Gasification of low rank coal

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Abstract

The use of domestic (high-ash) low rank (sub-bituminous and lignite) coal reserves contributes to the security of energy supply, and therefore high-ash coal is expected to remain as a key energy source in several countries (e.g. India, Turkey, Greece) for at least the next 30-40 years. However, the use of high-ash coals for energy production, currently performed mainly via combustion processes, poses a number of technical and economic challenges, e.g. low efficiency and environmental issues. Gasification is an attractive option, since it allows a more efficient, more environmentally friendly conversion of the coal. In particular, gasification for power production (IGCC) or synthesis of Substitute Natural Gas (SNG) offers high efficiency, reduced emissions and potential for the implementation of CO_2 capture.

The Energy research Centre of the Netherlands (ECN) has modified its biomass gasification set-up to make it suitable for high to very high-ash (> 35% wt.) coal and other low rank coal. The modified gasifier is called the i-MILENA. The i-MILENA is an indirect gasifier where gasification of the solid fuel is separated from the combustion of the remaining char and tar. The i-MILENA gasification technology has a high cold gas efficiency and high methane yield, making it very suitable for application in combination with gas engines, gas turbines and the production of Substitute Natural Gas (SNG).

In 2012, the EU Optimash project was started with partners in France, Turkey and India. As part of this project the ECN i-MILENA gasifier was tested with various low-rank coals from Turkey and India. The tests showed that the i-MILENA technology can gasify coals with more than 50 wt% ash, producing a medium-calorific gas without the requirement of an Air Separation Unit (ASU). The Cold Gas Efficiency is approximately up to 70%, which is relatively high for coals with a high ash content. The fuel conversion is close to 100%, as a result of the separation of the gasification and combustion zones. The remaining ash contains no carbon. This makes disposal or application of the ash much easier.

Keywords: Indirect gasification, i-MILENA, high-ash coal, low rank coal, Substitute Natural Gas (SNG).

1. Introduction

Coal will continue to play a key role in the world's energy scenario, not only for power generation, but also for the production of fuels (coal-to-SNG) and chemicals. Globally, coal resources have been estimated at over 861 billion tons. Coal meets around 30% of the global primary energy needs and generates 42% of the world's electricity [2]. Around 45% of the world's coal is either high-moisture or high-ash. Due to their contribution to the security of energy supply, high-ash coals are currently used for power generation in several countries (India, China, Turkey, Czech Republic, Poland, South Africa, Romania) [3]. In addition, high-ash coal is expected to remain as a key energy source in several countries (e.g. India, Turkey) for at least the next 30-40 years due to the availability of large domestic reserves and the installed-capacity for electricity production [4]. However, the use of high-ash coals for power production, which is currently performed mainly via subcritical combustion processes, results often in inefficient operation of the power plants. Emissions of fly-ash in power plants as well as the ash disposal pose an ecological and environmental challenge [5]. Furthermore, gasification offers the option to use coal with minimal environmental impact by conversion of the coal into Synthetic Natural Gas (SNG), i.e. high-quality methane. SNG can be used locally with minimum pollution in decentralized CHP applications or can be used as transportation fuel in Natural Gas Vehicles (NGV's). During the production of SNG, part of the carbon in the coal is separated from the gas as CO_2. When this relatively pure CO_2 stream is sequestrated, the carbon footprint can be reduced to the same level as natural gas.

India is a good example of the status of high-ash coal. India's vast coal resources (58.6 billion ton of proven hard coal reserves, i.e. 7% of global reserves [6]) contribute to a large share of its energy supply (42% of the primary energy in 2008) as well as to the security of energy supply [7]. However, the coal-based electricity sector in India faces up to several challenges, including the low efficiency of thermal plants, and inadequate transmission and distribution networks [6]. The low average efficiencies of Indian coal-fired power plants (27.6% LHV [8]) are partly due to the widespread use of high-ash coals in subcritical cycles as well as to the use of coal-fired plants for peak load electricity production, among other factors [9]. Only 13% of the Indian coal resources is of coking quality; the remainder is high-ash steam coal [6], with typically 40% wt. ash and a low calorific value (~3600 kcal/kg), which is difficult to wash below 30% ash to make it suitable for power generation [8]. Since it is expected that coal will remain the dominant energy source for India in the short and medium term, it is necessary to improve the efficiency of electricity generation from coal to exploit the extensive domestic high-ash coal resources while reducing emissions [6]. For this, several strategies has been proposed: enhancement of the efficiency of the existing power plants, improvement of coal beneficiation processes (decrease of combustibles loss in the rejects, ash disposal, reduction of energy- and water consumption), and development of clean coal technologies such as supercritical and ultra-supercritical combustion cycles, and Integrated Gasification Combined Cycle (IGCC).

Different gasification technologies were developed for the conversion of coal into gas. Most processes are operated at relatively high temperature (>1200°C) because the typical high-quality coals are not very reactive and require a high gasification temperature to obtain sufficient conversion. The most well-known technologies, e.g. from Shell, Siemens, and General Electric, are based on Entrained Flow gasification. The technology consists of a practically empty pressurized reactor in which fine fuel particles (generally smaller than 50 micrometers) are introduced and converted at temperatures of 1500°C or higher to ensure (near) complete conversion. This requires the use of pure oxygen. Entrained flow gasification therefore comes with an Air Separation Unit (ASU) for oxygen production. The gas residence time is short, only a few seconds, thus requiring small fuel size at high temperature to have sufficient conversion efficiency. Entrained Flow gasification is the logical choice for the less reactive bituminous coals, but for reactive coals like lignite or high-ash coals this technology is not optimal. A high ash content of the coal will have a strong negative impact on the overall process. The heating and the melting of the ash will further increase the oxygen consumption.

Fixed-bed gasification technology is probably more suitable for low rank / high-ash coals. Most gasifiers are of the "dry bottom" type. This means that the ash is not molten in the bottom of the gasifier. Fixed-bed coal gasifiers use a steam / oxygen mixture as gasification agent. A high ratio of steam to oxygen helps to moderate the temperature such that the ash does not melt. Fixed bed gasifiers require well defined particles typically in the range from 6 to 50 mm. Smaller particles are not allowed in the reactor, because this decreases the permeability of the bed. The fuel requirements and need for an ASU are the most important disadvantages of this type of coal technology compared to the proposed allothermal gasification technology.

The most logical type of gasification technology for low rank / high ash coal is fluidized bed technology. Fluidized bed gasifiers can be separated in three main categories: Bubbling Fluidized Bed (BFB), Circulating Fluidized Bed (CFB) and indirect or allothermal twin-bed concepts. All fluidized bed gasifiers use a bed material, normally this is the ash of the coal, sometimes with additives to optimize the process. The purpose of the bed material is to distribute and transport the heat in the gasifier (which prevents local hot spots), to mix the fuel with the gasification gas and the produced gases and, in the case of a catalytically active material, to reduce the concentration of tars. **Fig. 1** shows the basic principles and differences of three types of fluidized bed gasifiers. One of the main advantages of fluidized bed gasification is its fuel flexibility. Fuel geometry is not a restricting issue as long as the particles are not too big. When coal is used with a very high ash content, the size of the fuel is restricted to a few mm, because the bed material will be made up by the ash from the coal.

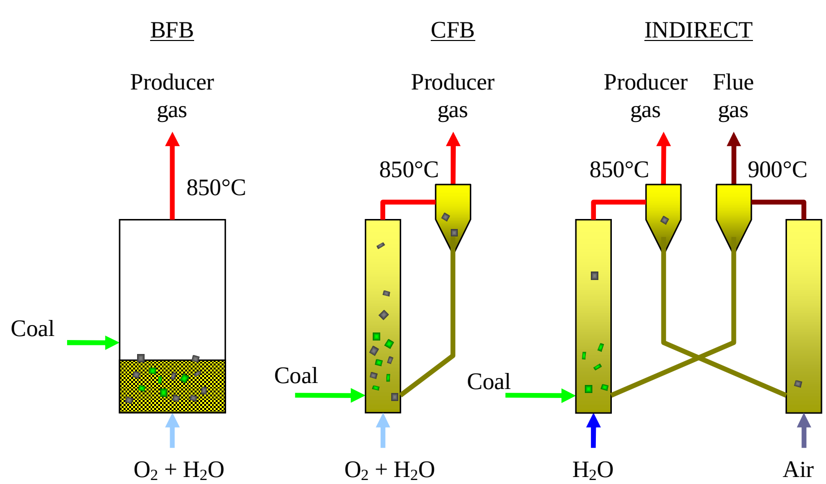

Fig. 1: Schematic comparison of BFB, CFB and indirect Gasification.

The Energy research Centre of the Netherlands (ECN) has developed a gasification technology called the MILENA technology [1]. The MILENA is an indirect gasifier where the gasification of the solid fuel is separated from the combustion of the remaining char and tar. The MILENA gasification technology has a high cold gas efficiency and high methane yield, making it very suitable for application of the gas in gas engines, gas turbines and the production of Substitute Natural Gas. The technology is relatively simple due to the atmospheric operation of the gasifier, the production of a N_2-free syngas without the need for an air separation unit, and the carbon-free residue that is produced. In addition, the integration of the gasifier and the combustor in the same vessel reduces the heat losses. The disadvantage of the atmospheric operation of the gasifier is the need for gas compression, but because of the high heating value of the gas the compression energy is much lower than would be required for a syngas containing no hydrocarbons.

In **Fig. 2** the principle of a direct gasifier (BFB or CFB) is given on the left and the right figure shows the principle of indirect gasification. The smart approach in indirect gasification is that pyrolysis and combustion are separated and a bed material is used to exchange heat between the two processes. This will lead to complete conversion of the fuel, contrary to direct gasification.

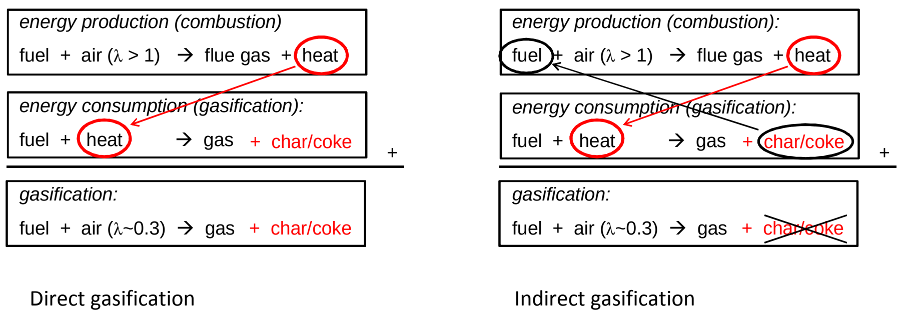

Fig. 2: Direct gasification (left) leaves a solid combustible residue called char or coke, whereas indirect gasification (right) uses this as an energy source within the process, leading to a much higher overall efficiency.

The indirect gasification concept of MILENA has more to offer than just the complete conversion of fuel. Below is a short list of the most prominent features of this technology:

- No air separation unit (ASU) needed – the combustion takes place in a separate vessel, hence air can be used and the product gas is essentially N_2 free.
- No carbon loss – the char/coke that remains after gasification is completely burned. The ashes are carbon free.
- High efficiency – because the gasification is decoupled from the combustion the gasification temperature can be much lower than direct gasification.
- Medium calorific gas – the product gas does not contain nitrogen, which increases the heating value substantially. This also offers the possibility to use the gas for more high-end applications (e.g. synthesis).
- Low pressure operation, thus, lower investment costs at low-medium scale facilities (< 100 MW).
- Reduced heat losses due to its integrated design.

The MILENA gasifier was developed for the gasification of biomass. The process is based on indirect or allothermal gasification. In one reactor the fuel is gasified or pyrolyzed using hot bed material. Because of the relatively low temperature (typically 850°C) of the pyrolysis process the conversion of the fuel is limited. The remaining char is combusted in a separate reactor. The heat from the combustion is used to heat the circulating bed material. In the biomass configuration the gasification takes place in the riser reactor where the residence time of the fuel is relatively short, but sufficient for the reactive biomass. If coal is used, the gasification takes place in the bubbling fluidized bed (BFB), where the residence time for the coal is much longer. Because the gasifier and combustion reaction are switched, this configuration is called the inverse MILENA or i-MILENA. A schematic layout of the i-MILENA gasifier is given in **Fig. 3**. Fuel is added to the bubbling fluidized bed zone (BFB) via a feeding screw. The BFB acts as steam gasification reactor, thus allowing longer residence times (in the order of 10 – 20 minutes) of the fuel particles compared to the MILENA concept in order to cope with less reactive (high ash) feedstock. The expected coal conversion is low (typical 60% carbon conversion). The remaining carbon is combusted in a riser reactor at a temperature of approximately 1000°C. Bed material (the ash of the coal) is used as heat carrier. The char flows from the BFB into the bottom part of the riser. Sufficient air is added to the riser to combust all the char and to have a vertical velocity of approximately 7 m/s. This velocity is high enough to carry the remaining solids (the ash) to the top of the reactor into the settling chamber. In the settling chamber the solids are separated from the flue gas and transported back into the BFB gasification zone via a downcomer.

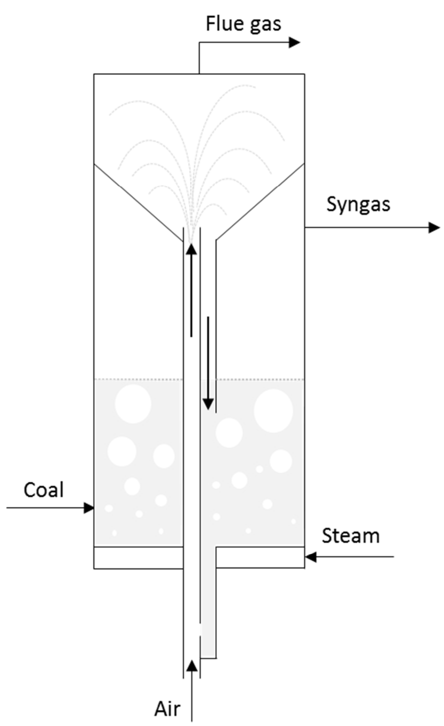

Fig. 3: Schematic layout of the i-MILENA gasification concept

If the coal conversion is too high in the BFB gasifier, the amount of char will be insufficient to generate the required heat and temperatures will drop. If the gasification temperature drops the yield of char will go up. An equilibrium temperature will set depending on the reactivity of the coal, but normally around 900 – 950°C in the gasifier and 980

– 1030°C in the combustor. Additional heat can be produced inside the BFB by adding a small amount of air as gasifying agent to the BFB and operating the system in a partly direct mode. Producer gas leaves the gasifier on the side and exits the system via a cyclone.

In this work, the development status of the i-MILENA technology is presented. Gasification tests using Indian Bilaspur high-ash sub-bituminous coal and German Heizprofi lignite as feedstock have been carried out in the 25-kW i-MILENA gasifier coupled to the OLGA tar removal system in order to assess the overall performance of the process.

2. Experimental

The goals of the test were to check if the carbon conversion is sufficient to run the process at commercial scale, to determine if the remaining ashes are free of carbon and to get an indication of the gas composition as well as a performance assessment of the OLGA system.

The lab-scale i-MILENA gasifier is coupled to a lab-scale gas cleaning installation. The gasifier and the gas cleaning system operate at atmospheric pressure. **Fig. 4** shows the lab-scale installation.

Fig. 4: Lab-scale i-MILENA (left) and OLGA (right) facility.

The main dimensions of the gasifier are: Riser / combustor diameter: 36 mm, gasifier / fluidized bed: 250 mm, overall height: 2 m. The diameter of the gasifier was later decreased to increase the velocity in the bubbling bed to have a better fluidization of the relatively large ash particles.

The reference feedstock used in the gasification tests is a high-ash Indian sub-bituminous high-ash coal from the Bilaspur region. Furthermore a test has been conducted with a low-ash German lignite from Heizprofi, to compare the process performance. **Table 1** summarizes the thermochemical properties of the fuels.

Table 1. Thermochemical properties of Indian high-ash coal and German lignite.

		Bilaspur	**Heizprofi**
Moisture (% wt., as received)		3.9	10
Ultimate analysis (% wt., dry basis)	C	31.9	60
	H	2.4	5.2
	N	0.86	0.59
	S	0.56	0.32
	O	11.2	29
Proximate analysis (% wt., dry basis)	Volatile matter	21.2	53
	Ash 550°C	54	4.4
Higher heating value (MJ/kg, as received)		12	24

The bed material used in the tests is mainly composed of coal ash, with a small amount of olivine that was used to start the process before coal ash was accumulated.

When an experiment is started it will take a while for the gasifier to fill the BFB with ash/coal and reach steady state operation. Good mixing in the BFB creates an average coal composition in the BFB where some particles are fully gasified and other just started, while ashes are returned to the BFB via the downcomer. Gasification is an endothermic process and heat has to be supplied to the BFB. Heat is created by removing part of the coal/bed material mix from the BFB and combusting it with air in the riser of the i-MILENA. The heated bed material ejected from the top of the riser fall back under gravity to the BFB via a downcomer. Flue gases leave the gasifier and exit the system via a flue-gas-cyclone. Heat can also be created inside the BFB by adding a little bit of air to the BFB and operating the system in a partly direct mode. Producer gas leaves the gasifier on the side and exits the system via a producer-gas-cyclone.

The overview of the i-MILENA experimental tests performed and the operating parameters are presented in **Table 2** and **Table 3**, respectively.

Besides composition of syngas and flue gas, production and composition of tars (measured through Solid Phase Adsorption) was assessed at different sampling points (after i-MILENA and after OLGA absorber) in order to determine the efficiency of tar removal in OLGA system. The concentration of H_2S was measured by micro-GC after i-MILENA.

Table 2. Overview of i-MILENA tests.

Feedstock	Test	Total test time (h)
High-ash Bilaspur sub-bituminous coal	1	6.6
	2	4.7
Heizprofi lignite	3	6.7

Table 3. Settings of i-MILENA tests.

		Test 1	Test 2	Test 3
i-MILENA gasifier	Coal flow rate (kg/h)	4.21	4.21	3.50
	Steam BFB (kg/h)	2	2	2
	Air BFB (NL/min)	0	0	0
	CO_2 purge (NL/min)	3	2	2
	Air riser (NL/min)	130	130	130
	Argon tracer gas (NL/min)	0	0	1
	T tracing reactor (°C)	900	900	900
	Temperature BFB (°C)	856	865	860
	Temperature riser (°C)	898	920	885
	Calcium sorbent (kg/h)	0	0	0
OLGA tar removal	Pump flow collector	8 Hz	8 Hz	-
	T profile	Standard	Standard	-
	Pump flow rate VIVA	25 Hz at 80°C	25 Hz at 80°C	-
	Pump flow rate absorber	5 Hz at 80°C	5 Hz at 80°C	-
	Pump flow rate stripper	25 Hz at 80°C	25 Hz at 80°C	-
	Stripper gas	16 NL/min air	16 NL/min air	-

3. Results and discussion

3.1 Producer gas and flue gas composition

Table 4 summarizes the composition of the producer gas and flue gas during i-MILENA tests with Indian high-ash coal and German lignite as feedstock. The methane concentration is high compared to entrained flow gasification processes. This is beneficial for the conversion of the gas into SNG.

Table 4. Producer gas composition, tar composition and flue gas composition during i-MILENA tests with Indian high-ash coal.

		Test 1	Test 2	Test 3
Producer gas composition				
CO	[% vol. dry]	17.1	17.4	26.3
H_2	[% vol. dry]	51.7	52.2	48.4
CO_2	[% vol. dry]	23.7	25.1	20.7
CH_4	[% vol. dry]	5.2	5.3	5.3
O_2 / (Ar)	[% vol. dry]	0.07	0.04	(1.32)[1]
C_2H_2	[% vol. dry]	0.071	0.070	0.055
C_2H_4	[% vol. dry]	0.482	0.377	1.43
C_2H_6	[% vol. dry]	0.011	0.006	0.080
C_6H_6	[ppmv dry]	2309	2574	3532
Toluene	[ppmv dry]	< 10	< 10	161
N_2	[% vol. dry]	3.8	1.2	0.5
H_2S	[ppmv dry]	3586	3229	1098
COS	[ppmv dry]	56	106	63
O_2	[% vol. dry]	8.9	7.5	5.0
CO_2	[% vol. dry]	10.4	11.3	12.9
C_xH_y	[ppmv dry]	1.6	9.2	17.5
CO	[ppmv dry]	32.3	1.5	130
NO	[ppmv dry]	278	253	153
NO_2	[ppmv dry]	29	14	6.0
NO_x	[ppmv dry]	0	0	0
N_2O	[ppmv dry]	9	14	4
SO_2	[ppmv dry]	6	0	0

[1] Argon concentration due to 1 NL/min argon added to producer gas

3.2 Tar composition and cyclone ash

A summary of results from SPA tar samples taken downstream the i-MILENA gasifier can be found in **Table 5**. Tars have been plotted according to the ECN classification, based on solubility:

Class 1: Unknowns, not measured by GC.

Class 2: Heterocyclic components (phenol, cresol).

Class 3: Aromatic components 1 ring: xylene, benzene and toluene. These components cannot be detected via SPA. Micro-GC analysis is thus used for these components.

Class 4: Aromatic components (2, 3 rings).

Class 5: Aromatic components (>3 rings).

Table 5. Summary of tar measurements after i-MILENA gasifier.

	Tar concentration (mg/Nm³ dry producer gas)		
	Test 1	Test 2	Test 3
Class 2	32	15	181
Class 3 (excl. toluene)	6	0	100
Class 4	3300	4010	8939
Class 5	910	1507	2261
Total unknowns	1184	1304	3337
Total tar (excl. toluene)	5432	6836	14819

The gas contains tars, but compared to MILENA biomass gasification the tar content (±40 g/Nm^3) is low due to the lower volatile content of coal with respect to biomass. Further reduction of tar is expected to be possible, but some kind of tar removal will be required at commercial scale. ECN normally uses the OLGA tar removal technology to remove tar. The bottom ash and fly-ash from the combustor were free of carbon.

3.3 Gasification carbon conversion

Table 6 shows the results of carbon conversion of the high-ash coal and lignite during the i-MILENA gasification stage. Carbon conversion in the gasifier section of i-MILENA was determined from a molar balance using argon as tracer gas and the producer gas composition. As can be seen, carbon conversion in the gasification section ranges between 50% and 69%. It was found that within the accuracies of the measurements the required conversion is achievable for operation of the process at commercial scale.

Table 6. Summary of results of carbon conversion in the BFB gasification section.

	Test 1	**Test 2**	**Test 3**
Gasification Carbon Conversion (%)[1]	N.A.	57	69
Gasifier T (°C)	856	869	860

[1] Overall (gasification & combustion section of i-MILENA) carbon conversion was close to 100%

The sampled bottom ash and fly-ash from the combustor (sampled in a later test) were free of carbon, see **Fig. 5**.

Fig. 5: Bottom ash, sieved > 1mm.

3.4 Performance of OLGA tar removal system

During i-MILENA tests, the producer gas was directed to the OLGA system to test the performance of tar removal under high-ash coal gasification (high dust load) conditions. The tar removal principle of OLGA is based on a multiple stage scrubber in which the gas is cleaned by scrubbing oil. In the first section of OLGA (the collector) the gas is gently cooled down by the scrubbing oil. Heavy tar particles condense and are collected, after which they are separated from the scrubbing oil and can be recycled to the combustion section of the i-MILENA. In the second stage (the absorber/stripper), lighter gaseous tars are absorbed by the scrubbing oil. In the absorber column the scrubbing oil is saturated by light tars. This saturated oil is regenerated in a stripper. Hot air is used to strip the tars out of the scrubbing oil. This air loaded with light tars can be further used as combustion air in the combustor of the i-MILENA gasifier.

Results of OLGA tar removal are shown in **Fig. 6.** As can be seen, OLGA proved to be able to remove 90-92% of class 4 tars, 99-100% of class 5 (heavy) tars, and 80-86% of the total tar production. It must be noticed that the OLGA system used in these experiments was not optimized for the typical i-MILENA conditions. Removal values are expected to be higher in an optimized system.

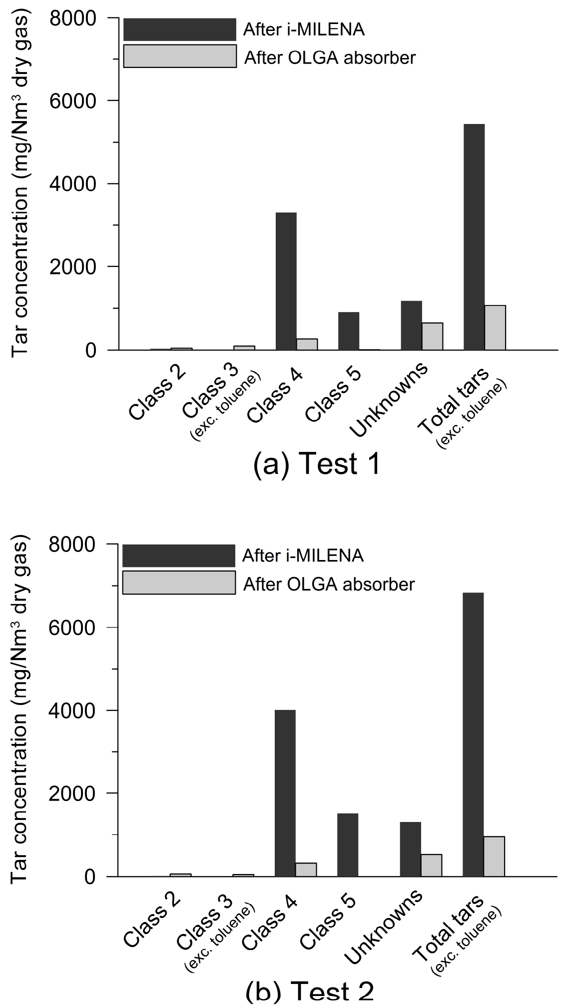

Fig. 6: Performance of OLGA tar removal system during i-MILENA tests with Indian high-ash coal.

4. Foreseen applications

At the moment there is no suitable commercial technology available for the gasification of high to very high-ash (> 35% wt.) low rank coal. ECN expects that the i-MILENA process becomes a commercial viable solutions for this type of coal for the following applications:

- (Co-)firing in boilers, scale > 1 $MW_{th.}$
- Gas engines for combined heat and power production, limited tar removal required, scale 2 – 20 MW_e.
- IGCC processes, tar removal required, scale > 6 MW_e.
- SNG production, preferably in combination with CCS, tar removal required, scale > 100 MW_{th}.

Because of the simplicity of the technology compared to conventional coal gasification processes it is expected that the technology can be used at relatively small scale. For small scale applications atmospheric operation is foreseen. For the large scale an operating pressure of 5 bar is foreseen, this reduces the size of the reactor.

With the technology maturing, the size of the installations will increase. It is foreseen that i-MILENA can be scaled up to 500 MW_{th}. At this scale, and larger, the amount of chemicals, methane and syngas are such that it is advantageous to develop a strategy to focus on co-production of chemicals, fuels and electricity. Removing some of the remaining impurities such as sulfur and chlorine components makes it possible to go for catalytic process, e.g. some of which are shown in **Fig. 7.**

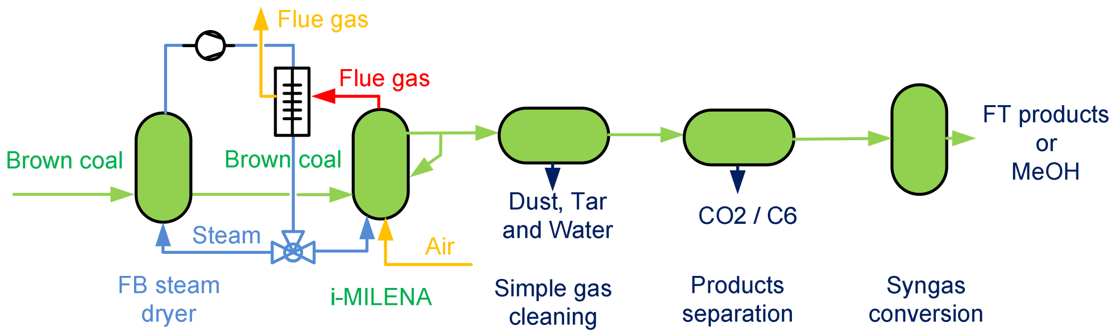

Fig. 7: (High ash) low rank gasification for synthesis of fuels & chemicals (100 – 1000 MWth).

5. Further development

A 800 kW_{th} pilot scale facility is available for further testing. ECN is looking for industrial partners to continue the development of the i-MILENA technology for (high ash) low rank coals. At the same time ECN is working on de scale-up for the MILENA gasifier for biomass with our commercial partner Dahlman (http://www.royaldahlman.com/renewable/). Several projects are under development ranging from 4 MW_{th} to 23 MW_{th} biomass / waste input.

6. Conclusions

The indirect i-MILENA gasification technology is a promising alternative for the efficient conversion of (high ash) low rank coal. Tests of high-ash sub-bituminous coal and low-ash lignite in the i-MILENA gasifier have shown that:

- Both low- and high-ash low rank coal (> 35% wt. ash) can be gasified in the i-MILENA gasifier with an overall complete conversion (white-grey ash in the flue gas cyclone), thus making ash disposal easier. In general, stable operation was achieved during the tests.
- Carbon conversion values in the gasification stage range between ~50-69%. The overall carbon conversion of the coal (gasification + combustion stages) is close to 100% and the calorific value of the gas is approximately 14 MJ/Nm^3 (HHV, dry).
- The OLGA system is able to remove the tars from the gas according to expectations.
- The technology is relatively simple due to the atmospheric operation of the gasifier, the production of a N_2-free syngas without the need for an air separation unit and the carbon-free residue that is produced. Steam consumption is relatively low, because the required conversion in the gasifier is relatively low, so there is

no need to maximize the steam to carbon ratio to accelerate the gasification reactions. The disadvantage of the atmospheric operation of the gasifier is the need for gas compression, but because of the high heating value of the gas the compression energy is much lower than would be required for a syngas containing no hydrocarbons.

- The lack of competing proven technology, especially in the medium scale range (1 - 500 MW_{th}), offers a good opportunity for the further development and demonstration of the technology for (very high ash, >35 wt% ash) low rank coal.
- Advanced high efficient coal gasification technology can contribute significantly to air quality improvement. If the technology is implemented well, CO_2 emissions can be reduced compared to coal combustion, especially when SNG is produced and the separated CO_2 is sequestrated.

7. Acknowledgements

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