

# Allothermal Gasification of High-Ash Coals

C.M. van der Meijden  
A. van der Drift  
B.J. Vreugdenhil  
A.J. Grootjes  
H. Bodenstaff  
G. Aranda Almansa

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## ALLOTHERMAL GASIFICATION OF HIGH-ASH COALS

Christiaan van der Meijden\*

Bram van der Drift

Berend Vreugdenhil

Sander Grootjes

Herman Bodenstaff

Guadalupe Aranda Almansa

Energy research Centre of the Netherlands, P.O. Box 1, 1755 ZG Petten, The Netherlands

\* Corresponding author: Phone: +31 224-564582, E-mail: [vandermeijden@ecn.nl](mailto:vandermeijden@ecn.nl)

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### ABSTRACT

The Energy research Centre of the Netherlands (ECN) has modified its biomass gasification set-up to make it suitable for high-ash coals. The modified gasifier is called the i-MILENA. The i-MILENA is an indirect gasifier. 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 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.

The experiments carried out with different types of high-ash and low-rank coals showed that the technology performs according to expectations. The next step in the development is the scale-up and demonstration of this technology at commercial scale.

## INTRODUCTION

Coal is still a major source of energy and is expected to continue to play an important role in the future energy infrastructure in the future as well. Coal resources are abundant compared to oil, but conventional conversion processes cannot compete with natural gas fuels plants, from an environmental point of view. Efficient and clean conversion of coal is required to be competitive with other fossil fuels. 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<sub>2</sub>. When this relatively pure CO<sub>2</sub> stream is sequestered, the CO<sub>2</sub> footprint can be reduced to the same level as natural gas.

Technology for the production of SNG from coal and lignite was demonstrated in the US in the 70's [1]. One large scale installation in North Dakota (U.S.) has been very successful until the recent drop in natural gas prices in the U.S. The CO<sub>2</sub> from this plant was used for Enhanced Oil Recovery (EOR). In China several new coal (lignite) to SNG installations are under construction at the moment [2].

Many countries have coal resources that are of very low quality. Ash concentrations of over 50wt% are not uncommon. This makes these reserves unsuitable for conventional combustion and gasification processes. At the moment, these coals are washed to separate the coal in a (very) high ash fraction and an usable fraction with a lower ash content. By this separation, part of the coal reserves are wasted. An alternative is to mix the high-ash coals with (imported) high-quality coals, to come to an acceptable average composition. Of course, this has a negative impact on the economics of a plant.

The Energy research Centre of the Netherlands (ECN) has developed a gasification technology called the MILENA technology [3]. The MILENA is an indirect gasifier for which 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 has been developed for biomass, but after a minor modification is suitable for high-ash coals and lignite as well. The modified MILENA gasifier is called the i-MILENA (inverse MILENA). An extensive test program was done to test different types of high-ash coals and lignite. The technology is now ready for further scale-up.

## **GASIFICATION**

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. Figure 1 shows the basic principles and differences of three types of fluidized bed gasifiers.

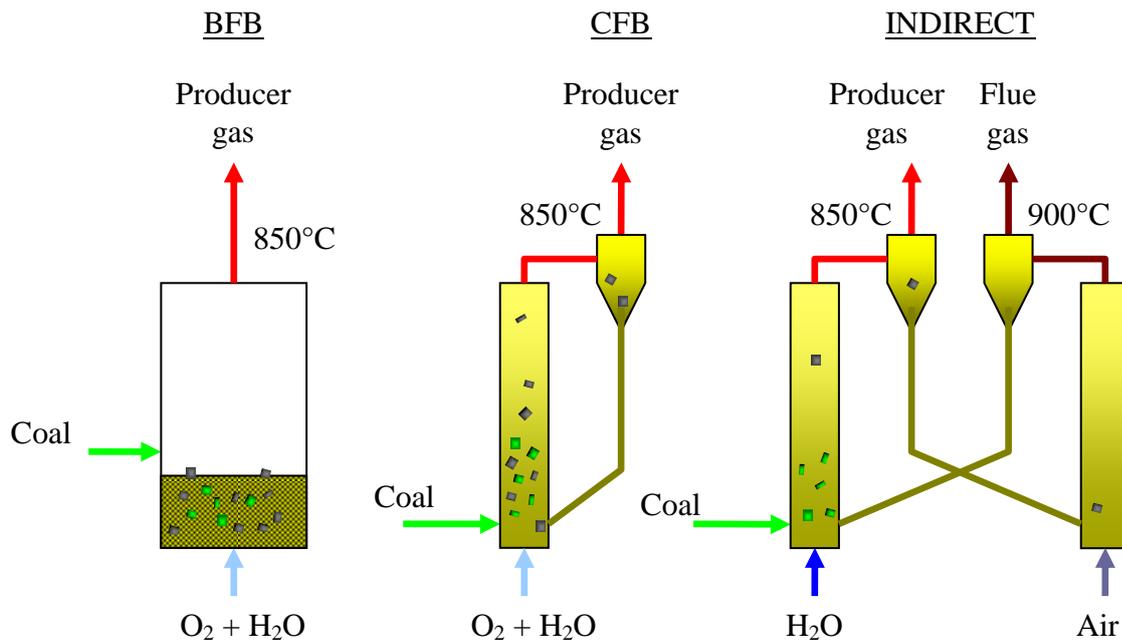


Figure 1: Schematic comparison of BFB, CFB and indirect Gasification.

In a BFB gasifier, the fuel is normally fed in or above the fluidized bed. The bed material is fluidized by a gas (air or an oxygen / steam mixture) entering the gasifier through nozzles distributed over the bottom of the reactor. The air is used in the bed to combust part of the gas and / or the char to produce the heat required for heating up the biomass and the endothermic gasification processes. The typical gas velocity in this gasifier is 1 – 2 m/s. BFB gasifiers are normally applied at a scale below 10 MW<sub>th</sub>. The reason for this scale limitation is probably the requirement of a good fuel distribution over the bed, which becomes more difficult when the gasifier is scaled up such that the diameter of the reactor increases.

At higher gas velocities, the bed material gets entrained and circulation of the bed material is required. This type of gasifiers is called Circulating Fluidized Bed (CFB). Typical velocities in the gasifier are normally between 3 and 10 m/s. The entrained bed material and the not completely converted fuel particles (char) are removed from the produced gas by a cyclone or another separation device. The particles are normally returned to the bottom of the gasifier via a 'non-mechanical' valve. This 'non-mechanical' valve can be a stand pipe which also serves the function of preventing gas leakage from the bottom of the gasifier riser into the solids outlet of the cyclone.

Separating the gasification of the biomass and the combustion of the remaining char creates the indirect or allothermal gasification process as shown in the right part Figure 1. The biomass fed to the 'gasifier' is converted into a gas and char (pyrolysis). The heat required for the heating of the biomass comes from the combustion reactor. This heat is transported by the circulating bed material. Char and bed material are separated from the gas by a solid gas separation device (e.g. a cyclone). The produced gas exits the gasifier to the gas cleaning system. The char and bed material are fed to the combustion reactor.

The char is combusted to produce the required heat for the gasification reactor. The heated bed material is returned to the gasifier reactor again.

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.

## **THE I-MILENA CONCEPT**

The MILENA gasifier was developed for the gasification of biomass (wood, etc.). The process is based on indirect or allothermal gasification. In one reactor the fuel is gasified or pyrolysed 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.

Figure 2 shows schematically the process. The coal is gasified in a bubbling fluidized bed (BFB) fluidized with steam at a temperature of approximately 930°C. The heat for the endothermic gasification reactions is added by the circulating hot bed material. The expected average residence time of the coal is in the order of 10 – 20 minutes. 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 combustor via a downcomer.

If the coal conversion is too high in the 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.

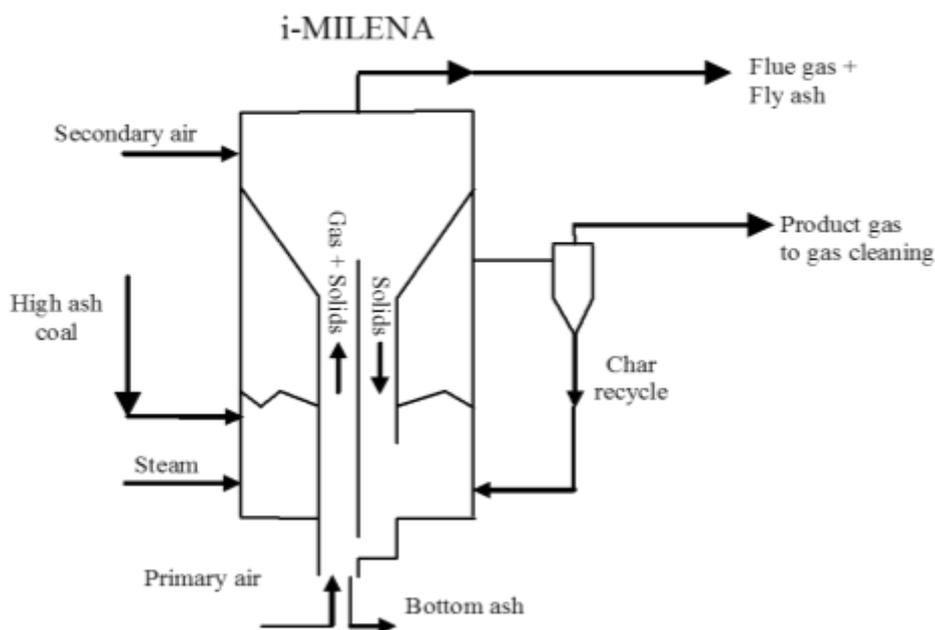


Figure 2: Simplified scheme of i-MILENA gasification process.

## EXPERIMENTAL SET-UP

The lab-scale i-MILENA gasifier is coupled to a lab-scale gas cleaning installation and a methanation unit. The gasifier and the gas cleaning operate at atmospheric pressure. Figure 3 shows the lab-scale installation.



Figure 3: Photos of 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: 2m. 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 coal properties are given in Table 1. The particles used in the lab-scale installation were milled and sieved. A fraction of 0.3 – 2 mm was used.

Table 1: Coal properties.

		High ash coal
moisture	[wt%]	3.9
volatile	[wt% dry]	21.2
ash	[wt% dry]	54
C	[wt% daf]	69.3
H	[wt% daf]	5.2
O	[wt% daf]	22.3
N	[wt% daf]	1.9
S	[wt% daf]	1.2
HHV	[kJ/kg daf]	27180

The main settings for the gasifier are given in Table 2. During the experiments the gas was sent to a combustor.

Table 2: Main settings during experiments.

	Settings
Coal feed (a.r.) [kg/h]	3.3
Steam [kg/h]	2
CO <sub>2</sub> [nl/min]	3
Primary combustion air [nl/min]	115

## EXPERIMENTAL RESULTS

An extensive test program was carried out in the lab-scale i-MILENA installation using Turkish and Indian coal with up to 50wt% of ash to check if the proposed concept is feasible. In this paper the results of the first tests with coal from India to prove the concept are described. After this tests the installation was slightly modified. The velocity in the BFB was increased to improve the fluidization of the relatively large ash particles. Figure 4 shows the changes in gas composition during the experiment. This first test was relatively short, because the installation was not yet equipped with a bottom ash removal system, so the larger ash particles accumulated in the system. This limited the duration of the test.

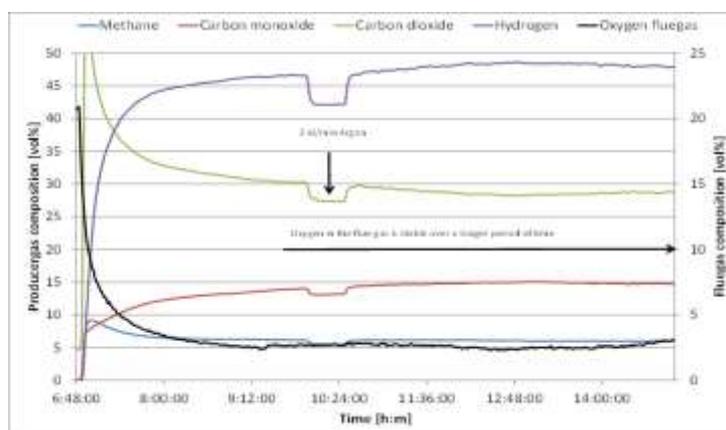


Figure 4: Gas composition during test with Indian coal

Table 3 shows the measured gas composition during the first two tests. The methane concentration is high compared to entrained flow gasification processes. This is beneficial for the conversion of the gas into SNG. Tars were measured during the second test only. During this test the gasification temperature was relatively low. It is expected that the tar content will drop when the gasification temperature is increased.

Table 3: Gas composition on dry basis and main process parameters.

	01-05-2012	02-05-2012
CO (vol%)	17.8	14.9
H <sub>2</sub> (vol%)	47.2	48.3
CO <sub>2</sub> (vol%)	28.1	28.5
CH <sub>4</sub> (vol%)	5.1	6.0
N <sub>2</sub> (vol%)	2.7	2.6
C <sub>2</sub> H <sub>y</sub> (vol%)	0.7	1.3
C <sub>6</sub> H <sub>6</sub> & C <sub>7</sub> H <sub>8</sub> (ppmv)	2900	3200
H <sub>2</sub> S & COS (ppmv)	4050	4900
Tar (mg/Nm <sup>3</sup> )	n.m.	9066
Combustor temp. (°C)	903	889
Gasifier temp. (°C)	888	853
O <sub>2</sub> combustor (vol%)	3.2	2.5
CO <sub>2</sub> combustor (vol%)	14.6	16.1

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. The required carbon conversion to have sufficient carbon in the combustor to generate the heat for the gasification process was calculated for the tested coal to be around 58 – 62%. From the measured flue gas and producer gas composition the carbon conversion was calculated. It was found that within the accuracies of the measurements the required conversion is achievable.

The measured gas composition showed that the heating value of the gas is relatively high. The gas contains tars, but compared to MILENA biomass gasification the tar content is low (typical value 40 g/nm<sup>3</sup>). Further reduction of tar is expected, but some kind of tar removal will be required at commercial scale. ECN normally uses the OLGA tar removal technology [4].

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



Figure 5: Bottom ash, sieved > 1 mm.

## FORESEEN APPLICATIONS

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

- (Co-)firing in boilers, scale > 1 MW<sub>th</sub>.
- Gas engines for combined heat and power production, limited tar removal required, scale 2 – 20 MWe.
- Combined Cycles using gas turbines for power production, tar removal required, scale > 6 MWe.
- SNG production, preferably in combination with CCS, tar removal required, scale > 100 MW<sub>th</sub>.

Because of the simplicity of the technology compared to the “standard” 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 bars is foreseen, this reduces the size of the reactor.

## FURTHER DEVELOPMENT

ECN will finalize the lab-scale experimental work in 2014. The steam to coal ratio will be minimized to reduce the steam consumption and other coal will be tested.

A 800 kW<sub>th</sub> pilot scale facility is available for further testing. ECN is looking for industrial partners to continue the development of the i-MILENA technology for low rank / high ash coals and lignite.

A the same time ECN is working on de scale-up for the MILENA gasifier for Biomass with our commercial partner Dahlman ([www.dahlman.nl](http://www.dahlman.nl)). Several projects are under development ranging from 4 MW<sub>th</sub> to 23 MW<sub>th</sub> biomass / waste input.

## CONCLUSIONS

The experimental program performed in the ECN lab-scale i-MILENA facility showed that this technology is suitable for conversion of high-ash coal into a combustible gas with a high efficiency. The carbon conversion of the coal is close to 100% and the expected calorific value of the gas is approximately 14 MJ/nm<sup>3</sup> (HHV, dry).

The technology is relatively simple due to the atmospheric operation of the gasifiers, the lack of pure oxygen as gasification agent 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 MWth), offers a good opportunity for the further development and demonstration of the technology for high ash coal (>35 wt% ash).

Advanced high efficient coal gasification technology can contribute significantly to air quality improvement. If the technology is implemented well, CO<sub>2</sub> emissions can be reduced compared to coal combustion, especially when SNG is produced and the separated CO<sub>2</sub> is sequestered.

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**ECN**

Westerduinweg 3  
1755 LE Petten  
The Netherlands

P.O. Box 1  
1755 LG Petten  
The Netherlands

T +31 88 515 4949  
F +31 88 515 8338  
info@ecn.nl  
www.ecn.nl