

# Catalytic Purification Process for the Upgrading of Landfill Gas to Fuel Cell Quality

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## Introduction

The main driving force for the utilization of landfill gas (LFG) is to avoid greenhouse gas emissions and to lower gas emissions with extremely high ozone depletion potential (ODP). The potential use of a complex matrix like landfill gas [1] (see Table 1) as feeding fuel for the so-called molten carbonate fuel cells (MCFC) imposes the need for new upgrading technologies in order to meet the much tougher feed gas specifications of this type of fuel cells in comparison to gas piston engines [2]. Due to the high number of impurities and potential catalyst poisons contained in LFG, the chosen purification process must be robust and able to tolerate peak concentrations of these impurities.

Table 1: Determination of the concentrations of major and some minor components present in raw LFG at two landfill sites.

Major Components	Landfill 1 [Vol%]	Landfill 2 [Vol%]
Methane	54	53
Carbon dioxide	35	24
Oxygen	0,89	0,78
Nitrogen	11	23
Minor Components	Landfill 1 [ $\mu\text{g}/\text{m}^3$ ]	Landfill 2 [ $\mu\text{g}/\text{m}^3$ ]
Sulfur-containing comp.	230.560	45.088
BTX	89.000	59.800
FCC	1.570	187
CC	600	542
Silicon-organic comp.	35.000	19.180

Figure 1(a and b) shows the damages caused by the utilization of LFG on engine parts and a reformer catalyst of an MCFC.



Fig. 1(a): Damages caused by acid corrosion and the deposition of micro-crystalline silica ( $\text{SiO}_2$ ) on a cylinder head of a gas engine after approx. 3 500 h of operation.  
(b) SEM image of  $\text{SiO}_2$ -deposition on the surface of a Ni-catalyst reformer after a few hours of operation.

Current state-of-the-art technologies, such as adsorption with active carbon [3], absorption with solvents [4] and chilling methods [5] fail to deliver a gas with the gas specifications of an FC. The following concept describes a new catalytic purification process for LFG conditioning, which may be supposed to be more competitive than state-of-the-art technologies and summarizes some lab-scale and field-test results.

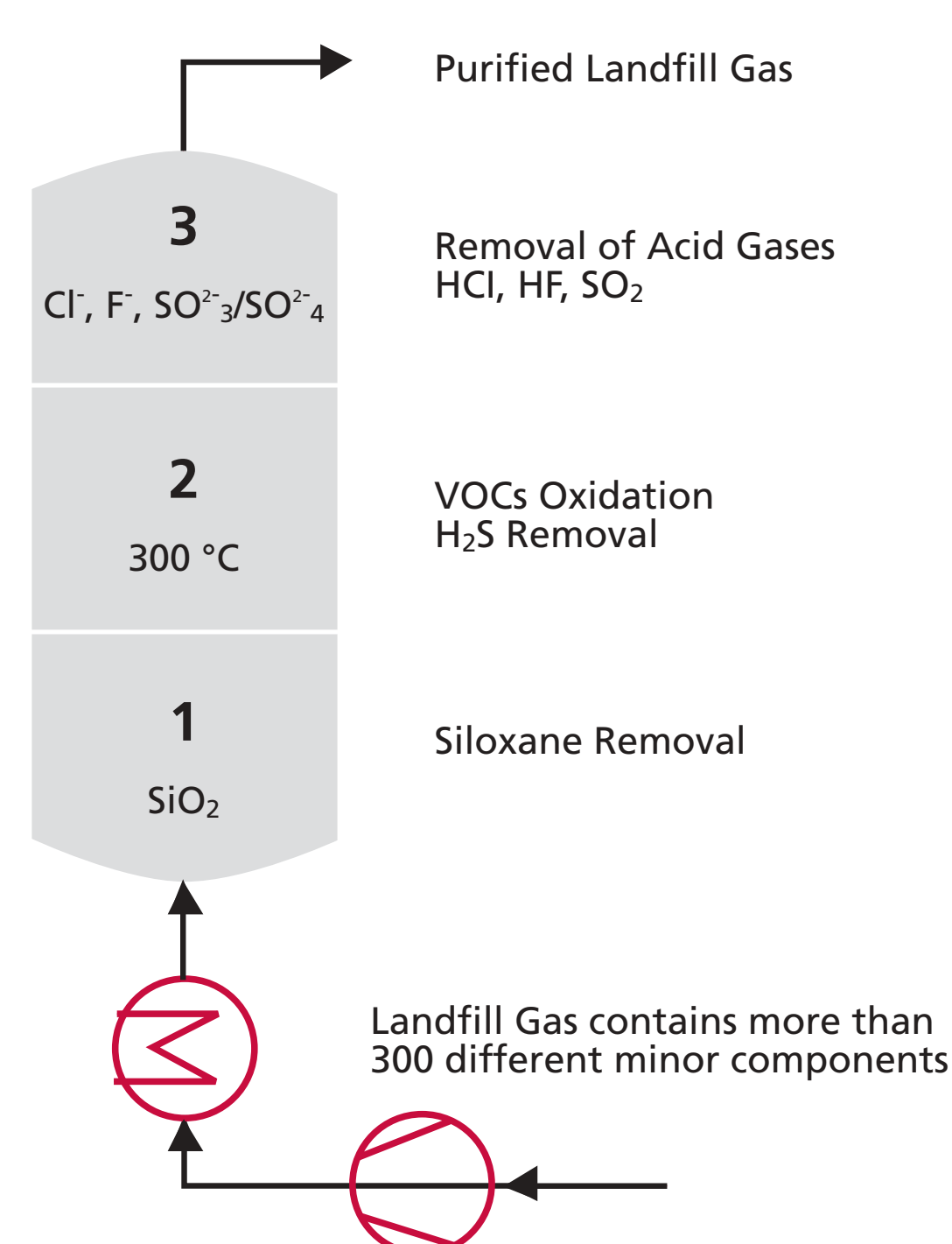


Fig. 2: Simplified sketch of the three-step catalytic process for LFG conditioning.

Figure 2 shows a simplified sketch of the three-step purification process. In the first step, the removal of siloxanes take place; in the second step the VOCs and sulfur-containing compounds are oxidized to ( $\text{CO}_2$ ,  $\text{SO}_2$ ,  $\text{HCl}$  and  $\text{HF}$ ) without compromising the methane content. In the last step, the acid gases are removed with alkalized materials, which is a state-of-the-art technology.

## Results

A screening of different materials carried out in order to identify a suitable and cheap material, which abates siloxanes selectively. The materials were tested with LFG that was additionally enriched with the siloxanes D4 (octamethylcyclotetra-siloxane) and L2 (Hexamethyldisiloxane) as model compounds to shorten the test times.

Figure 3 shows the conversion curves for the decomposition of D4 at 300 °C. It is clear that the activated alumina exhibits the highest conversion. The initial conversion rate is 100 % and decreases gradually due to the irreversible deactivation of the active centres of the activated alumina. For the case of L2 the deactivation was slower since D4 produces twice as much  $\text{SiO}_2$  per molecule. After 35 h the conversion rate for D4 dropped to approx. 9 % and 20 % for L2.

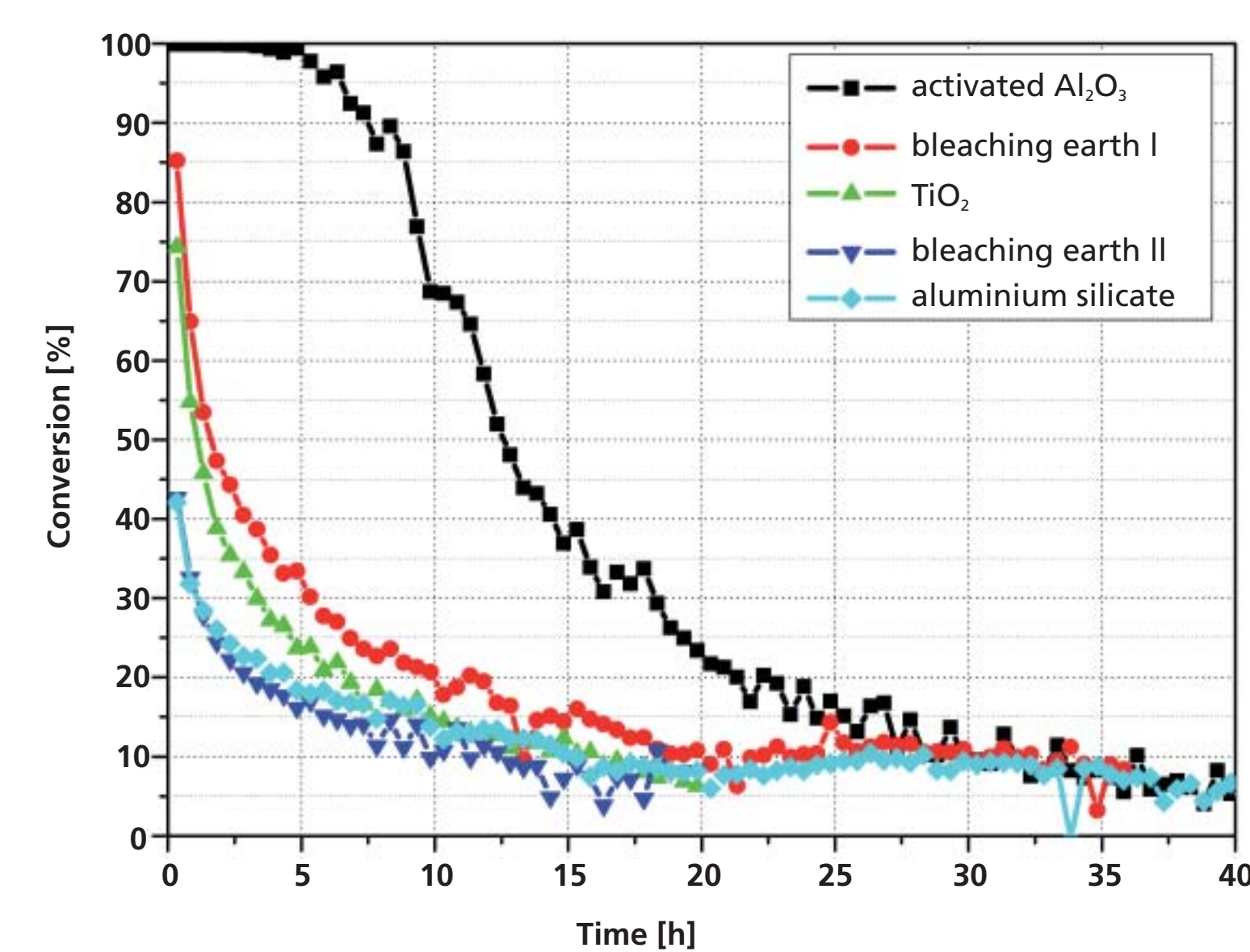


Fig. 3 Siloxane removal performance of various materials at 300 °C with an enriched LFG.

Figure 4 shows the conversion of aromatic and halogenated model compounds monitored with a GC-FID/ECD and a CI-MS. It is clear that the conversion lies around 100% and no deactivation through poisoning is observed. The fluctuations in conversion observed correlate to short-time declines of oxygen concentration in the feeding gas. The decrease in activity observed after nearly a month of operation is assigned to the breakthrough of siloxanes in the first reactor, which result in irreversible poisoning of the vanadium-based catalyst.

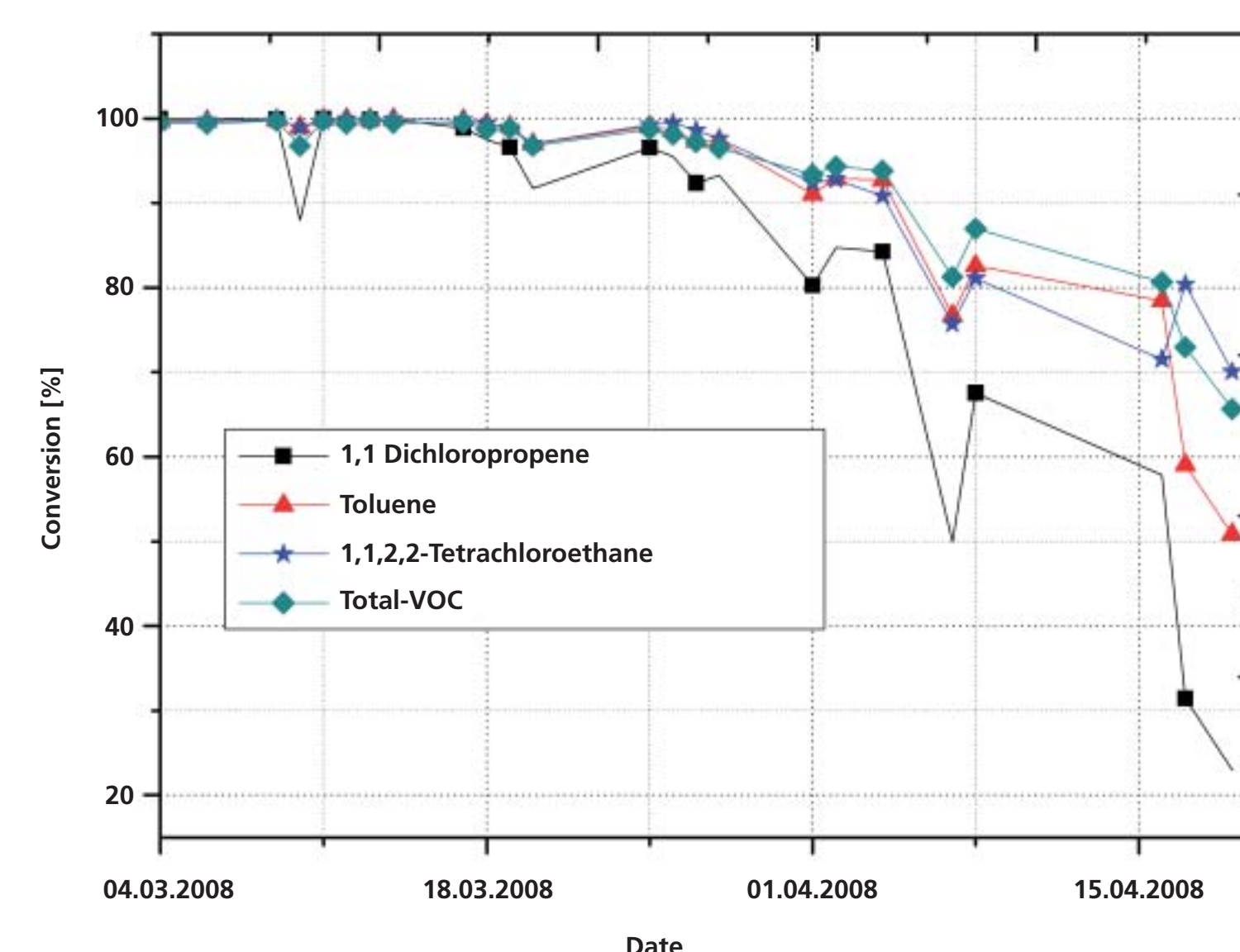


Fig. 4 Quantification of VOCs in raw and purified LFG by GC coupled with FID and EC detectors.

## Conclusions

- Identification of suitable catalysts for the removal of LFG impurities:
- $\text{V}_2\text{O}_5/\text{TiO}_2$ -catalyst exhibits good activity for the oxidation of both organic and inorganic model compounds.
- Commercial activated alumina shows selectivity towards siloxane removal and higher capacities than activated carbons.
- Identification of optimal process conditions.
- Relatively low-cost consumables with good performance.

## References

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