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Landfill gas pretreatment for fuel cell applications

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Abstract

The US Environmental Protection Agency (EPA) has proposed regulations [1] to control air emissions from municipal solid waste landfills. If these regulations are adopted, they would require waste methane mitigation in order to prevent emission into the atmosphere and reduce the effect on global warming. One potential use of the waste methane is in a device which produces energy, the fuel cell. This device would reduce air emissions affecting global warming, acid rain, and other health and environmental issues. By producing useable energy, it would also reduce our dependency on foreign oil. This paper discusses the US EPA program underway at International Fuel Cells Corporation to demonstrate landfill methane control, and the fuel cell energy recovery concept. In this program, two critical issues needed to be addressed: (i) a landfill gas cleanup method that would remove contaminants from the gas sufficient for fuel cell operation, and (ii) successful operation of a commercial fuel cell power plant on that lower-heating value waste methane gas.

Program description

International Fuel Cells Corporation (IFC) was awarded a contract by the US EPA to demonstrate methane control with energy recovery from landfill gas using a commercial 200-kW phosphoric acid fuel cell. IFC is conducting a three-phase program to show that this concept is economically and environmentally feasible in commercial operation. Work was initiated in January 1991 on Phase I that consisted primarily of a conceptual design, cost, and evaluation study. The Phase II work addressed the issue of contaminant removal from the gas. This consisted of construction and testing of a landfill gas cleanup pretreatment module designed to remove those contaminants. Phase III of this program, which is scheduled to begin in October 1993, has as its goal the demonstration of methane control and the fuel cell energy recovery concept at Penrose Station, an existing landfill gas-to-energy facility owned by Pacific Energy Corporation in Sun Valley, CA.

Methane mitigation and the fuel cell advantage

There are several methods of reducing the quantity of methane emissions from landfills. These include: flaring; combustion combined with heat recovery; conversion to electricity; conversion to pipeline gas, and conversion to vehicular fuel.

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Of these methods, flaring is the least desirable since it converts the methane into carbon dioxide (CO_2), another greenhouse gas, and it results in no useful by-products such as heat or electricity. While either combusting the methane with heat recovery or converting it to electricity also produces CO_2, there is a total CO_2-emission reduction that is realized. The fuel cell, itself, emits less CO_2 per kW than any other heat recovery or electrical generating equipment. In addition, by producing electricity this way at a landfill, less electricity needs to be generated by a central utility station and therefore less CO_2 is emitted from that utility.

The fuel cell method of converting the methane to electricity offers a number of other advantages:

(i) as shown in Fig. 1, the fuel cell emits very few pollutants compared with other electric generators;

(ii) it produces electricity at 40% efficiency, based on the lower heating value, and

(iii) the fuel cell is factory constructed and truck transportable; therefore, as a landfill age increases and it is no longer economical to utilize the methane, the fuel cell can be easily moved to a new site.

Commercial 200-kW landfill gas fuel cell

The landfill gas-to-energy concept would incorporate a 200-kW commercial phosphoric acid fuel cell. The basis for the landfill gas fuel cell would be the PC25 natural gas fuel cell power plant commercially produced by ONSI Corporation [2], an IFC subsidiary.

This unit is a packaged, truck-transportable, fuel cell power plant which has been in natural gas commercial service since early 1992. Today, power plants operate for 26 utility customers in 11 countries on 3 continents. These power plants have accumulated over 100 000 h of operation in commercial service, with an overall availability of 92%. They produce electricity at 40% efficiency, based on the lower heating value, and exhibit an overall efficiency of 85% when fuel cell waste heat is utilized in cogeneration applications. Their air emissions are lower than the background air quality in many US cities. These measured characteristics, when incorporated in the landfill gas fuel

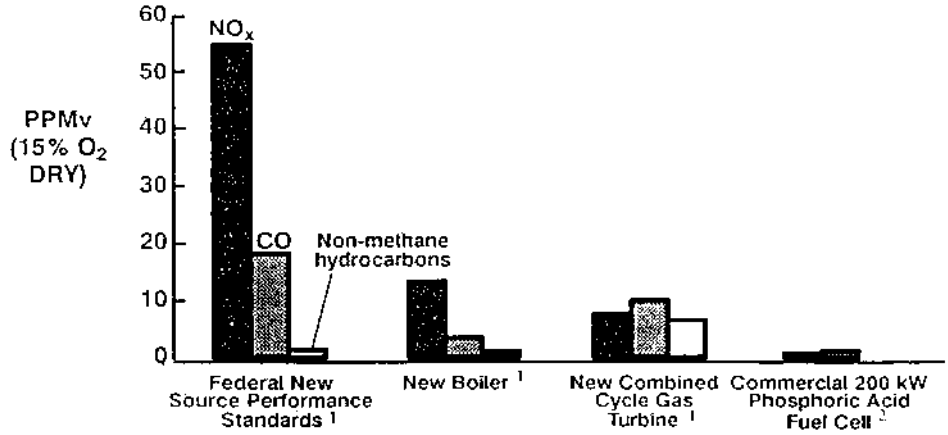

Fig. 1. Power plant emissions comparison (natural gas). 1. From Staff Recommendations for Generic Power Plant Emissions Factors, California Energy Commission, Aug. 1989. 2. Source: ONSI Corporation.

cell concept, would verify the estimated emissions benefits generated in the Phase I study.

Landfill gas availability and characterization

In Phase I of the contract, municipal solid waste landfills in the USA were evaluated. From this evaluation, the potential power generation market available for fuel cell energy recovery was estimated to be 4370 MW. The evaluation also identified a market niche segment, based on potential power rating, for a 200-kW fuel cell power plant module. This segment contained 1700 sites with a combined potential power rating of 1010 MW.

The assessment concluded that these sites are ideally suited for fuel cell operation. The commercial 200-kW fuel cell can provide a generating capacity tailored to the site because of its modular nature. Sites in this range could also be served by competing options, such as a gas turbine, which exhibit poorer emission characteristics (Fig. 1).

The Phase I study also characterized landfill gas contaminants. Contaminant levels for one site (i.e., the Penrose Landfill, located in Sun Valley, CA) are shown in Table 1. (The data in this Table are based on a number of years of measurements taken at the Penrose site by a variety of methods; the values shown are 'worst case' scenarios, i.e., 'high values'.) This characterization, reported in the Phase I Final Report [5], was used to design the gas pretreatment module, built and tested in Phase II.

Gas pretreatment module

One essential element of Phase II was the construction and testing of a gas cleanup system at the Penrose Landfill site. Landfill gases consist primarily of CO_2, CH_4, O_2, plus trace amounts of sulfides, organic halides, and non-methane hydrocarbons. The specific contaminants in the landfill gas of concern to the fuel cell are sulfur and halides. Both of these ingredients can 'poison' and therefore reduce the life of the power plant's fuel processor. The fuel processor is the unit which converts CH_4 in the gas stream into H_2 and CO_2 over a catalyst bed. The catalyst in this bed can react with the halides and sulfides and lose its activity.

Process

The system designed to remove fuel cell contaminants is shown in Fig. 2. Hydrogen sulfide is first removed by adsorption on a packed bed. Several materials including zinc oxide, activated carbon, and carbon impregnated with various compounds to increase sulfur capacity can be used for this purpose. This bed is not regenerable on site, but must be removed to another site if regeneration is desired.

Two-stage, low-temperature condensation followed by activated carbon adsorption are included in the process steps used to remove the heavy and chlorinated hydrocarbons. The first stage condenser is designed to operate at slightly above the freezing point of water followed by another condenser which is designed to operate at below 0 °C. To prevent freezing of water in the second condenser, a dehydration bed is located between the condensers. This bed is designed to reduce the dew point of the gas to

TABLE 1

Raw landfill gas contaminants and concentrations for Penrose test site

Landfill gas trace contaminants	Design raw gas concentration level (ppm, by volume)
Aromatics	
Benzene	2
Chlorobenzene	1
Ethylbenzene	13
Styrene	0.5
Toluene	35
Xylenes	22
Total	73.5
Halogenated hydrocarbons	
Cis-1,2-dichloroethene	5
Dichloroethane	3
Dichloroethene	3
Methylene chloride	12
Tetrachlorethylene	6
Trichloroethylene	70
Trichlorofluoroethane	0.6
Vinyl chloride	1.4
Total	101
Hydrocarbons	
Hexane	297
Isobutane	95
Isopentane	963
n-Pentane	198
Octane	81
Total	1634
Sulfides	
Dimethyl disulfide	0.02
Dimethyl sulfide	8
Ethyl mercaptan	5
Hydrogen sulfide	103
Methyl mercaptan	5
Total	121

significantly below the freezing point of water prior to its entering the second condenser. These dehydration beds are designed to be regenerated by heating a purge gas flowing through the bed. Two desiccant modules operate in parallel so that one is always operational while the other is being regenerated.

Dry landfill gas is then fed to the second stage refrigeration condenser. This condenser is operated to condense a mixture of hydrocarbons, aromatics, and halogenated hydrocarbons. Condensates are collected and transferred to the enclosed flare for thermal destruction. If the second stage condenser is ineffective in removing hydrocarbon contaminants the downstream carbon adsorption unit, whose temperature is controlled by the second stage condenser, is conservatively sized to remove all heavy hydrocarbon and halogen contaminant species. Two activated carbon beds operate in parallel so one is always operational when the other is being regenerated. Finally, the gas passes

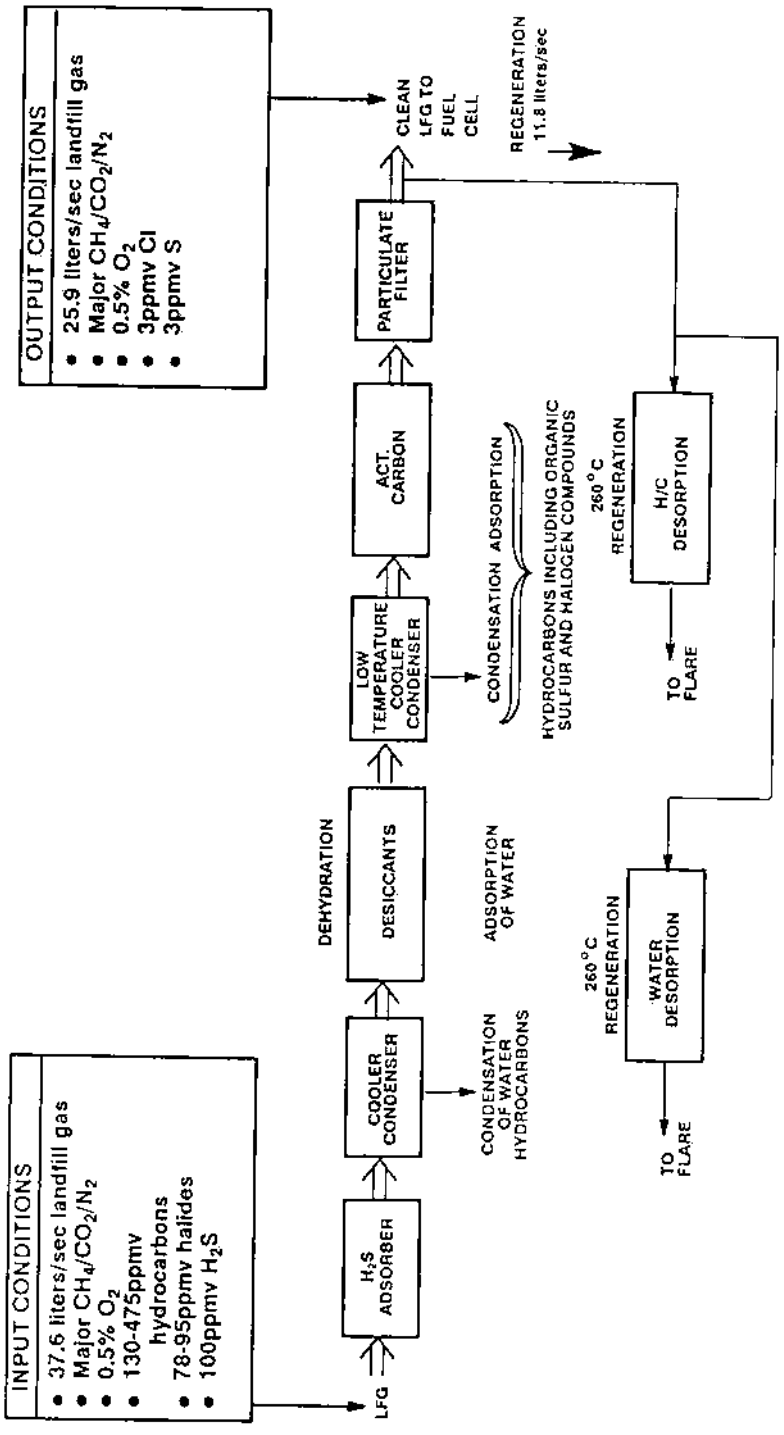

Fig. 2. Landfill gas pretreatment system.

through a particulate filter and is warmed indirectly by an ambient air-finned tube heat exchanger before being fed to the fuel cell unit. The process operating pressure is designed to remain steady at 2.43 kg/cm^2 with only nominal pressure loss across the equipment. Thus, the process can be controlled easily without any critical pressure control problems.

Initially, it was planned to remove H_2S downstream in the process using a bed of zinc oxide. This bed was to be located after the bed of activated carbon. However, initial field testing of this configuration showed that the H_2S was converted to carbonyl sulfide (COS) upstream of the zinc oxide bed. The zinc oxide bed will not effectively remove the generated COS. However, as shown in Table 2, other impurities present in the landfill gas, which are believed detrimental to the fuel cell, were effectively removed by the unit.

Laboratory testing to identify the cause of the COS formation showed that the COS is formed by the reaction of H_2S with the CO_2 present in the gas stream according to:

$$CO_2 + H_2S \longrightarrow COS + H_2O \tag{1}$$

The amount of COS formed may be predicted by use of the equilibrium equation:

$$K_{eq} = \frac{[COS][H_2O]}{[H_2S][CO_2]} \tag{2}$$

$$[COS] = \frac{K[CO_2][H_2S]}{[H_2O]} \tag{3}$$

It was found that a large fraction of H_2S in the inlet gas stream is converted to COS. This high conversion is believed due to the removal of product water by absorption on the desiccant material. This removal lowers the concentration of water in the gas phase, reducing the denominator in eqn. (3) and resulting in an increase in the concentration of COS.

Based on these laboratory results, the unit has been modified to remove H_2S upstream of the process, i.e., prior to the desiccant bed. The modified unit with upstream removal of H_2S is currently undergoing testing at the Penrose Site.

TABLE 2

Gas pretreatment unit test results (without upstream H_2S removal)[a]

Compounds	Raw Penrose landfill gas	At carbon bed exit
CH_4 (%)	43	Not measured
CO_2 (%)	39	Not measured
N_2 (%)	17	Not measured
O_2 (%)	1.2	Not measured
C_3–C_6 alkanes (ppm)	92	Not measured
H_2S (ppm)	83	None detected
COS (ppm)	None detected	76
Organic sulfur compounds (ppm)	9	None detected
Organic halogen compounds (ppm)	24	None detected
Other NMOCs (ppm)[b]	170	0.02

[a]Analysis by gas chromotography and mass spectrometry.
[b]Non-methane organic compounds.

Summary

Methane emissions from landfills and other sites are potential contributors to global warming. Conventional methods to mitigate these emissions, such as flaring, produce other greenhouse gases such as CO_2. By operating a fuel cell at a landfill site, CH_4 is destroyed while efficiently generating electric power and lowering CO_2 emissions. In order to operate a fuel cell on landfill gas, the gas must be 'cleaned up' or purified. A landfill gas cleanup pretreatment module was designed, constructed, and is undergoing testing at a landfill site. Initial results indicate that the unit removed all impurities detrimental to the fuel cell with the exception of H_2S. The unit was modified to remove this compound and testing is continuing. Based on successful completion of this testing, it is anticipated that operation of a commercial fuel cell power plant on the cleaned-up CH_4 gas will help demonstrate the economic and environmental feasibility of this concept.

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