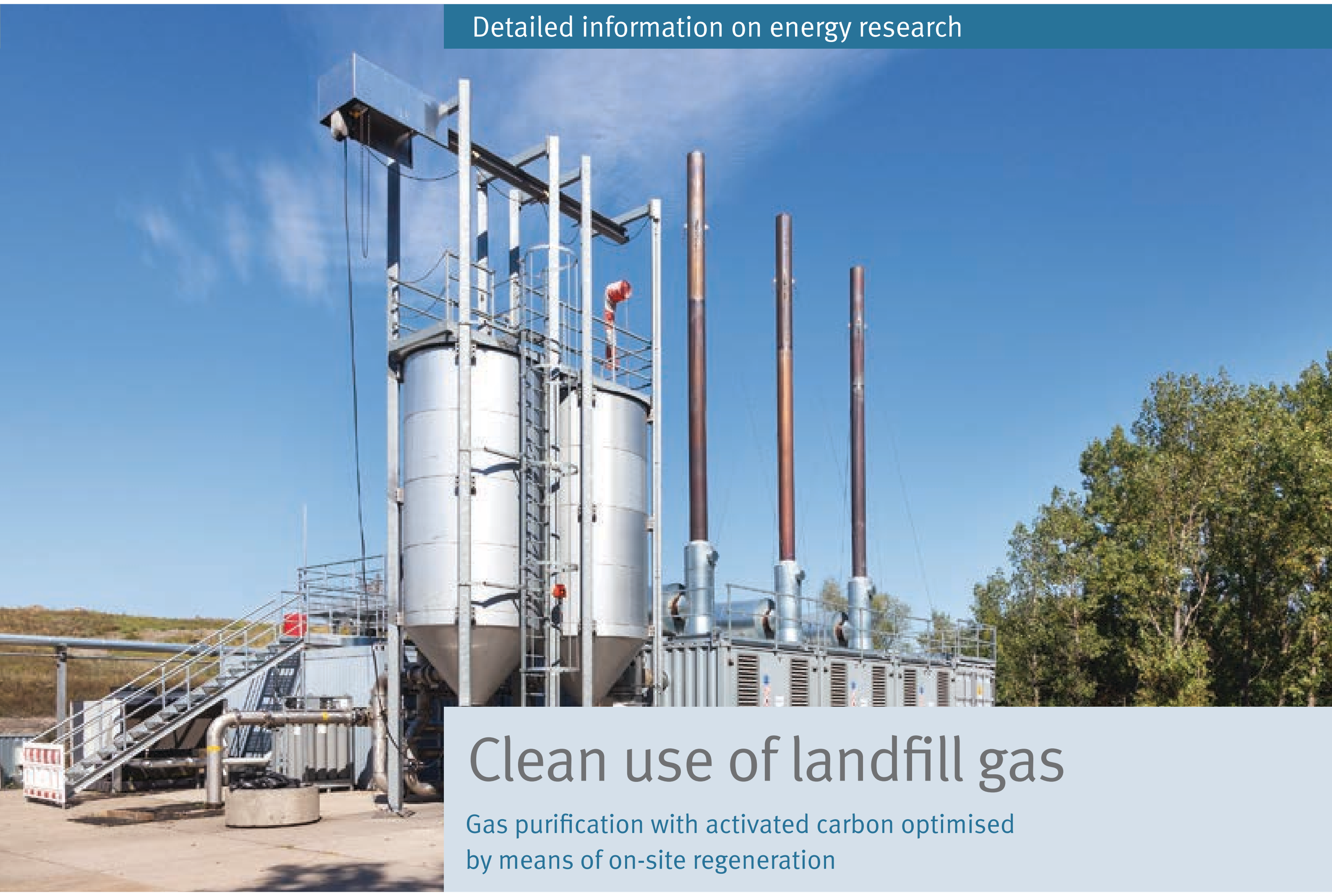

Clean use of landfill gas

Gas purification with activated carbon optimised by means of on-site regeneration

BINE Information Service

In the production of landfill gas, other gases are produced alongside the desired methane that damage motors and turbines. To protect these plant components, trace compounds of organic silicon contained in the gases are separated beforehand, e.g. by means of adsorption. The high consumption of the adsorbent, however, endangers the efficiency of the process. A research team has developed an on-site regeneration method for the adsorbent. This can increase the efficiency of the energy recovery of landfill gas.

Biogas is produced in landfill sites, sewage treatment plants and agricultural biogas facilities. It is produced through the biological decomposition of organic waste under exclusion of oxygen. Its primary constituents are carbon dioxide and up to 60 per cent methane. Owing to the high methane content, biogas is used for energy generation. Trace compounds of organic silicon contained in the gases, however, damage motors and turbines. In the combustion process within the engine compartment, these are converted into silicon dioxide owing to the prevailing high temperatures and deposited there. This leads to greater wear of the components and results in reduced operating hours. An upstream gas purification method is consequently employed. Various technical solutions exist, such as scrubbing, refrigeration drying, adsorption with activated carbon and molecular filtering. Adsorption with activated carbon is the most commonly used method.
A research project conducted by Siloxa Engineering and Fraunhofer UMSICHT investigated how the gas treatment process could be made economically and technically more efficient. Two method variants were tested in this regard, one involving activated carbon adsorption with subsequent regeneration and the other catalytic separation from activated aluminium oxide. Activated carbon absorbs not only the trace elements of organic silicon, but also other organic trace compounds pre-

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sent in abundance in landfill gas and non-hazardous for engine operation, such as hydrocarbons, terpenes and sulphur compounds. The result is poor utilisation of the adsorbent. For this reason, the activated carbon must be frequently exchanged. This high rate of consumption results in added costs, which to a certain extent endanger the economic utilisation of the landfill gas.
As a solution, the research group has successfully tested an on-site regeneration method for the activated carbon. The idea is to increase the service life of the activated carbon and consequently to reduce the number of exchanges by regenerating the adsorbent on site. The combination of thermo and vacuum desorption lends itself to this endeavour. In this process, the absorbed compounds are cast out by generating a vacuum with simultaneous temperature increase.
In the test plant, the desorbent was collected using an adsorber filled with activated carbon, termed a control filter. Disposal of the desorbent using a high-temperature gas burner is envisaged for the industrial implementation of the process.

Test plant at the landfill site

The researchers developed a test plant in which two methodological approaches can be investigated: The catalytic separation of trace compounds of organic silicon from activated aluminium oxide (cat-ox process) and adsorptive purification through activated carbon with subsequent regeneration (activated carbon process).
The research team worked with a small 0.6-Nm³/h test bench and a large 100-Nm³/h test plant (Fig. 1). The two of these were connected to one another and were supplied with crude gas via a partial flow from the main landfill gas line. The test plant was situated at the Vereinigte Ville landfill site.
The small test bench was used by the researches to perform several test cycles consecutively using small quantities of adsorbent.
Activated carbon and activated aluminium oxide were investigated in an isolated fixed-bed reactor in the 100-Nm³/h circuit. This involved close scrutiny of the adsorptive properties and the transformation of trace elements of organic silicon under practical conditions.

Catalytic separation

The cat-ox process as a concept involves the separation of organic silicon compounds from activated aluminium oxide at temperatures of 250 to 350 °C. The organic silicon compounds on the surface of this material react to form silicon dioxide. This solid reaction product is deposited, however, in the pores of the material. The aluminium oxide is consequently deactivated.
The team at Fraunhofer UMSICHT also tested other possible materials for catalytic separation under laboratory conditions in the preliminary stages. Aluminium oxide, however, proved to be the most suitable. Calcium oxide (CaO) and magnesium oxide (MgO) are ruled out as the carbon dioxide contained in the landfill gas reacts to form a carbonate layer on the materials, which reduces the reactivity. Titanium dioxide and vanadium pentoxide showed good reactivity over extended periods, though are ten to twenty times as expensive as aluminium oxide.
In the large test plant, the landfill gas is initially heated to max. 350 °C via a thermal oil heater. It is then fed into a reactor filled with activated aluminium oxide. Through reactive adsorption, the compounds of organic silicon on the aluminium oxide are converted to silicon dioxide. The purified landfill gas is cooled to approx. 50 °C in the heat exchanger and fed back into the main gas line of the landfill site.
The tests show that the process is essentially suitable for separating organic silicon compounds. Results to date lead researchers to conclude that the operating costs in this regard are roughly comparable to those of the adsorptive activated carbon procedure with on-site regeneration. Further technical considerations, however, need to be addressed before use in practice. These include for instance questions around how effective heat

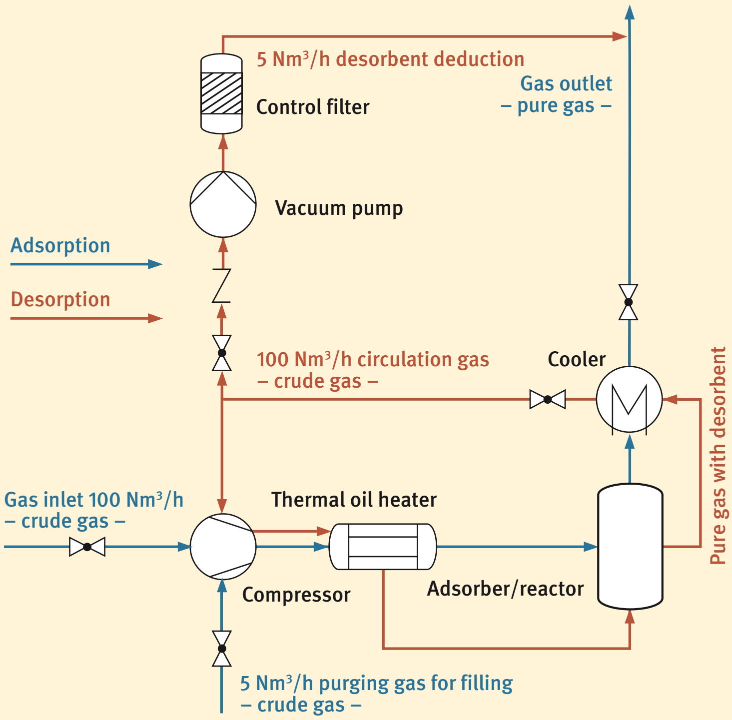

Fig. 1 *Diagram illustrates the test structure of the 100-Nm³/h plant at the Vereinigte Ville landfill site.*

Fig. 2 *Silicon dioxide deposits on the cylinder head of a gas motor after 3,500+ operating hours resulting from the combustion of organic silicon trace compounds in the engine compartment.*

Fig. 3 *The research team tested both catalytic separation on activated aluminium oxide (cat-ox process) and adsorptive purification using activated carbon with subsequent regeneration (activated carbon process).*

Fig. 4 *The two materials, aluminium oxide and activated carbon, were each examined by the research team in an isolated fixed-bed reactor with landfill gas.*

transfer to the reactor can be achieved and how dust deposition can be prevented in plant components behind the reactor.

Adsorptive purification with subsequent regeneration

The approach of the activated carbon procedure in the large test plant involves initially concentrating the landfill gas and heating it to max. 70 °C. The gas is then fed through an adsorber filled with activated carbon. Trace elements of organic silicon contained in the gas are adsorbed on the activated carbon. The purified gas is cooled before being fed back into the main line of the landfill site.

The desorption of the activated carbon occurs through a combination of thermal regeneration and vacuum desorption. In the process, a partial flow of the gas is heated to max. 160 °C and fed into the adsorber. A vacuum pump at the outlet of the adsorber ensures a vacuum of 700 mbar. The gas filled with desorbent is cooled to approx. 80 °C in a heat exchanger and fed through a control filter, which is an adsorber filled with activated carbon. The gas is then fed back for activated carbon adsorption.

The field tests have shown that the on-site regeneration of activated carbon is partially achievable in the method and constant residual loading is attained. As expected, however, the activated carbon exhibited very limited retention of polar organic compounds such as trimethylsilanol, meaning that additional purification must be considered for these for technical application. The proportion of trimethylsilanol in the total load of organic silicon compounds can be up to 50 per cent in the landfill gas. The research team recommended as a solution a process of pre-purification by means of seepage water washing. A system such as this has already been implemented by industry partner Siloxa in Ihlenberg in Mecklenburg-Western Pomerania, Germany and separates virtually all contained trimethylsilanol and other polar organic silicon compounds.

Recommendations for a gas purification plant

The research group developed a theoretical procedural concept for a gas purification plant with a throughput of 1,000 Nm^3/h: A process of seepage water washing is proposed as a pre-purification step. In the process, the landfill gas is cooled to 4 °C in order to enhance the absorptive capacity.

If the landfill gas has a temperature of 41 °C, a moisture saturation of 100 %, a pressure of 1,013 mbar and if an air temperature of 35 °C prevails, the energy requirement at a throughput of 1,000 m^3/h would be approx. 18 kWhel. Two alternately operated adsorbers are located downstream for adsorption and desorption. The desorbent is ultimately disposed of via a high-temperature gas burner.

By integrating an upstream gas purification process of this nature, modern gas motors can achieve a rate of efficiency of approx. 43 % in the conversion to electricity of landfill gas.

For this procedural concept, the research team also produced an operating costs analysis for an average silicon load present in the landfill gas. On the basis of this, the specific operating costs were estimated to be 1.06 euro cents per kWh_{el}. This is considered to be a significant saving over a conventional adsorption plant involving the exchange of activated carbon, the specific operating costs of which were calculated to be 1.65 euro cents per kWh_{el} according to the research team's data. Further tests are needed under boundary application-like conditions to develop the market viability for the combination of activated carbon procedure and seepage water washing.

Biomass exploitation potential

Landfill gas as a resource contributes 0.3 per cent to electricity production from renewable energies in Germany. It is grouped under biomass, which after wind energy currently accounts for the second largest proportion of power generation from renewable resources. This data has been calculated for 2013 by the Centre for Solar Energy and Hydrogen Research Baden-Württemberg on behalf of the German Federal Ministry for Economic Affairs and Energy.
In addition to landfill gas, biowaste, sewage gas and biogas, the biomass category also includes biogenic liquid and solid fuels. Biogas makes the largest contribution to energy generation from biomass. According to figures released by the German Biogas Association, biogas contributed around four per cent to gross electricity consumption in Germany in 2013. In total, 7,850 biogas plants generated approx. 24 terawatt-hours of electricity. For 2014, a sharp decline is expected in the construction of new biogas plants according to the study. Just 37 megawatts of additional capacity is forecast. This comes in response to an amendment to the German Renewable Energy Sources Act of 2014, which sees a reduction in funding.

Research projects for gas purification

The German Federal Ministry for Economic Affairs and Energy is funding research projects for biomass, in particular through the „Biomass Energy Utilisation“ support programme. Gas purification is currently addressed by the following two schemes, among others: The “Improved Biogas Upgrading to Biomethane by Ultrasonic–Stimulated CO_2 Desorption at Low Temperatures” (CaviCap) project is concerned with alternative methods for the separation of gas components. Under the project management of engineering and consulting firm GICON, a prototype is being developed to separate CO_2 from biogas with ultrasonic induced desorption. Prevalent amine scrubbing is found to be uneconomical in small-scale biogas plants owing to its temperature level. The project’s participants therefore are working on a new method to enrich methane through CO_2 separation in the existing temperature range.
In the “Biowaste Methane” joint research project under the management of Fraunhofer UMSICHT, the project team is analysing the potential of biomethane from biowastes. The results are aimed at plant operators, disposal companies and local politicians. The study is focussed around investigating the need to adapt existing gas purification technologies for the treatment of biogas from waste materials sourced from bio-waste containers. The research team illustrates cost and proceeds structures in this regard as a basis for plant configuration and optimal supply. Recommendations are also presented for dealing with plant-related, organisational and economic obstacles.

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