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Partial oxidation of landfill leachate in supercritical water: Optimization by response surface methodology

Yanmeng Gong, Shuzhong Wang*, Haidong Xu, Yang Guo, Xingying Tang

Key Laboratory of Thermo-Fluid Science and Engineering, Ministry of Education, School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an, Shaanxi 710049, China

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ABSTRACT

To achieve the maximum H_2 yield (GY_{H_2}), TOC removal rate (TRE) and carbon recovery rate (CR), response surface methodology was applied to optimize the process parameters for supercritical water partial oxidation (SWPO) of landfill leachate in a batch reactor. Quadratic polynomial models for GY_{H_2} , CR and TRE were established with Box–Behnken design. GY_{H_2} , CR and TRE reached up to $14.32 \text{ mmol}\cdot\text{gTOC}^{-1}$, 82.54% and 94.56% under optimum conditions, respectively. TRE was invariably above 91.87%. In contrast, TC removal rate (TR) only changed from 8.76% to 32.98%. Furthermore, carbonate and bicarbonate were the most abundant carbonaceous substances in product, whereas CO_2 and H_2 were the most abundant gaseous products. As a product of nitrogen-containing organics, NH_3 has an important effect on gas composition. The carbon balance cannot be reached due to the formation of tar and char. CR increased with the increase of temperature and oxidation coefficient.

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1. Introduction

Sanitary landfill is widely employed in the disposal of municipal solid waste throughout the world with landfill leachate formation being an inevitable consequence. In the past several decades, some conventional landfill leachate treatments have been developed. However, a combination of chemical, physical and biological steps or an advanced method is required to meet the strict quality standards for direct discharge of leachate into surface water.

Supercritical water (SCW, $T > 647.15 \text{ K}$, $P > 22.12 \text{ MPa}$) presents complete miscibility with oxygen and organics, making SCW a very suitable medium for the oxidation of organics (Bermejo and Cocero, 2006). Supercritical water oxidation (SCWO) has been regarded as a promising technology to treat landfill leachate effectively in recent years (Gong and Duan, 2010; Wang et al., 2011; Williams and Onwudili, 2006). During SCWO process, organics can be degraded into CO_2 and H_2O finally in a few minutes or even several seconds via homogeneous oxidation reaction with oxidant in SCW. Gong and Duan (2010) reported that chemical oxygen demand (COD) and biochemical oxygen demand (BOD) removal efficiencies of landfill leachate could reach up to 99.23% and 98.06%, respectively, at 703 K and 30 MPa in a continuous transpiring-wall SCWO reactor. It has been verified by Williams and Onwudili (2006) that almost complete oxidation of the organic components of the leachate could be achieved under SCWO

conditions. Moreover, MnO_2 was an effective catalyst in destruction of landfill leachate (Wang et al., 2011). To date, Aquarden Technologies in Denmark has developed an operational SCWO-based solution for on-site treatment of leachate and recognized that SCWO technology could be instrumental in resolving the landfill leachate treatment challenge (Aquarden website, 2015).

Typically, in a SCWO system, landfill leachate is preheated above critical temperature of water before it reacts with oxidant in a subsequent reactor, as shown in Fig. 1. It has been confirmed that significant tar and char will be generated in the preheating process of high concentration feedstock (Antal et al., 2000; Guo et al., 2013; Matsumura et al., 2006b). Sricharoenchaikul (2009) observed that the portion of tar and char reached 57.6% at 773 K and 30 MPa in the supercritical water gasification (SCWG) of 10 wt.% black liquor, whereas the portion decreased to 24.6% at 923 K. Actually, landfill leachate is a high concentration organic wastewater, in which the COD concentration could reach up to $35,000\text{--}50,000 \text{ mg L}^{-1}$ (Renou et al., 2008). Thus, it can be predictable that the leachate preheater and SCWO reactor might be subject to plugging by tar and char in a long running system, because once tar and char form they are difficult to be converted (Chuntanapum and Matsumura, 2010).

Some research suggests that the plugging problem caused by tar and char might be solved by increasing heating rates of feedstock (Matsumura et al., 2006a; Sinağ et al., 2004; Watanabe et al., 2005), adding catalyst (Matsumura et al., 2005; Xu and Lancaster, 2008) or adding a small amount of oxidant (Guo et al.,

* Corresponding author.

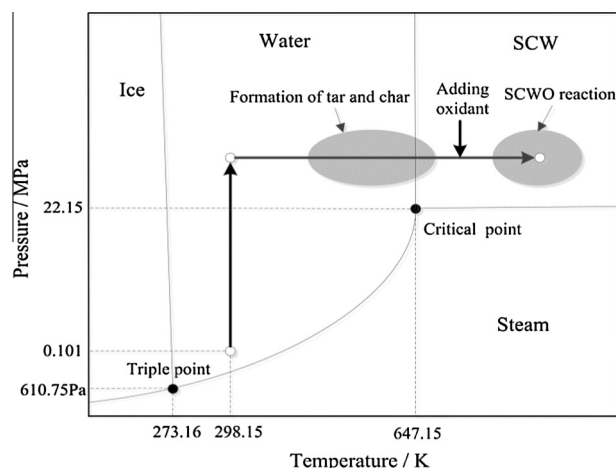


Fig. 1. Reaction fields in SCWO process for landfill leachate.

2010; Jin et al., 2010; Xu et al., 2011). The heat resistance exists inevitably in the process of external heating method, showing that it is difficult to receive a rapid heating of landfill leachate (Jin et al., 2010). Although Na_2CO_3 , K_2CO_3 and NaOH can be used to inhibit the formation of char and tar in SCWG of biomass, these alkaline catalysts may cause corrosion and plugging of reactor (Sinağ et al., 2004). Supercritical water partial oxidation (SWPO) involves carrying out oxidative reactions in SCW in the presence of sub-stoichiometric quantities of an oxidant. The key advantage of SWPO process is the use of partial oxidation in-situ to flash heat the feedstock through the sensitive temperature range, resulting in less char or tar formation and high hydrogen yield (Johanson et al., 2001).

To date, little attention has been paid to partial oxidation of landfill leachate in SCW. In this work, landfill leachate SWPO was performed in a batch SCWO reactor investigating the effects of oxidation coefficient, reaction time, temperature and pressure on the process with response surface methodology (RSM). Another major goal was to obtain the optimum operating parameters to achieve the maximum hydrogen yield and TOC removal rate and minimum generation of tar and char.

2. Materials and methods

2.1. Leachate characteristics

Landfill leachate was collected in November from a municipal landfill site on Xi'an Shaanxi Province, China, whose characteristics are listed in Table 1. The landfill is received only municipal solid waste since it was in operation in 1994 with the total volume of 49 million m^3 . Hydrogen peroxide solution with mass fraction of 30% served as the oxidant.

2.2. Apparatus and procedure

Fig. 2 illustrates the experimental apparatus in this study. The batch reactor was made of Hastelloy-C276 alloy with a net capacity of 350 mL which was designed to a maximum temperature and pressure of 923 K and 32 MPa, respectively. The temperature was

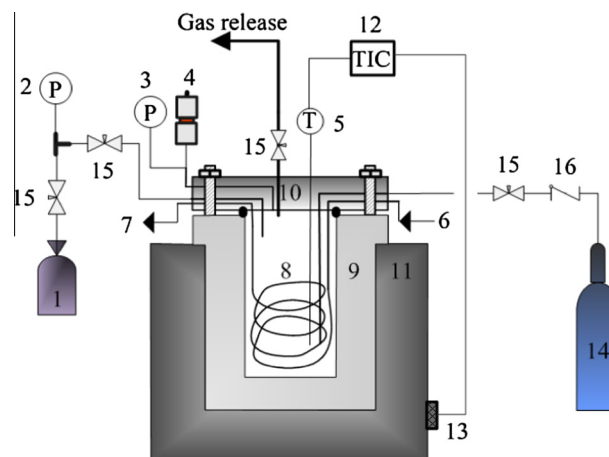


Fig. 2. Schematic diagram of the batch reactor (1) gas bag; (2) low-pressure gauge; (3) high-pressure gauge; (4) safety valve; (5) thermocouple; (6) cooling water inlet; (7) cooling water outlet; (8) cooling coil; (9) kettle; (10) kettle cover; (11) electric heater; (12) temperature controller; (13) power source; (14) nitrogen cylinder; (15) needle valve; (16) check valve.

monitored by a thermocouple and adjusted by a temperature controller automatically. A high-pressure gauge was applied to determine the inner-pressure of reactor when the reactor was heating, and another low-pressure gauge was used to measure the pressure of products after the reactor was cooled to room temperature. The measuring ranges of these two pressure gages were 40 MPa and 1.6 MPa, with accuracy of ± 0.64 MPa and ± 25.6 kPa, respectively. A dip tube was extended to the bottom of reactor for purging the whole reactor by N_2 .

The experiments were performed using the following procedure for safety and operation convenience. According to the thermodynamic property of water, the volumes of landfill leachate and H_2O_2 solution can be calculated for a given oxidation coefficient, temperature and pressure. At the beginning of each experiment, quantitative landfill leachate and H_2O_2 solution were charged into the reactor before the reactor was sealed. The whole system was purged by N_2 for 10 min to sweep the air. Afterward, the switch of heating power was turned on and the temperature controller was adjusted to the selected values. As soon as the temperature was reached the setting value, the reactor was held for a specified time. When the desired reaction time has elapsed, the electric heater was stopped immediately and the reactor was cooled to room temperature rapidly by cooling water. Gaseous and liquid products were collected and analyzed at the end of each experiment. In this work, we repeated each experiment three times.

2.3. Experimental analysis

The analysis of gaseous products occurred on a gas chromatograph (GC-112A, Shanghai) with a thermal conductivity detector (TCD) and a $3 \text{ m} \times 3 \text{ mm}$ TDX-01 packed column for the detection of H_2 , CO , CH_4 and CO_2 . Helium served as the carrier gas, with the flow rate and pressure of $18.5 \text{ mL} \cdot \text{min}^{-1}$ and 0.42 MPa, respectively.

The total organic carbon (TOC) and total carbon (TC) of the liquid products were monitored by a TOC analyzer (ET1020A, EURO

Table 1
Leachate characteristics.

pH	COD ($\text{mg} \cdot \text{L}^{-1}$)	$\text{NH}_4\text{-N}$ ($\text{mg} \cdot \text{L}^{-1}$)	TN ($\text{mg} \cdot \text{L}^{-1}$)	TC ($\text{mg} \cdot \text{L}^{-1}$)	TOC ($\text{mg} \cdot \text{L}^{-1}$)
8.04 ± 0.02	6633 ± 330	393 ± 54	3800 ± 82	9650.3 ± 467.2	4079.0 ± 376.0

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