Bioenergy recovery from landfill gas: A case study in China

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Abstract Landfill gas (LFG) utilization which means a synergy between environmental protection and bioenergy recovery was investigated in this study. Pressure swing adsorption technology was used in LFG purification, and laboratory experiment, pilot-scale test, and on-site demonstration were carried out in Shenzhen, China. In the laboratory experiment, A-type carbon molecular sieve was selected as the adsorbent by comparison of several other adsorbents. The optimal adsorption pressure and adsorption time were 0.25 MPa and 2 min, respectively, under which the product generation rate was 4.5 m³/h and the methane concentration was above 90%. The process and optimization of the pilot-scale test were also reported in the paper. The product gas was of high quality compared with the National Standard of Compressed Natural Gas as Vehicle Fuel (GB18047-2000), when the air concentration in feed gas was under 10.96%. The demonstration project was composed of a collection system, production system, and utilization system. The drive performance, environmental protection performance, and economic feasibility of the product gas — as alternative fuel in passenger car, truck, and bulldozer — were tested, showing the feasibility technology for LFG utilization.

Keywords landfill gas (LFG), compressed purified landfill gas (CPLG), pressure swing adsorption (PSA), alternative vehicle fuel, demonstration project

1 Introduction

The major components of landfill gas (LFG) are methane (CH₄) and carbon dioxide (CO₂), which result from anaerobic decomposition of the biodegradable organic fraction of waste in landfill sites. Both CH₄ and CO₂ are greenhouse gases (GHG). According to the report of the Intergovernmental Panel on Climate Change (IPCC), the increasing concentrations of GHG in the atmosphere tend to warm the surface of the earth, and the global average surface temperature has increased over the 20th century by about 0.6°C [1]. The atmospheric concentration of CH₄ and CO₂ has increased by 151% and 31%, respectively since 1750, and the concentrations continue to increase [2]. Slightly more than half of the current methane emissions are anthropogenic and 5%–20% of the methane emission from anthropogenic source is from waste landfill [3]. Therefore, reducing the GHG emission from landfills contributes to stabilize GHG concentrations in the atmosphere that would prevent serious anthropogenic interfer-ences with the climate [4,5].

To reduce the GHG emission, LFG collection and treatment is compulsory. For example, EU requires the Member States that LFG should be collected from all landfills receiving biodegradable waste and LFG must be treated and used [6]. The United States requires that LFG and liquid effluents should be continuously collected and treated for 30 years after closure of the landfill [7].

The generation of municipal solid waste (MSW) in China reached 1.521×10⁸ tons in 2007, which was nearly 4.9 times of that in 1980, and showed an annual increase at rate of about 6%. The disposal rate of MSW was 62.0%, 80.9% of which was transported into sanitary landfills [8]. As landfill is the most important disposal method of MSW, the GHG generated from landfills is increasing with the growth of MSW generation and the development of disposal rate.

In order to mitigate the possible impact on global warming, collecting and flaring LFG can obviously reduce the GHG emission, because the global warming potential (GWP) of CO₂ (GWP = 1) is far lower than that of CH₄ (GWP = 23). However, CH₄ is also an important gas fuel with high calorific value and is the main component of natural gas. With a calorific value varying between 15 and 20 MJ/m³ with the CH₄ component in the range of 45% to 60%, LFG can also be regarded as one kind of renewable bioenergy resource.

The main LFG utilization methods include direct heating, electricity generation, purification for pipeline...
quality gas, vehicle fuel, or fuel cell, depending on the CH$_4$ concentration of final products primarily [9–11]. Electricity generation and direct heating are the most adopted options in LFG utilization projects of the existing landfills in the world [12]. Though fuel cell is promising electricity generating technology in the future for its high efficiency, its high cost makes it currently infeasible in economy [13]. Purification for automobile fuel can produce alternative fuel for transportation vehicles, which can reduce the consumption for non-renewable fossil energy resources and GHG emission. As oil price increases rapidly, LFG purification for automobile fuel is of high additional value.

The purification technologies of LFG include physical absorption, chemical absorption, adsorption separation, and membrane separation [14]. Pressure swing adsorption (PSA), one kind of adsorption separation technology, was used in our research on LFG purification. Compared with the other technologies, PSA has three advantages: 1) the adsorbent can be used for a long period, which is important for continuous industrial production; 2) it operates at normal temperature and relatively low pressure, which means low energy consumption; 3) the application in chemical engineering industry has been proven successful [15].

The paper reports the research progress from laboratory experiment to demonstration project. The laboratory experiment determined adsorbents and technological conditions of PSA. The pilot-scale test was completed at Yulongkeng Landfill and Xiaping Sanitary Landfill in Shenzhen, China, in which the relationship among operation conditions, the CH$_4$ concentration in product gas, and the recovery rate of CH$_4$ were studied. The demonstration project was located at Xiaping Sanitary Landfill, in which the LFG generation is in peak period. The project was done in 2006, and the treatment capacity was 500 m$^3$ LFG/h.

## 2 Laboratory research

### 2.1 Selection of adsorbents

The LFG purification aims to separate CH$_4$ from a mixture of CO$_2$, N$_2$, and O$_2$, and the removal of CO$_2$ is the first step in the study. The common adsorbents used in chemical engineering industry for gas purification are zeolite molecular sieve (ZMS) and carbon molecular sieve (CMS). CMS, as an important adsorbent, appeared in the 1970s [16]. In this study, 3A, 4A, 5A and 13X ZMS were compared with M-type and A-type CMS. The saturated adsorption capacities and the PSA separation effect of different adsorbents were tested. The influence of various operation conditions, including air pressure and CH$_4$ concentration of feed gas, adsorption time and amount, and energy consumption were investigated.

As shown in Fig. 1, with the increase of flow rate and inlet pressure, the breakthrough time decreased gradually, and the penetration curve became steep. In the range of low flowrate, the adsorption of CO$_2$ increased much slower along with the increase of inlet pressure, which indicated that the controlling step was external diffusion. With the increase of intake flowrate, the influences of the external diffusion decreased while internal diffusion gradually became the controlling step in the mass transfer process of CO$_2$ between solid phase and gas phase. When the intake flowrate was excessively high, the CO$_2$ adsorption decreased. The relationship between saturated adsorption and inlet pressure of different adsorbents are shown in Fig. 2. At the same saturated adsorption, the pressure of 3A, 4A, and 5A ZMS was much higher than that of the other three at the same flowrate, and the adsorption capacities of 3A, 4A, and 5A ZMS were lower than those of M-type and A-type CMS.

Considering saturated adsorption, flowrate, and inlet pressure, A-type CMS was selected as the adsorbent for PSA experiment with the optimal flowrate at 267 mL/min. In the range of 200 mL/min to 300 mL/min, the mass transfer zone, breakthrough time, inlet pressure and the moving speed of the mass transfer zone were about 2 m, 0.5 min, 2 kg/cm$^2$, and 2 m/min, respectively. The experiment using pure CH$_4$ at the same operation condition showed that the moving speed of the mass transfer zone was about 3 m/min, and the mass transfer zone was only about 12 cm, which was much shorter than that of CO$_2$. These significant differences made it possible to separate CH$_4$ from CO$_2$ during the PSA process.

### 2.2 PSA experiments

The PSA penetration curves of the selected A-type CMS in the PSA experiment are shown in Fig. 3. When the inlet flowrate was below 1 m$^3$/h, the penetration time increased significantly with the increasing flowrate. At a constant flowrate, with the increase of the inlet pressure, the CO$_2$ adsorption increased, and the penetration time was prolonged. Due to the increase of gas density, the treatment capacity of feed gas increased gradually at high pressure. At the same time, the recovery rate of CH$_4$ decreased significantly with the increase in inlet pressure. Therefore, proper pressure was important for the PSA adsorption, and the optimal pressure was determined to be around 2.7 atm.

The separation efficiency of the mixture of CH$_4$ and CO$_2$ under various conditions, including temperature, pressure, flowrate, and concentration ratio, was analyzed, in order to optimize the operation parameters for the pilot-scale test. The experimental results are shown in Fig. 4, and the equipment is shown in Fig. 5.

As shown in Fig. 4, when the CH$_4$ concentration in feed gas was low, the recovery rate was high. When the CH$_4$ concentration was above 40%, the recovery rate was insensitive to the concentration of feed gas, which was between 60% and 70%. There was an optimal adsorption...