance N_2O production. Therefore, the efficiency of recovery and utilization of energy could be enhanced by stripping the produced methane and N_2O simultaneously. Meanwhile, this advanced anaerobic technology can be applied to remove specific emerging compounds and their toxicity efficiently, thereby achieving effective anaerobic energy recovery and emerging compounds control. In addition, if the organic matters were concentrated in the anaerobic stage, or just the anaerobic hydrolysis/acidification was applied, effective removal of emerging compounds would be still achieved. Meanwhile, organic matters could be converted into solid substance and then recovery and utilization of energy would be achieved through anaerobic digestion.

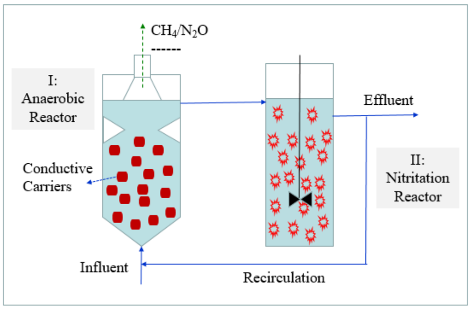

Figure 3. A new wastewater treatment concept with anaerobic treatment and nitritation-based processes for simultaneous energy recovery and emerging compounds control.

The nitrogen management process includes mainly nitritation, anaerobic ammonia oxidation or partial denitrification and so on. The core technology is nitritation, and through nitrite accumulation provides electron acceptor for N_2O production and nitrogen removal. In the nitritation stage, as anaerobic effluent contains low concentrations of organic carbon, activities of heterotrophs would be enhanced. Thereby, emerging compounds removal would be enhanced through activities of both heterotrophs and AOB. As a result, high-efficient control of emerging compounds can be achieved and nitrogen can also be effectively removed.

6. Conclusion

Sustainable development of wastewater treatment requires the energy and resource recovery from wastewater and also control of emerging compounds. Under anaerobic conditions, methanogenesis and/or N2O production could be achieved simultaneously, and some types of emerging compounds removal could be enhanced. During nitritation, nitrogen and emerging compounds removal could be also simultaneously achieved. A new wastewater treatment concept was proposed to promote the recovery of energy and removal of emerging compounds, including advanced anaerobic treatment through enhancing DIET and nitritation-based nitrogen management.

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