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# Supercritical water gasification of landfill leachate for hydrogen production in the presence and absence of alkali catalyst

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## ABSTRACT

Gasification of landfill leachate in supercritical water using batch-type reactor is investigated. Alkali such as NaOH, KOH,  $K_2CO_3$ ,  $Na_2CO_3$  is used as catalyst. The effect of temperature (380–500 °C), retention time (5–25 min), landfill leachate concentration (1595 mg L<sup>-1</sup>–15,225 mg L<sup>-1</sup>), catalyst adding amount (1–10 wt%) on hydrogen mole fraction, hydrogen yield, carbon gasification rate, COD, TOC, TN removal efficiency are investigated. The results showed that gaseous products mainly contained hydrogen, methane, carbon dioxide and carbon monoxide without addition of catalyst. However, the main gaseous products are hydrogen and methane with addition of NaOH, KOH,  $K_2CO_3$ ,  $Na_2CO_3$ . In the absence of alkali catalyst, the effect of temperature on landfill leachate gasification is positive. Hydrogen mole fraction, hydrogen yield, carbon gasification ratio increase with temperature, which maximum value being 55.6%, 107.15 mol kg<sup>-1</sup>, 71.96% is obtained at 500 °C, respectively. Higher raw landfill leachate concentration leads to lower hydrogen production and carbon gasification rate. The suitable retention time is suggested to be 15 min for higher hydrogen production and carbon gasification rate. COD, TOC and TN removal efficiency also increase with increase of temperature, decrease of landfill leachate concentration. In the presence of catalyst, the hydrogen production is obviously promoted by addition of alkali catalyst. the effect of catalysts on hydrogen production is in the following order: NaOH > KOH >  $Na_2CO_3$  >  $K_2CO_3$ . The maximum hydrogen mole fraction and hydrogen yield being 74.40%, 70.05 mol kg<sup>-1</sup> is obtained with adding amount of 5 wt% NaOH at 450 °C, 28 MPa, 15 min.

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## 1. Introduction

The sanitary landfill method for the ultimate disposal of solid waste material is still widely used because it is inexpensive (Zhao et al., 2010). Landfill leachate is generated from the leaching of municipal solid waste and excess rainwater percolating through the waste layer (Christensen et al., 2001). Landfill leachate is a kind of organic wastewater containing high concentrations of chemical oxygen demand (COD), total organic carbon (TOC), total nitrogen (TN) and heavy metals. In the past several decades, some conventional landfill leachate treatments have been developed. However, due to its complexity, a combination of chemical, physical and biological steps is required to treat landfill leachate for discharge into surface water (Ye et al., 2017; Bakraouy et al., 2017; Liao et al., 2014). However, the focus of traditional landfill leachate treatments is high removal efficiency of organic matters and

nitrogen-containing compounds in landfill leachate. Wet biomass in landfill leachate has not been utilized effectively to produce clean energy. Therefore, developing a new resource utilization process to treat landfill leachate is attractive.

Supercritical water can dissolve organic compounds because of its special physical and chemical properties. The solubility and the diffusion coefficients of supercritical water can be easily controlled by adjusting the reaction temperature and pressure (Cheng et al., 2016). Supercritical water gasification (SCWG) is an advanced technology which can obtain hydrogen- rich gaseous products from wet biomass without any drying process (Li et al., 2011). High solubility of the intermediates in the reaction medium significantly inhibits tar and char formation that are one of the main drawbacks of conventional gasification.

Many reports have presented the biomass gasification in supercritical water, such as microalgae (Cherad et al., 2016), grass (Nanda et al., 2016a,b), sludge (Gong et al., 2014, 2017) and fruit waste (Nanda et al., 2016a,b) to produce hydrogen. However, few studies on gasification of landfill leachate in supercritical water are reported. Gong (Gong and Duan, 2010) had reported the

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**Table 1**  
The characteristics of landfill leachate used in experiments.

Parameter	COD	TOC	NH <sub>3</sub> -N	TN	pH
Unit	mg L <sup>-1</sup>	mg L <sup>-1</sup>	mg L <sup>-1</sup>	mg L <sup>-1</sup>	/
Range	42,536–44,073	14,860–16,120	2008–2275	1703–2279	5–7

treatment of landfill leachate using supercritical water oxidation (SCWO). During SCWO process, COD removal efficiency of landfill leachate could reach up to 99.23%. Williams (Williams and Onwudili, 2006) had also verified almost complete oxidation of the organic components of the leachate in SCWO system. Partial oxidation of landfill leachate under supercritical water condition to produce hydrogen was reported (Gong et al., 2015). The results showed that hydrogen gas yield, TOC removal rate and carbon recovery rate of 14.32 mmol gTOC<sup>-1</sup>, 82.54% and 94.56% was achieved under optimum conditions, respectively.

Suitable catalysts in SCWG can accelerate the gasification reaction, which can promote hydrogen production. Various kinds of homogeneous and heterogeneous catalysts including metal (Mastuli et al., 2017), metal-oxide (Peng et al., 2017) and alkali (Ge et al., 2017) have been investigated for SCWG of real biomass and model compounds. Alkali such as NaOH, KOH, K<sub>2</sub>CO<sub>3</sub>, Na<sub>2</sub>CO<sub>3</sub>, is a homogeneous catalyst which can be mixed with the feedstock uniformly and promote water-gas shift reaction thus to increase the hydrogen yield (Ding et al., 2014).

In this study, landfill leachate was gasified under supercritical water condition. NaOH, KOH, K<sub>2</sub>CO<sub>3</sub>, Na<sub>2</sub>CO<sub>3</sub> catalyst was added into SCWG process. The effect of temperature, retention time, landfill leachate concentration, catalyst loading on landfill leachate gasification gas component, gas yield, carbon gasification rate, TOC, COD and TN removal efficiency is investigated.

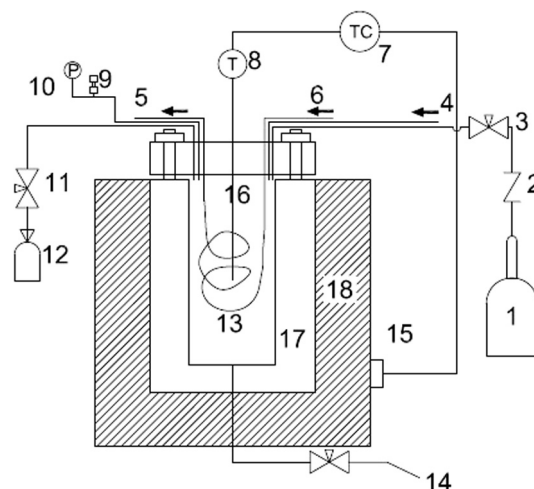
## 2. Materials and methods

### 2.1. Materials

The landfill leachate used in this experiment was collected from municipal solid waste landfill site on Zhengzhou, China. The solid waste deposited in the landfill was domestic garbage such as food waste, paper, plastic and so on. The landfill leachate generated from landfill was collected by pipe and stored in regulating reservoir. The landfill leachate used in this experiment was collected from reservoir with artificial sampler, which was young with the age of less than 5 years. The characteristics of landfill leachate are shown in Table 1. Nitrogen was used to check the air tightness and eliminate the air in device before experiment, which purity was up to 99%. All the other chemicals used in experiments were purchased with analytical purity.

### 2.2. Supercritical water gasification system

The scheme diagram of experimental apparatus was shown in Fig. 1. Supercritical water gasification reactor was produced by Nantong HuaAn supercritical extraction Co. Ltd, which was a batch type with volume of 600 mL. The reactor was made of stainless steel HC276. The highest temperature and pressure for reactor was 650 °C and 60 MPa. For the safety, the operation pressure was fixed under 40 Mpa. The reactor shell was wrapped in insulation. The inside temperature was regulated and monitored by the temperature control device with ±1 °C precision. The pressure of the reactor was shown in pressure gauge. The cooling coil was fixed in the reactor and to be used for cooling the reactor rapidly with water to ambient temperature at the end of each experiment.



**Fig. 1.** Scheme diagram of supercritical water gasification system.

1.nitrogen cylinder 2.check valve 3.pressure reducing valve 4.inlet of leachate 5.outlet of cooling water 6.inlet of cooling water 7.temperature control 8.thermocouple 9.safety valve 10.pressure gauge 11.valve 12.gas collecting 13.cooling coil 14.liquid product 15.thermocouple 16.reactor 17.heating coil 18.insulation wall

The experimental steps are as follows: before the experiment, nitrogen is injected into the reactor to avoid the effect of air on the gasification result. According to the temperature and pressure required in the experiment, landfill leachate and catalyst are mixed and charged into the reactor with high pressure metering pump. Then, the reactor is electrical heated with the average heating rate of 3 °C/min (If the reaction temperature is set at 500 °C, it takes about 158 min from room temperature 25 °C to 500 °C). The pressure in reactor increase with rise of temperature, which is determined by the volume of feed solution and temperature. When temperature and pressure reach the set point, this point is recorded as the starting point of the reaction. The setting operation condition was held for 5–25 min. After the fixed reaction time is reached, the gasification is suspended, then the cooling water is opened, and reaction temperature, pressure and reaction time are recorded. The effluent is cooled down to room temperature rapidly with cooling water. Gaseous products are collected into gasbag for analysis. When the pressure inside reactor decrease to normal level, flushing the reactor using nitrogen, then residual mixtures in reactor are collected into beaker. The mixtures in beaker are separated by filtration. The liquid and solid phase products are analyzed by Gas Chromatography–Mass Spectrometry and infrared spectrum, respectively. The experiments are repeated at different conditions.

### 2.3. Analytical methods

The characteristics of landfill leachate before the supercritical water gasification and the liquid effluent after reaction are specified with the value of COD, TOC and TN, which indicated the removal of organic pollutants and nitrogen-containing compounds. Measurement for the concentration of COD is made using China standard methods. TOC and TN are monitored by N/C 2100 TOC analyzer.

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