COAL GASIFICATION PILOT PLANT FOR HYDROGEN PRODUCTION. PART A: COAL GASIFICATION AND SYNGAS DESULPHURIZATION

Giovanni RAGGIO, Alberto PETTINAU^{*}, Alessandro ORSINI, Marcella FADDA SOTACARBO S.p.A.– Società Tecnologie Avanzate Carbone – c/o Centro Servizi C.N.I.S.I. 09010, Portoscuso (CA), ITALY

Daniele COCCO

University of Cagliari – Department of Mechanical Engineering – piazza d'Armi, 09123 Cagliari, ITALY

Paolo DEIANA

ENEA – Centro Ricerche della Casaccia – via Anguillarese 301, 00060 S.Maria di Galeria (RM), ITALY

> Maria Luisa PELIZZA, Massimo MARENCO Ansaldo Ricerche – corso Perrone, 25 – 16152 Genova, ITALY

ABSTRACT

Nowadays, the need for a diversification in primary energy sources has raised an increasing interest in coal, which allows a larger price stability and represents a reliable primary source from a strategic point of view.

Coal gasification processes, in particular, allow an efficient use of this energy source, with a low environmental impact. These processes, moreover, allow hydrogen (and other environmental-friendly fuels) and electric power co-production.

To this effect, Sotacarbo, together with Ansaldo Ricerche, ENEA and the Department of Mechanical Engineering of the University of Cagliari, is developing a pilot plant to test the use of gasification technologies for the combined production of hydrogen and electric power in medium and small scale commercial plants. In particular, the pilot plant is made up of two up-draft fixed-bed gasifiers, respectively 700 and 35 kg/h of coal, the latter equipped with a syngas treatment line, including the depulverization, desulphurization, water-gas shift conversion, CO₂ separation and hydrogen purification sections.

This paper reports the main results, achieved by a calculator simulation, of a preliminary analysis carried out to assess the main operating parameters of the gasification process and the conventional syngas desulphurization section. In particular, the effects of a number of the main process operating parameters on the gasification system and on downwards treatment sections have been analyzed. The gasification air enrichment with oxygen is, among the parameters, particularly interesting.

Key-Words: Coal gasification, Sulcis coal, Pilot plant, CO-shift, CO₂ removal, Hydrogen production

^{*} Corresponding author:

Phone: +39 0781 509047 – Fax: +39 0781 508349 E-mail: apettinau@sotacarbo.it

NOMENCLATURE

- ESP Electrostatic Precipitator
- IGCC Integrated Gasification Combined Cycle
- LHV Lower Heating Value
- MDEA Methyl-diethanolamine, (HO-CH₂-CH₂-)₂ N-CH₃
- MEA Monoethanolamine, HO-CH₂-CH₂-NH₂
- PSA Pressure Swing Adsorption
- WGS Water-Gas Shift conversion process

INTRODUCTION

Nowadays, the need to release energy production from the use of oil and natural gas as primary energy sources and, in general, to diversify such sources in order to assure the supplying, is making coal more and more interesting. This fuel, widely available in the world and distributed more uniformly than oil and natural gas, allows a great price stability and represents a secure source from a strategic point of view [1].

Moreover, the increasing interest in environmental problems has recently led to the development of clean coal technologies, designed to enhance both the efficiency and environmental acceptability of coal extraction, preparation and use, in particular for power generation [2].

Among clean coal technologies, gasification is particularly interesting since it allows both power generation (in Integrated Gasification Combined Cycles power plants, IGCC) and environmental-friendly fuel production, with a particular reference to hydrogen.

All over the world, gasification processes, due to the low flexibility of synthesis gas (syngas) production, are, so far, mainly used in large-scale IGCC power plants in order to supply base energy load. But in a short-term future, the possibility to use syngas to produce hydrogen could make gasification technologies very interesting for medium and small-scale industrial application.

As to this possibility, Sotacarbo, together with Ansaldo Ricerche, ENEA and the Department of Mechanical Engineering of the University of Cagliari, is developing an integrated gasification process for combined production of hydrogen and electrical energy, to be used in medium and small-scale commercial plants. The research project concerns the development of a gasification and syngas treatment pilot plant, which will be located in the Sotacarbo Research Centre at Carbonia, near Cagliari. The plant includes a pilot-scale (700 kg/h) coal gasifier and a laboratory-scale (35 kg/h) coal gasifier; in particular, the latter is equipped with a syngas treatment process for hydrogen production. The research project is co-funded by the Italian Ministry of Education, University and Scientific Research (MIUR) and the total cost is estimated in 11 million euros.

This paper reports the main results of the preliminary analysis and the estimated performances, based on a computational simulation, of the gasification and cold syngas desulphurization sections. The preliminary analysis of the syngas treatment section, based on the hot desulphurization, CO shift-conversion, CO_2 removal and H_2 separation processes, is reported in the second part of this work [3].

PILOT PLANT CONFIGURATION

The simplified flow scheme of coal gasification process considered by Sotacarbo for hydrogen production is shown in figure 1. The pilot plant includes a fixed-bed up-draft gasifier, a water scrubbing unit, a sulphur compound removal process, a CO-shift conversion process integrated with a CO_2 separation system, a hydrogen separation section and, finally, the power generation section.

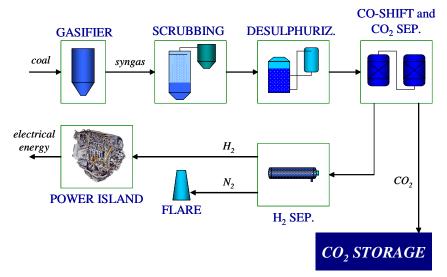


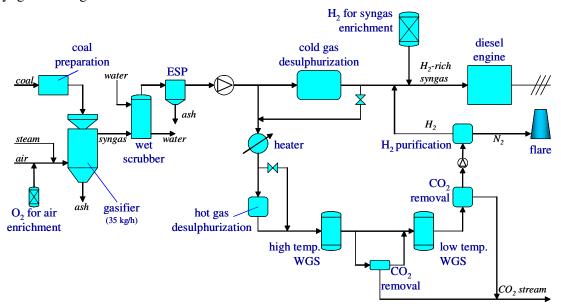
Figure 1 – Simplified scheme of the Sotacarbo process.

As already mentioned, on the basis of the previous flow scheme, Sotacarbo and the other partners of the project are currently developing the final design of the overall pilot plant, which will be used for the R&D activities required by the development and the optimization of the different sections of the integrated process.

In order to test different plant solutions and different operating conditions, during this first phase of the research project a very flexible and simple layout for the pilot plant has been considered. The experimental results obtained by means of this pilot plant will constitute the basis of the second phase of the research project, which will lead to the realization of the demonstrative unit of the Sotacarbo coal-to-hydrogen process.

The currently layout of the Sotacarbo pilot plant includes two fixed-bed up-draft Wellman-Galusha gasifiers: a 700 kg/h pilot gasifier and a 35 kg/h laboratory-scale gasifier; both gasifiers are equipped with a wet scrubber for syngas cooling (to about 50 °C) and dust and tar removal. In particular, the 700 kg/h pilot gasifier will only be used for the optimization of the gasification technology, especially with reference to the operating conditions (i.e. steam/coal mass ratio, oxygen/coal mass ratio, oxygen purity, etc.). On the other hand, the 35 kg/h laboratory-scale gasifier will be equipped with the overall syngas treatment process, in order to produce the hydrogen for the power generation section.

Figure 2 shows the simplified scheme of the Sotacarbo laboratory-scale coal gasification plant. As mentioned, the coal gasification section includes the gasifier and the wet scrubber, as well as a wet electrostatic precipitator (ESP), which allows to achieve a fine particulate and tar removal (the design concentrations are 1000 mg/Nm³ for the particulate and 200 mg/Nm³ for the tar). The clean gas leaves the ESP at atmospheric pressure and, by



means of a compressor which increases the pressure to about 1.4 bar, feeds the downstream syngas treating sections.

Figure 2 – Simplified scheme of Sotacarbo laboratory-scale experimental plant.

According to the design conditions, downstream the compressor, the syngas is splitted into two streams: the main stream, about 80% of the produced syngas, is sent to a cold gas desulphurization process, whereas the secondary stream, that is the remaining 20% of the produced syngas, is sent to a hot gas desulphurization process, which is followed by the hydrogen production section. In particular, the cold gas desulphurization process is based on a hydrogen sulphide (H_2S) absorption process (which uses metil-dietanolamine, MDEA, as sorbent) and it is directly followed by the power generation section, represented by an internal combustion engine (diesel cycle). The secondary syngas treatment line includes a dry hot gas desulphurization process (which employs metal oxide-based sorbents) followed by an integrated CO-shift and CO₂ absorption system and a hydrogen purification system, based on the PSA (Pressure Swing Adsorption) technology. In order to test the operation of the diesel engine with the overall clean gas produced by the coal gasification process, the coal gas desulphurization unit has been designed for the overall syngas mass flow. On the other hand, the size of the secondary syngas treatment line, even if much smaller than the size of commercial scale plants, should give reliable experimental data for the scale-up of the future plants.

The laboratory-scale gasifier is designed to operate the gasification with enriched air (simply by using an oxygen tank). Moreover, the possibility to test the internal combustion engine with hydrogen enriched mixtures has been considered. In order to produce hydrogen enriched fuels, the hydrogen produced by the hot gas treatment line can be mixed with the clean syngas from the cold gas desulphurization process; otherwise, it is possible to operate the hydrogen enrichment simply by using a hydrogen tank located upstream the diesel engine.

Furthermore, a suitable portion of the clean syngas produced by the cold desulphurization process can be splitted upstream the diesel engine and fed to the integrated CO-shift and CO_2 absorption system, in order to compare the performances of both cold and hot syngas desulphurization processes for hydrogen production.

In order to ensure a full plant flexibility, as well as simplify the management of the experimental pilot plant, the different cooling and heating devices are not fully integrated. However, the aforementioned layout, if necessary, can be easily modified without significant costs.

COAL GASIFICATION SECTION

As already mentioned, the pilot plant includes two up-draft fixed-bed Wellman-Galusha gasifiers, developed and manufactured by Ansaldo Ricerche S.p.A.

Coal, which will be bought in big bags for the experimental tests, is drawn out from the storage area through a heaver and, through a tackle, charged in a particular hopper in order to empty big bags. Then, coal is drawn out from this hopper, weighted and sent to the two gasifiers through a conveyer belt.

The gasifier (figure 3) is characterized by four main operating zones, where the coal drying, devolatilization, gasification and combustion processes take place. As the coal flows downwards, it is heated by the hot raw gas that moves upwards, coming from the gasification and combustion zones [4-5]. The gasification agents (air and steam) are introduced into the reactor near the bottom, so that they are pre-heated by cooling the bottom ashes, which are removed through the coal grate.

In order to distribute the coal as uniformly as possible and optimize the gasification process, the gasifier is equipped with an internally cooled stirrer (in order to keep a low metal temperature), which is characterized by two degrees of freedom: an axial rotation and a vertical translation. Furthermore, the gasifier is equipped with a cooling water jacket, in order to operate an accurate temperature control.

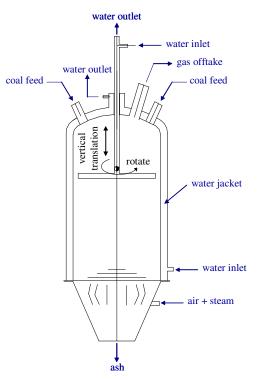


Figure 3 – Simplified scheme of 700 kg/h pilot gasifier.

Since the 700 kg/h pilot gasifier does not include the syngas desulphurization section, it will be only fuelled with low sulphur coals (with a sulphur content lower than $0.5\div0.6$ % wt.). As already mentioned, the pilot gasifier will be used to set up the gasification technology and to develop an automatic process for plant regulation and control, required to scale-up and commercialize the gasification process.

The 35 kg/h laboratory-scale gasifier, instead, will be fuelled by several coals and, in particular, with high-sulphur Sulcis coal, whose ultimate analysis is shown in table 1. Moreover, in the laboratory-scale gasifier, the possibility to enrich the gasification air with oxygen will be investigated.

Ultimate analysis [% wt.]	
Carbon	53.17
Hydrogen	3.89
Nitrogen	1.29
Sulphur	5.98
Oxygen	6.75
Chlorine	0.10
Moisture	11.51
Ash	17.31
LHV [MJ/kg]	20.83

Table 1 – Sulcis coal ultimate analysis.

The performances of the gasification section and the preliminary design of the main syngas treating equipments have been carried out starting from a gasification equilibrium model, developed by the Department of Mechanical Engineering of the University of Cagliari and implemented with Fortran language [6].

Input data		
Sulcis coal mass flow [kg/h]	35.0	
Steam/coal mass ratio	0.26	
O_2 /coal mass ratio	0.44	
O ₂ molar fraction	0.2073	
Dry-based syngas composition		
СО	0.3103	
CO_2	0.0254	
H ₂	0.1838	
N ₂	0.4302	
CH_4	0.0297	
H ₂ S	0.0148	
COS	0.0008	
Ar	0.0051	
H ₂ O	0.0763	
Gasifier performances		
Syngas mass flow [kg/h]	105.12	
Syngas volumetric flow [Nm ³ /h]	102.2	
Syngas LHV [MJ/kg]	6.6414	
Cold gas efficiency	39.65	
Syngas outlet temperature [°C]	288.5	
Gasifier yield [Nm ³ /kg]	2.70	

Table 2 – Main gasification parameters for Sulcis Coal.

The simulation model allows to estimate the syngas composition and the main gasifier performances according to coal composition and the main operating parameters of the gasification section. Table 2 shows the syngas composition and the gasifier performances here assumed as design conditions. In particular, table 2 refers to the use of Sulcis coal, atmospheric air (characterized by an oxygen molar ratio of 0.2073) as an oxidant agent and to an equilibrium temperature of 1000 °C in the gasification and combustion zone of the gasifier.

Coal gasification with air enrichment

As already mentioned, the 35 kg/h laboratory-scale gasifier will be equipped with an air enrichment system using pure oxygen in order to test the performance of the coal gasification process with increasing values of the oxygen content. The performance evaluation of the laboratory-scale gasifier has been carried out by means of the previously described equilibrium model, even with reference to the Sulcis coal of table 1. Figure 4 shows the mass flow and the LHV of the raw gas produced by the gasifier as a function of the oxygen purity (that is the oxygen molar fraction of the air fed to the gasifier); similarly, figure 5 shows the CO, H₂ and CH₄ molar fraction even as a function of the oxygen purity. Figures 4 and 5 refer to the same steam/coal mass ratio (equal to 0.26), whereas the oxygen/coal mass ratio has been decreased in order to keep the coal gas temperature constant (to 288 °C).

Figure 4 shows that, as expected, the coal gas mass flow decreases by enriching the air stream, due to the absence of the diluting nitrogen; for the same reason, the LHV of the coal gas increases with the oxygen purity. In particular, a remarkable mass flow reduction, by about 35-40%, can be achieved by enriching the air up to 40-45% of oxygen, with an LHV of about 9.5-10 MJ/kg. Obviously, as shown in figure 5, the air enrichment leads to the increase of the H₂, CO and CH₄ contents of the produced coal gas.

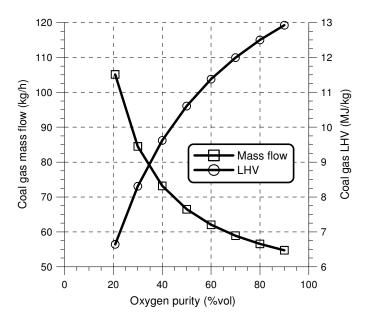


Figure 4 – Mass flow and LHV of the coal gas as a function of the oxygen purity.

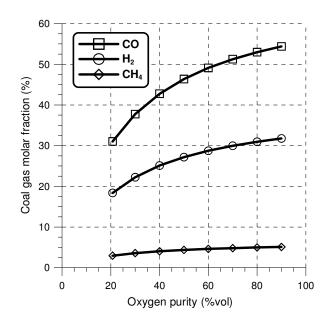


Figure 5 – Molar fraction of CO, H_2 and CH_4 as a function of the oxygen purity.

Finally, figure 6 demonstrates that the coal gasification process carried out with enriched air requires lower values of the oxygen/coal mass ratio, mainly due to the lower mass flow, and also to the lower sensible heat at the outlet of the gasifier; therefore, the use of enriched air also requires lower values of the air/coal mass ratio.

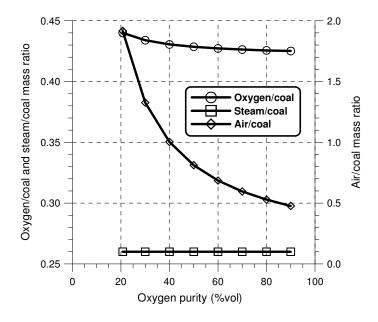


Figure 6 – Oxygen/coal, steam/coal and air/coal mass ratios as a function of the oxygen purity.

COLD SYNGAS DESULPHURIZATION PROCESS

According to the design conditions, 80% of the depulverized syngas produced by the laboratory-scale gasifier is sent to the cold gas desulphurization section, in order to reduce the

hydrogen sulphide concentration down to the maximum values allowed by the CO-shift conversion processes. As a matter of fact, the catalysts commonly used by the CO-shift reactors can be poisoned by H_2S concentrations higher than about 100 ppm (that is higher than the typical emission standards). On the other hand, the lower amounts of the COS (carbonyl sulphide) are mainly removed by means of the raw gas wet scrubber.

The cold syngas desulphurization system here considered (shown in figure 7), is based on a chemical-physical H_2S absorption, carried out by a mixture of water and MDEA (methyl-diethanolamine), which is highly selective for hydrogen sulphide.

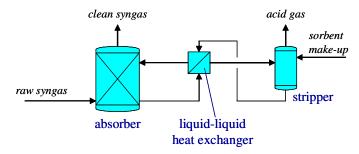


Figure 7 – Simplified scheme of cold syngas desulphurization system.

The absorption process is mainly governed by the H₂S chemical dissociation:

$$H_2S \Leftrightarrow H^+ + HS^-$$

which is highly influenced by the absorption pressure and temperature, as well as by the pH value of the solvent solution (that is by the MDEA concentration). The MDEA captures the H^+ ions released by the H_2S dissociation, according to the following reaction:

$$MDEA + H^+ \Leftrightarrow (MDEA)H^+$$

which promotes the H_2S dissolution. The previous reactions are highly reversible and the ionic species remain in solution without generating stable compounds.

The saturated sorbent exiting from the bottom of the absorber is sent to the stripping column at about 90 °C. The temperature increase allows to release H^+ ions, which react with the HS⁻ ions, thus leading to the formation of the H₂S. Water and sorbent vapours are separated (through a condenser) from the H₂S-rich gas stream in order to reduce the sorbent make-up. Finally, the acid gases are sent to a proper treatment system [7-8].

The preliminary analysis of the cold syngas cleaning process has been carried out by using the ChemCAD 5.2 simulation software [9]. In particular, raw syngas is saturated and depulverized in the wet scrubber and is subsequently sent to the absorber. This is considered as a plate column, which operates at about 30 °C, in which syngas (about 80 kg/h) has a countercurrent contact with a solvent, composed by MDEA (50 %) diluted in water. The regeneration process takes place in a stripper, which is a plate column equipped with a condenser (in order to reduce the solvent make-up) and a reboiler (in order to maintain the regeneration temperature at about 100 °C). In order to optimize the power consumption in the reboiler, according to the simplified scheme shown in figure 7, the regeneration system is equipped with a liquid/liquid heat exchanger, in which the lean solution (from the stripper) is

cooled by heating the rich solution (from the absorber). The main operating parameters of the H_2S removal process are shown in table 3.

H ₂ S absorption process		
Raw syngas mass flow [kg/h]	81.297	
H2S concentration in feed syngas [mole %]	1.295	
Absorption temperature [°C]	30	
Regeneration temperature (reboiler) [°C]	~100	
Minimum ΔT in liquid/liquid heat exchanger [°C]	10	
Solvent composition:		
MDEA	50%	
Water	50%	

Table 3 – Main cold syngas desulphurization operating parameters.

Figure 8 shows the expected H_2S removal efficiency in function of the liquid/gas mass ratio. The analysis has been carried out with different values of the reboiler power consumption, by considering the syngas produced by the air-blown gasification process (that is without any air enrichment).

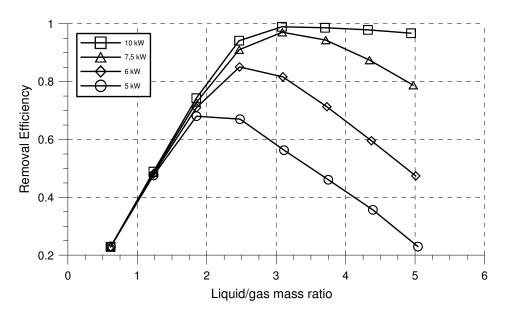


Figure $8 - H_2S$ *removal efficiency.*

The analysis shows that, for a given power consumption of the reboiler, the increase of the liquid/gas mass ratio involves an increase of the absorption efficiency, a maximum value, which is followed by a reduction. As a matter of fact, by increasing the liquid mass flow, a reduction of the stripped gas mass flow takes place, with a consequently pH reduction in the lean solution and a decrease of the H_2S separation efficiency.

On the other hand, the increase of the stripper power consumption involves an increase of the removal efficiency. In particular, a power of about 10 kW and a liquid/gas mass ratio of about 3 are required in order to achieve the design desulphurization efficiency (about 99.5%).

Moreover, a further increase of the power consumption allows only to achieve a little improvement of the desulphurization efficiency.

Acid gas treatment system

In conventional IGCC power plants, the acid gases exiting from the stripping system are treated by means of a Claus-SCOT process in order to recover almost pure sulphur [10]. However this process, due to its high capital cost, is not suitable for an experimental plant, as well as for small-scale commercial applications.

Therefore, Sotacarbo, in its research project, has considered a different acid gas treatment process, where sulphur compounds are separated by producing sulphuric acid. In particular, the H_2S -rich gas is firstly fed to a combustor, where the H_2S is converted to SO_2 ; the flue gases are sent to a proper reactor, where the SO_2 is catalytically converted to SO_3 . Finally, the gas is sent to a water scrubber, where the SO_3 reacts with water to form sulphuric acid, according to the following chemical reaction:

$$SO_3 + H_2O \Leftrightarrow H_2SO_2$$

This treatment process is very similar to that used by the SNOXTM process, developed for the desulphurization of the flue gases produced by conventional combustion power plants [11]. The main advantages of this technology are the low capital costs and the simple plant management.

CONCLUSIONS

The increasing interest in hydrogen production from low rank fuels through gasification processes has led Sotacarbo, together with Ansaldo Ricerche, ENEA and the Department of Mechanical Engineering of the University of Cagliari, to develop an integrated gasification process for the co-production of hydrogen and electrical energy, to be used in medium and small-scale commercial plants. In this framework, a research project, concerning the development of a gasification and syngas treatment pilot plant, has been funded by the Italian Ministry of Instruction, University and Scientific Research. The pilot plant will be located in the Sotacarbo Research Centre at Carbonia, near Cagliari.

In particular, the pilot plant, which will be composed by two gasification sections (a pilot 700 kg/h gasifier and a laboratory-scale 35 kg/h gasifier) and by a syngas treatment line, will be used to carry out a number of experimental tests in order to choose the most suitable technologies and optimize each section and the integrations of the overall plant.

As to the gasification section, this paper reports the effects of the oxidant composition (in particular the oxygen concentration) with respect to syngas properties and in the gasification operating parameters (such as oxygen/coal, air/coal and steam/coal mass ratios). In particular, a remarkable syngas mass flow reduction, of about 35-40%, can be achieved by increasing the oxygen purity beyond 40-45%, with a LHV increase to about 9.5% and with a rise of the H_2 , CO and CH₄ concentration in the raw syngas. Moreover, the air enrichment requires a careful regulation of the oxygen/coal and steam/coal mass ratios in order to maintain constant the gasifier outlet temperature.

Finally, as far as the conventional desulphurization process is concerned, the influence of the liquid/gas mass ratio on the H₂S removal efficiency has been evaluated. In particular, a

power of about 10 kW and a liquid/gas mass ratio of about 3 are required in order to achieve the design desulphurization efficiency.

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