

Tar in Biomass Producer Gas, the Energy research Centre of The Netherlands (ECN) Experience: An Enduring Challenge

Luc P. L. M. Rabou,* Robin W. R. Zwart, Berend J. Vreugdenhil, and Lex Bos

Energy research Centre of The Netherlands (ECN), Post Office Box 1, 1755 ZG Petten, The Netherlands

Received July 8, 2009. Revised Manuscript Received August 20, 2009

Biomass gasification at temperatures below 1300 °C yields producer gas with a range of heavy hydrocarbons. These compounds, collectively known as tar, cause fouling and emission problems in equipment using the producer gas. This paper gives an overview of the work performed at the Energy research Centre of The Netherlands (ECN) on tar measurement, tar prevention, tar cracking, and tar removal. Much of the work has been performed in cooperation with partner institutes and industry. Measurement techniques discussed are the tar guideline, solid-phase adsorption (SPA) method, and tar dew point analyzer. On the subject of tar prevention, the effects of operating conditions, fuel composition, and bed materials in fluidized-bed gasifiers are covered. Tar cracking results are presented for catalytic materials, high-temperature treatment, and the use of plasma. ECN research on tar removal involves among others the development of the water-based GASREIP system and the oil-based OLGA technique.

Introduction

Growing concern about the effect of CO₂ emissions and dwindling reserves of fossil fuels, especially of the most highly valued oil and natural gas, push the interest in renewable sources of energy. Among them, biomass is the only one that can deliver heat and power and act as carbon source for the production of transportation fuels and chemical products. Although the public debate has recently focused on competition between food and energy, it is likely there will be sufficient biomass available to meet both the world food demand and a substantial fraction of the world energy demand.¹

Both the concerns about competition with food production and the relatively high cost of biomass, when compared to fossil fuels, make it of paramount importance to aim at high conversion efficiency. For many purposes, gasification of biomass is an essential first step toward that goal. The second step is gas cleaning, designated by several people as the Achilles heel of the process.^{2,3} More in particular, that quote refers to tar removal from biomass producer gas.

The Energy research Centre of The Netherlands (ECN) has been involved in biomass gasification and gas cleaning for more than 15 years. Many of the successes and failures during that period have only been described in confidential reports or published in Dutch. Some of the results have been presented to a limited audience at conferences. The present paper gives an overview of tar research at ECN, with the emphasis on physical removal of tar from biomass producer gas, and describes negative as well as positive results. It may assist other researchers and operators of gasification installations from repeating errors that we made and in directing their efforts toward more promising concepts.

*To whom correspondence should be addressed. E-mail: rabou@ecn.nl.

(1) Smeets, E. M. W.; Faaij, A. P. C.; Lewandowski, I. M.; Turkenburg, W. C. *Prog. Energy Combust. Sci.* **2007**, *33*, 56–106.

(2) Reed, T. Personal communication, 1998, according to ref 13.

(3) Kiel, J. H. A. In Proceedings of the 12th European Biomass Conference, Amsterdam, The Netherlands, 2002; pp 465–468.

History of ECN Research on Biomass Gasification. Biomass gasification research at ECN started in the first half of the 1990s with the use of a 300 kW_{th} downdraft fixed-bed gasifier originally designed for coal gasification.^{4,5} In 1996, the 500 kW_{th} circulating fluidized-bed (CFB) biomass gasifier “Bivkin” was built.^{5,6} It has been used since for most of the work reported here. Part of the work has been performed using smaller lab-scale gasifiers: a 5 kW_{th} bubbling fluidized bed (BFB), a 25 kW_{th} slow-pyrolysis unit, and the 25 kW_{th} “Milena” gasifier, which can be used both in BFB mode and as an indirect (allothermal) gasifier.^{7,8} After a final 700 h endurance test in 2006, the Bivkin CFB gasifier has been decommissioned to make room for an 800 kW_{th} Milena indirect gasifier, which was officially taken into operation in September 2008.

Early gasification research at ECN concentrated on the pretreatment, feeding, and gasification properties of various biomass streams available and high-temperature gas cleaning.^{4,9} The original intent was to gather information for the construction of a 30 MW_e biomass integrated gasification combined cycle (BIGCC) installation.¹⁰ When plans for the

(4) Kiel, J. H. A.; Bos, A.; den Uil, H.; Plaum, J. M. In Proceedings of the 3rd International Conference on Combustion Technologies for a Clean Environment, Lisbon, Portugal, 1995; pp 1–33.

(5) Schenk, E. P.; van Doorn, J.; Kiel, J. H. A. In Proceedings of the Conference on Biomass Gasification and Pyrolysis, Stuttgart, Germany, 1997; pp 129–138.

(6) Schenk, E. P.; van Doorn, J.; Bodegom, M.; van der Drift, A. In Proceedings of the 10th European Conference and Technology Exhibition, Biomass for Energy and Industry, Wurzburg, Germany, 1998; pp 1757–1760.

(7) van der Drift, A.; Boerrigter, H.; van der Meijden, C. M. In Proceedings of the 14th European Biomass Conference, Paris, France, 2005; pp 628–630.

(8) van der Meijden, C. M.; van der Drift, A.; Vreugdenhil, B. J. In Proceedings of the 15th European Biomass Conference, Berlin, Germany, 2007; pp 868–872.

(9) Pierik, J. T. G.; Curvers, A. P. W. M. Logistics and pretreatment of biomass fuels for gasification and combustion. Report ECN-C-95-038, 1995.

(10) van Ree, R.; Oudhuis, A. B. J.; Faaij, A. P. C.; Curvers, A. P. W. M. Modelling of a biomass-integrated-gasifier/combined-cycle (BIG/CC) system with the flowsheet simulation programme Aspen Plus. Report ECN-C-95-041, 1995.

North-Holland BIGCC were canceled, attention was shifted to smaller installations for the combined production of heat and power using gas engines. This required gas cooling to lower temperatures, which prompted the need to deal with tar. Gas cleaning became even more demanding for applications, such as fuel cells and the production of substitute natural gas (SNG).

Tar problems can be circumvented by gasification at temperatures high enough to break down all hydrocarbons, from methane to complex tar compounds. That option can be used to produce syngas for the production of liquid transportation fuels by the Fischer–Tropsch process. It is also applied in the Buggenum coal gasification power plant, when part of the coal (10–30%) is substituted by biomass. However, the use of a high-temperature pressurized entrained flow (EF) gasifier exchanges tar problems for more elaborate biomass pretreatment and feeding problems. In that respect, biomass gasification research at ECN has come full circle. ECN is again studying biomass pretreatment and feeding. In cooperation with industry, ECN is developing the BO2 torrefaction process to improve the properties of the biomass.^{11,12} It involves a mild heat treatment that increases the heating value, reduces the hygroscopic nature, and improves the grindability. Torrefaction also produces a gas with some reactive organic compounds. Handling this gas again requires knowledge about the properties of tar.

Tar Definition and Measurement

Tar can be defined as “the organics, produced under thermal or partial-oxidation regimes (gasification) of any organic material. Tar is generally assumed to be largely aromatic”.¹³ However, different interpretations of this definition and the use of various measurement methods led to confusion. The members of the Gasification Task of the IEA Bioenergy Agreement, U.S. Department of Energy (DOE), and DGXVII of the European Commission agreed to develop a tar measurement protocol. Experts decided to

define tar as “hydrocarbons with molecular weight higher than benzene”.¹⁴ In subsequent papers, the slightly broader term “organic compounds” was used.^{15,16} ECN led the international consortium developing the tar measurement protocol.^{17–22} This eventually led to a CEN technical specification on the sampling and analysis of tar and particles in producer gas.²³

Tar compounds can be classified into primary, secondary, and tertiary tars. That classification refers to the process conditions in which the compounds are formed. Primary tars are formed by decomposition of the building blocks of biomass. Primary tar compounds contain oxygen in significant amounts. Secondary and tertiary tars are formed by destruction of primary tar compounds and recombination of fragments. In these processes, oxygen and some hydrogen are removed. Updraft gasifiers produce largely primary tar; downdraft gasifiers produce tertiary tar; and fluidized-bed gasifiers produce a mixture of secondary and tertiary tar compounds. Tar concentrations in biomass producer gas are in the order of 100 g/Nm³ for updraft gasifiers, 10 g/Nm³ for fluidized-bed gasifiers, and 1 g/Nm³ for downdraft gasifiers. Because most of the work at ECN involves the use of fluidized-bed gasifiers, our main experience is with secondary and tertiary tar compounds.

The distinction between secondary and tertiary compounds is not always clear, as can be seen from the overlap between lists of secondary and tertiary compounds given by Milne et al.¹³ In that respect, a classification based on the number of rings is less ambiguous. At ECN, we use either the ring classification or a scheme based on physical properties (see Table 1).²⁴

Although the tar guideline allows for accurate measurement of tar in biomass producer gases, ECN often uses the more convenient solid-phase adsorption (SPA) method.²⁵ The SPA method is useful for compounds from phenol to pyrene.²⁰ At ECN, the contributions of about 35 components are evaluated from the GC spectrum. Contributions of unidentified compounds are estimated by comparison to those from identified compounds and reported as “unknowns”. Because the contribution of “unknowns” can be quite substantial, we have decided more recently to subdivide the “unknowns” into five groups, which correspond approximately to the number of rings (≤ 2 , 3, 4, 5, and ≥ 6). Because the SPA result for toluene is unreliable, we exclude toluene from tar class 3. Instead, we measure the toluene concentration by gas chromatography (GC) and treat toluene as one of the gaseous components in the producer gas.

The concentrations of separate tar compounds have limited practical usefulness. For operators of installations, it is more

(11) Bergman, P. C. A.; Boersma, A.; Kiel, J. H. A. In Proceedings of the 15th European Biomass Conference, Berlin, Germany, 2007; pp 78–82.

(12) Kiel, J. H. A.; Verhoeff, F.; Gerhauser, H.; Meuleman, B. In Proceedings of the 16th European Biomass Conference, Valencia, Spain, 2008; pp 48–53.

(13) Milne, T. A.; Evans, R. J.; Abatzoglou, N. Biomass gasifier “tars”: Their nature, formation, and conversion. Report NREL/TP-570-25357, 1998.

(14) Maniatis, K.; Beenackers, A. A. C. M. *Biomass Bioenergy* 2000, 18, 1–4.

(15) Abatzoglou, N.; Barker, N.; Hasler, P.; Knoef, H. *Biomass Bioenergy* 2000, 18, 5–17.

(16) Simell, P.; Ståhlberg, P.; Kurkela, E.; Albrecht, J.; Deutsch, S.; Sjöstrom, K. *Biomass Bioenergy* 2000, 18, 19–38.

(17) Neeft, J. P. A.; Knoef, H. A. M.; Zielke, U.; Sjöstrom, K.; Hasler, P.; Simell, P. A.; Dorrington, M. A.; Greil, C. In Proceedings of the 1st World Conference on Biomass for Energy and Industry, Sevilla, Spain, 2000; pp 582–585.

(18) Neeft, J. P. A.; Knoef, H. A. M.; Buffinga, G. J.; Zielke, U.; Sjöstrom, K.; Brage, C.; Hasler, P.; Simell, P. A.; Suomalainen, M.; Dorrington, M. A.; Greil, C. In Proceedings of the 5th International Conference on Thermochemical Biomass Conversion, Telfs, Austria, 2001; pp 162–175.

(19) van Paasen, S. V. B.; Kiel, J. H. A.; Neeft, J. P. A.; Knoef, H. A. M.; Buffinga, G.-J.; Zielke, U.; Sjöstrom, K.; Brage, C.; Hasler, P.; Simell, P. A.; Suomalainen, M.; Dorrington, M. A.; Thomas, L. Guideline for sampling and analysis of tar and particles in biomass producer gases. Report ECN-C--02-090, 2002.

(20) Neeft, J. P. A.; van Paasen, S. V. B.; Knoef, H. A. M.; Buffinga, G.-J.; Zielke, U.; Sjöstrom, K.; Brage, C.; Hasler, P.; Simell, P. A.; Suomalainen, M.; Dorrington, M. A.; Thomas, L. In Proceedings of the 12th European Biomass Conference, Amsterdam, The Netherlands, 2002; pp 469–472.

(21) Coda, B.; Zielke, U.; Suomalainen, M.; Knoef, H. A. M.; Good, J.; Liliedahl, T.; Unger, C.; Ventress, L.; Neeft, J. P. A.; van de Hoek, H. W.; Kiel, J. H. A. In Proceedings of the 2nd World Biomass Conference, Rome, Italy, 2004; pp 793–796.

(22) van de Kamp, W.; de Wild, P.; Zielke, U.; Suomalainen, M.; Knoef, H.; Good, J.; Liliedahl, T.; Unger, C.; Whitehouse, M.; Neeft, J.; van de Hoek, H.; Kiel, J. In Proceedings of the 14th European Biomass Conference, Paris, France, 2005; pp 791–794.

(23) DD CEN/TS 15439:2006. Biomass gasification. Tar and particles in product gases. Sampling and analysis. ISBN 978 0 580 50444 0, 2007 (a draft version of the standard and background documents can be found at www.tarweb.net).

(24) Kiel, J. H. A.; van Paasen, S. V. B.; Neeft, J. P. A.; Devi, L.; Ptasiński, K. J.; Janssen, F. J. J. G.; Meijer, R.; Berends, R. H.; Temmink, H. M. G.; Brem, G.; Padban, N.; Brammer, E. A. Primary measures to reduce tar formation in fluidised-bed biomass gasifiers: Final report SDE project P1999-012. Report ECN-C--04-014, 2004.

(25) Brage, C.; Yu, Q.; Chen, G.; Sjöstrom, K. *Fuel* 1996, 76, 137–142.

Table 1. Classification of Tar Compounds

tar class	class name	properties
1	GC undetectable	very heavy, 7- and higher ring compounds
2	heterocyclic	cyclic hydrocarbons with heteroatoms, (highly) water soluble, e.g., phenol, cresol, and pyridine
3	light aromatic	compounds that usually do not pose problems regarding condensation or water solubility, e.g., toluene, styrene, and xylene
4	light polyaromatic	2- and 3-ring compounds that condense at intermediate temperatures at relatively high concentrations, e.g., naphthalene, phenanthrene, and anthracene
5	heavy polyaromatic	4–6-ring compounds that condense at high temperatures at low concentrations, e.g., fluoranthene, pyrene, chrysene, perylene, and benzo(<i>ghi</i>)perylene

important to know the tar dew point, i.e., the temperature at which tar condensation begins upon cooling of producer gas. For a single tar compound, the dew point can be calculated from

$$22400 \frac{C}{M} \frac{T}{273} \frac{1}{p_{sv}(T)} = 1 \quad (1)$$

Here, T is the absolute temperature, C is the compound concentration in g/Nm^3 , M is the molecular weight, and $p_{sv}(T)$ is the saturated vapor pressure at temperature T . For a mixture of tar compounds, the dew point can be calculated in a similar way by taking the sum of separate contributions (Implicitly, it has been assumed that tar vapors behave as ideal gas and that tar compounds show ideal mixing behavior. The latter approximation can be justified by the fact that usually only a few compounds within neighboring classes contribute significant fractions to the sum). The ECN website www.thersites.nl explains the concept and offers an option to calculate the dew point from concentrations of separate tar compounds.

ECN has also developed, in cooperation with Michell Instruments, a tar dew point analyzer. It allows for a quick and easy measurement of the tar dew point and can be useful to prevent tar-related problems in downstream equipment.^{26–28}

In essence, it involves the measurement of the change in light reflection by a mirror when it is heated or cooled in a temperature region around the tar dew point.

Tar Prevention

The first step to solve tar problems is to prevent or reduce tar formation when possible. ECN has investigated the effect of fuel properties and operating conditions on tar formation in fluidized-bed gasifiers. The work has been performed in cooperation with the universities of Eindhoven (TU/e) and Twente (UT) and with the research institutes KEMA and TNO-MEP. Results have been presented at a number of

(26) van Paasen, S. V. B.; Boerrigter, H.; Kuipers, J.; Stokes, A. M. V.; Struijk, F.; Scheffer, A. Tar dewpoint analyser; for application in biomass gasification product gas. Report ECN-C--05-026, 2005.

(27) van Paasen, S. V. B.; Boerrigter, H.; Kuipers, J.; Struijk, F.; Stokes, A. In Proceedings of the 14th European Biomass Conference, Paris, France, 2005; pp 823–826.

(28) Vreugdenhil, B. J.; Kuipers, J. In Proceedings of the 16th European Biomass Conference, Valencia, Spain, 2008; pp 659–662.

(29) van Paasen, S. V. B.; Bergman, P. C. A.; Neeft, J. P. A.; Kiel, J. H. A. In Proceedings of the 12th European Biomass Conference, Amsterdam, The Netherlands, 2002; pp 597–599.

(30) van Paasen, S. V. B.; Kiel, J. H. A. Tar formation in a fluidised bed gasifier: Impact of fuel properties and operating conditions. Report ECN-C--04-013; 2004.

(31) van Paasen, S. V. B.; Kiel, J. H. A. In Proceedings of the 2nd World Biomass Conference, Rome, Italy, 2004; pp 952–955.

(32) van Paasen, S. V. B.; Kiel, J. H. A. In Proceedings of the 6th International Conference on Science in Thermal and Chemical Biomass Conversion; Bridgwater, A. V., Ed.; Victoria, British Columbia, Canada, 2004; pp 542–556.

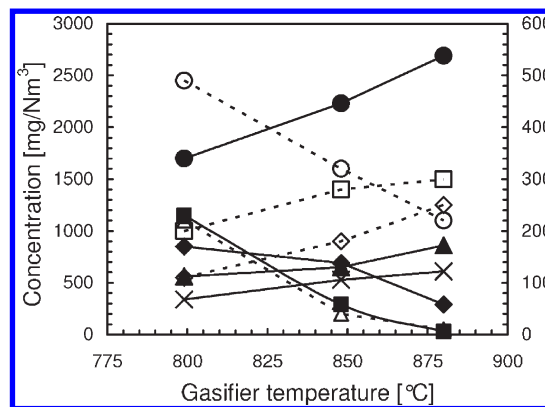


Figure 1. Effect of the CFB gasifier temperature on the concentration of tar compounds in producer gas. (Left axis) Naphtalene (●), phenol (■), xylene + styrene (◆), acenaphthylene (▲), and phenanthrene (×). (Right axis) Methylnaphtalene (○), fluorene (□), fluoranthene (◇), and cresol (△).

conferences and in ECN reports.^{24,29–33} The results can be summarized as follows: (1) The composition of dry fuel has little effect on the tar amount and composition. Lignin produces slightly more tar than cellulose, but the tar dew point is similar. In general, fuel ash content is not relevant.^{24,30,32} In the next section, some exceptions will be discussed. (2) A higher moisture content of fuel reduces the amount of tar formed. The tar dew point decreases slightly.^{24,30,32} (3) The temperature has the most marked effect on tar amount and composition. In fact, it is also the main factor that determines the differences between tar from updraft, fluidized-bed, and downdraft gasifiers. With increasing temperature, OH groups disappear first, followed by CH_3 groups. A higher temperature promotes polymerization, resulting in compounds with a larger number of rings. The total amount of tar decreases, but the concentrations of class 4 and 5 compounds increase (see Figure 1). Because the heavier tar compounds have lower vapor pressures, the tar dew point rises with the gasifier operating temperature.^{24,29–31} (4) Increasing the gas residence time in a hot zone has a similar effect to but smaller than increasing the temperature.^{24,29,31} (5) The addition of dolomite or olivine to the gasifier bed material reduces the amount of tar. Dolomite becomes active at lower temperatures than olivine. At 900 °C, olivine approaches the activity of dolomite.^{24,33} Concentrations are reduced in all tar classes. (6) Biomass char is able to reduce the amount of tar in producer gas, but too much char is required for practical use in gasifiers.

Although some measures have limited effect on the total tar content, they may still be useful to change the tar composition,

(33) Devi, L.; Smits, B. A.; Ptasinski, K. J.; Janssen, F. J. J. G.; Bergman, P. C. A.; Kiel, J. H. A. In Proceedings Bioenergy 2003, Jyväskylä, Finland, 2003; pp 418–420.

e.g., to prevent wastewater contamination or obtain tar more compatible with washing liquids.

A more drastic reduction of the tar content in producer gas can be obtained by a two-stage gasification process, given the name CASST (clean air sustainable syngas technology) at ECN. In the first stage, biomass is converted into pyrolysis gas and char by moderate heating in an inert atmosphere. In a second stage, char is gasified with steam at high temperature. Combustion of the pyrolysis gas covers the heat requirement of both stages. If pyrolysis is performed at 350 °C, a 10-fold reduction in tar content is achieved. Analysis of a CASST combined cycle showed that the efficiency could be close to that of a combined cycle with an air-blown CFB gasifier.³⁴ Actual tests showed the concept to be less ideal. CASST was abandoned, but some of the ideas and test results were invaluable for later developments.

Tar Cracking

Much of the work by ECN on tar cracking has been performed in cooperation with TU/e and the companies HoSt and BTG. The effects of catalysts, high temperature, and plasma have been studied. Some results are presented below.

Catalytic Tar Cracking. Nickel and dolomite are the materials studied most often for use as catalysts in downstream tar crackers. Tar reduction by the use of dolomite and olivine, as mentioned in the previous paragraph, is in fact tar cracking applied within the gasifier. Because we had observed low tar concentrations in producer gas during some of our gasification experiments, we decided to investigate the use of ash rich in Ca and Mg, as an alternative to dolomite. We used ash from paper residue sludge heated in air to 600 °C and ash from gasification of chicken manure. The latter ash contains a significant amount of char.

For a test of the tar cracking activity, we filled a reactor tube with active material sandwiched between pebbles. In the case of dolomite, the active material was sandwiched between sand and pebbles. We placed the reactor tube in a furnace, heated to 750 or 900 °C. We fed a sidestream of producer gas from the 5 kW_{th} BFB gasifier to the reactor tube and used the SPA method to determine the tar content and composition up- and downstream of the furnace.

Figure 2 shows the fractions of tar compounds broken down within about 1 s of contact with active material. At 750 °C, ash from chicken manure appears to be more active than dolomite. Ash from paper sludge shows little activity, except for 5-ring components. However, that may be a spurious result, because concentrations of 5-ring components are close to the detection limit. At 900 °C, the activities of dolomite and ash from chicken manure are equal. Ash from paper sludge does show tar cracking activity but considerably less than the other two materials.

Although the ash of chicken manure shows interesting activity for catalytic tar cracking, two remarks should be made. First, we have observed large variation in the tar content of producer gas from chicken manure without obvious differences in ash composition. Hence, a component or factor that we have not looked for may be responsible for the catalytic activity. Second, ash will have to be densified or sintered to produce material suitable for use in large-scale

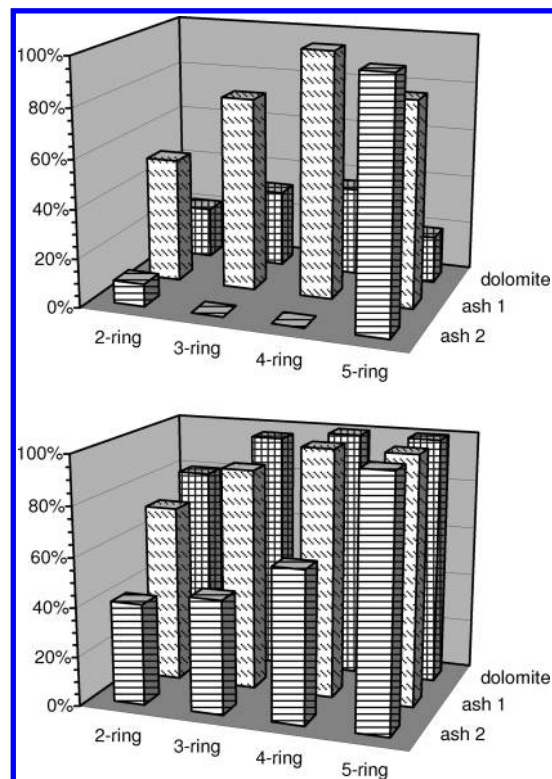


Figure 2. Fractions of tar compounds with 2–5 rings removed from producer gas by contact for about 1 s with dolomite, ash from chicken manure (ash 1), and ash from paper sludge (ash 2). The temperature was 750 °C (upper graph) or 900 °C (lower graph).

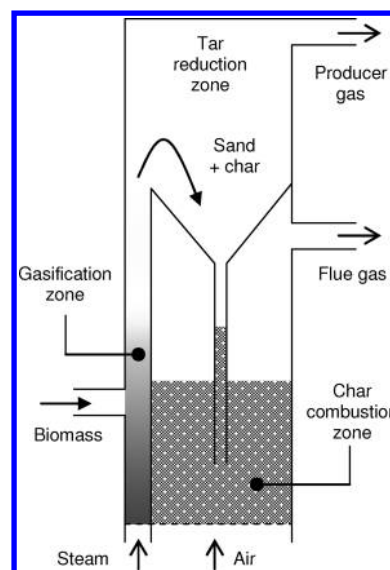


Figure 3. STAR concept for biomass gasification with tar reduction by char.

reactors. This may well reduce the catalytic activity or offset any price advantage when compared to dolomite.

Char from biomass fuels with low ash content shows some activity for tar cracking too. Because BFB and CFB gasifiers produce a considerable amount of char, we considered two options to use the char for tar reduction. The first option, given the name STAR concept, was to add a char hold-up chamber to a Battelle-type gasifier (see Figure 3). Experiments showed that the tar content of producer gas

(34) den Uil, H. In Proceedings of the 1st World Conference on Biomass for Energy and Industry, Sevilla, Spain, 2000; pp 578–581.

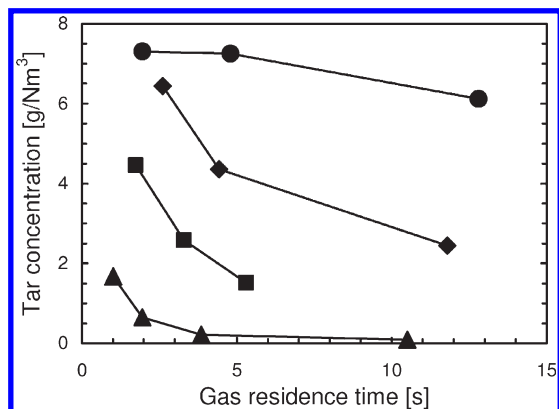


Figure 4. Effect of the tar cracking temperature and residence time on the tar concentration in producer gas. Temperatures were 900 °C (●), 1000 °C (◆), 1075 °C (■), and 1150 °C (▲).

could be reduced by 60–80%. However, the required conditions of at least 1.5 s of contact in a hot zone containing 1.5 kg of char/m³ are difficult to realize in practice.

The second option, given the name TREC (tar reduction by char), was developed from the STAR concept. It consists of a granular bed, which acts as a particle filter. It removes fly ash and char from producer gas, which flows in the radial direction through the bed.³⁵ Char deposited on the granules acts as a catalyst for tar cracking. The effectiveness of TREC can be enhanced by the use of catalytically active granules. The granular bed is kept moving perpendicular to the gas flow. Thus, granules are refreshed constantly to keep the pressure drop low. At 900 °C operating temperature, the TREC module reduced the tar content by 75%. The tar dew point dropped from 350 to 170 °C. However, the pressure drop over the TREC reactor increased steadily because of soot formation.

A slightly modified version with olivine as bed material proved successful in a test in which producer gas was fed to a solid oxide fuel cell (SOFC).³⁶ When used downstream of an air-blown BFB gasifier, it reduced the tar content to about 25% of the original value and reduced the tar dew point to below 100 °C.

Thermal Tar Cracking. Experiments on thermal tar cracking have been performed at lab scale with producer gas from a BFB gasifier.³⁷ The producer gas was pumped through a quartz tube of 75 mm in diameter. The tube was heated over a length of 750 mm. The residence time of producer gas in the heated zone was varied by the pumping speed. Figure 4 shows the total tar concentration after heat treatment at temperatures from 900 to 1150 °C for various residence times. At the highest temperature, the tar concentration is reduced to 20% after 1 s. After 4 s residence time, less than 3% remains. At 1075 °C, a reduction to 20% takes 5 s; at 1000 °C, that level is not reached even after 12 s.

Figure 5 shows the relative contributions of groups of compounds with 2, 3, 4, or 5 and more rings. With increasing temperature, the relative contribution of compounds with 2

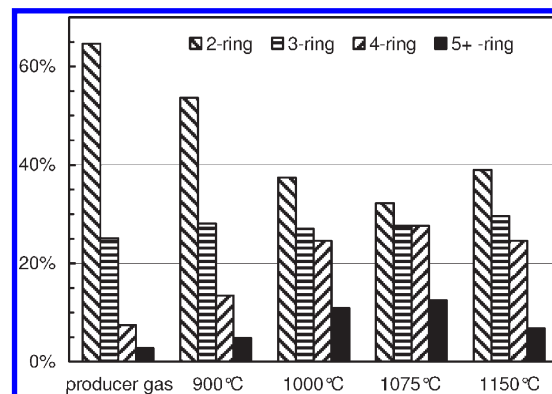


Figure 5. Contributions of compounds with 2–5 rings to total tar for tar in producer gas and after tar cracking at different temperatures (averaged over residence times).

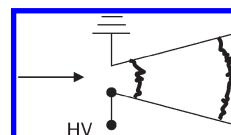


Figure 6. Schematic representation of glid-arc discharge used for tar removal from producer gas.

rings decreases and the contributions of compounds with 4 and more rings increase. At the highest temperature, the trend appears to reverse, but that is probably due to the low total amount of tar left. At that point, only a few compounds remain above the detection limit. The trend toward compounds with a larger number of rings eventually leads to the formation of soot. However, tar compounds are not the only source of soot. Lighter hydrocarbons are reformed or broken down too. At 1150 °C, toluene and ethylene disappear even quicker than tar. Benzene and ethane are a little more stable, with about 10% remaining after 5 s of residence time. Methane is the most stable compound, from which nearly 25% remains after 10 s.

Plasma Tar Cracking. ECN tested the effectiveness of a glid-arc plasma for tar removal. The plasma is produced by an electrical discharge between electrodes that form a gap that widens from the base to the tip of the electrodes.³⁸ An arc discharge starts at the point where the electrodes are closest. The arc moves along the electrodes, stretching to fill the widening gap, until it breaks and disappears. The discharge creates a plasma of energetic electrons, ions, and radicals that can break down tar compounds in producer gas flowing through the plasma, as indicated in Figure 6.

Tests have been performed with electrode configuration types I and II. In both cases, the results were rather disappointing. Less than 50% of the total tar content was removed, even at plasma energy densities corresponding to 25% of the producer gas energy content. Figure 7 shows the result at a producer gas temperature of 600 °C. At 800 °C, tar removal was slightly more effective; at 400 °C, it was slightly worse. Similar to the case of thermal cracking, a fraction of the lighter hydrocarbons was destroyed too.

The tests at ECN led to the conclusion that major improvements were needed to make plasma tar cracking a

(35) van der Drift, A.; Carbo, M. C.; van der Meijden, C. M. In Proceedings of the 14th European Biomass Conference, Paris, France, 2005; pp 566–568.

(36) Broust, F. Green fuel cell. SOFC fuel cell fuelled by biomass gasification gas. Publishable final activity report of EU project SES6-CT-2004-503122, 2008.

(37) Houben, M.; Verschuur, K.; de Lange, H.; Neeft, J.; Daey Ouwens, K. In Proceedings of the 12th European Biomass Conference, Amsterdam, The Netherlands, 2002; pp 581–584.

(38) Czernichowski, A. In *Non-thermal Plasma Techniques for Pollution Control. Part B: Electron Beam and Electrical Discharge Processing*; Springer-Verlag: Berlin, Germany, 1993; NATO ASI series G, Vol. 34, pp 371–387, ISBN 3-540-57174-4/0-387-57174-4.

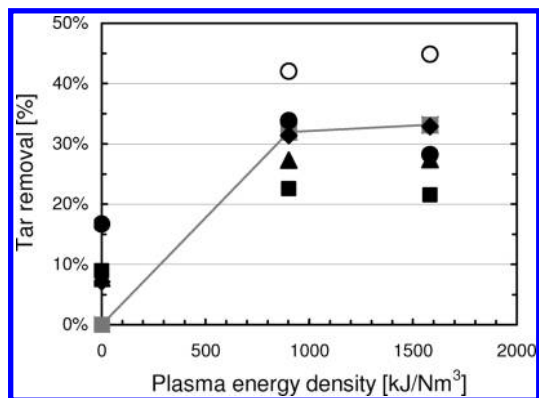


Figure 7. Tar removal efficiency of glid-arc type II for producer gas at 600 °C. Total tar (gray square and line), 1-ring compounds (O), 2-ring compounds (◆), 3-ring compounds (▲), 4-ring compounds (■), and 5-ring compounds (●).

practically useful technique. To that end, more fundamental research is performed at TU/e. One line of research is aimed at plasma generation by high-voltage pulsed corona discharge.³⁹ A second line of research involves plasma generation by flamelets created by oxygen injection.⁴⁰ Research on these subjects is still continuing, with financial support from the Energy Research Subsidy (ERS) program of the Dutch Ministry of Economic Affairs.

Tar Scrubbing with Water

The first attempts at ECN to remove tar by scrubbing involved the use of water as the scrubbing agent. The aim was to clean producer gas sufficiently for use in a gas engine. As producer gas contains at least 15% water by volume; the water needed for scrubbing can be recycled from condensate that forms upon cooling of the producer gas to the temperature of the environment. Most of the tar has to be removed from the condensate before the water can be reused. Disposal of excess water requires more thorough cleaning.

A successful example of water scrubbing for tar removal and gas engine operation on cleaned producer gas is the Harboøre installation.⁴¹ There, an updraft gasifier is used that produces mainly primary tar compounds that are more water-soluble than tar compounds produced by the fluidized-bed gasifiers at ECN. Hence, we thought separation of water and tar should be more easy in our case. It turned out to be more complicated than expected.

Tests at ECN have been performed with a rotating particle separator (RPS) and with three versions of GASREIP, an acronym for gas cleaning (both tar and NH₃) and power production. The gas and water cleaning systems were delivered by HoSt. The same company previously built the 500 kW_{th} CFB Bivkin gasifier, which supplied producer gas for the tests. Most of the dust was removed from the producer gas upstream of the RPS or GASREIP by cyclones.

RPS. The RPS contains a rotating cylinder from which the central part is blocked and the outer ring is filled with narrow

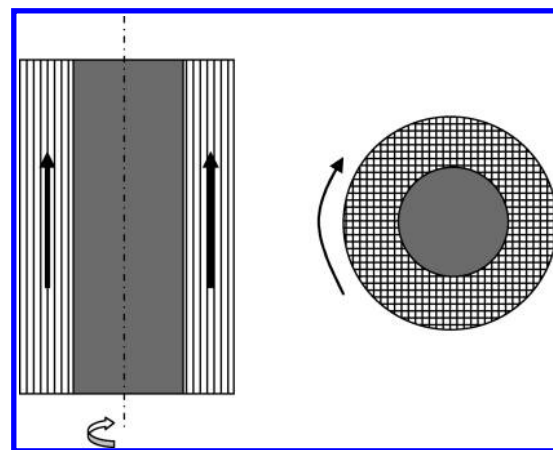


Figure 8. Cross-sections through RPS along and perpendicular to the axis of rotation.

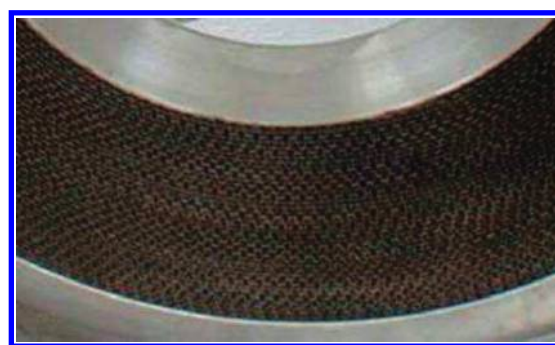


Figure 9. Section of RPS showing the narrow channels.

channels (see Figures 8 and 9).⁴² Gas flows through the narrow channels. The rotation generates a centrifugal force that drives particles or droplets in the gas to the wall. The channels can be cleaned by, e.g., gas pulses or a water spray. For the test at ECN, the top of the RPS was continuously sprayed with water to flush tar droplets and dust from the channel walls. The operating temperature of the RPS was 40–50 °C.

Tests were performed with gas produced by a CFB gasifier operating at 850 °C. Wood pellets with about 10% moisture were used for fuel. The RPS operated at 3000 rpm and was flushed with water at 200 L/h. The full gas flow of 190 Nm³ h⁻¹ produced too high of a pressure drop over the RPS. Measurements have been taken at about 25% of the maximum flow. The RPS reduced the dust content in the producer gas from 340 to 15 mg/Nm³. The tar content was decreased from 8 to 4.5 g/Nm³. The calculated tar dew point was 52 °C.^{43,44} That result showed that cleaned gas contained only tar vapor; i.e., droplets of condensed tar were effectively removed. Unfortunately, however, tar droplets were not removed from the channels of the RPS. Post-test inspection showed that the bottom end of the RPS was almost completely blocked.

(39) Pemen, A. J. M.; van Paasen, S. V. B.; Yan, K.; Nair, S. A.; van Heesch, E. J. M.; Ptasinski, K. J.; Neeft, J. P. A. In Proceedings of the 12th European Biomass Conference, Amsterdam, The Netherlands, 2002; pp 844–847.

(40) Houben, M. P. Analysis of tar removal in a partial oxidation burner. Ph.D. Thesis, Eindhoven University of Technology, Eindhoven, The Netherlands, 2004.

(41) Teislev, B. In Proceedings of the 12th European Biomass Conference, Amsterdam, The Netherlands, 2002; pp 1027–1029.

(42) Brouwers, B. *Chem. Eng. Technol.* **1996**, *19*, 1–10.

(43) van der Meijden, C. M.; Neeft, J. P. A.; van der Ploeg, F. B. Roterende deeltjes scheider (RDS) voor reiniging van biomassa stookgassen. Report ECN-C--01-118, 2001.

(44) Neeft, J. P. A.; van der Meijden, C. M.; Rabou, L. P. L. M.; Klein Teeselink, H.; Brenneisen, L. M.; van der Ploeg, F. B.; Dinkelbach, L. In Proceedings of the 12th European Biomass Conference, Amsterdam, The Netherlands, 2002; pp 593–596.

GASREIP A. The gas cleaning of the first GASREIP setup consisted of a scrubber for tar and NH_3 removal, followed by a sawdust filter for the removal of tar aerosols (see Figure 10). Cleaned gas was sent to a gas engine. Water from the scrubber passed a settling tank to separate heavy tar, a candle filter for further cleaning, and a stripper to remove NH_3 . Most of the water was then reused in the scrubber. In the scrubber, acid was added to the water to remove NH_3 more effectively from the producer gas. In the stripper, base was added to drive NH_3 from the water.

It soon turned out that the gas cleaning was insufficient for producer gas from a CFB gasifier. Cleaned producer gas still contained 2.3 g/Nm^3 tar.⁴⁵ Although that value was lower than the result obtained with the RPS, it was considered still too high for use of the gas in a gas engine. When connected to a fixed bed gasifier, the installation managed to bring down the tar concentration from 0.6 to 0.2 g/Nm^3 . A gas engine operated on cleaned producer gas for a test period of 6 h. Post-test inspection showed severe fouling of the inlet gas filter of the engine. The fouling could be due to the formation of gas channels in the sawdust filter. Through those channels, tar aerosols could easily have escaped capture and reached the gas engine.

GASREIP B. A second setup was designed with an additional scrubber and a wet electrostatic precipitator (ESP) to

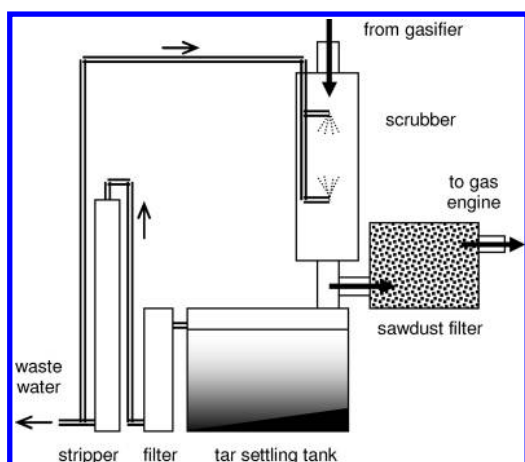


Figure 10. Scheme of GASREIP A configuration for producer gas cleaning (not to scale).

remove tar aerosols more effectively. The modified scheme is shown in Figure 11. Water for the second scrubber is cooled in a heat exchanger (not shown in the figure) to improve the tar removal and NH_3 capture.

The lower operating temperature of the second scrubber leads to further condensation of water from the producer gas and, thus, to continuous refreshment of the scrubber water. The ESP contains electrodes at high voltage (HV) to charge tar and water droplets and remove them from the gas by a strong electric field. The water cleaning system has also been changed. The candle filter upstream of the stripper has been replaced by a sand bed filter. Downstream of the stripper, an active carbon filter has been added.

The GASREIP B configuration succeeded in bringing down the tar content of producer gas from 10 to 3 g/Nm^3 when the CFB gasifier operated at $800 \text{ }^\circ\text{C}$ and to 1.4 g/Nm^3 when the CFB gasifier operated at $880 \text{ }^\circ\text{C}$.⁴⁴ The difference is caused by the larger contributions of class 2 and class 3 tar compounds at the lower gasifier operating temperature. That can also be seen in Figure 12, which shows the performance of the scrubbers and ESP for separate tar classes. Downstream of the second scrubber, there is still a significant contribution from class 5 tar compounds, probably condensed on dust particles or droplets. The ESP removes that contribution almost completely. The calculated tar dew point downstream of the ESP was about $60 \text{ }^\circ\text{C}$. A gas engine operated successfully for 70 h on clean producer gas.⁴⁶

The ESP was a commercial device designed for a 5 times larger flow than delivered by the ECN gasifier. The gas residence time was about 12 s. When part of the ESP channels were blocked and the residence time was reduced to about 4 s, the ESP still worked well.⁴⁷ In another test, the mixture of tar and water produced by the ESP was recycled to the gasifier and injected into the air inlet. Measurements showed that at least 70% of the tar was broken down.⁴⁸ Continuous recycling of the ESP effluent to the gasifier would increase the tar content of producer gas by 30–50%.

Although the gas cleaning worked well, there remained operational problems to be solved. The gas pipe between the scrubbers became clogged by a deposit of tar and dust. Tar and dust formed a thick foam on top of the water in the tar settling tank. The tar load to the active carbon filter was high. The wastewater contained 0.2 g/m^3 benzene, 0.8 g/m^3 toluene, and about 4 g/m^3 heavier tar compounds. These

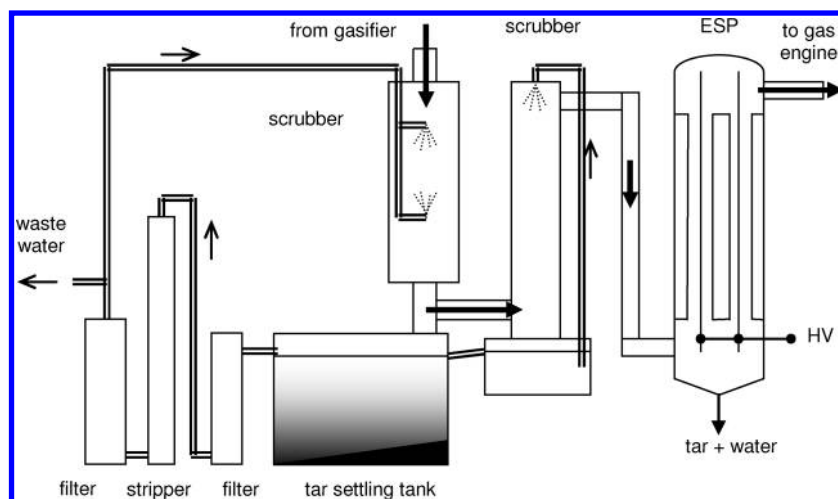


Figure 11. Scheme of GASREIP B with two scrubbers and ESP (not to scale).

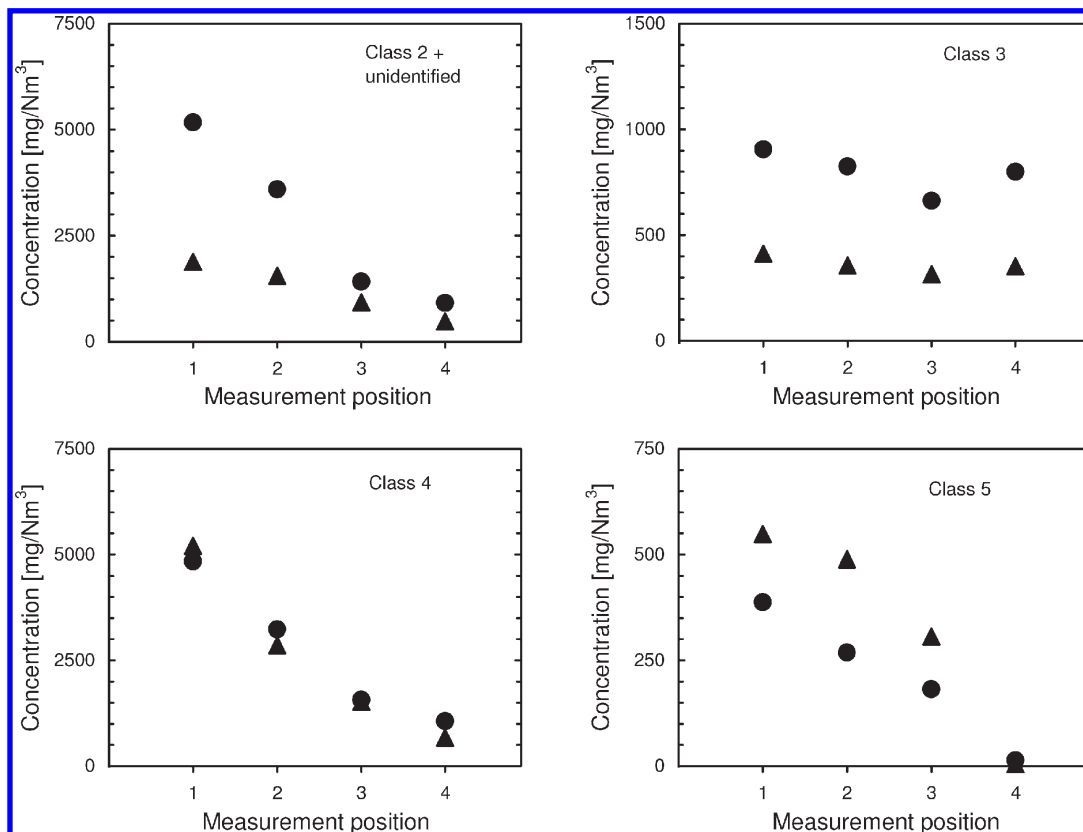


Figure 12. Contributions of different tar classes to tar concentration in producer gas from CFB gasifier operating at 800 °C (●) or 880 °C (▲). Measurement position 1, raw gas; 2, downstream first scrubber; 3, downstream second scrubber; and 4, downstream ESP.

figures should be considered indicative only, because most contributions were close to the detection limit of the analysis method applied. However, they exceeded the allowed limit for disposal to the sewer by a factor 10.

GASREIP C. The third setup was designed to solve some of the problems encountered in the second one described above. The second and third cyclone for dust removal were replaced by a single new one to obtain better and more reliable dust removal. New scrubbers were installed with metal packing for improved contact between gas and water. The first scrubber was operated in counterflow with water cooled by a cooling tower. The second scrubber was intended for NH₃ removal only. The ESP was positioned between both scrubbers to minimize the tar load to the second scrubber. The tar settling tank was enlarged and divided into sectors to improve the settling process. The settling tank was insulated and slightly heated, to reduce water and tar viscosities and reduce the solubility of benzene and toluene. Figure 13 shows a scheme of the GASREIP C system.

The system has been tested for 50 h in total over 5 test periods lasting 5–18 h each. Producer gas cooled from 300 to 25 °C when 18 °C water was used in the tar scrubber. The tar concentration downstream of the ESP was reduced to

0.7 g/Nm³. Remaining tar consisted only of class 2, 3, and 4 compounds with appreciable vapor pressures at 20 °C. As a result, the calculated tar dew point was only 21 °C. Hence, tar condensation downstream of the gas cleaning could easily be prevented by moderate heating of the gas line to, e.g., 30 °C.

The GASREIP C configuration was more effective in tar removal than the previous versions, but it still suffered fouling problems. Despite the use of a very open metal packing, the pressure drop over the tar scrubber quickly increased because of clogging by tar. Post-test inspection also showed naphthalene deposit on the coolest parts of the scrubber, e.g., the water spray nozzle and top flange (see Figure 14). Some tar was visible on the walls of the ESP, but the thickness of the layer was negligible in comparison to the diameter of the channels.

After 50 h testing, the water in the tar settling tank contained 0.6 g/m³ benzene and 0.1 g/m³ toluene. These values were 25 times lower than before with GASREIP B. Part of the difference was caused by dilution by the larger initial volume of fresh water. Still, the difference was large enough to prove that moderate heating can drive off most of the benzene and toluene from the water and, thus, reduce the load to the active carbon filter. Water of the NH₃ scrubber, which operated at 10–20 °C, contained 6 g/m³ benzene and 0.6 g/m³ toluene. These compounds were completely removed by the stripper, which operated at 50–60 °C.

Tar Scrubbing with Oil

The experience with the GASREIP systems led to the conclusion that mixing of dust, tar, and water should be prevented. That can at least partially be realized with a scrubber operating above the water dew point of the gas using

(45) Dinkelbach, L.; van der Meijden, C. M.; Brenneisen, L. M.; Strating, S. D.; Geleijn, M.; Wubbe, R. GASREIP GASREIniging en Prime mover design Fase A. Report ECN-C--00-084 (Report 2EWAB00.34), 2000.

(46) Rabou, L. P. L. M.; van der Meijden, C. M.; Brenneisen, L. M.; de Kant, E.; Klein Teeseling, H.; Wubbe, R. In Proceedings of the 12th European Biomass Conference, Amsterdam, The Netherlands, 2002; pp 442–444.

(47) van Paasen, S. V. B.; Rabou, L. P. L. M.; Bar, R. In Proceedings of the 2nd World Biomass Conference, Rome, Italy, 2004; pp 781–784.

(48) Rabou, L. P. L. M. *Fuel* 2005, 84, 577–581.

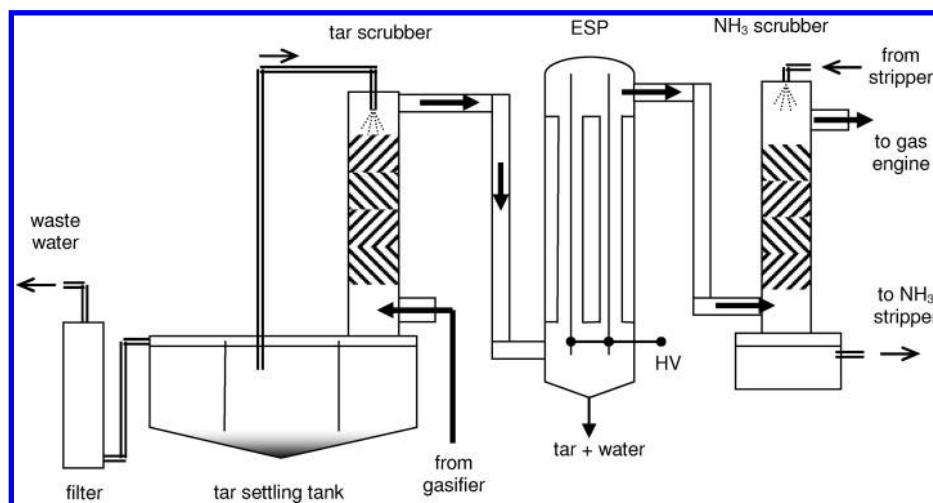


Figure 13. Scheme of GASREIP C with tar scrubber, ESP, and NH_3 scrubber (not to scale).

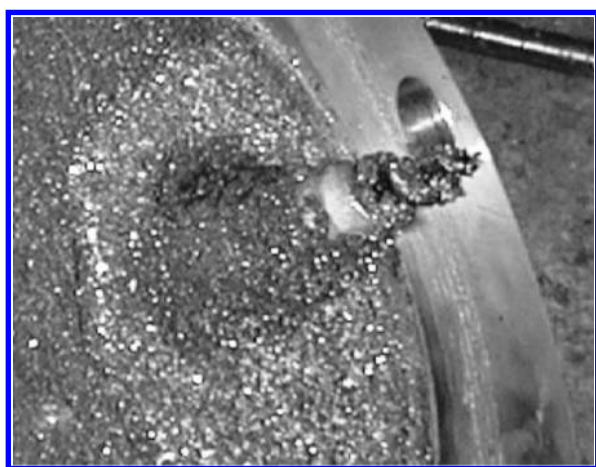


Figure 14. Naphtalene deposit on top flange and spray nozzle of the tar scrubber.

a scrubbing liquid compatible with tar. If the right scrubbing liquid is chosen, it can absorb tar from producer gas down to a vapor pressure far below that of saturated vapor. Usually that suffices to prevent tar condensation in downstream equipment. The technology developed by ECN was given the name OLGA.⁴⁹

The first laboratory OLGA facility had a capacity of $2 \text{ Nm}^3 \text{ h}^{-1}$ producer gas. Figure 15 shows a simplified flow scheme. It consists of a collector, absorber, and stripper. The collector quenches the producer gas with oil and cools the gas to a temperature above the water dew point. Part of the tar condenses and mixes with the scrubbing oil. The absorber removes tar vapor and part of the benzene and toluene by absorption in oil at a constant temperature. The stripper operates at higher temperature to drive the absorbed hydrocarbons from the absorber oil. The system contains a cooler for collector oil, to remove heat absorbed from the producer gas, and a heat exchanger, to reduce the heat duty in the absorber–stripper loop. Measured tar concentrations downstream of OLGA correspond to a tar dew point below $0 \text{ }^\circ\text{C}$.

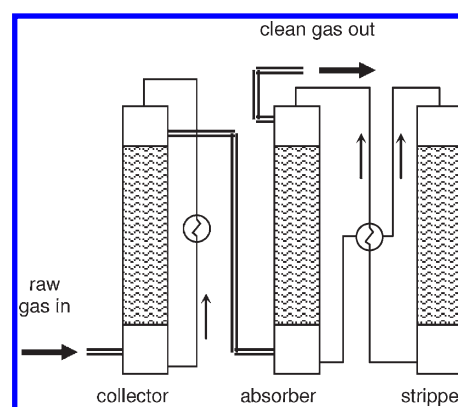


Figure 15. Simplified flow scheme of the OLGA tar removal system.

Tar compounds captured in the collector oil can be recycled to the gasifier or used as fuel in another installation. Tar removed from the absorber oil by the stripper can be used in a similar way. If air is used as a stripping medium, it is logical to consider direct use of the tar-laden air in the gasifier. If steam is used as a stripping medium, direct use in an air-blown gasifier is less straightforward but use in an O_2 /steam-blown gasifier can be considered.

The laboratory installation has been modified several times but is still used regularly. Originally, it was used only downstream of the 5 kW_{th} BFB gasifier. Later on, it was also connected to the $25 \text{ kW}_{\text{th}}$ Milena gasifier, which can be used in BFB or indirect mode. In total, the facility has been operating for more than 2000 h.

When laboratory tests proved successful, ECN started a cooperation with the company Dahlman for further development and marketing of the OLGA technology. In 2003, a pilot installation with a capacity of $200 \text{ Nm}^3 \text{ h}^{-1}$ was built downstream of the $500 \text{ kW}_{\text{th}}$ CFB Bivkin gasifier.⁵⁰ Figure 16 shows the OLGA pilot installation hovering above the site where it was mounted. The GASREIP system was reused downstream of OLGA for further producer gas cooling, water condensation, and NH_3 removal.

(49) Bergman, P. C. A.; van Paasen, S. V. B.; Boerrigter, H. In *Proceedings of the Expert Meeting on Pyrolysis and Gasification of Biomass and Waste*; Bridgwater, A. V., Ed.; CPL Press: Strasbourg, France, 2003.

(50) Boerrigter, H.; van der Drift, A.; van der Meijden, C. M.; van Paasen, S. V. B.; Pels, J. R.; Rabou, L. P. L. M.; Verhoeff, F. In *Proceedings of the 2nd World Biomass Conference*, Rome, Italy, 2004; pp 891–893.



Figure 16. Mounting of the OLGA pilot installation at ECN.

The OLGA process was originally designed for essentially dust-free producer gas. That required the use of an efficient upstream dust removal system, e.g., a hot gas filter. To reduce the investment and operating costs, we investigated the performance downstream of a cyclone that removes only the larger dust particles. Tests in the lab-scale facility indicated that the remaining dust could be removed effectively without fouling of the packing in the OLGA columns.⁵¹ Captured dust had to be separated from the scrubbing oil to limit the increase in viscosity. More problematic was the formation of tar aerosols, promoted by the presence of fine dust. The absorber captured only part of the aerosols formed in the collector. As a result, the tar dew point downstream of OLGA increased. The problem was solved by the use of an ESP downstream of the collector. The modified OLGA system performed successfully in an endurance test of 700 h, providing clean producer gas to a gas engine and micro gas turbine.^{52,53}

In 2006, a 2000 Nm³ h⁻¹ OLGA demo system was installed and tested successfully downstream of a special type of

(51) Boerrigter, H.; van Paasen, S. V. B.; Bergman, P. C. A.; Konemann, J. W.; Emmen, R. In Proceedings of the 14th European Biomass Conference, Paris, France, 2005; pp 990–993.

(52) Verhoeff, F.; Rabou, L. P. L.; Mvan Paasen, S. V. B.; Emmen, R.; Buwalda, R. A.; Klein Teeselink, H. In Proceedings of the 15th European Biomass Conference, Berlin, Germany, 2007; pp 895–900.

(53) Rabou, L. P. L. M.; Grift, J. M.; Conradie, R. E.; Fransen, S. *Energy Fuels* 2008, 22, 1944–1948.

updraft gasifier in Moissannes, France.⁵⁴ The pilot OLGA system at ECN is now connected to an 800 kW_{th} Milena indirect gasifier for tests due in 2009. Although these gasifiers produce tar compounds or tar concentrations that differ from the BFB and CFB gasifiers used in the early stage of the development, they still allow for application of the OLGA technology for tar removal.

Research at ECN has moved from producer gas cleaning for gas engines to the production of SNG with the use of catalysts. The broader scope of gas conditions and more demanding applications pose challenges to the researchers involved in the OLGA development. They have to make the OLGA process more versatile and suitable for industrial operation and, not the least important, reduce investment and operating costs.

Conclusion

Over 15 years of research by ECN on biomass gasification and producer gas cleaning has led to the conviction that tar removal from producer gas is best performed by oil scrubbing at a temperature at which water remains in the vapor phase. Tar reduction by measures taken in the gasifier or tar cracking in downstream reactors can reduce the tar load and simplify tar removal. However, for most applications tar removal remains a necessity. Water can be used for tar scrubbing, but mixing dust, tar, and water makes it hard to keep systems running.

Research at ECN has culminated in the patented OLGA process, which can reduce the tar dew point in producer gas to below 0 °C. The process has been developed for BFB and CFB gasifiers but can be adapted to other types of gasifiers. Although we did once proclaim the tar problem solved, tar remains an enduring challenge. However, the focus has shifted, with research aimed at improvements that make the OLGA process more versatile, rugged, and economic.

Acknowledgment. The work reported here was a joint effort of many people at ECN and partner institutes. Many of their names can be found in the list of references. Here, we also thank the people who remained unnamed but who designed, built, operated, and maintained the installations, who took care of the software and data acquisition, who developed analytical methods and performed the analyses, or who surveyed the safety. We thank the companies HoSt and Dahlman, for their important contributions to the development of GASREIP and OLGA, and all of the other companies involved in one or more of the ECN projects on producer gas cleaning and application. Financial support for the work was obtained from the Dutch Ministry of Economic Affairs, the EWAB programme of The Netherlands Agency for Sustainable Development and Innovation (SenterNovem), and the Agency for Research in Sustainable Energy (SDE).

(54) Koneman, H. W. J.; van Paasen, S. V. B. In Proceedings of the 15th European Biomass Conference, Berlin, Germany, 2007; pp 873–878.