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TAR MEASUREMENT STANDARD: A JOINT EFFORT FOR THE STANDARDISATION OF A METHOD FOR MEASUREMENT OF TARS AND PARTICULATES IN BIOMASS PRODUCER GASES

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B. Coda[1*], U. Zielke[2], M. Suomalainen[3], H.A.M. Knoef[4], J. Good[5], T. Liliedahl[6], C. Unger[7], L.Ventress[8], J.P.A. Neeft[9], H.W. v.d.Hoek[10] and J.H.A. Kiel[1]

[1]: Energy research Centre of the Netherlands, ECN Biomass, P.O. Box 1, 1755 ZG Petten, The Netherlands
[2]: Danish Technological Institute (DTI), Kongsvang Allé 29, DK-8000 Aarhus, Denmark
[3]: Technical Research Centre of Finland (VTT), P.O. Box 1601, 02044 VTT Espoo, Finland
[4]: BTG Biomass Technology Group, P.O. Box 217, 7500 AE Enschede, The Netherlands
[5]: Verenum, Langmauerstrasse 109, 8006 Zürich, Switzerland
[6]: Kungl Tekniska Högskolan (KTH), Teknikringen 42, 10044 Stockholm, Sweden
[7]: Fraunhofer UMSICHT, Osterfelder Strasse 3, 46047 Oberhausen, Germany
[8]: Casella Group, Stoke Orchard, GL52 4RZ Cheltenham, United Kingdom
[9]: SenterNovem, PO Box 8242, 3503 RE Utrecht, The Netherlands
[10]: NEN, Netherlands Standardisation Institute, Postbus 5059, AV 2600, Delft, The Netherlands

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[1]: Energy research Centre of the Netherlands, ECN Biomass, P.O. Box 1, 1755 ZG Petten, The Netherlands, phone: +31-224-564359, e-mail: coda@ecn.nl
[2]: Danish Technological Institute (DTI), Kongsvang Allé 29, DK-8000 Aarhus, Denmark
[3]: Technical Research Centre of Finland (VTT), P.O. Box 1601, 02044 VTT Espoo, Finland
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ABSTRACT: The Project "Tar Measurement Standard" focuses on the Standardisation of a Guideline for the measurement of organic contaminant (called 'tar'). This Guideline provides a set of procedures for the measurement of organic contaminants and particles in producer gases from biomass gasifiers. The procedures are designed to cover different air or oxygen blown gasifier types (updraft or downdraft/fixed bed or fluidised bed gasifiers), operating conditions (0 - 900°C and 0.6 - 60 bars), and concentration ranges (1 mg/m_n^3 to 300 g/m_n^3). Although several institutes have now used this Guideline (developed in a previous EU project), it does not have the status of an international standard yet. The overall objective of the project is to remove this obstacle by standardising the Guideline. The result will be a CEN Standard. As acceptance and use by others is considered to be essential, dissemination and internalisation of the Standard forms part of this project. This paper deals with the current status of the project, which started at the beginning of 2003.
Keywords: Tar, Measurement, Gasification, Standardisation

1 INTRODUCTION

So far no well-developed and standardised measurement method exists for tars in biomass-producer gases, and different sampling and analysis methods are currently being used. In a previous EU-project (ERK6-CT1999-20002), a ´Guideline´ [1, 2] for tar measurement and analysis was developed in order to remove this obstacle. The measurement principle is based on the discontinuous sampling of a gas stream containing particles and condensable organic compounds. The sampling train is shown schematically in Figure 1.
Several institutes are now using this Guideline allowing for a much more effective exchange and comparison of tar data. Activities to expand the use of the Guideline and transfer it into a European (CEN) standard method have been recently started in the framework of a new 3-year EU-project called "**Tar measurement Standard**".

This method has the strength of extensively covering the determination of a broad range of organic compounds that might occur in biomass-producer gas. One of the major achievements of the previous project was to tackle the issue of 'tar definition', which had been a topic of discussion in the scientific community for a long time. In the Guideline, two types of organic compounds can be determined that can generally be named "tar" [3]. These compounds are divided into two different groups - the gravimetric tar and a number of individual organic compounds (GC-detectable tars). **Gravimetric tar** is defined as the evaporation residue at conditions given and set in the Guideline (temperature, pressure, duration). **Individual organic compounds** are not defined, but those to be expected in biomass producer gases are listed in a compound list in the Guideline including chemical abstract service (CAS) registry numbers. This list is compiled from experimental data on the presence of compounds found in biomass producer gases from updraft, downdraft and fluidised bed gasification. Individual compounds are usually characterised by means of GC.

Individual compounds analysis and gravimetric analysis give complementary information one of each other; in principle they can be performed separately and the

user is free to choice either one or both methods, depending on the type of information needed, such as, for example, the determination of condensation point of tar based on dew point calculations [4]. As the Figure 2 shows, standard GC analysis is not generally able to cover organic compounds that are larger than coronene (i.e., approximately 7 rings), while in the gravimetric tar determination, larger compounds than 3 rings will be also determined. The upper limit on gravimetric tars is given by molecular sizes. Therefore, for some operating conditions, a certain overlap between the two types of analysis might occur [5]. The Guideline method foresees also the possibility to quantify the 'heavy' gravimetric (non-GC-detectable) tars by subtraction of the GC-detectable fraction of the total gravimetric tars.

The project activities are divided in two groups of tasks: 1) performing the necessary R&D tasks for the achievement of the standard; 2) standardisation at CEN level.

2 MAIN R&D ACTIVITIES

In order for the Guideline to achieve the status of a European Standard, it is mainly necessary to collect data on accuracy and reproducibility of the method. These are performed in 2 groups of activities:

1) Round Robin Test on GC and gravimetric analysis
2) Parallel measurement campaign

The first set of activities aims at gathering data on accuracy and reproducibility of analysis methods, while by means of the second activity the whole method (sampling+ analysis) is tested.

2.1 Round Robin Test (RRT)

A Round Robin test has been lately carried out in order to assess the accuracy and reproducibility of the analytical procedures.

The composition of tar depends on the gasification process. In principle tar can be divided into two groups:

– Low temperature tar, which is formed in the updraft gasifier and consists mostly of polar compounds
– High temperature tar, which is formed in downdraft and fluidised bed gasifiers and consists mostly of non-polar compounds.

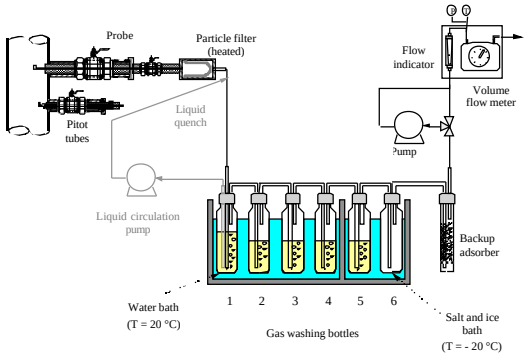

Figure 1 The sampling method *The Guideline sampling set-up: atmospheric and isokinetic sampling train for tar and particles with removable probe and pitot tubes for flow measurement.*

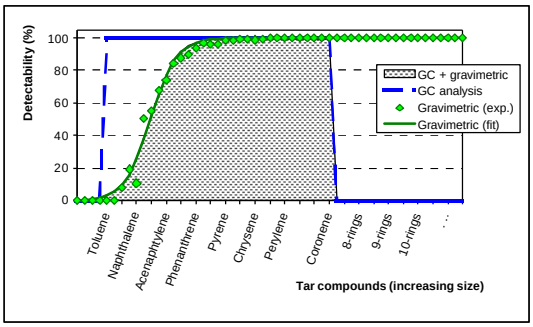

Figure 2: *Detection of tar compounds for GC and gravimetric determination.*

Round Robin of gas chromatographic method was divided into two phases, synthetic samples and real samples, in order to simplify the determination/tracing of possible problems, errors and uncertainty sources.

DTI has organised a Round Robin Test consisting of two rounds. In the first round synthetic samples with about 10 tar compounds representing updraft and downdraft or fluidised bed gasifiers were analysed gas chromatographically by the participating laboratories. On the basis of the results of round 01, real samples from updraft and fluidised bed gasifiers have been analysed gas chromatographically and gravimetric (residue of evaporation) in round 02. The samples represent both types of tar at typical concentration levels for raw

and clean gases in the Round Robin test and parallel measurement. For the GC analysis, a selection of compounds was made that could be most representative for the composition spectrum respectively of fluidised bed gasifier and updraft gasifier. Round Robin tests will be reported according to ISO 5952. From the results of the RRT, data on accuracy and reproducibility on the analysis of the selected organic compounds will be derived.

The participation of several institutes/laboratories was required in order to collect adequate amount of data. In addition to the project partners, also other institutes/laboratories active in tar measurement have been invited to participate. The RRT was performed in the period March 2003- April 2004. In March 2003 16 laboratories, which take an active part in gasification of biomass and tar measurement, were asked about their interest to participate in a Round Robin Test on tar analysis. Finally the first part of the RRT included results from 6 laboratories. In august 2003 received 9 laboratories l5 real tar samples, included one blind sample, in connection with round 02. Results from 6 laboratories were delivered. A third part concerning the gravimetric tars analysis has been conducted with 7 laboratories, and it has been closed in april.

The final results of the RRT exercise are still being evaluated. As an overall preliminary result, it can be stated that statistical results of GC analysis differed between FB gasifier samples and updraft gasifier samples. The scatter of the results was larger for updraft tars than for FB gasifier tars. The reason for that might be related to different degree of suitability of GC-FID technique for polar compounds than for apolar compounds. By use of GC-FID, which was used by most of participating laboratories, a satisfactory peak separation for the updraft gasifier tar spectrum is not achievable. For this type of compounds, the results of the round robin show that the GC-MS technique is a more suitable technique. The type of column in the GC is a critical issue. The use of the appropriate GC technique/column for the analysis of different type of compounds (polar vs. apolar) is currently evaluated.

2.2 Parallel measurement campaign

The parallel measurement campaign is a continuation of the RRT because information of the accuracy and reproducibility of the sampling method can be gathered. The parallel measurement campaign is performed in the year 2004. Extensive preparation of the parallel measurement campaign have been performed in 2003 and beginning 2004. The preparation concerned the choice of the gasifier, how many institutions should be involved, and what to compare and analyse. At the time of writing this paper, a first parallel measurement campaign has been successfully carried out in Denmark, at the commercial scale updraft gasifier of Harbøore. 4 institutions have participated in the parallel measurement campaign (DTI, UMSICHT, ECN and KTH). Results will be analysed in the following period.

The second parallel measurement campaign will be performed in fall 2004 at a fluidised bed gasifier. This planning will allow the major R&D activities of the project to be finished at the end of the second year of the project according to the initial project planning.

2.3 Other interesting R&D activities

The validity of the Guideline method for low tar concentrations is a topic that has not yet been completely investigated. The Guideline method is planned to cover the tar concentrations ranging from 1 mg/m^3_n to 300 g/m^3_n. This range was tested during the previous project except the range from 1 mg/m^3_n to 1000 mg/m^3_n. Therefore the validity of the Guideline method in the tar concentration range from 1 mg/m^3_n to 1000 mg/m^3_n should be still determined. This task is checked in RRT 2 with tars samples from a clean gas of an CFB gasifier, and it will be (partly) accomplished during the parallel measurements.

The Petersen column, developed by DTI, is a new type of sampling collector, is an alternative to the sampling train of the Guideline method [1]. Other institutes are currently testing the performance of the Petersen column, which largely decreases the handling of the solvent. DTI and VTT have tested the Petersen column with good correspondence to the results of the sampling train.

3 EUROPEAN STANDARDISATION

Under CEN, a 'Task Force" (TF) has been established in 2003 to work at the topic "Measurement of Organic Contaminants – 'tar'- in Biomass Producer Gases'. The TF (number 143) is opened to representatives of each country affiliated to CEN, and each representative (or each national committee, if there is more than 1 representative per country) has the right of vote. The Experts of the "Tar Measurement Standard project" act as national representatives in the TF.

CEN had issued a formal call for participation to all the member states of CEN through the corresponding national institutes. A dissemination action has been undertaken within the project to invite technical experts in the biomass (gasification) field and to let experts aware of this call for participation. So far, apart from the countries to which the project partners belong (Finland, The Netherlands, Denmark, Sweden, UK, Switzerland), Germany has nominated an official delegate who has joined the work of the TF from the very beginning. At this moment, Portugal has also nominated a national expert; representatives from France, Italy and Austria have also applied to their national standardisation institute to become national experts and their candidature is currently being evaluated at their respective national standardisation bodies. Other CEN-nation members are still invited to join the Task force.

There are different types of standard:

- TS (Technical Specification)
- TR (Technical Report)- not normative
- EN (European Standard) – highest degree of standard.

For each standard type a different procedure applies in terms of drafting process and in terms of voting/ administrative procedures at CEN and normative values at EU level.

Decision on which procedure to follow was taken in the first official meeting of the CEN TF 143 in June 2003. The Task Force decided to go for a TS type of standard, and to publish a separate TR report as appendix to the TS standard. The CEN TF 143 is going to discuss a third version of the Standard in Rome.

The CEN TF 143 has established official liaisons with other CEN Task Forces like the CEN Task Force of standardisation of solid Biofuels, (CEN BT 335); especially liaisons with institutions such as ASTM (the American Standardisation Institute), already established, and ISO, are important to initiate activities aiming at the later internationalisation of the Standard and acceptance as a measurement method at a broader level. Currently, at international level, there are no other similar activities on tar measurement like the current European initiative. In the US, a similar standardisation procedure might be initiated later, when the European standardisation activity will be about to deliver its final results [6].

4 CONCLUSIONS AND EXPECTED RESULTS

The primary result of the present project will be a standardised Guideline for tar measurements, aimed to reduce the technical and non-technical risks for implementation of biomass based CHP-systems in the future. With the aid of a good and standardised measurement technique, the performance of the gasifier, gas cleaning train and engine or turbine generator set can be monitored to learn about and suppress the technical risks. With this knowledge consensus about tolerances (maximum allowable concentrations of tars) for trouble free operation of gas engines, gas turbines and gas cleaning equipment can be defined. The result is that manufacturers of gas cleaning equipment and gas engines/turbines can give guarantees, which can improve the realisation of biomass CHP systems. The final CEN Standard is expected to be ready by the end of 2005. Comparison of tar measurement data between developers of gasification technology is therefore finally officially allowed.

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