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Strategies for sustainable wastewater treatment based on energy recovery and emerging compounds control: a mini-review

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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 reduction and control of ecological toxicity is the focus for emerging compounds. Under anaerobic conditions, especially with the enhancement of direct interspecies electron transfer, methanogenesis and/or nitrous oxide production could be achieved separately or simultaneously, and the removal efficiency of some types of emerging compounds could be improved. 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 the anaerobic treatment process by incorporating directly electron transfer enhancement and nitritation-based nitrogen removal. The proposed strategy provided a new technology concept 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 of human health and ecological safety. Therefore, it has been considered as one of the most important technologies in the previous century. Removal of pollutants including organic carbon, nitrogen, and phosphorus has been historically considered during development of wastewater treatment, corresponding to the wastewater treatment stages of 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 [1].

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 a 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, the ecological risk and toxicity control of emerging compounds should be the near future focus.

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

With the sustainable development of wastewater treatment, concepts are gradually changing from the 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 [2,3]. For energy recovery, the main approach includes converting organic matters in wastewater into energy through anaerobic treatment directly. 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 main focuses, 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. [2] 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. [3] 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.

The removals of emerging compounds during wastewater treatment are important because of their toxicities, especially from the perspectives of environmental risk and ecological health. The effect and removal mechanisms of emerging compounds such as pharmaceutical and personal care products (PPCPs) in wastewater treatment (i.e., review from Luo et al. [4] have been evaluated systematically. Carballa et al. [5] examined the removal of PPCPs and endocrine disrupting chemicals (EDCs), and found that biodegradation of activated sludge contributed to the removals of PPCPs and EDCs. Furthermore, the biodegradation pathway of emerging compounds and the occurrence of their transformation products in wastewater have attracted intensive attention as well as their environmental risk and ecological toxicities [6]. Therefore, in future, effective control technologies 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. [7] 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 was a significant component. Therefore, maintaining the healthy and safe development of ecosystem becomes the final target from the perspective of sustainable development. As shown in Fig. 1 [8], a new concept for sustainable wastewater treatment is proposed so as to achieve an ecosystem-based wastewater treatment. The new concept includes three levels of energy, material, and ecosystem. The internal support is energy, which includes energy flow

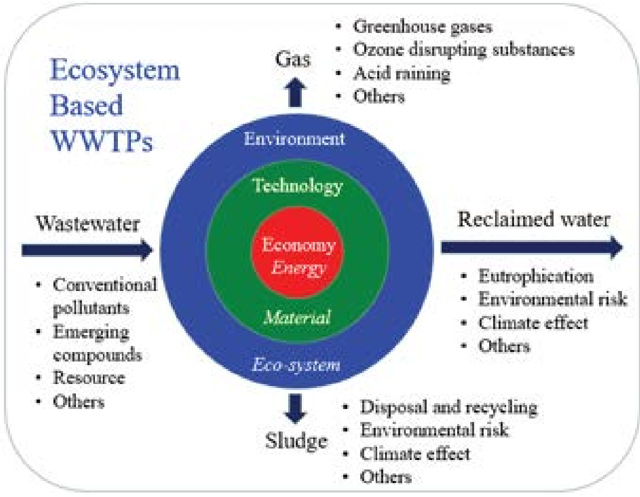

Fig. 1. The proposed framework for the ecosystem-based wastewater treatment plants [8].

such as electricity for system 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 environment, 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. Therefore, wastewater treatment process includes not only nutrients removal, but also the support of sustainable development of ecosystem.

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 enhanced electron transfer

Due to the low organic concentration in municipal wastewater, there are some key problems when the 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 [9]. To date, the anaerobic treatment technology based on anaerobic membrane bioreactor is intensively developed and investigated. For the anaerobic membrane bioreactor, mechanism of membrane fouling and its control strategy are still key research aspects so as to reduce the operating cost [10,11]. 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 [12].

Anaerobic treatment includes several steps, such as hydrolysis, acidification, and methane production, which require different types of microorganisms to work 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 [13]. 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 [13].

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 [13]. Conductive materials mainly shortened the lag phase of methane production and increased the methane production rate [13]. The acceleration of conductive materials depends on many factors. For example, dosing ferroferric oxide increased the methanogenic efficiency of the tryptone acclimated anaerobic sludge while for the starch acclimated anaerobic sludge, the facilitation was less affected [14]. During facilitation, conductive materials were proposed to act as electron conduits which transported the electron between syntrophic bacteria and methanogen, and both activities of the electron transport chain and extracellular electron transfer ability were enhanced [15]. Microbial and metabolic mechanisms of anaerobic treatment are still required for 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 chemical 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. In activated sludge treatment, 17α-ethinylestradiol was readily biodegraded under aerobic condition in wastewater treatment [16]. For biodegradable compounds, various transformation products were observed, and their occurrence in activated sludge attracted more and more attention such as ibuprofen and hydroxylated metabolites. Although anaerobic biodegradation attracts less attention, it showed effective removals of emerging compounds including pharmaceuticals and hormone. More specifically, some emerging compounds which cannot be degraded by aerobic processes, can be degraded easily under anaerobic conditions, such as sulfamethoxazole (Fig. 2) [17]. Meanwhile, dosing redox mediator or conductive materials further improved the anaerobic treatment efficiency of emerging compounds (Fig. 2) [17]. Besides sulfamethoxazole, effective removals of tylosin, kemicitine, oxytetracyline, and amoxicillin were observed under anaerobic condition [18]. Toxicity of emerging compounds is mainly related to their concentration, and the health risk on human and environmental risk of emerging compounds were reported in previous studies. In addition, after biodegradation, their transformation products are usually complicated and perform similar or higher toxicity than their parent compounds, so their ecological effects after wastewater treatment cannot be ignored. Recently, Völker et al. [19] 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.

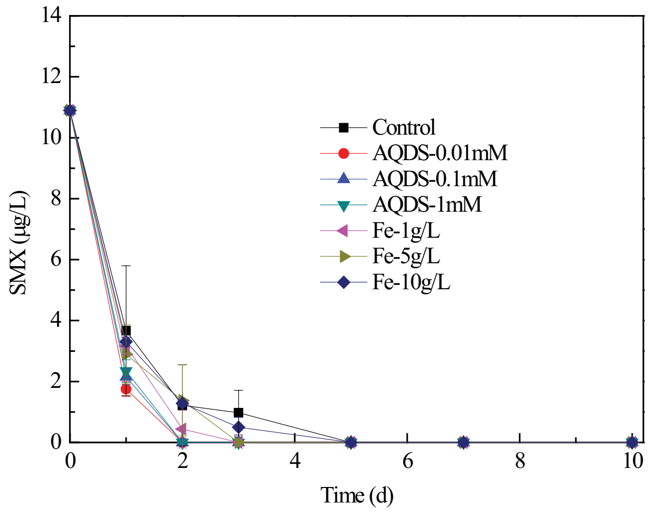

Fig. 2. Example of effect of dosage of anthraquinone-2,6-disulfonate (AQDS) or ferroferic oxide (Fe) on anaerobic degradation of sulfamethoxazole (SMX) [17].

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

Effective management of nitrogen is very important for energy and resource recovery from wastewater. To date, the 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 the 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 the energy consumption; (2) reduction in the 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 [20–23].

The ammonia monooxygenase (AMO) gene in AOB makes it possible to simultaneously cometabolize certain types of emerging compounds. For example, the biological removal of 17a-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 [24,25]. Fernandez-Fontaina et al. [26] also concluded that under high ammonia nitrogen loading rates, emerging compounds could be efficiently removed via biological processes, with the AMO-based cometabolism being the possibly dominating metabolic pathway. Batt et al. [27] 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 the nitrifying activated sludge was 50%, while it was only 1% in the common activated sludge. The high biomass concentrations and long sludge retention times such as membrane bioreactors enhanced the biodegradation of emerging pollutants [28].

For responding bacteria in wastewater treatment, *Proteobacteria* have a significant contribution in the removal of antimicrobial and anti-inflammatory substances [29]. Alphaproteobacteria and Betaproteobacteria were reported to increase in the presence of carboxylic PPCPs [30,31]. Biodegradation of triclosan, bisphenol A, and ibuprofen by *Nitrosomonas europaea* was investigated by Roh et al. [32], and 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* could be inhibited by the degradation products or the toxicity of triclosan [32]. Tran et al. [33] studied the 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 concentrations. In addition, the existence of heterotrophs promoted certain PPCPs removal [33]. In particular, acclimation of heterotrophs that can degrade emerging compounds through cometabolism or directly deserves further investigation, especially under oligotrophic nitrifying conditions. For biodegradation of PPCPs, cometabolism is supposed to be the dominating biodegradation process because of low amounts of PPCPs, which cannot serve as the growth substrate [34].

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 the burning agent or rocket oxidizer. In particular, the energy generation efficiency can be promoted during the cocombustion of methane and N_2O mixture. Therefore, N_2O production by denitrification is an effective way for energy recovery. 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 h [35]. 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. 3) [36]. Therefore, coupling denitrification for N_2O production is not only a technology to effectively control nitrogen, but an ideal way to achieve energy generation from nitrogen during wastewater treatment.

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

Based on the research results earlier, a new wastewater treatment concept coupling energy recovery and emerging compound control is proposed (Fig. 4). This new technology mainly includes anaerobic treatment and high-efficient nitrogen removal processes. The two processes are carrying out under the optimal condition separately, which would improve the system performance.

Anaerobic treatment includes methanogenesis, denitrification for N_2O production, and enhanced emerging compounds removal. Under anaerobic conditions, the methanogenesis process can be enhanced by dosing conductive

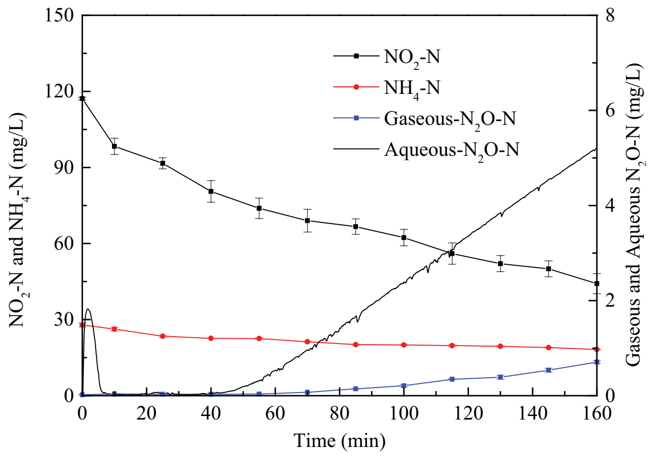

Fig. 3. Example of nitrous oxide (N_2O) production in a typical denitrifying sequencing batch reactor reaction cycle with starch as the organic carbon and nitrite as the electron acceptor [36].

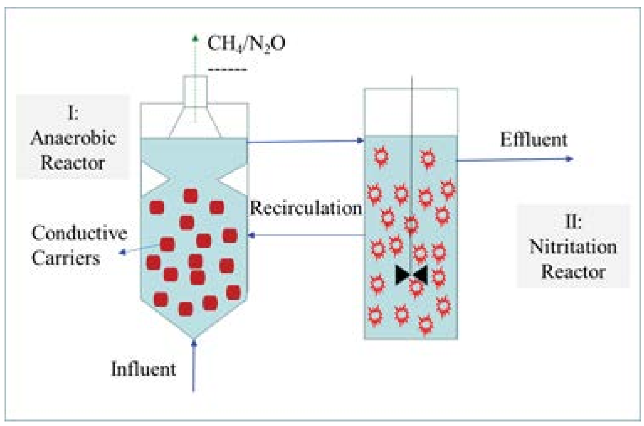

Fig. 4. A new wastewater treatment concept with anaerobic treatment and nitritation-based processes for simultaneous energy recovery and emerging compounds control.

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 the recovery and utilization of energy would be achieved through anaerobic digestion.

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, providing electron acceptor for N_2O production and nitrogen removal. In the nitritation stage, as anaerobic effluent contains low concentrations of organic carbon, activities of oligotrophic heterotrophs may 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. Conclusions

Sustainable development of wastewater treatment requires energy and resource recovery from wastewater and also control of emerging compounds. Under anaerobic conditions, methanogenesis and/or N_2O 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 and nitritation-based nitrogen management processes.

Acknowledgments

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