Click here to return to Contents Page

CCT 2005
Second International Conference on Clean Coal Technologies for our Future
10-12 May 2005 • Castiadas (Cagliari), Sardinia, Italy • www.cct2005.it

# SOTACARBO R&D project for hydrogen and clean fuels production from coal gasification and $CO_2$ removal

*Carlo Amorino, Enrico Maggio, Alessandro Orsini, Francesco Repetto, Maria Luisa Pelizza, Federica Pratola, Giuseppe Girardi, Antonio Calabrò, Giorgio Cau, Daniele Cocco*

# SOTACARBO R&D project for hydrogen and clean fuels production from coal gasification and $CO_2$ removal

Carlo Amorino; Enrico Maggio, Alessandro Orsini

(Sotacarbo SpA – Centro servizi Consorzio NISI, Portoscuso)

Francesco Repetto; Maria Luisa Pelizza, Federica Pratola

(Ansaldo Ricerche SpA– Corso Perrone 25, Genova)

Giuseppe Girardi; Antonio Calabrò

(ENEA - La Casaccia, Via Anguillarese, 301 Roma)

Giorgio Cau; Daniele Cocco

(Dipartimento di Ingegneria Meccanica, Piazza D'Armi, Cagliari)

**10-12 May 2005**

Castiadas (Cagliari), Sardinia, Italy

## ABSTRACT

Increasing interest in hydrogen production and availability of huge world coal reserve are making the possibility to produce hydrogen from coal gasification.

To this end, Sotacarbo in co-operation with Ansaldo Ricerche, ENEA and the University of Cagliari – Department of Mechanical Engineering, has developed a research project for design, construction and testing on a pilot plant for hydrogen and other environmental value fuel gas production, from Sulcis coal gasification. The project has been funded by Ministry of Education, University and Research (MIUR). The test facilities will be located in the Sotacarbo Research Centre, which is under construction in Carbonia, South East Sardinia (Italy).

The test facility is designed to increase the environmental valency of coal through the integration of gasification and suitable downstream syngas treatment sections for power and hydrogen enriched stream production which is employable as energetic carrier itself or into the internal combustion engine as first attempt or potentially into fuel cells.

The main purpose of the R&D Project is the development and application of innovative technologies for carbon dioxide separation and hydrogen enrichment sections together with the application of known technologies ( such as particulate, tar and sulphur compounds removal) to syngas streams.

## INTRODUCTION

The production of hydrogen from coal syngas is considered a pre-combustion $CO_2$ capture technology that removes the carbon content of fossil fuels and produces a $CO_2$-rich by-product stream. In this approach, coal is gasified to produce a syngas that, after cleaning up, has high concentration of $CO_2$ and $H_2$. Carbon monoxide is reacted with steam in a catalytic shift conversion reactor to produce $CO_2$ and more $H_2$. This system can be used for both $H_2$ production (or other clean fuel such as Methanol and, with increasing interest, Dimethyl-Ether) for electricity generation and high concentration $CO_2$ capture.
The aim of the present research project is studying, developing and tuning processes for production and treatment of coal syngas through design, construction and testing of a pilot platform fed with Sulcis and imported coal.

The project has been partially funded, in December 2003, through the national law 297/99, by MIUR, the Italian Ministry of Education, University and Research.
The total cost are estimated in about 11,36 million of Euro shared between the partners.

After preliminary activities carried out in the last months of 2003, the project practically started in January 2004 and the conclusion is planned within 2008. Actually it is being performed by a partnership made up of Sotacarbo, as project coordinator, with the collaboration of Ansaldo Ricerche, ENEA and the University of Cagliari - Department of Mechanical Engineering.

**Sotacarbo** is a limited company established in 1987, in accordance with the Italian law 351/85, which aims to develop new and advanced clean coal technologies. The present shareholders are ENEA and the Sardinia regional government and since 1989 the company has represented Italy in the international organization IEA The Clean Coal Centre.

Sotacarbo is constructing, together with Carbonia town Administration, a new research centre in the former coal mining area of Serbariu, near Carbonia, Southern Sardinia, Italy. This will be the Italian reference point for the research of new technologies for coal utilisation.

**Ansaldo Ricerche (ARI)** is a high tech engineering company based in Genoa. ARI develops, tests, evaluates and optimises new and intelligent technologies which operate reliably, economically and in an environmentally friendly way. Among other research topics of concern, one of the main activities at ARI focuses on solid fuel processing through combustion/gasification with integrated innovative technologies.

**ENEA** is an Italian Agency for New technologies, Energy and Environment. With its Energy Department and Energy Plant Processes Division, ENEA develops advanced technologies for energy production with high efficiency and low environmental impact, and carries out test and check activities in operating industrial plants.

**DIMECA** is the Department of Mechanical Engineering of Cagliari University. DIMECA carries out fundamental and applied researches on mechanical and industrial engineering through both consolidated methodology of experimental studies and numerical techniques for industrial design, analysis and simulation.

## GENERAL OBJECTIVES

The MIUR research project, in addition to the design, construction and testing of a pilot platform, also includes:

- the construction of experimental structures of coal technologies Research Centre able to accommodate analytical laboratories and the pilot platform,
- a high level training activities for technicians, engineers and researchers to be utilised in the Research Centre.

**Sotacarbo Research Centre**

The Coal Research Centre will be locate in an existing building of Serbariu former coal mine, that Carbonia town Admisitration is restoring. In particular the Centre will accommodate the seat of Sotacarbo company, analytical laboratories, the pilot platform and future test plants.
Sotacarbo Research Centre have a covered area of about 2500 $m^2$ in a land of about 1 hectare and can accommodate 800 $m^2$ for offices, 840 $m^2$ for Common facility area (conference hall, exhibition rooms etc), 340 $m^2$ for Mechanic shop area and then 885 $m^2$ for arranging 16 analytical laboratories.

**Researcher training**

In order to train researchers and technicians in the sector of coal technologies, to be utilised in research and development projects, Sotacarbo organised and managed, with the collaboration of Cagliari University and the support of specialised companies, a high training course dressed to selected graduate and qualified people living in Sardinia. The training course, which is started in March 2004 and finished in November 2004, allowed to individuate 8 researchers and 5 technicians that will take on the company .

**Pilot platform description**

The present phase of the research project focuses its attention on design, contruction and demonstration of a pilot platform tailored for increasing environmental fuel quality through coal gasification and syngas treatment process for power and hydrogen production.
The proposed experimental facility consists of an integrated system made up of two main plants:

1. Pilot scale gasification system
2. Integrated lab scale power cycle system

The first includes:

- pilot scale coal gasifier (700 kg/h),
- syngas wet scrubber
- flare.

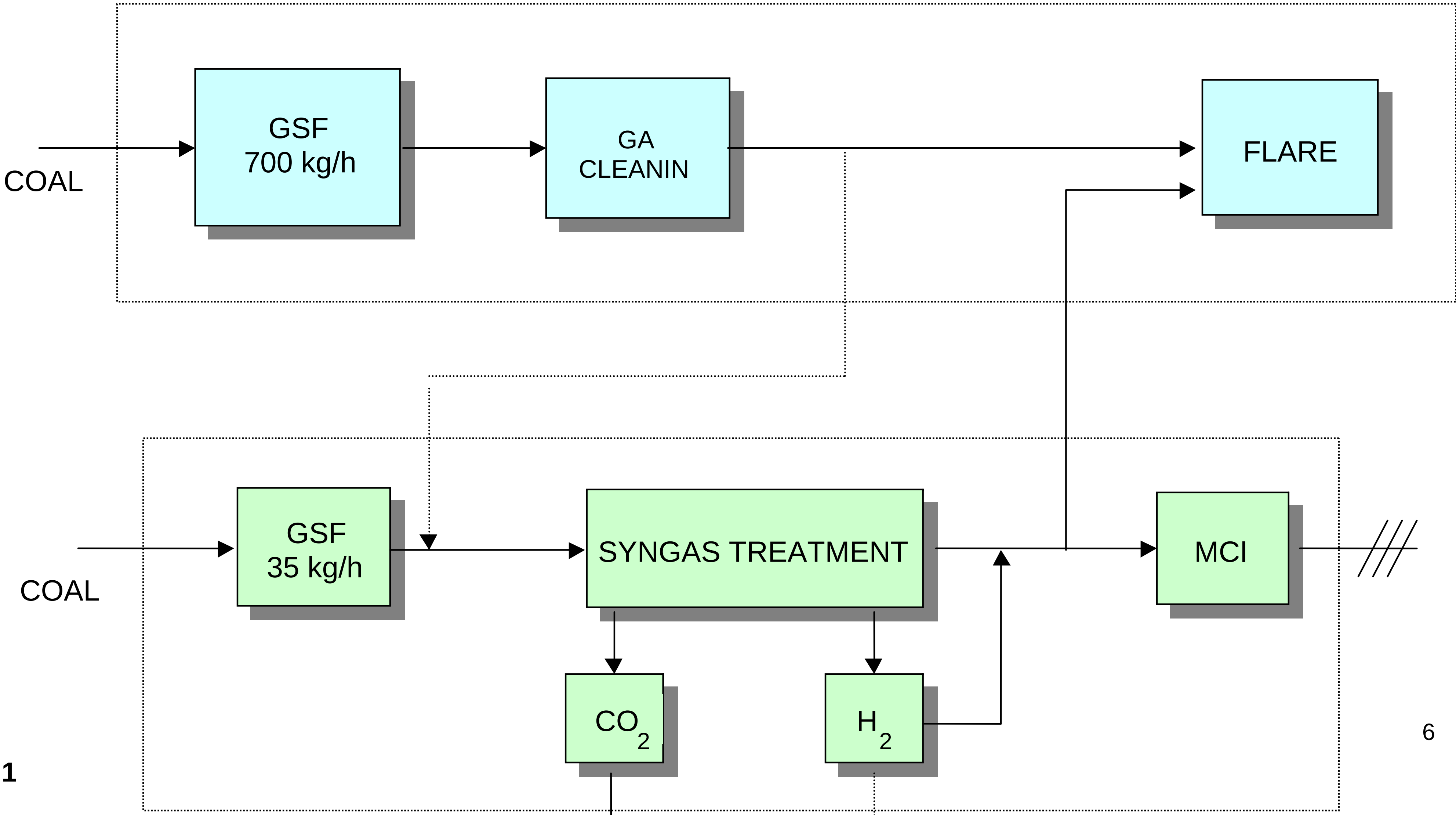
The latter consists of the following sections:

- lab scale gasifier (35 kg/h),
- fly ash and tar removal,
- desulphurisation,
- shift conversion,

- carbon dioxide separation,
- hydrogen purification
- internal combustion engine.

The lab and the pilot scale plants are both located in the platform but they can operate independently one to the other. (fig. 1 )

**PILOT PLATFORM**




6

**Fig. 1**

The pilot scale gasifier design is optimised in terms of fluid dynamics, feeding system, ash removal system and inlet automation, in order to achieve an efficient gasification of coal. Its size makes it suitable for process optimisation and calculation of scaling up evaluation, but unsuitable for ease and flexible experimental operation.

The lab scale plant is designed to achieve a representative syngas composition of the one coming out from the pilot scale gasifier, and allows a more flexible plant management at lower operating costs. The laboratory scale gasifier results hence of particular interest for:

- testing and implementation of the down stream syngas cleaning system (particulate and tar removal, desulphurization),
- testing and implementation of hydrogen purification section (shift conversion and carbon dioxide separation)
- experimental data collection.

In the pilot platform, the environmental performances of the overall system are optimised in terms of noise and vibration pollution and exhaust/effluent gas and liquid blown or drained into the ambient.

**Pilot scale gasification system (700 kg/h)**

In this plant , an updraft, countercurrent pilot scale gasifier (700 kg/h), based on fixed bed Wellman Galusha technology, has being designed for coal feeding. The gasifying agent (air) is introduced from the reactor bottom and flows upward toward the top. The solid fuel is uniformly fed from the reactor top through a multipoint injection, moves downwards and is extracted from the bottom. A diagram of an updraft gasifier is presented in Fig. 2. Four sequential processes occurs in the reactor on going from the top to the bottom: drying, pyrolysis, gasification and combustion. The latter process, consisting in the partial combustion of the feed with the oxygen into the air stream, is needed to sustain the endothermic processes taking places in the upper part of the reactor (i.e. gasification, pyrolysis and drying), giving the energy necessary to sustain the gasification. Water contained in the feed leaves the top of the reactor as a fraction of gaseous compound produced. The mixture of air and steam introduced in the reactor bottom is uniformly distributed throughout the reactor by a moving grate, which also ensure the collection of ashes at the reactor bottom. The gaseous stream flowing upwards in the reactor gets richer in gases produced by gasification of the solid fuel.

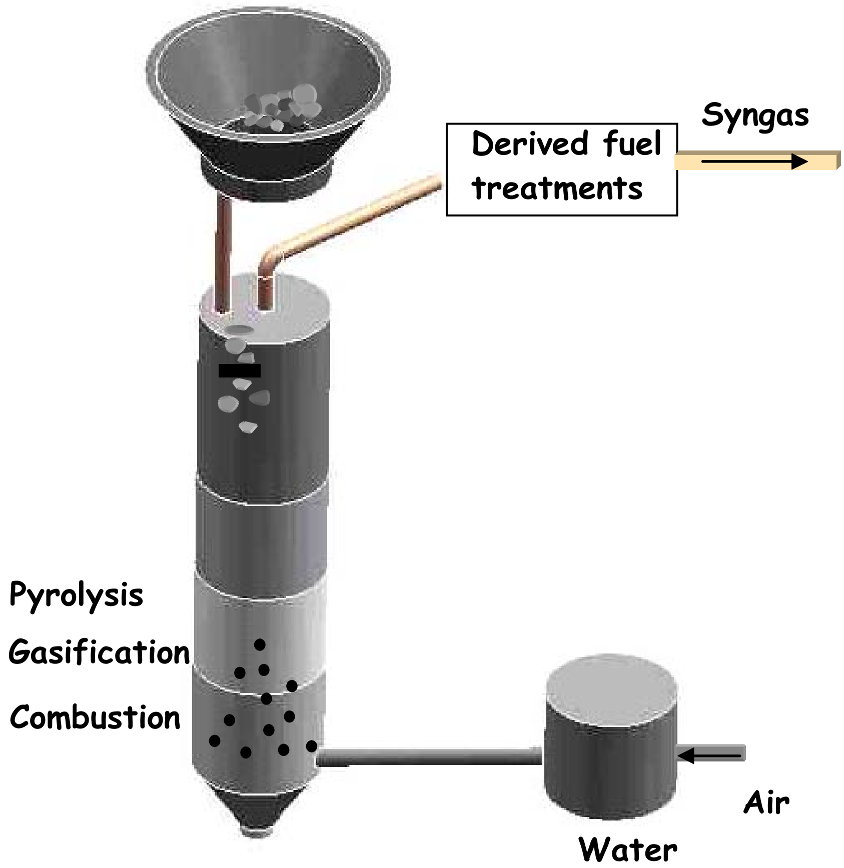



**Fig 2** *Counter current, fixed bed gasifier with oxygen enrichment*

Updraft gasifiers may process fuels with high water content (up to 50 % of mass fraction), without requiring any pre-drying.

Syngas coming from the pilot gasifer is sent to a wet scrubber where dust and HCl in it contained are abated reducing also the fuel gas temperature.

The cleaned gas is sent to a flare .

In a first phase of testing, the pilot plant will be used for testing only coal with low sulphur content.

A stream of syngas can be sent to lab plant.

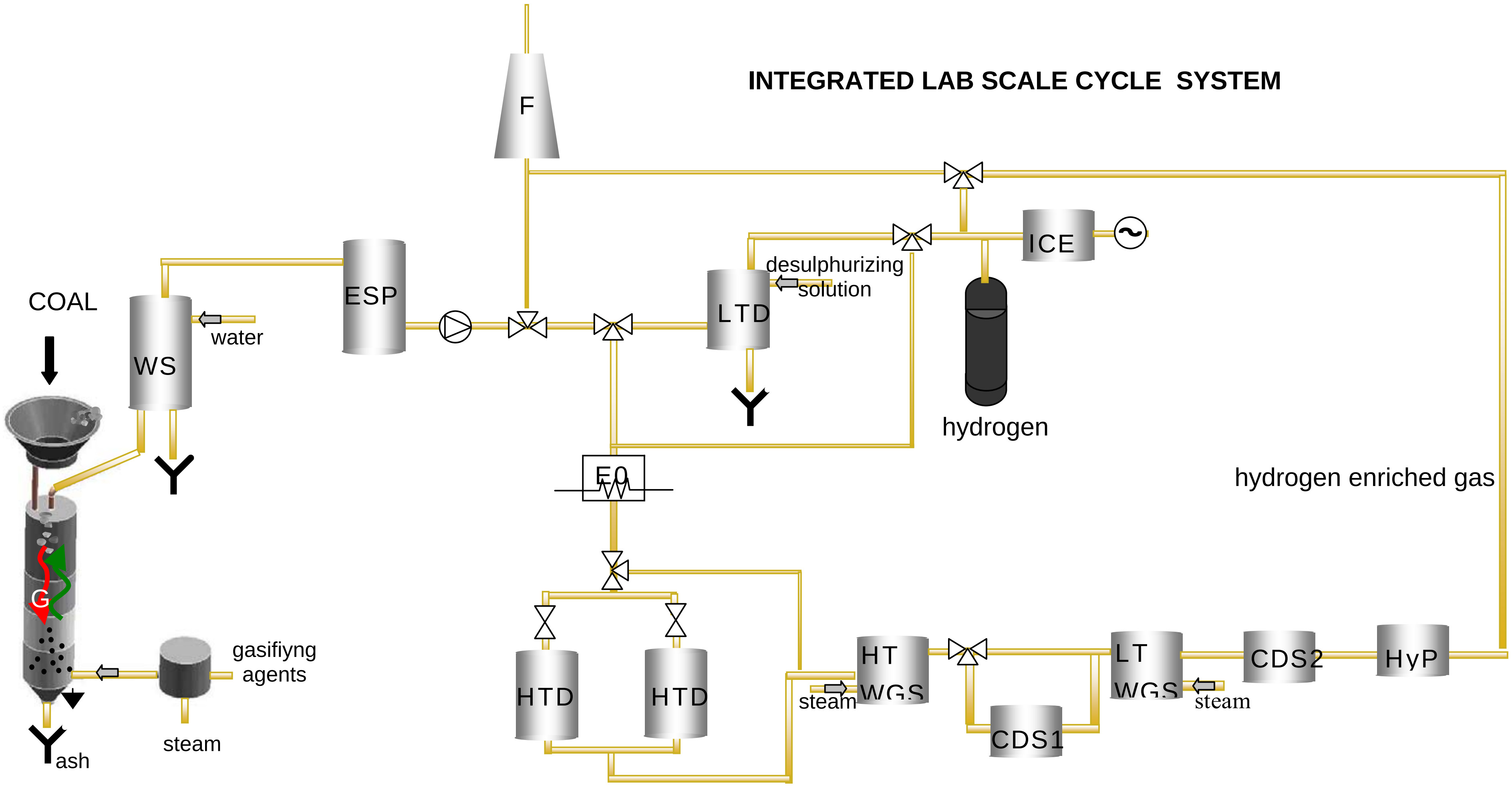
**Integrated lab scale power cycle system**

The present part of the project aims at the development and demonstration of an innovative concept for production of power and hydrogen enriched stream, employable as energetic carrier itself or potentially into fuel cells.
The novelty of the project consists of environmentally and cost effective abatement technologies developed and tested for syngas cleaning, both for power production in internal combustion engines and for high purity hydrogen stream production.
In this project, the integrated part of gas cleaning and treatment of the research proposed is designed for the lab scale (35 Kg/h) instead of the pilot scale (700 Kg/h) gasifier thus providing higher level in the plant management flexibility during transient conditions and reduced investment and operating costs.
With reference to Figure 3 which shows the conceptual integrated lab scale power plant scheme, the syngas coming out from the gasifier, subjected to a gas cleaning section, consisting of a scrubber and an electrostatic precipitator, is split in two streams:

1. 80 % of total gas flow is sent to a power production line which is made up of:
   - a cold desulphurisation system
   - an internal combustion engine for power production.
2. 20% of the total flow rate is sent to a hydrogen production line, which is made up of:
   - a hot desulphurisation system
   - two water gas shift sections (high and low temperatures)
   - two $CO_2$ separation stages to obtain an enriched hydrogen gas stream
   - a Hydrogen purification system;

The system also allows the possibility of alternating/including the cold desulphurisation system, using a downstream by-pass located in the hydrogen production line.
Syngas in excess can be sent to a flare together with air, at 20 °C, and auxiliary fuel to allow combustion process and removal of syngas.
As safety measure, in emergency situation or during normal start up and shut down operation, the lab plant is equipped with a nitrogen system for inertisation of gasifier, scrubber, electrostatic precipitator and connection piping.
Following is reported a description of the single components of the system.

## INTEGRATED LAB SCALE CYCLE SYSTEM




**Legenda:**

**CDS:** Carbon dioxide separation
**E0**. Heater
**ESP:** Electrostatic precipitator
**G:** Gasifier

**HTD:** High temperature desulphurization
**HTWGS:** High temperature water gas shift
**ICE:** Internal combustion engine
**LTD:** Low temperature desulphurization

**LTWGS:** Low temperature water gas shift
**HyP**: Hydrogen poprpion
**F:** Flare
**WS**: Water Scrubber



**Fig. 3**

*Lab scale gasifier*

In order to achieve higher yield in hydrogen produced, a system for providing air enriched in oxygen will be tested to reduce the amount of inert gases to be separated in the downstream sections. Saturated steam is added to the air to improve temperature control.

The updraft gasifier will be loaded with Sulcis and imported coal and other types of coal where available. In Table 1 is reported the proximate and ultimate analysis of two reference coals.

**Table 1** *Proximate and ultimate analysis of Sulcis and South African coal*

| Sulcis Coal | | | South African Coal | | |
|---|---|---|---|---|---|
| **Proximate analysis** | **Mean value (wt%)** | **Variability range (wt%)** | **Proximate analysis** | **Mean value (wt%)** | **Variability range (wt%)** |
| Moisture | 11, 51 | 9÷13 | Moisture | 8 | 4÷12 |
| Volatile matter | 38,56 | 38÷40 | Volatile matter | 23 | 20,7 ÷ 34 |
| Ash | 17,33 | 15÷19 | Ash | 15 | 3 ÷ 16 |
| Fixed carbon | 32,59 | 32÷37 | Fixed carbon | 54 | 48 ÷ 56 |
| **Ultimate analysis** | | | **Ultimate analysis** | | |
| Humidity | 11,52 | 9 ÷ 13 | Humidity | 8 | 4 ÷ 12 |
| Ash | 17,31 | 15 ÷ 19 | Ash | 15 | 3 ÷ 16 |
| Total carbon content | 53,22 | 50 ÷ 54 | Total carbon content | 65,88 | 63,04 ÷ 78,9 |
| Combined hydrogen | 3,89 | 3 ÷ 5,1 | Combined hydrogen | 3,71 | 3,18 ÷ 6 |
| Nitrogen | 1,29 | 1 ÷ 1,.3 | Nitrogen | 1,5 | 1,16 ÷ 1,69 |

| | | | | | |
|---|---|---|---|---|---|
| Sulphur | 5,98 | 5,6 ÷ 8,5 | Sulphur | 0,55 | 0,43 ÷ 1 |
| Oxygen | 6,75 | 5 ÷ 9,5 | Oxygen | 5,36 | / ÷ / |
| Chlorine (ppm) | 1000 | n.d. | Chlorine (ppm) | 0,05 | 0,04 ÷ 0,25 |
| Inherent moisture | 6,77 | 5,58 ÷ 7,57 | Inherent moisture | 2,66 | 5,58 ÷ 7,57 |
| **Low Heating value** | **Mean value (MJ/kg)** | **Variability range (MJ/kg)** | **Calorific value** | **Mean value (MJ/kg)** | **Variability range (MJ/kg)** |
| LHV | 20,83 | 19,67 ÷ 21,35 | LHV I | 24,79 | 0,99 ÷ / |

Typical values expected for syngas composition when using coal as feedstock are reported in Table 2 (Hobbs *et al.*) (1):

**Table 2** *Syngas composition coming out from the gasifier*

| | |
|---|---|
| Flow rate | 90 Nmc/h |
| Composition | % (v/v) dry |
| CO | 28 – 30 |
| H2 | 14 – 16 |
| CO2 | 0.8 – 0.9 |
| N2 | 50- 52 |
| CH4 | 1.3- 1.4 |
| H2S | 1.7 |
| Water content | saturation |
| Particulate | 0.85 kg/h |
| Tar | 1.5 kg/h |
| Temperature | 250 °C |
| Pressure | atmosferic |

*Scrubber (WS)*

Raw syngas produced in the updraft gasifier is treated using a scrubber with water to reduce tar and chlorides (which may be contained in the syngas depending on the composition of the feedstock) in order to obtain a dedusted gas which can be further treated in the downstream sections. This unit allows a temperature reduction of the syngas coming out from the gasifier (about 200-250 °C) together with scrubbing of acidic compounds, such as hydrochloric acid and the condensation of the condensable organic compounds dragged by the gas stream. Scrubbing will be realized in a series of packed towers, where any trace of tar, not retained in the precipitator, will be removed from the gas together with any water soluble component.

*Electrostatic precipitator (ESP)*

Raw gas is then treated to reduce its particulate and tar content. For this purpose, an electrostatic precipitator is used for the wet removal of tar and ash, which allows, with high efficiency (up to 99.9%) and low pressure drops, the reduction of particulate and tar dragged in the syngas as aerosol particles.

The coal syngas coming out from electrostatic precipitator is compressed by a blower and divided in two stream: 20% to the hydrogen production line and 80% to the power production line

***Power production line***

*Low temperature desulphurisation (LTD)*

The stream of 80% of main gas flow from the electrostatic precipitator is cooled at about 30° C and fed in an absorber where come into contact in counter-current with a solution of water and tertiary amines that remove hydrogen sulphide and part of $CO_2$. The cleaned syngas coming out from the absorber can be directly sent to the internal combustion engine (ICE). In order to enrich the $H_2$ concentration in the syngas, the plant is also equipped with a hydrogen make up from a cylinder.

In the case of COS concentrations in the syngas not negligible, a further treatment for COS removal is scheduled to be located in upstream the desulphurisation absorber. Moreover, because COS hydrolysis reaction occurs at 300-350 ° C, it is convenient to heat the overall syngas flow from blower and then split it into the two stream: 20% to the hydrogen production line and 80% to the power production line

*Internal combustion engine (ICE)*

An internal combustion engine specifically designed for clean syngas feeding is used for the conversion of syngas into energy. The plant is provided with an hydrogen enrichment, through a bottle of hydrogen, in order to test the engine operation with high hydrogen concentration fuel.

***Hydrogen stream production***

*Heat exchanger (EO)*

As above mentioned, in the case of very low COS concentration in the syngas, 20% of the overall syngas flow coming out from the electrostatic precipitator, through the blower, is sent to an heat exchanger for rising the temperature up to 350 °C before feeding it into the high temperature desulphurisation section.

*High temperature desulphurisation (HTD)*

Hydrogen sulphide together with traces of sulphur compounds other than sulphur oxide, such as COS, are removed by using regenerable sorbent based on metal oxide. To this end two fixed bed reactors are used, each operating in turn in the oxidation and regeneration phases. Regeneration occurs in the reactors by oxidation with external air, when the acid gases ($SO_2$) concentration in the outlet stream reaches the established upper limit value.

*Water gas shift conversion sections with by-pass of $CO_2$ removal*

In order to obtain a high hydrogen conversion yield, the shift reaction is realized in two steps at high and low temperatures (HT-WGS & LT-WGS). Between the high and the low stage, a by-pass for $CO_2$ removal is introduced.

After cleaning, saturated or superheated steam is added to the syngas to realize the catalyzed shift conversion of carbon monoxide in carbon dioxide with production of hydrogen according to equation (1):

$$CO + H_2O \leftrightarrow CO_2 + H_2 \qquad (1)$$

The amount of steam added depends on the syngas composition. The gas produced after the shift conversion is constituted by hydrogen, carbon dioxide, steam, nitrogen and traces of carbon monoxide. The shift conversion cannot be considered as an innovative process, as it is widely applied in the chemical and petrochemical industry, but its application to coal syngas has not been tested yet.

The CO- Shift section is made up of two parts:

1. a high temperature reactor , operating at 300-350°C,

2. a second reactor, operating at lower temperature (200-250°C)

The heated syngas coming from the absorber is mixed with steam and is fed into the first shit reactor where occurs about 90% of conversion according to the reaction . The flow coming out from the first reactor is split in two streams: one is sent to the second shift reactor and the other one is fed to a $CO_2$ capture system (CDS1)where temperature and dioxide carbon concentration are abated. Then the two stream are mixed before to be fed (at 200-250 °C) to the second shift reactor where the hydrogen concentration in the syngas is some more increased.

The by-pass of $CO_2$ removal (described in the next section) is designed for minimizing the energetic cost of the shift conversion sections, since it causes in the shift reaction (see equation 1) a reduction in the vapor consumption. In fact the $CO_2$ removal section between the high and low temperature water shift sections determines a decrease of $CO_2$ concentration in the gas stream getting into the low temperature water gas shift. This causes the reaction equilibrium to move toward the products with a reduction in the vapor consumption for the achievement of the same outlet hydrogen enrichment level in the gas stream.

Syngas coming out from the second shift reactor is finally sent to a further stage of $CO_2$ removal.

*Carbon dioxide separation (CDS2)*

Gas from the shift conversion undergoes further processing for the separation of carbon dioxide from hydrogen. Chemical processes, based on absorption with amines, is employed to remove carbon dioxide from the gas stream containing hydrogen using a reactor with hydrophilic and hydrophobic polymeric membranes.

The process includes the following sections:

- Syngas diffusion into the amine solution through hydrophilic membrane
- $CO_2$ absorption into the amine solution
- Purified syngas separation through hydrophobic membrane

*Hydrogen purification (HyP)*

The hydrogen purification section can allow to test and compare different purification system (such as membrane and Pressure Swing Absorption ).

## Configuration flexibility

The lab plant configuration allows, through a suitable system of valves and by-pass, to:

- send 100% of syngas flow (100%) coming out from the gasifier, to the cold desulphurization system and then to the internal combustion engine.
- send 20% of syngas flow from the cold desulphurization system, to the CO-shift section, by-passing the hot gas desulphurization treatment.

## Blank tests on single plant sections

Following the design and the installation of the single components of the system in the proposed pilot platform, a first blank experimental test grid is proposed. These blank tests will be run on each single section of the plant, independently on to the other, either at ambient temperature or at the operating design conditions using simulated gas. The objective of these experimental tests is the demonstration of the single plant sections in terms of:

- Control of mechanical and electrical components and operational control system, at ambient temperature
- Achievement of the expected yields at operating temperature.
- Start-up and shut-down.

## Operating test on the integrated pilot platform

After a first experimental campaign on pilot and lab scale gasifier for the evaluation of syngas composition for different types of coal, the experimental lab scale system will be tested as a whole.

## Conclusions

The research project will allow to gain highly useful knowledge and experience in improving processes for producing clean fuel gas and hydrogen in a small-medium scale plant, from Sulcis and commercial coal.

Some of the main objectives are:

- Improve and optimise the coal gasification process regarding both the automation and feeding system.
- Improve gasifier performance in terms of different types of coal fed, composition of the gasifing agents in terms of nitrogen content, oxygen enrichment, steam ratio and possible use of carbon dioxide as inert gas instead of nitrogen.
- Test the cleaning and desulphurization techniques (hot and cooled) for the achievement of a syngas suitable for feeding in the internal combustion engine for power production.

- Develop and validate simulation models, to collect experimental data and to gain engineering experience for the scaling-up of pilot plant sections.
- To qualify and quantify CO-Shift catalysts and to verify the reactor performances in different operating conditions.
- To evaluate, from a technical and economical point of view, the process of hydrogen separation process from syngas using special membranes.
- To evaluate and compare $CO_2$ separation technologies based on physical and/or chemical processes.

## References

De Jong W., Andries J., Hein K.R.G. (1999) Coal/biomass gasification in a pressurised fluidised bed reactor. Renewable Energy, 16, 1110-1113

M. L. Hobbs, P.T. Radulovic, D. Smoot (1992) Prediction of effluent composition for fixed bed coal gasifier.