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Research paper

Comparison measurements of tar content in gasification systems between an online method and the *tar protocol*

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ABSTRACT

One of the major obstacles for the application of biomass gasification systems is the relatively high tar content of the producer gas that can inhibit its use in engines or turbines or in further processes like methanation or conversion to other secondary fuels or chemicals. Moreover, tars are difficult to define and also because of the different attempts to do so remains the determination of its content challenging. Nowadays conventional wet chemical and other standard methods for the determination of tars are very time consuming and do not allow continuous online monitoring of the gas quality. Furthermore, the execution by different users can affect the results. One approach to avoid these disadvantages is an automatic system that monitors the tar concentration in the producer gas online during the gasification process.

Such an automatic system was developed at IFK in the past with the goal of commercialization and moreover better comparability of the results from tar measuring. To show its accuracy and advantages over standard methods, comparative measurements with the standard method DIN CEN/TS 15439 at an electrically heated lab scale gasifier at different settings have been performed at IFK over several hours. Then, the results of both measurement techniques were compared to each other. The results show a stable operation of the gasifier and therefore also the tar production and its fast detection with the online system. Furthermore, the comparison of the results from both measurement techniques shows good agreement. A satisfying validation of the online method could be conducted.

1. Introduction

The so-called tars in the producer gas are one, if not the major problem for the application of gasification systems. Even if no general and comprehensive survey is available, the technical and economical problems related to tars have caused a cancellation of several investments in the past. Tars consist of various organic compounds. There exist different definitions of tars and originally they are defined as condensed hydrocarbons only. Nevertheless, in the field of biomass gasification, tars are commonly specified as a complex mixture of condensable hydrocarbons, which range from single to multi-ring aromatic compounds along with other, oxygen-containing and complex polycyclic aromatic hydrocarbons, in gaseous or condensed form [1–3]. Because of the different definitions in literature and fields of research, a detailed definition of tars for this work is given in chapter 4 (Tar definition). All the tar forming hydrocarbons are undesirable in gasification producer gases because of the various problems associated with their polymerization, condensation (tar formation out of gaseous substances) and following deposition [2–4]. They cause problems in the downstream process equipment like engines and turbines used in applications of the producer gas or in catalysts in gas cleaning and upgrading equipment [1,5,6]. The determination of their content in the producer gas and its online monitoring over time for process control is, therefore, of major importance in research and industry.

The detection of tars appears to be a complex undertaking due to the different definitions and the large variation in the tar properties resulting from different designs and operational parameters of gasification systems. Examples of different tar species distribution are given in e.g. Refs. [3,7–9]. The conditions under which tars are created and can be detected or sampled depend a lot on operational parameters of the gasification plant. The peripheral equipment of the gasifier system, such as heated gas pipes, condensers and particle filters, as well as the experience of the user can also influence the results of tar measurements [10–12]. This is why

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standardized tar measuring methods are required for comparability.

Through the collaboration of different international experts, a first technical guideline for sampling and analysis of tars for biomass gasification, the "Tar Protocol" [11,13,14] could be developed and released under the framework of the European Committee for Standardization (CEN). The test method is based on the principle of cold trapping in impinger trains with a solvent and subsequent laboratory analysis. Other methods based on cold trappings on solid adsorbents have also been developed, e.g. the SPA method [15], but all these methods are time consuming - with sampling times in the range of one to a few minutes and up to 1 h - and require specific knowledge and instrumentation. Therefore, their analysis is expensive and especially does not allow online monitoring of the gas quality. The sample preparation with subsequent analysis is in a time frame in the order of minimum 1 h up to one day, if it is done immediately. This is one reason why tar is not controlled on a routine basis and therefore remains problematic until today.

To avoid the drawbacks of those offline tar measurements, different research groups across the world are working on online tar measurement methods today. A brief overview with some advantages and disadvantages of those techniques is given already in Ref. [16]. The techniques will therefore only be summarized here. They are:

- The Laser-Inducted Fluorescence Measurement (LIF) by TU Berlin/TU Munich [17],
- The Molecular Beam Mass Spectrometry by NREL [18],
- The Time-Of-Flight Mass Spectrometry (TOF-MS) with laser ionization by the University of Rostock and KIT [19],
- The Online Gas Chromatography/Mass Spectrometry (GC/MS) with electron and laser ionization by TU Berlin [20],
- The Ion-Molecule Reactions Mass Spectrometry (IMR-MS) by CEA [21], and
- The Photo Ionization Detection (PID) by BTG Netherlands and KTH [22]

Furthermore, another online technique based on a FID was originally developed at the *Institute of Combustion and Power Plant Technology* (IFK) and a first prototype with the goal of commercialization was set-up in cooperation with the industrial partner *Ratfisch Analysensysteme GmbH* in the past [23,24]. But technical challenges and constraints inhibited the scientific and commercial success and forced both partners to a further development of the technique [16].

2. Objectives

The first part of the resumed work was to further develop the existing but defective functional prototype to a robust, accurate, low-cost and fast continuous online measurement device that meets the needs for industrial applications. This further developed measurement device meets the requirements for numerous applications like:

- Monitoring the general gasification process (tar peak tracing) and the efficiency of e.g. tar scrubber and catalytic tar reformer.
- Optimization of tar scrubber facilities and therefore waste water and solvent optimization.
- Downstream process monitoring to decrease maintenance of equipment or before gas engines to enlarge motor life.
- Fundamental tar research projects.

To achieve the above mentioned operation purposes, the specific advantages of the device are:

- High sampling rate (around 60–90 s per measurement cycle). Usable for semi-continuous tar concentration monitoring at high molecular measuring range.
- Light, robust and applicable in rough plant environment with easy handling for measuring at different measurement points.
- User friendly interface without additional IT-infrastructure.
- Easy maintenance and removal of tar and dirt deposits.

Commissioning of the further developed prototype and basic laboratory tests with defined tar species are published in Ref. [16]. The aim of the experimental scientific work presented within this paper is the comparison of the results of measurements between the online measurement device and the conventional wet-chemical measurement technique (The *Tar Protocol*, DIN CEN/TS 15439) over a time frame of several hours without interruption of the measuring procedure. The tests were done at an air blown fluidized bed gasifier to proof the reliability and accuracy of this further developed measurement device.

3. Analyzer technique and measurement principle

The measurement principle of the current, further developed device is generally based on the previous TA120-3 FID functional prototype which was originally developed at IFK in the 1990s and improved over time by *Ratfisch Analysensysteme GmbH*. The principle was published already in detail in the past [1,23–26]. Another detailed description of the measurement principle and all changes made in hard- and software compared to previous development versions of the device are also described in Ref. [16], including some results of laboratory based basic experiments. The scope of this section is therefore to give only a brief, but for the understanding of this work still sufficient overview of the method.

The general measurement principle is shown in Figs. 1 and 2. It is based on a differential measurement of the organically bound carbon in the sample gas of two sample loops with an equal volume. At the beginning of each measurement run, the sample loops are filled consecutively with sample gas (sampling phase, Fig. 1). In a second step, nitrogen flushes the sample gas consecutively to the FID (analyzing phase, Fig. 2, step 1: analysis of sample loop 1 (solid line), step 2: analysis of sample loop 2 (dashed line)). FID are well suited for the determination of the hydrocarbon content, since they have a high sensitivity and linearity. Further is stated by Holm, that the signal is with sufficient accuracy directly proportional to the

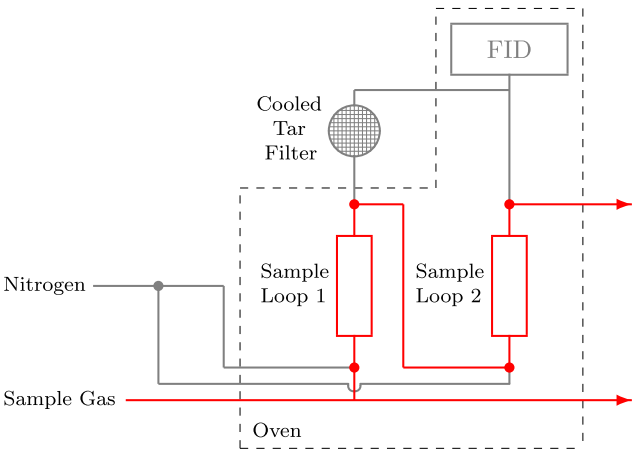

Fig. 1. Basic principle of the FID tar measurement system: Sampling phase.

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