



Energy upcycle in anaerobic treatment: Ammonium, methane, and carbon dioxide reformation through a hybrid electrodeionization–solid oxide fuel cell system



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ARTICLE INFO

Article history:

Received 8 November 2016

Received in revised form 22 February 2017

Accepted 23 February 2017

Keywords:

Anaerobic digestion

Electrodeionization

Solid oxide fuel cell

Biogas

Ammonia

Electricity

ABSTRACT

To create possibilities for a more sustainable wastewater management, a novel system consisting of electrodeionization (EDI) and solid oxide fuel cells (SOFCs) is proposed in this study. This system is integrated with anaerobic digestion/landfills to capture energy from carbonaceous and nitrogenous pollutants. Both EDI and SOFCs showed good performances. EDI removed 95% and 76% ammonium-nitrogen ($\text{NH}_4^+\text{-N}$) from diluted (0.025 M) to concentrated (0.5 M) synthetic ammonium wastewaters, respectively, accompanied by hydrogen production. SOFCs converted the recovered fuels, biogas mixtures of methane and carbon dioxide, to electricity. Under the optimal conditions of EDI (3.0 V applied voltage and 7.5 mm internal electrode distance (IED), and SOFCs (750 °C operating temperature), the system achieved 60% higher net energy output as compared to conventional systems. The estimated energy benefit of this proposed system showed that the net energy balance ratio is enhanced from 1.11 (existing system) to 1.75 (this study) for a local Hong Kong active landfill facility with 10.0 g L⁻¹ chemical oxygen demand (COD) and 0.21 M $\text{NH}_4^+\text{-N}$. Additionally, an average of 80% inorganic ions (heavy metals and nutrient elements) can be removed from the raw landfill leachate by EDI cell. The results are successful demonstrations of the upgrades of anaerobic processes for energy extraction from wastewater streams.

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1. Introduction

Energy extraction from wastewater stream has gained increasing attention to eliminate environmental threats and offset fossil fuel consumption [1–4]. The most readily adaptable approaches are anaerobic treatment, e.g., anaerobic digestion (AD) or landfill, converting wastewater or municipal solid waste into biogas (approximately 60% methane (CH_4)/40% carbon dioxide (CO_2) volume/volume [v/v]) and ammonium-rich fermentation broth/leachate (400–8000 mg L⁻¹ $\text{NH}_4^+\text{-N}$) [5–8]. Although biogas is well established as a fuel for electricity generation via combined heat and power (CHP) or cogeneration with gas engines [9–11], the electricity conversion efficiency is usually limited to around 30% [12,13]. Ammonium-rich wastewater, discharged without proper

treatment, brings severe environmental impacts [14–17]. Further improvements and updates are hence necessary and urgent.

Conventionally, $\text{NH}_4^+\text{-N}$ in fermentation broth/leachate can be removed through adding an alkali to raise the pH level over its pK_a value (9.25) followed by physicochemical methods such as microwave radiation, air stripping (AS), and heating [18–22]. Biological nitrogen removal processes such as nitrification–denitrification, nitrification shortcut, and anaerobic ammonium oxidation have also been applied [21,23]. These approaches require expensive chemicals and consume intensive energy [18–22]. Ammonia (NH_3) can be used as an alternative fuel to hydrogen (H_2) [24]. Upon decomposition, NH_3 produces two harmless gases (nitrogen [N_2] and H_2), and simultaneously releases 320 kJ mol⁻¹ chemical energy. Theoretically, this process produces approximately 10% more energy than H_2 oxidation (285 kJ mol⁻¹) [25–27]. Since NH_3 has special properties (incombustibility, incomplete decomposition, toxicity, and solubility), its energy potential has not been highly recognized yet [28,29]. For wastewater in which NH_3 exists in form of NH_4^+ ion, the low conversion efficiency from NH_4^+ ion to

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Nomenclature

EDI	electrodeionization	AS	air stripping
SOFC	solid oxide fuel cells	IED	internal electrode distance
COD	chemical oxygen demand	EBR	energy balance ratio
CHP	combined heat and power		

NH_3 gas, high cost, and NO_x emissions also reduce the motivation to explore its energy potential [30].

SOFC is a promising energy conversion technology. It is capable of electrochemically converting a variety of gas fuels (H_2 , NH_3 , CH_4 and other hydrocarbon compounds) into electricity. Also, it has an energy conversion efficiency as high as 50% [31,32]. The upper right-hand panel of Fig. 1 illustrates the power generation mechanism of SOFCs. (I) When CH_4 is fuel, it reforms with H_2O or CO_2 and produces H_2 and CO at the anode under the high-temperature condition. (II) These reformed gases react with oxygen ions (O^{2-}) and produce CO_2 and H_2O , and release electrons (e^-). (III) The released electrons flow through the external circuit and they are collected by the current collector to produce electrical power. (IV) Once electrons reach the cathode, they are accepted by O_2 molecule and O^{2-} ions are produced again. In this cycle, O_2 and gas fuels are consumed for electricity generation.

SOFCs have some advantages for energy harvesting from wastewater. As SOFCs generate electrical power in a straightforward way through electrochemical reactions and does not go through thermodynamic cycles, its power generation efficiency is not limited by the Carnot efficiency [33]. Although carbon deposition is an issue for SOFCs fed with pure CH_4 , this issue might be solved for biogas because its reformation with H_2O or CO_2 generates gases instead of solid carbon [34–36]. As such, biogas is poten-

tially a good fuel for power generation systems using SOFCs [37]. Another potential fuel for SOFCs is NH_3 gas. When NH_3 is fed into SOFCs, it is thermally decomposed into N_2 and H_2 . Subsequently, H_2 goes through the same reaction process with other gas fuels [38]. Studies have found that carbon deposition can be prevented by adding NH_3 to CH_4 as well [31,39]. Therefore, this novel system of integrating SOFCs with AD and landfill facilities is proposed to convert NH_3 and biogas into electrical power for simultaneous pollution control.

The main issue with this system is that the aqueous NH_4^+ in fermentation broth/leachate requires an additional conversion step to NH_3 gas so as to be fed into SOFC. As aforementioned, the existing approaches for NH_3 gas recovery are both uneconomic and environmentally unfriendly [40–42]. Hence, advanced technologies are required. EDI is an attractive technology because it has an excellent selectivity for target ion. Ion migration is driven by electric potential gradients rather than by physical pressure [43,44]. As shown in the bottom right panel of Fig. 1, directional movement leads to accumulation of target ions such that the concentrated ions can be harvested with low energy consumption. As Mondor et al. reported, EDI has been used to produce fertilizers from swine manure with $1.0 \text{ kW h kg}^{-1} \text{ NH}_3$ energy input, saving $1.8 \text{ kW h kg}^{-1} \text{ NH}_3$ compared to $2.8 \text{ kW h kg}^{-1} \text{ NH}_3$ required for AS [45]. Moreover, the applied voltage leads to water splitting,

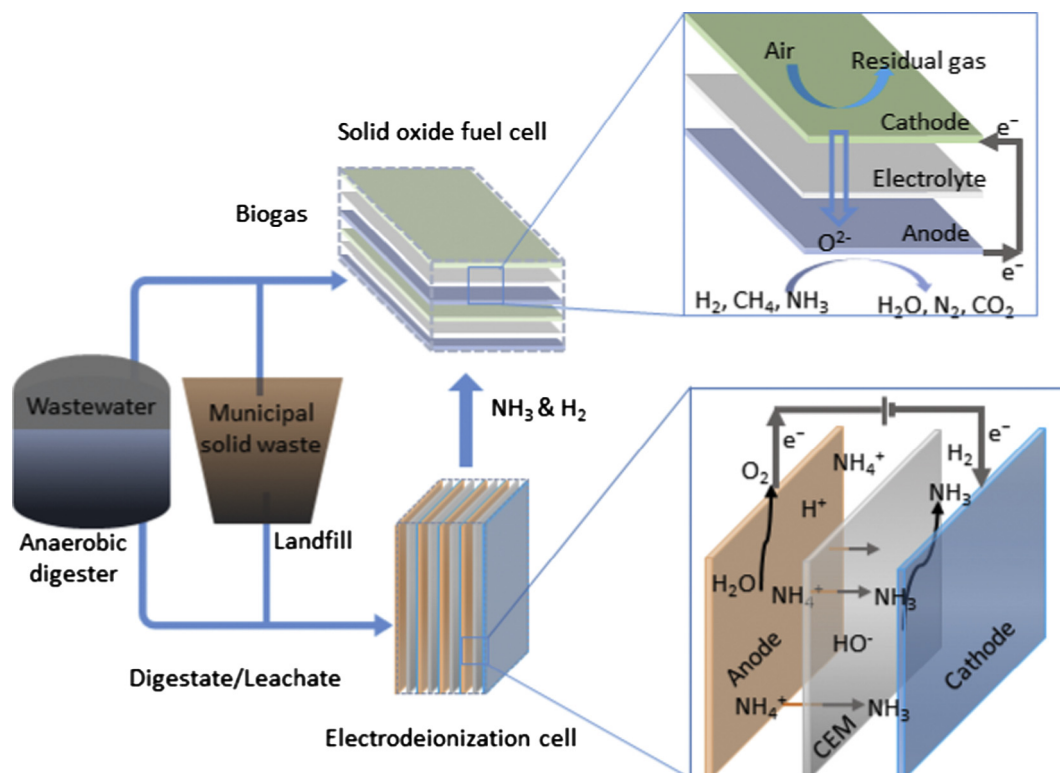


Fig. 1. Schematic diagram of the EDI-SOFC system integrated with anaerobic treatment.

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