

LANDFILL GAS PRETREATMENT FOR FUEL CELL APPLICATIONS

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Introduction

The U.S. Environmental Protection Agency (EPA) has proposed regulations¹ 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 U.S. 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: 1) a landfill gas cleanup method that would remove contaminants from the gas sufficient for fuel cell operation, and 2) 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 U.S. 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, California.

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
- Conversion to vehicular fuel

Of these methods, flaring is the least desirable since it converts the methane into carbon dioxide (CO₂), 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 CO₂, there is a total CO₂ emission reduction that is realized. The fuel cell, itself, emits less CO₂ per kilowatt-hour 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 CO₂ is emitted from that utility.

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

- As shown in Figure 1, the fuel cell produces very few pollutants compared to other electric generators;
- Its electrical efficiency, shown in Figure 2, is higher than that of conventional generators; and
- 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 Corporation², 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 hours of operation in commercial service, with an overall availability of 92 percent. They produce electricity at 40 percent efficiency, based on the lower heating value, and exhibit an overall efficiency of 85 percent when fuel cell waste heat is utilized in co-generation applications. Their air emissions are lower than the background air quality in many U.S. cities. These measured characteristics, when incorporated in the landfill gas fuel cell concept, would verify the estimated emissions benefits generated in the Phase I study.

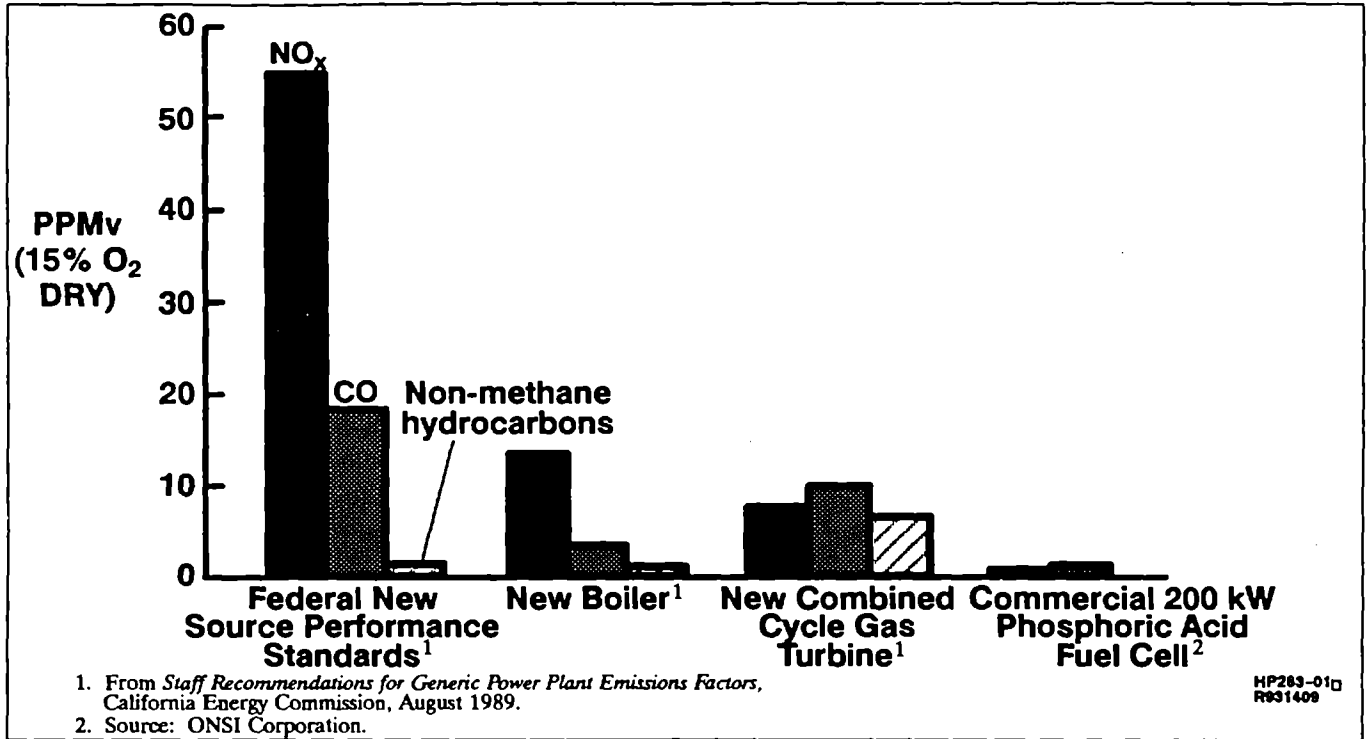


Figure 1. Power Plant Emissions Comparison (Natural Gas)

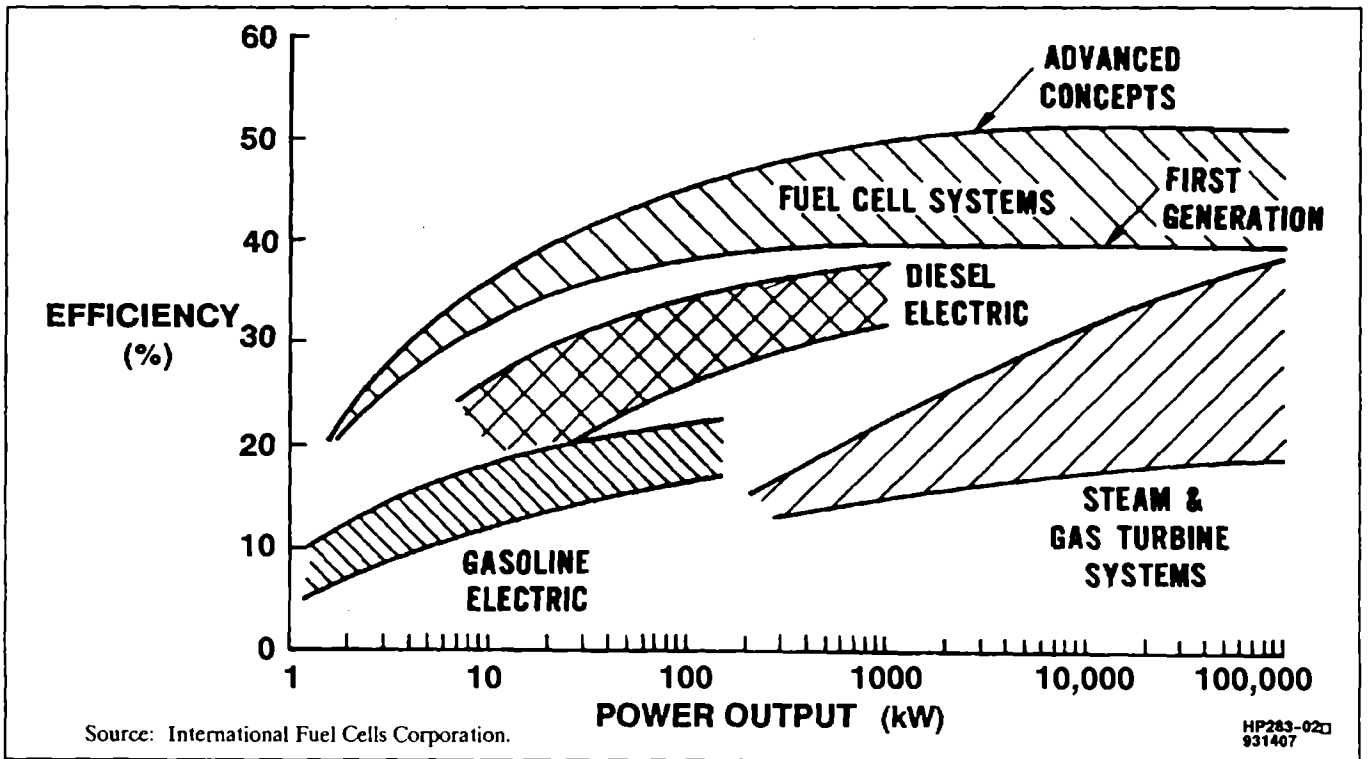


Figure 2. Power System Efficiency Comparison Based on Lower Heating Value

Landfill Gas Availability and Characterization

In Phase I of the contract, Municipal Solid Waste (MSW) landfills in the U.S. were evaluated. From this evaluation, the potential power generation market available for fuel cell energy recovery was estimated to be 4,370 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 1,700 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 (Figure 1).

The Phase I study also characterized landfill gas contaminants. Contaminant levels for one site (i.e., the Penrose Landfill, located in Sun Valley, California) are shown in Table I. (The data in the 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 Report³, was used to design the Gas Pretreatment Module, built and tested in Phase II.

Gas Pretreatment Module

One essential element of Phase II was construction and testing of a gas cleanup system at the Penrose Landfill site. Landfill gases consist primarily of CO₂, methane, and oxygen 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 methane in the gas stream into hydrogen and CO₂ over a catalyst bed. The catalyst in this bed can react with the halides and sulfides and lose its activity.

TABLE I	
Raw Landfill Gas Contaminants and Concentrations for Penrose Test Site	
Landfill Gas Trace Contaminants	Design Raw Gas Concentration Level (ppm - by volume)
<u>Aromatics</u>	
Benzene	2
Chlorobenzene	1
Ethylbenzene	13
Styrene	0.5
Toluene	35
Xylenes	22
Total	73.5
<u>Halogenated Hydrocarbons</u>	
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
<u>Hydrocarbons</u>	
Hexane	297
Isobutane	95
Isopentane	963
n-Pentane	198
Octane	81
Total	1634
<u>Sulfides</u>	
Dimethyl Disulfide	0.02
Dimethyl Sulfide	8
Ethyl Mercaptan	5
Hydrogen Sulfide	103
Methyl Mercaptan	5
Total	121

Process

The system designed to remove fuel cell contaminants is shown in Figure 3. 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.

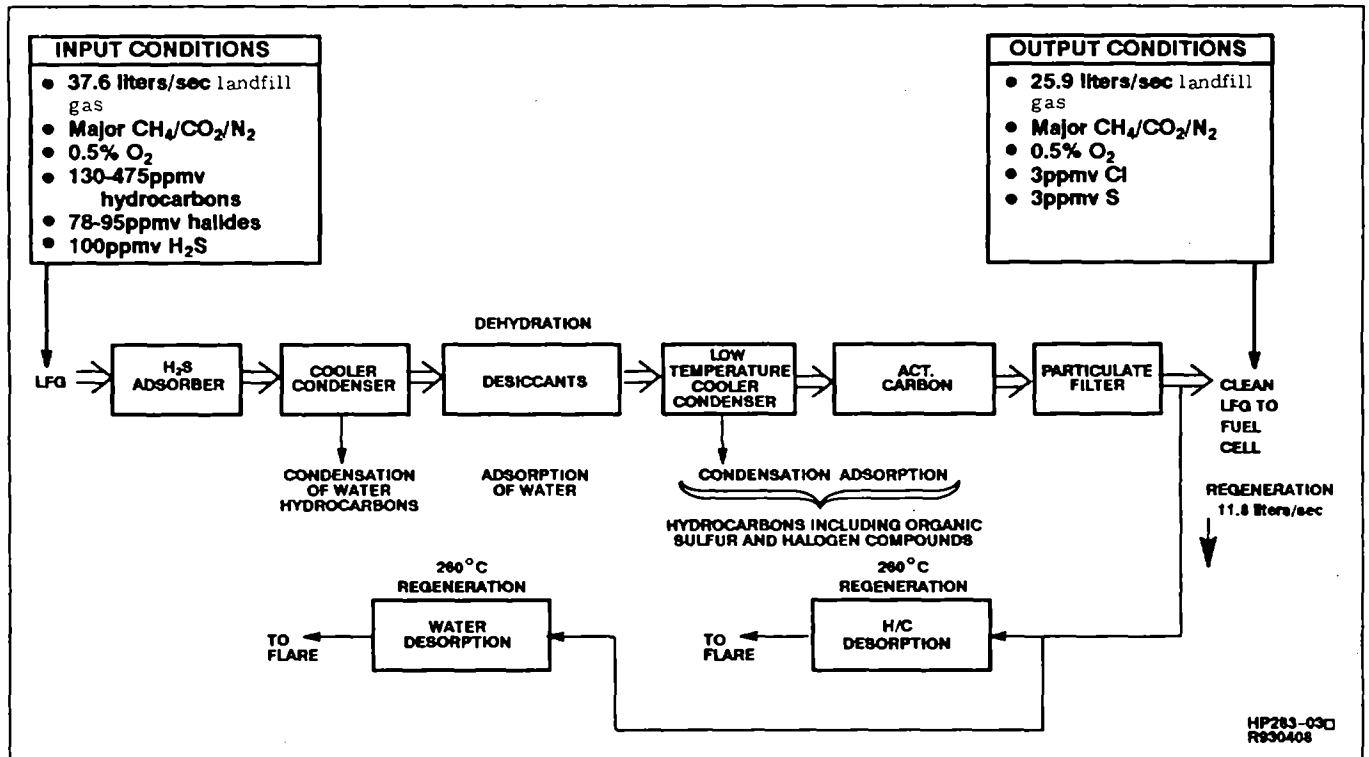


Figure 3. LFG Pretreatment System

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 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 contami-

nants, 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 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/cm² 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 hydrogen sulfide 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 hydrogen sulfide 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 II, 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 hydrogen sulfide with the CO₂ present in the gas stream according to



**Table II. Gas Pretreatment Unit Test Results*
(Without Upstream H₂S Removal)**

	<u>Raw Penrose LFG</u>	<u>At Carbon Bed Exit</u>
CH₄ (%)	43	Not Measured
CO₂ (%)	39	Not Measured
N₂ (%)	17	Not Measured
O₂ (%)	1.2	Not Measured
C₃ - C₆ Alkanes (ppm)	92	Not Measured
H₂S (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)	170	0.02

* Analysis by gas chromatography and mass spectrometry
** Nonmethane organic compounds

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

$$K_{eq} = \frac{[\text{COS}] [\text{H}_2\text{O}]}{[\text{H}_2\text{S}] [\text{CO}_2]} \quad (2)$$

$$[\text{COS}] = \frac{K [\text{CO}_2] [\text{H}_2\text{S}]}{[\text{H}_2\text{O}]} \quad (3)$$

It was found that a large fraction of the hydrogen sulfide 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 equation (3) and resulting in an increase in the concentration of COS.

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

Summary

In 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 carbon dioxide. By operating a fuel cell at a landfill site, methane is destroyed while efficiently generating electric power and lowering carbon dioxide emissions. In order to operate a fuel cell on landfill gas, the gas must be purified or "cleaned up." 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 hydrogen sulfide. 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 methane gas will help demonstrate the economic and environmental feasibility of this concept.

References:

1. U.S. Federal Register, May 30, 1991. Part III Environmental Protection Agency, 40 CFR Parts 51, 52 and 60: Standards of Performance for New Stationary Sources and Guidelines for Control of Existing Sources, Municipal Solid Waste Landfills, Proposed Rule, Guideline and Notice of Public Hearing. Washington, D.C.: United States Government Printing Office.
2. ONSI Corporation. 1993. The PC25 Fuel Cell Power Plant: Product Brochure. South Windsor, Connecticut.
3. Sandelli, G. J. January 1992. "Demonstration of Fuel Cells to Recover Energy From Landfill Gas, Phase I Final Report: Conceptual Study." EPA - 600-R-92-007 (NTIS PB92-137520).

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16. ABSTRACT The paper discusses the U.S. EPA's 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 are being addressed: (1) a landfill gas cleanup method that would remove contaminants from the gas sufficient for fuel cell operation, and (2) successful operation of a commercial fuel cell power plant on that lower-heating-value waste methane gas. (NOTE: The EPA has proposed regulations 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 that produces energy, a 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 U.S. dependency on foreign oil.)		
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