Project no.: **SES6-CT-2003-502743**

Project acronym: **ISCC**

Project full name:
Innovative In Situ CO_2 Capture Technology for Solid Fuel Gasification

Instrument type:
Specific targeted research or innovation project

Priority name:
Sustainable energy systems

Publishable final activity report

Period covered: from 1.01.04 to 31.12.06

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Start date of project: 1.01.2004

Duration: 36 months

Project coordinator name: Dr. Roland Berger

Project coordinator organisation name:
University of Stuttgart, Institute of Process
Engineering and Power Plant Technology (USTUTT)

Content

1 Project execution

1.1 Project objectives

The three-year project aimed to develop a new process for upgrading high moisture low rank brown coals yielding three valuable products:

- A fuel gas consisting of mainly hydrogen to be used in power generation
- A purge gas stream containing >95% CO_2, ready for transportation to storage (CO_2 capture >90%) or chemical fixation
- A pre-calcined feed for a cement kiln consisting of CaO, coal ash and required additional minerals

The ISCC technology is based on the **L**ime **E**nhanced **Gas**ification (LEGS) reaction. The LEGS reaction combines steam gasification of low rank, high moisture brown coal, with the high temperature removal of CO_2 by using high temperature efficient sorbent materials (e.g. lime). The combination of the gasification and the in situ CO_2 capture shifts the reaction towards H_2 production in the gas stream. The CO_2 laden sorbent material must be regenerated in an additional regeneration step before being recycled back into the gasifier. The high number of carbonation-calcination cycles experienced by the sorbent particle places high demands on the necessity for a robust, mechanically and chemically stable sorbent.

The two core-sub-processes of the ISCC process are:

LEGS-reaction: The combined gasification/reforming step to produce pure H_2

$$Coal + Steam + CaO \rightarrow CaCO_3 + H_2 + char$$

Regeneration: The sorbent regeneration via char combustion or indirect heating to produce pure CO_2

$$CaCO_3 + char + O_2 \rightarrow CaO + CO_2$$

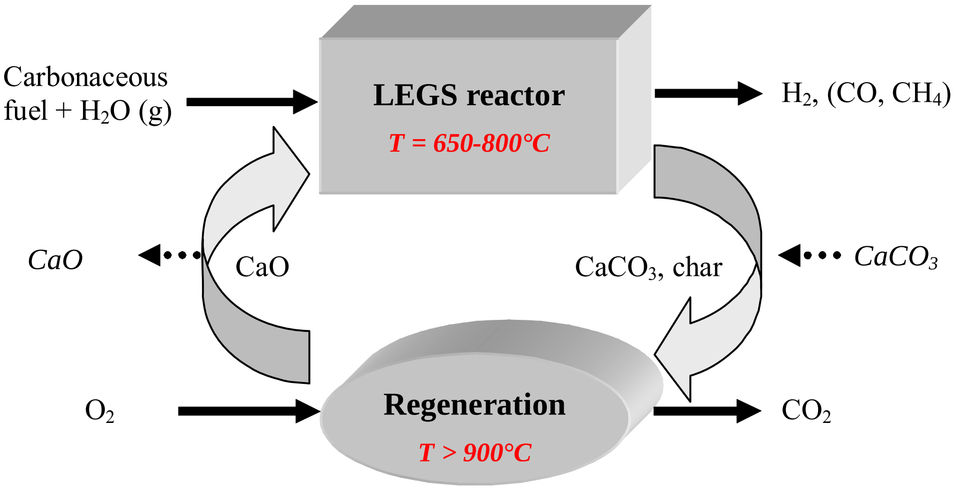

Figure: Schematic representation of the ISCC process

1.2 Contractors involved

The consortium of the project consists of fourteen partners from seven countries. Universities, research centres, SMEs, and industrial partners are represented in the ISCC consortium. The following table provides an overview on the participating contractors.

Role	No.	**Name**	Short Name	Country
Universities				
CO	1	University of Stuttgart – Insitute of Process Engineering and Power Plant Technology	USTUTT	Germany
CR	2	National Technical University of Athens / Laboratory of Steam Boilers and Thermal Plants	NTUA/ LSB	Greece
CR	5	Politechnika Wrocławska – Wrocław University of Technology	WUT	Poland
CR	9	Energy research centre, University of Ulster	UU	UK
CR	10	Technical University of Brandenburg - Cottbus	BTU Cottbus	Germany
SME				
CR	8	IVE Weimer	IVE	Germany
CR	11	SCS-Technology	SCS	Austria
Industry				
CR	6	Public Power Corporation of Greece	PPC	Greece
CR	13	Kopalnia Węgla Brunatnego "Turów" Spółka Akcyjna	KWB Turów	Poland
CR	14	Vattenfall Europe Mining AG	VE Mining AG	Germany
Research centres				
CR	3	Center for Solar Energy and Hydrogen Research	ZSW	Germany
CR	4	Glowny Instytut Gornictwa -Central Mining Insitute	GIG	Poland
CR	7	Technical Reseacrh Centre of Finland	VTT	Finland
CR	12	Consejo Superior de Investigaciones Científicas	CSIC	Spain

1.3 Work performed and end results

In the following sections the major results of the project are described. It is intended to give a good overview of the project and not to go into the details. For more details please contact the authors.

1.3.1 Definition of feed characteristics and process requirements (WP 1)

The basic flow sheet of the ISCC process used for combined power and cement production (CPCP) is illustrated in Figure 1. This concept managed the deactivated lime from process carbonation-calcination thus the concept is more cost effective and improve the efficiency of the whole process. The regeneration method is not determined for this basic description.

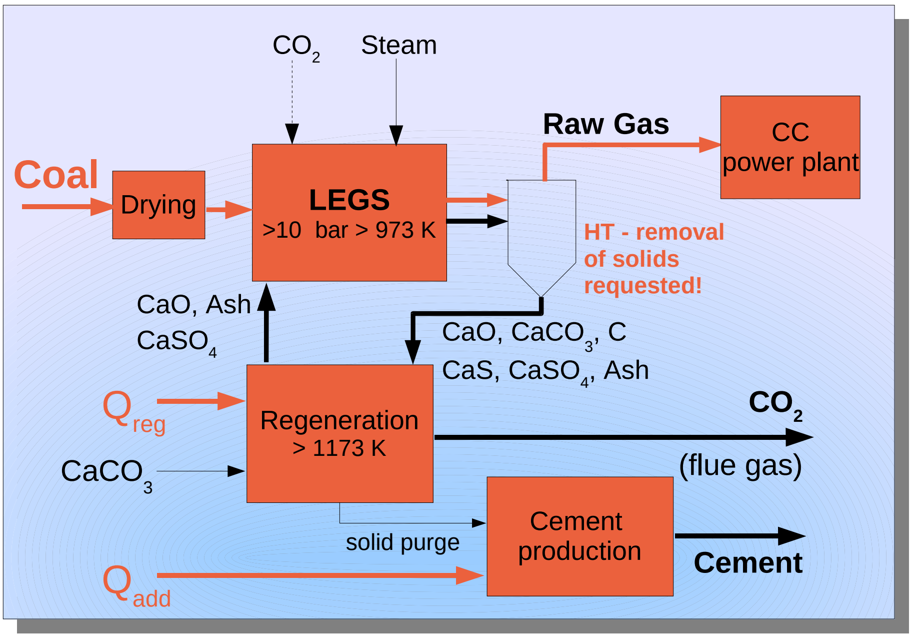

Figure 1: Basic Process Design CPCP

Regarding Figure 1 two main process units can be identified. In the LEGS gasification reactor a Hydrogen rich gas is produced. The simplified LEGS reaction equation can be described as

Fuel + a CaO + b H_2O ➔ x H_2 + (a-z) $CaCO_3$ + y C + z CaS (1)

Due to the strongly exothermic CO_2 absorption during steam reforming a slightly exothermic gasification reaction can be adjusted leading to minimised energetic losses and a simple reactor design. In addition to the main reactions steam reforming, water gas shift reaction and CO2 absorption summarised in equation (1) further reactions as the formation of tars and ammonia must be taken into account in a detailed examination. Furthermore the removal of sulphur by CaO must be investigated in more detail.

Beside the Hydrogen rich gas a solid product consisting of limestone, CaO, char, ash, gypsum and CaS is produced during the LEGS reaction. The solid product has to be separated from the gas and must be processed in a further process unit. This process can be described as a modified lime kiln with high temperature input and simultaneous CaS oxidation.

(a-z) $CaCO_3$ + y C + z CaS + c O_2 + heat ➔ (a-z) CaO + z $CaSO_4$ + ? CO_2 + ? H_2O (2)

One additional advantage of the process is that the remaining char from the gasification can supply a part of the heat demand of regeneration. Regarding equation (1) and (2) it is clear that a purge of solids is required to remove the gypsum from the system. Furthermore this purge is required for ash removal as well.

Beside the two main processes the pre-treatment of fuel and the gas cleaning have to be considered as well.

Fuel:

In general, suitable fuel inputs for the ISCC gasification process can be characterised as follows:

- Carbonaceous energy carriers to enable the exothermic CO_2 - absorption
- Solid or liquid leading to an upgrading to an easy combustible and clean gas

- Humidity in the fuel may decrease steam addition
- Ashes are suitable cement feedstock

Based on these qualities Lignite and other low C coals seem to be perfect LEGS inputs

Sorbent:

The ideal sorbent for the ISCC process would be one fulfilling the following conditions:

- Able to react with CO_2 at sufficiently high temperatures to allow high gasification yields (*in situ*).
- High capture capacity of CO_2 (kg of CO_2/ kg of sorbent)
- Low heats requirements for regeneration (low calcination heat and low fraction of inactive mass).
- Good cycle stability in many sorption-desorption cycles and/or low cost.

Natural limestones (or their derived CaO as CO_2 sorbent) fulfil very clearly the first condition. Within the project it was shown, that they also fulfil the other requirements, especially low price and abundant availability. In this respect, there are few sorbents, if any, cheaper than crushed limestone.

Furthermore, the ideal capture capacity of CO_2 of a particle CaO is 78.6 w%. This means that even when molar Carbonation conversions are as low as, for example 10%, the capture capacity of particles of CaO is still comparable to some synthetic sorbent being proposed for adsorption systems (Yong et al 2002).

A reactivation method for lime would be advantageous to improve the carbonation conversion of the sorbents during many cycles of the ISCC process. This would be as well good for the economy of this technology. Despite the fact that the cycle stability of some lime resources is poor, it can be compensated with a sufficiently high make up flow of fresh limestone as will be seen below. The weakest point is that the heat requirements for regeneration are unavoidably high because the calcination heat of $CaCO_3$ is high. The temperatures for regeneration are also very high in some cases and this can lead to unacceptable sorbent deactivation and even sintering.

The resulting average activity of limestone estimated based on experimental results is illustrated in Figure 2. Details can be found in the Deliverable D 1.1 of the ISCC project.

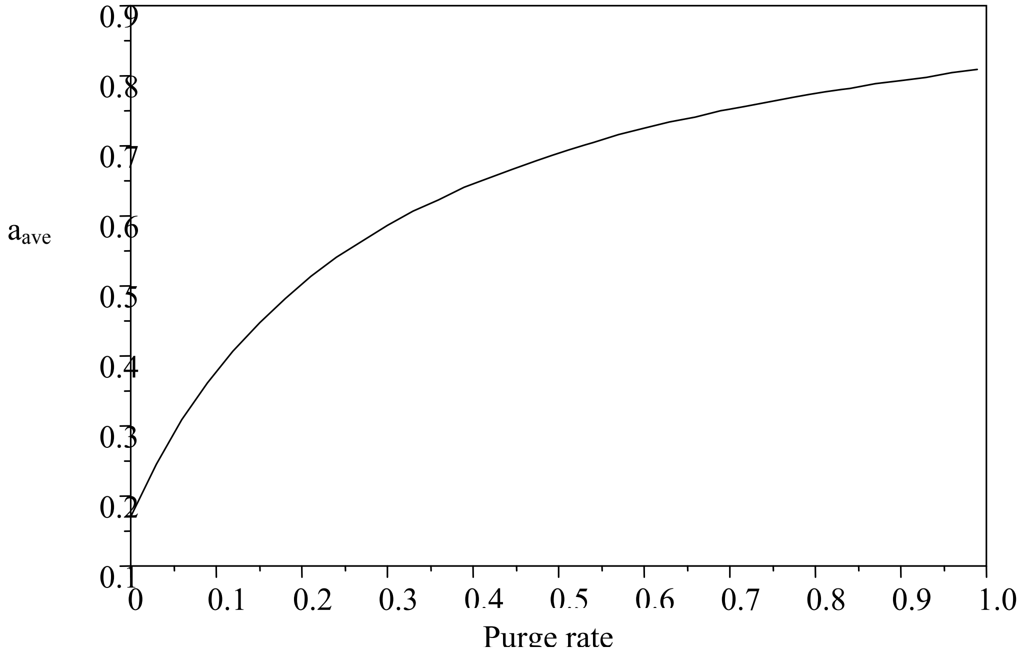

Figure 2: The average CaO activity a_{ave} as function of the purge rate x^{SRP}

The sorbent characteristics are described in more detail in section 1.3.2
The in deep investigation of calcination-carbonation cycle under variable conditions have shown the opportunity of sorbent reactivation by spontaneous changing the carbonation process parameters after the sorbent have seen approx. 20 - 25 cycles.
On the base obtained preliminary results it seems that the new concept of the operation the LEGS process can be formulated. The example of parameters sensitivity for this type processes is presented in the Figure 3. The arrow introduced into the original figure shows the area of the most promising developments in cutting the total costs of carbon dioxide capture.

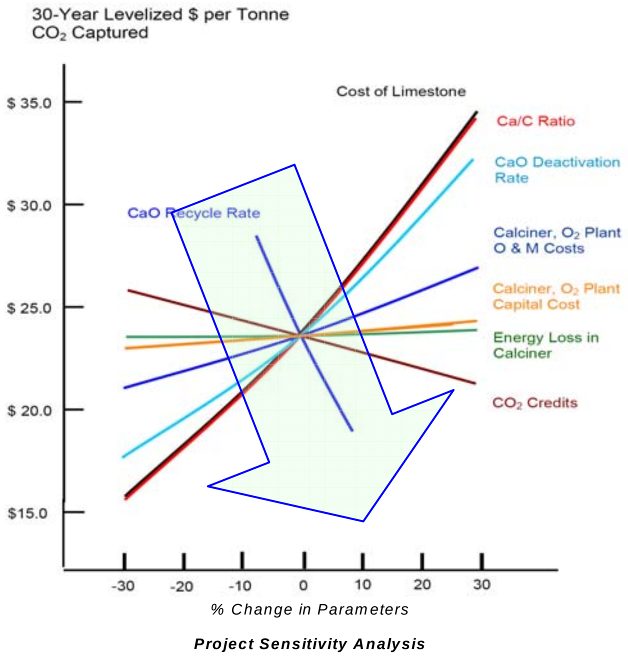

Figure 3: Economics of CO_2 Capture using the Calcium Cycle with a Pressurized Fluidized Bed http://www.neillandgunter.com/downloads/AFM-economics_of_CO_2_Capture.pdf

The considerable improvement of the LEGs process could be achieved by the regular (say after each 25 C-C cycles) targeted swing in the parameters of the carbonization cycle when the capture potential of the sorbent approaches the minimal capacity acceptable in the process. That might be carried out by the radical change of the process parameters, with parallel use of gas containing high concentration of carbon dioxide introduced in the system.
The novelty of the idea follows from the fact, that the process improvement will be explored through the change in operation parameters of gasification with only minor interference into the design of the fluidized bed system.
The European geological overview of lignite and sorbent material for ISCC process was evaluated. Details can be found in the Deliverable D 2.b of the ISCC project. More Information on this is available on request from Partner WUT. This deliverable provides geological overview of lignite and sorbent material for ISCC process in Europe see Figure 4.
Data concerning the calcium sorbent and lignite resources and partly the data about its composition were collected for Austria, Bulgaria, Czech Republic, France, Greece, Germany, Hungary, Italy, Poland, Romania, Slovakia, Slovenia, Spain, Turkey, UK.

It is presented in an interactive map which allows a quick overview of the local resource all over Europe and which can be easily enhanced with further data.

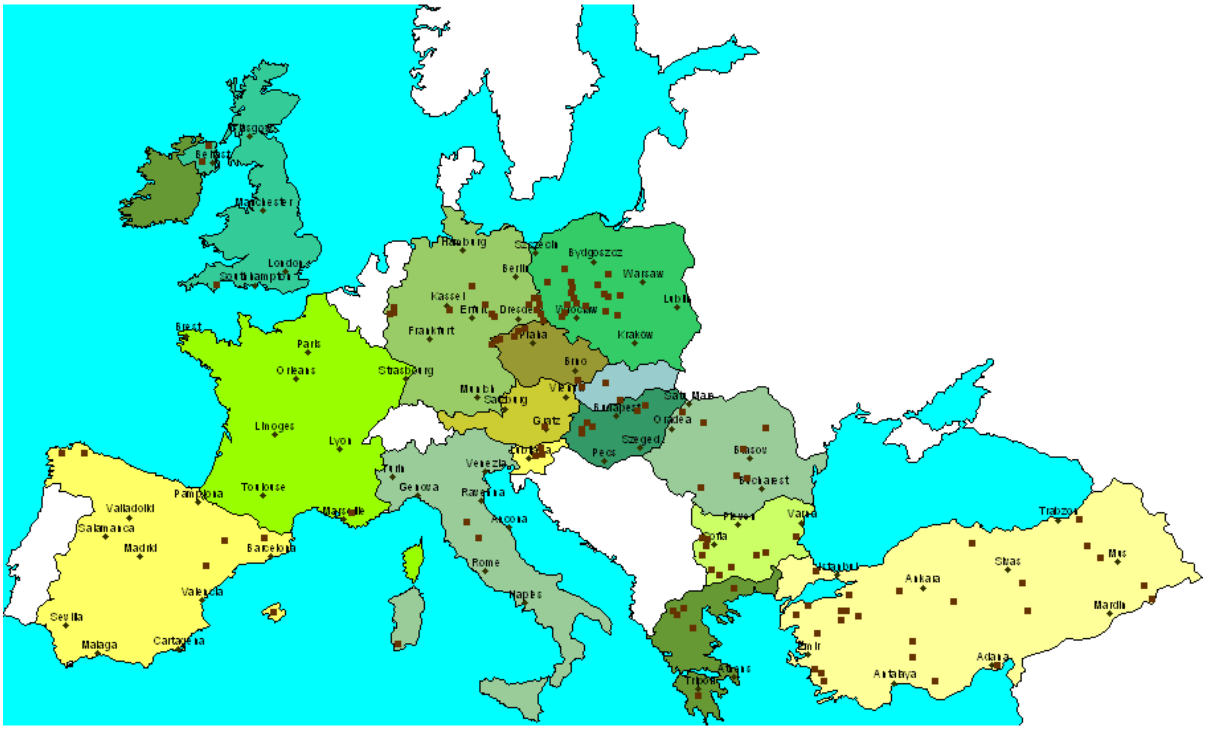

Figure 4: The European geological overview of lignite and sorbent material for ISCC process

Solid Product:
Requirements for Portland cement chemical constituents are summarized in Table 1.

Table 1: Requirements for Portland cement chemical constituents

Parameter	Unit	MIN	MAX
SiO_2	m-%	16,0	26,0
Al_2O_3	m-%	4,0	8,0
Fe_2O_3	m-%	2,0	5,0
Mn_2O_3	m-%	0,0	3,0
TiO_2	m-%	0,0	0,5
CaO	m-%	58,0	67,0
MgO	m-%	1,0	5,0
$K_2O + Na_2O$	m-%	0,0	1,0
SO_3	m-%	0,1	2,5
P_2O_3	m-%	0,0	1,5
Loss on Ignition	m-%	0,5	3,0

For the monitoring and control of production the following formula has been found to be useful for adjusting the composition of the raw meal on the basis of laboratory analyses of the components (proportioning formulas):
Lime Standard according to Kühl:

$$LST = \frac{CaO}{2.8 \cdot SiO_2 + 1.1 \cdot Al_2O_3 + 0.7 \cdot Fe_2O_3} \cdot 100$$

A high lime standard normally causes high cement strengths. The following lime standards are characteristic for Portland cements:

90 to 95	Standard Portland cement
95 to 98	High early strength cement

Product Gas:
Higher gasifier pressures result in higher Methane yields, increasing the Carbon content of the product gas and thus decreasing the gasifier Carbon capture. As Carbon conversion increases, the Carbon capture will naturally decrease because more of the previously "unconverted" Carbon is partitioned into the gas phase as CO, CO_2, and CH_4.
However, increasing the Steam/Carbon ratio in the gasifier input decreases the Methane content in the product gas (Figure 5) thus increasing the Carbon capture.

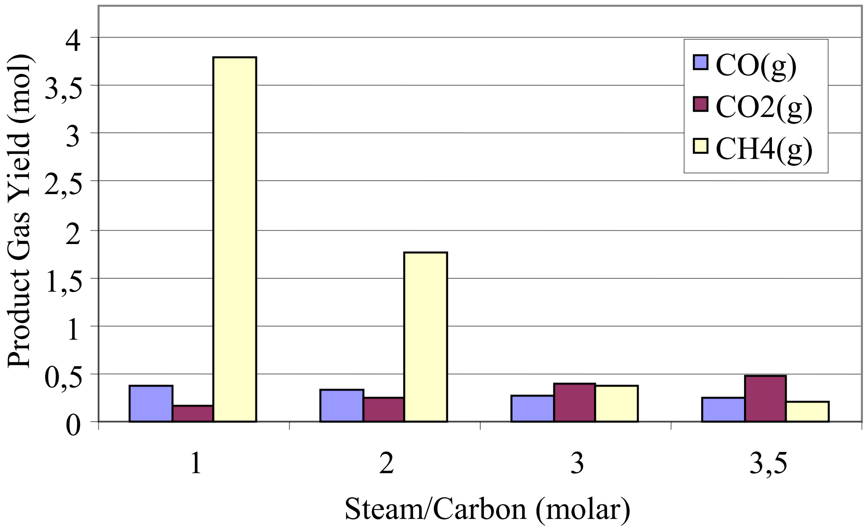

Figure 5: Carbon content in product gas as a function of S/C 20 bar, 750 °C and 80 % Carbon conversion

Regeneration Flue Gas:
The analysis of existing and expected CO_2 sequestration techniques revealed that the properties and quality of produced Carbon Dioxide ranges widely in composition depending on the sequestration approach. For the purpose of the ISCC project it has been decided that several cases have to be considered. They are summarized in Table 2.

Table 2: Requirements for product gas from regeneration

	Case 4.2.1 CO_2 Sequestration in Coal Beds	Case 4.2.2 CO_2 for Transport in Pipelines	Case 4.2.3 Solid CO_2 Storage in Insulated Depository	Case 4.2.4 CO_2 for Integrated Transport with H_2	Case 4.2.5 CO_2 Mineral Sequestration	Case 4.2.6 CO_2 Biological Sequestration
Temperature Range [C]	-50; +50	-50; +50	<-78.0	pipeline regulations	ambient to elevated up to 250	Ambient
Pressure Range [MPa]	7.0 -15.0	7.0 -15.0	normal, to hydrostatic	5.0 -15.0	normal to elevated	Normal

Form	compressed supercritical gas , liquid	compressed supercritical gasa, liquid	solid	supercritical gas	0.1- 10.0 liquid, supercritical gas	Gas
Compositions	Regulated >99.95 when from pipeline, or from truck Broad, when compressed fue gases used,	Regulated >99.95	>99.99	99.999 S< 1(targeted 10.0) ppm	Broad, regulated by the way of delivery	Broad, no content of hazardous regulated component
Purity Requirements	Liberal,	Particulates< 10mg/m^3	Liberal, regulated by transport way to solidification plant	high, comparable with Hydrogen	Liberal Regulated by transport requirements	High, no contain of poisons

1) For underground sequestration in aquifers the quality requirements are regulated according to the transport way from the source to the sequestration site. The pipeline quality, for example requires the very dry gas, because of highly corrosive properties of wet CO_2.

1.3.2 Basic investigations CO_2 absorbent properties and mechanical performance (WP 2)

1.3.2.1 Introduction

This section presents a synthesis of the results discussed in Workpackage 2 Deliverables, completed by CSIC, ZSW and VTT. Natural limestones have been the main focus of investigation, because they are the cheapest and most widely available materials for the purpose of the ISCC process. Test have been carried out in a range of laboratory installations and devices in the different institutions, including different termogravimetric equipment, small fixed and fluidised beds reactors both at atmospheric and high pressure. Electron microscopy and mercury porosimetry have been the techniques employed to follow the textural changes in the sorbent as reaction progresses. Several (4-6) scientific papers in specialized journals have been published or are under evaluation.

1.3.2.2 The sorbent performance requirements in the ISCC process.

The first limit for the CO_2 capture in the ISCC gasifier and for the CO_2 regeneration in the calciner, is given by the equilibrium of the carbonation reaction. This equilibrium is represented in Figure 1, in the interval of temperatures of interest in this project (600-1150˚C). According to this equilibrium, the ideal conditions for high capture efficiencies of CO_2 in the gasifier are high total pressures and low temperatures. But it is known that low gasification temperatures (below 750 ºC) may present rate limitations (low conversion of the fuel during gasification) and therefore, high total pressures (10 atm or higher) are required if capture levels are to be high. On the other hand, high total pressures will demand very high calcination temperatures during the regeneration (atmosphere very rich in CO_2 requires T> 1000 ºC, see Figure 6). This has emerged as a critical point for the sorbent performance perspective. Alternatives such as the use of lower pressure in the calciner, steam to reduce the CO_2 partial pressures, or even the use of air instead of O_2 in the regenerator (with subsequent need to capture CO_2 downstream the regenerator) are discussed in section 1.3.1 and 1.3.3.

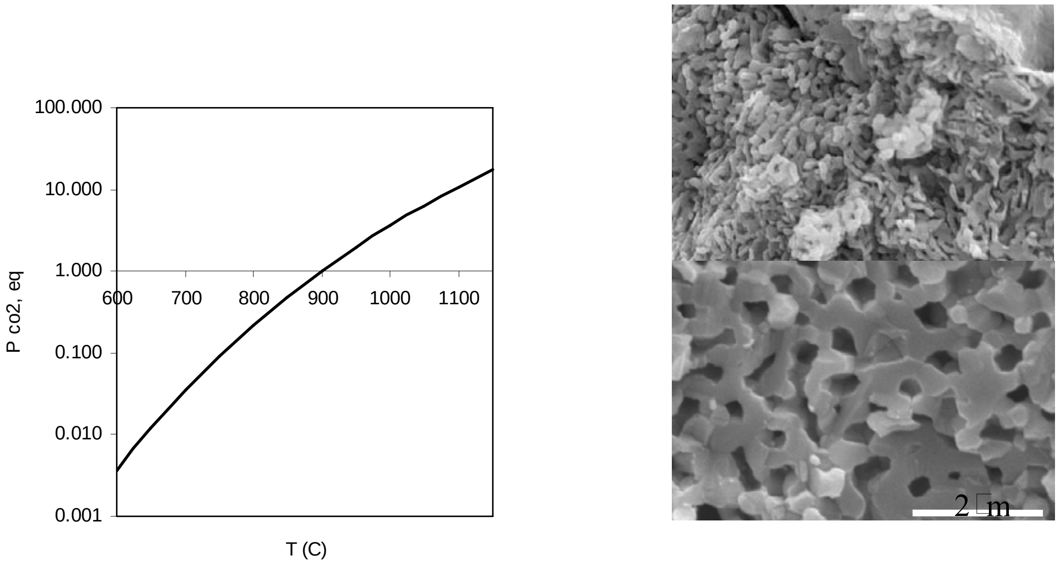

Figure 6: Left: The equilibrium pressure of CO_2 over CaO in the range of temperatures relevant for the ISCC process. Right: View of a calcined particle of limestone (1 cycle) and after 30 carbonation-calcination cycles

Once the key operating conditions are chosen for carbonation and regeneration, a critical sorbent performance issue for the ISCC process is that the effective removal of CO_2 from the gasifier requires availability of a sufficient amount of “active CaO”. The concept “active CaO” was introduced to account for the fact that only a fraction of the CaO is able to react with CO_2 to produce $CaCO_3$. All the available data, from the literature and from this project, shows that the capture capacity of lime decreases rapidly with the number of carbonation/ calcination cycles. For fluidised bed systems at high pressure, like those to be used in the ISCC process, this limiting conversion determines the overall performance of the reactor system (see 1.3.1). The measurement of these conversions as the number of cycles increases have therefore been one of the main task in workpackage 2, and the key results are summarised in the next paragraphs. Reactivation methods have the potential to restore activity in the sorbent, and some of the main routes have also been explored in this work (although with no much success as indicated below).

The following variables have been used to investigate the cyclic carbonation and calcination reactions in both the termogravimetric analyser and fixed bed reactors at CSIC, ZSW and VTT: effect of sorbent type and the number of cycles (typically 100 and up to 500), effect of particle size, effect of calcination temperature, effect of (long) calcination times, effect of CO_2 partial pressure during carbonation and during calcination, effect of steam, effect of total pressure. Figure 7 shows an example of conversion curves vs. time and one of the test rigs used in the project. This curve belongs to a long series of cycles from an experiment carried out with Piaseck limeston (from Czatowice). Many different series of long duration tests like this one have been obtained to see how the operating conditions affect to the conversion achieved along the cycles. These thousand of thermograms have been used to derive and validate empirical models to fit the sorbent performance results, and apply them to overall model codes in WP1 and WP5.

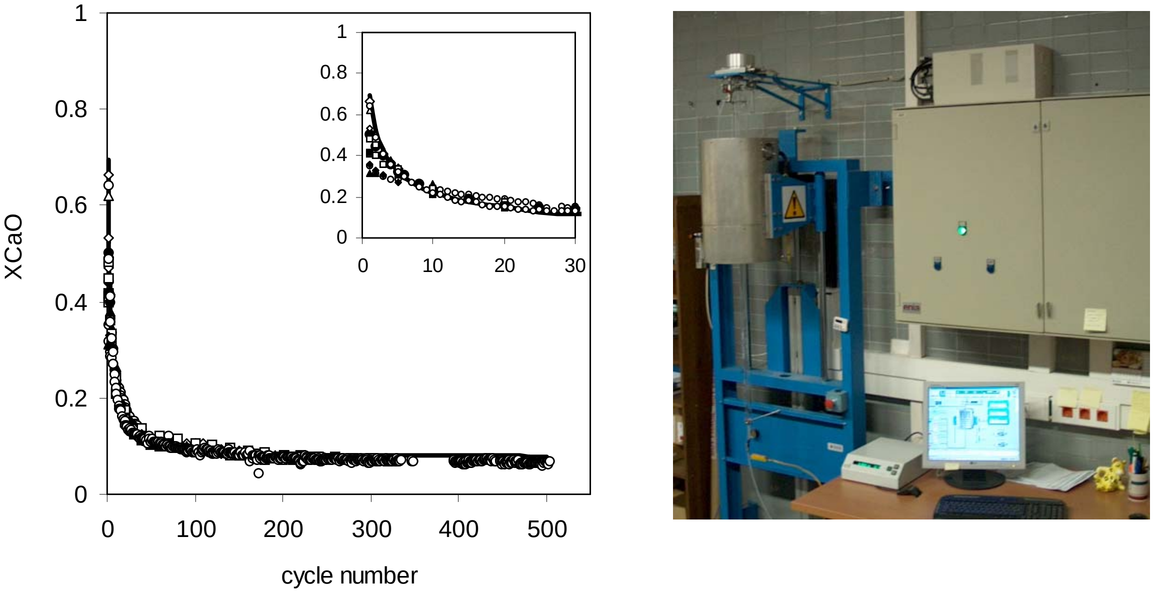

Figure 7: Example of a series of experimental CaO conversion vs. cycle number, and experimental installation designed and built at CSIC under this project for multicycle sorbent testing

Studies on textural analysis were also carried out at CSIC to explain the deactivation mechanism for the CaO along the cycles, finding two key mechanism: grain growth by sintering associated to a loss of available surface of reaction, combined with a product layer thickness measured to be around 50 nm thick, that drastically slows the progress of carbonation. In some circumstances, pore closure was also taking place together with an overall shrinkage of the particle.

Mechanical properties of the sorbent along cycles were investgated by ZSW. The mechanical stability of selected sorbent was characterised by detection of the extent of attrition after several hours with defined fluidisation in a FB (fluidized bed) reactor. Beside mechanical load, the sorbent material was exposed to several absorption/desorption cycles. The extent of attrition was measured by sieve analysis as particle mass < 200 µm generated during the test run. Several pre selected sorbent were tested. The measured ab-/desorption capacity correlates with results of TGA (Thermo Gravimetric Analysis). The first calcination was identified as the main reason for attrition of the sorbent under fluidzed bed conditions. To estimate the extent of attrition it was distinguished between “spontaneous” and “steady state” attrition. Supplying new sorbent to the process (e.g. $CaCO_3$ at the regenerator) it can be assumed that the amount of “spontaneous” attrition will be found in the flue gas of the calciner, while the “steady state” attrition will be found in the product gas from the gasifier also.

The search of precalcination conditions at CSIC to develop a more stable sorbent did not result in any improvement of sorbent performance along cycling. Differences were only observed in the first few cycles but they are not relevant to justify a continuous precalcination process in the ISCC process. The same negative conclusion applies to the use of additive salts (NaCl and Na_2CO_3 on limestones) that did not translate into any improvement of sorbent performance under the conditions tested. In contrast, reactivation with water is a very promising technique because its simplicity and because it has shown potential for practical application. Results were positive both in reactivating highly cycled solids (after a hundred of carbonation calcination cycles) and with materials deactivated in the first calcination trough extended calcination times and/or calcination temperatures up to 1100. Although results have been encouraging, the challenge is to avoid excessive particle fragmentation during hydration.

More controlled hydration processes, with steam instead of liquid water, need to be explored in the future. Testing of synthetic sorbents, recently reported in the literature to perform much better than limestones, was also conducted. The results were the same as those reported by the original authors (always at mild calcination conditions) but failed when repeated at conditions relevant for the ISCC process (calcination temperatures over 900ºC).
In conclusion, the laboratory studies in WP2 have demostrated that natural limestones should be a suitable high temperature sorbents for the ISCC systems despite their limited performance (even without a reactivation step). This is because it is affordable to use a large make up flow of limestone (specially if it is necessary to purge ash and sulphur compounds as discussed in section 1.3.1) and because the system can rely (at least in part) on a stable residual capture capacity (7-8% for high cycle numbers of Figure 2) of the sorbent, that remains even at the most demanding calcination conditions.

1.3.3 Regeneration process development (WP 3)

1.3.3.1 Introduction

The first task of WP 3 started with a screening of heat transfer methods showing the borders of technically and economically feasibility. Based on that, some of the initial ideas for the regeneration process had to be rejected and the experimental work was concentrated on the remaining options. During the ongoing ISCC process development the parameter frame conditions of gasification and regeneration became more clarified. Operating at elevated pressure helps in the gasifier to increase conversion rate and CO_2 absorption, but it is disadvantageous for the regeneration step. One of the project aim was to produce a concentrated CO_2 stream (> 95 %) in the regenerator. Equilibrium calculations showed that along with high pressure and pure CO_2 atmosphere very high regeneration temperatures were required. Under that challenging process conditions (up to 1000 °C and above) little experience was available which had to be generated in experimental work mainly at ZSW and VTT. In task 3.3 IVE evaluated the remaining regeneration options concerning their performance in an integrated ISCC concept.

1.3.3.2 Requirements on the regeneration process and boundary process conditions

Within the ISCC process the regeneration part has to fulfill the following requirements:
Recovery of lime
Flue gas containing > 95% CO_2
Energetic efficiency > 80 % compared to theoretical heat demand of the regeneration reaction
The cycle behaviour of the CO_2 sorbents is in principle influenced by the regeneration operation conditions temperature, surrounding gas composition (CO_2 concentration, steam, etc.), particle size conversion degree, etc. The most important requirements for the suitability of an absorbent for high temperature CO_2 removal are:
high reaction rate (in temperature range of 600-700°C)
chemical stability (reversible CO_2 uptake/release),
mechanical stability (attrition resistance) and
thermal stability (resistance for sintering at regeneration temperature)
In summary, there are several variables to be optimized in the calciner
calcination temperature and pressure:
atmosphere and steam content
The equilibrium calcination temperature is dependent on the CO_2 pressure illustrated in Figure 8. It is assumed that calcinations temperature should be at least 50 K above the equilibrium condition. This assumption was confirmed by regeneration experiments where temperature and reaction rate was determined for elevated CO_2 pressures in an lab scale fixed

bed reactor. The determined experimental start temperature for CO_2 absorption was closed to the equilibrium temperature, because high CO_2 partial pressure supports absorption. For the experimental determination of desorption the reactor temperature had to be increased significant above equilibrium.

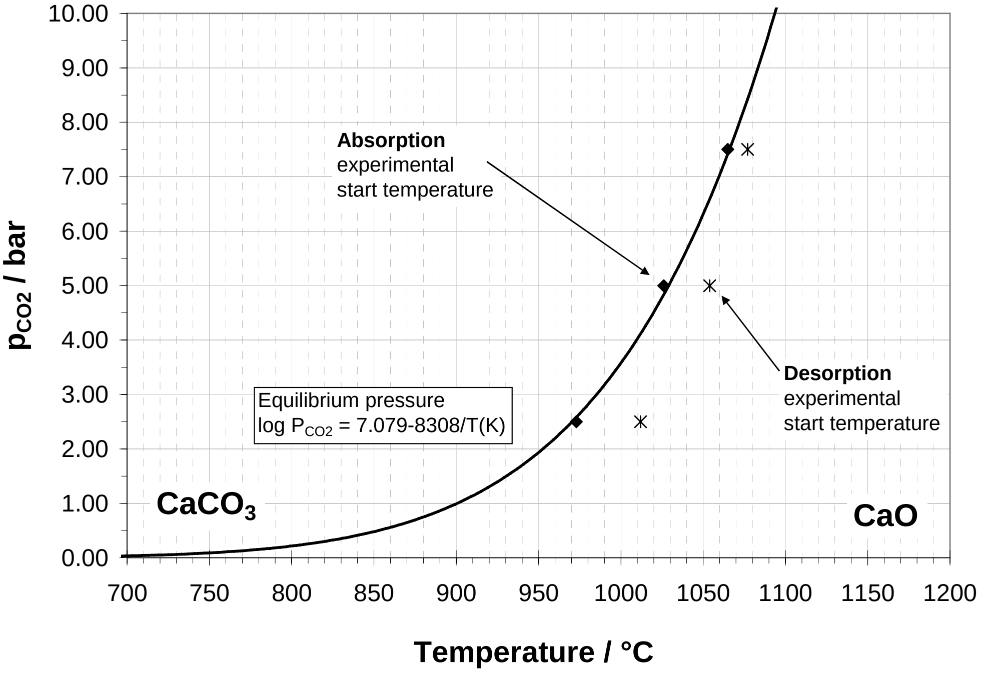

Figure 8: Equilibrium pressure of CO_2/CaO-System and experimentally determined start temperatures of ab-/desorption at different CO_2 pressures (atmosphere 100% CO_2)

Attempting the regeneration at high pressure (burning part of the fuel with O_2 in a FB) the maximum temperature tested before the ISCC project is 1060 °C (in the Acceptor Gasification Process). Experiments carried out with Thermo Gravimetric analyser at ZSW with calcite by varying the regeneration temperature show that with increasing regeneration temperature the sorbent performance (cycle stability) decreases. Figure 9 shows the experimental results. One can see that the CO_2 absorption capacity decreases more rapidly when the regeneration is carried out at higher temperatures. It is assumed that the sintering of pores at higher regeneration temperature causes the degradation of the sorbent.

However, the improvement of cycle stability due to lower regeneration temperature is not yet very clear, e.g. the results of CSIC obtained in the ISCC project so far are very similar (in terms of sorbent performance, see Figure 10) to those obtained by Curran et al (1967) during the testing of the Acceptor Gasification Process, where regeneration was carried out at 1060 ^{o}C with performance similar to CSIC results at lower temperatures.

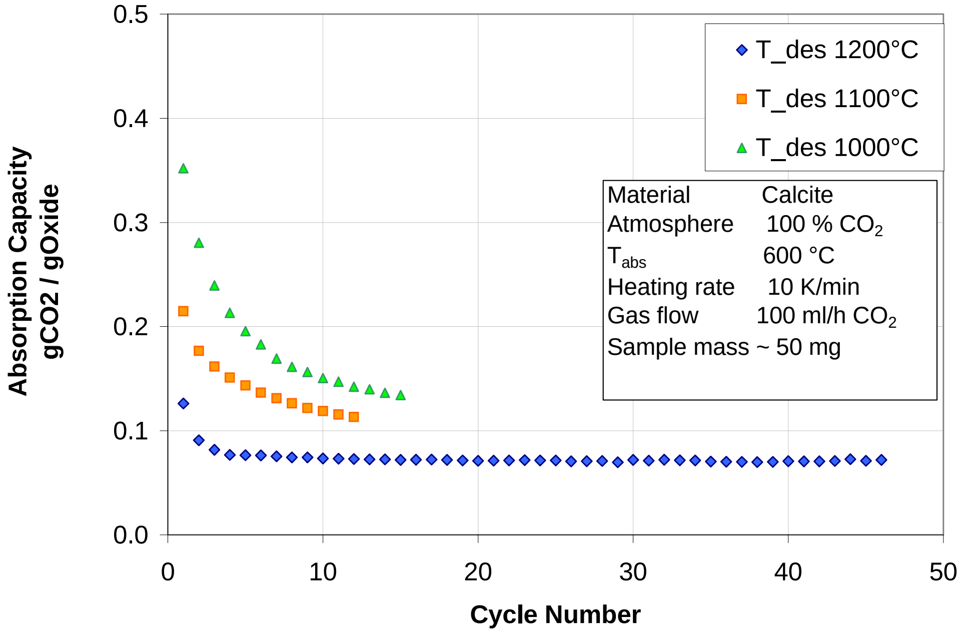

Figure 9: ZSW experiment; decay of absorption capacity of calcite with 1000 °C, 1100 °C and 1200 °C regeneration temperature (atmospheric, 100% CO_2)

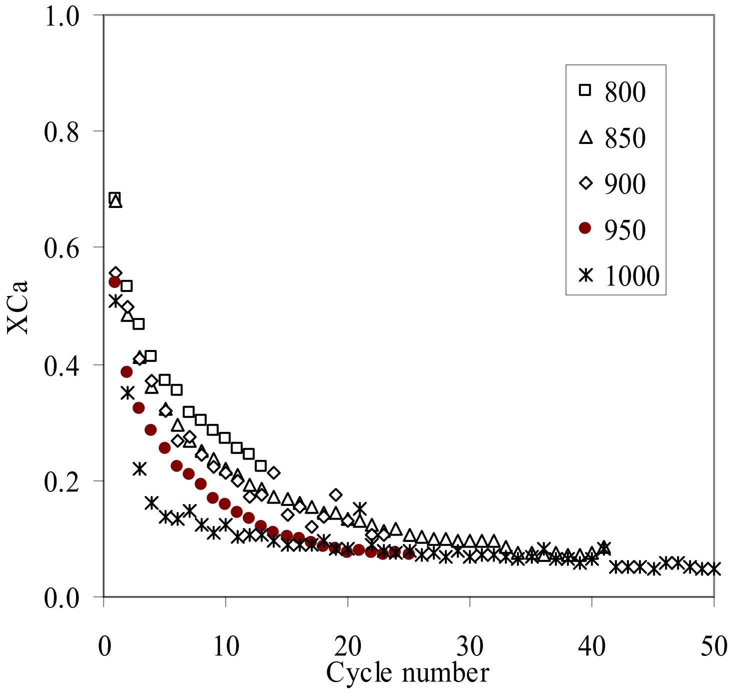

Figure 10: CSIC experiments; decay of absorption capacity (Xca is molar conversion of Ca to $CaCO_3$) of a limestone at different calcination temperatures. Carbonation T=650C, 10%vol CO_2, 10 min; calcinations at 10%vol CO_2, 10-20 min

However, all results show clearly that regeneration temperature should be as low as possible to avoid increased sintering effects.

On the other hand, the regenerator must be operated at same pressure level as the gasifier. The system cannot include pressurising or depressurising of hot solid streams because today there is no technology available. Concepts which would require cooling, pressurising and reheating of solid recycling flows are most probably not enough energy efficient or would require novel heat and material transfer methods in cooling and heating of solid output and input streams. Therefore the minimum operation pressure is determined by the gasification reaction and the maximum pressure by the regeneration process. To obtain a higher maximum pressure steam can be used to reduce the CO_2 partial pressure and thus temperature during regeneration.

1.3.3.3 Applicable solutions

Based on these results and conclusions a concept for oxyfuel regeneration was developed by IVE for a 20 bar gasification reaction.

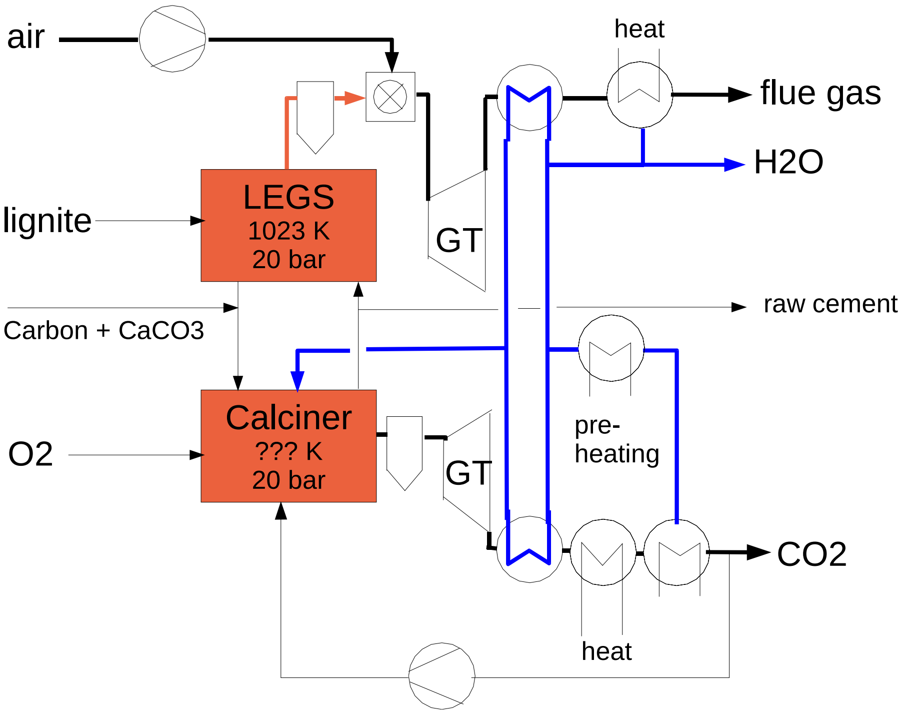

Figure 11: Concept for $CaCO_3$ regeneration using the oxyfuel approach

In order to reach the required high concentration of CO_2 directly in the effluent gas, combustion has to be carried out using oxygen combustion. To reduce the required regeneration temperature, steam is used for reduction of the CO_2 partial pressure. As a further restriction, melting of bed material must be avoided. For a mixture of $Ca(OH)_2$ and $CaCO_3$ in combination with steam, eutectic melting was observed at a temperature of only 650 °C if the steam pressure exceeds 13 bar [Curran, 1967]. Therefore the process design should restrict the steam pressure to maximum 12 bar in the regeneration unit. The resulting required operation conditions can be arranged by additional recycling of CO_2 to the regenerator input.

Then the solid output stream of the gasifier is combusted by a mixture of O_2/steam and recycled CO_2. The small pressure drops of the cyclones and the fluidised-bed can be overcome by the dipleg design. This type of gasification and combustion system has been demonstrated e.g. by the American Silvagas process and in the previous CO_2 acceptor gasifier.

This process concept has the following advantages:

1. The heat and mass transfer principle is sound and does not require "exotic" new materials or methods. Heat losses are also avoided as hot bed material is recycled without any additional cooling or heating stages.
2. The CFB reactors are typically high-throughput units and are suitable to large size.

3. The heating value of char is directly utilised for calcination of $CaCO_3$, which means that the process energetic efficiency is increased. In addition CaS formed during gasification from CaSO4 is quickly oxidised at high regeneration temperatures under reformation of CaSO4. This reaction is strongly exothermic with a reaction enthalpy of > 900 kJ/mol CaS at standard conditions. Because there is an enrichment of Sulfur compounds in the solid loop the CaS/CaSO4 reactions must be taken into account for calculation of energetic efficiency.

Basic assumptions for the simulations done by IVE using scilab software :

- Output streams gasifier from Table 6.1, related lignite input 100 kg
- Compressor efficiency = 0,9
- Gas turbine isentropic efficiency = 0,95
- Energy demand of fan for gasifier product recycling neglected
- Pure Carbon as additional fuel for regeneration
- Temperature gradient at pinch point 25 K
- Combustion chamber temperature 1400 °C
- Air temperature 15 oC
- Pressurized O_2 at the same temperature as compressed CO_2 available as input.

Regarding Figure 8 and assuming a process temperature 50 K above equilibrium conditions the required calcinations temperature is in the range of 1050 -1100 °C. The CO_2 pressure in the wet regeneration flue gas is 8 bar, the equilibrium pressure at 1100 °C is 10,7 bar. About 82 kg of O2 are required as regeneration input with 1,2 MJ electricity demand for O_2 generation [Wang, 2004]. The 96 Vol.-% CO_2 product gas is at a pressure of 1 bar available. To avoid eutectic melting of the bed due to high steam pressure 35 % of the dry flue gas has to be recycled to the regenerator input.

1.3.3.4 Conclusions

On the one hand the activities in WP 3 (screening, experimental, evaluation) showed the upper limitation of regeneration process conditions. On the other hand this knowledge is available to develop and evaluate first applicable solutions for the regenerator even for a pressurized ISCC system up to 20 bar. Further process evaluation can be found in WP 5.

1.3.4 Pilot Scale Experiments of Gasification and Regeneration Process (WP 4)

The major objectives of this work package was to test the feasibility of the pressurized gasification and calcination process. The work done is quite extensive. Therefore, the following information can just give the most important results.

Pressurised gasification tests in a BFB (Bubling Fluidized Bed) with a CaO bed material

Gasification experiments under pressure were conducted with the calcined limestone Rübeland.The process requires the continuous fresh flow of hot calcined lime to- and the continuous removal of carbonated lime from the gasifier in order to satisfy the heat balance (for the endothermic steam gasification) and to maintain an adequate CaO activity in order to capture CO2. This is not possible in the IVD stand-alone PFB gasifier; only semi-batch experiments are possible (constant CaO bed and continous feeding of fuel over time).

As described in Deliverable 1.1b, the temperature determines the partial pressure of carbon dioxide necessary for a desired capture efficiency. However, it is not feasible to conduct semi-batch experiments at high pressure due to experimental constraints. When executing a gasification experiment under pressure, some parameters are held constant: a) the gas velocity so proper fluidisation is achieved, b) the steam-to-carbon ratio so equilibrium CO_2 partial pressure exceeded (and absorption occurs), c) the bed mass which is limited by the bed diameter. Therefore, the duration of the CO_2 sorption period is:

$$\tau_{absorption} \propto \frac{M_{CaO}}{Pressure}$$

Therefore, for small fluidised bed facilities, only a maximum pressure of 6 bar is feasible if quasi-steady state conditions are to be achieved.

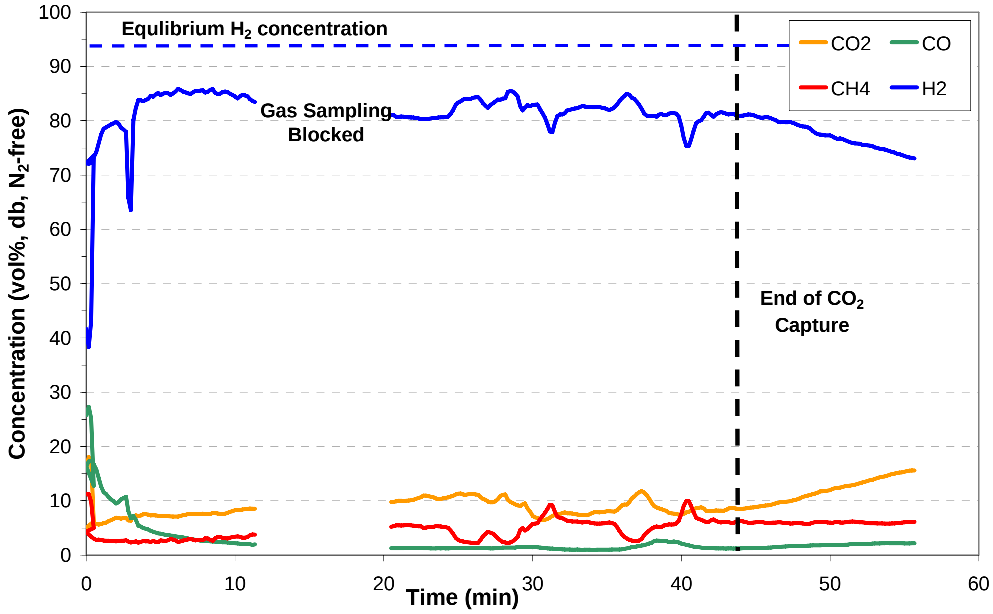

Figure 12: Run IVD-P#9. Cycle 1, P_{ave} = 3.5 bar, T_b = 700°C, WHVS = 0.65 kg daf/h-kg CaO_o , S/F = 5 kg/kg daf (Data: USTUTT)

As shown in Figure 12, the hydrogen concentration approaches equilibrium when using a freshly calcined lime bed. The results are very good considering the relatively small height of the bed (static height is 30 cm) which translates to a small gas-solid residence time of less than 1 s.

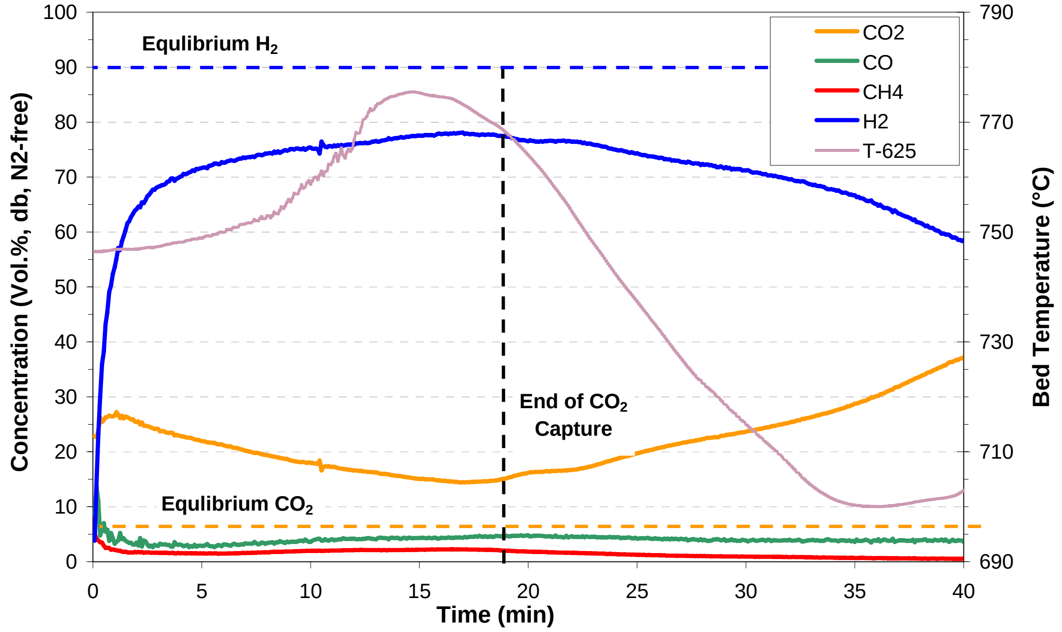

Figure 13: Run VTT-P4. P_{ave} = 6.0 bar, T_b = 750-770°C, WHVS = 3.4 kg daf/h-kg CaO_o , S/F = 7.5 kg/kg daf (Data: VTT)

The problem when operating at higher pressures is highlighted in Figure 5. The duration of CO_2 capture at 6 bar is only approximately 15 minutes compared to that of over 50 minutes at

3.5 bar. However, no significant steady state is achieved. The reduction in carbonation time is attributed to the much higher WHSV (3.4 h^{-1} compared to 0.65 h^{-1}). A quantifiable method to evaluate the experiment results is to compare the measured gas concentrations to equilbrium. Equilibrium concentrations were calculated using HSC 5.1 based upon the experimental conditions. The approach of the experimental hydrogen concentration to equilibrium is 88.2% and 86.2% for gasification at 3.5 bar and 6.0 bar respectively.

Gas species, including non-condensable hydrocarbons, sulphur compounds (H_2S and COS) and condensable hydrocarbons ("tars") were collected and subsequently measured. The results of run VTT-P4 show that small amount of unsaturated non-condensable hydrocarbons are tars present. The relatively small amounts of both condensable and non-condensable hydrocarbons are attributed to the feedstock, which in this case was Vattenfall SP char. A char feedstock was selected because the fuel is fed from the top of the reactor and it was observed that the volatiles from the coal did not come in direct contact with the bed, and therefore little CO_2 absorption occurred. Values of H_2S and COS were below the detection limit (< 0.5 ppmv).

Table 3: Overview of parameters and major gas components for run VTT-P4

Parameter		Gas Species	Vol% - N_2 free
Bed Temp	750-770°C	H_2	77.7
Pressure	6.0 bar	CO_2	14.8
WHSV	3.4 kg daf/h-kg CaO_o	CO	4.4
Steam / Fuel	7.5 kg/kg daf	CH_4	2.2
$X_{CaO}^{\dagger}$	14.7 mol%	C_xH_y	0.7
Hydrocarbon	**ppmv - N_2 free**	**Tar**	**mg/Nm^3 N_2-free**
C_2H_6	364	Toluene	128
C_2H_4	364	Naphthalene	366
C_2H_2	0	Biphenyl	29
C_3H_8	0	Phenanthrene	60
C_3H_6	17	Total Tar	583
C_3H_4	0		

† X_{CaO} is the mol% of CaO converted to $CaCO_3$

The Gasification-Calcination Cycle

In a continuous process, the CaO bed material will have an average maximum activity depending on the process purge rate (Deliverable 1.1b). The implications of this decrease in bed activity is discussed in detail in Deliverable 4.4. Cyclic experiments were conducted to evaluate the performance of the lime bed as a function of bed activity since the average bed activity in the continuous ISCC process will be below 30%. All semi-batch experiments lasted over 50 minutes in total coal feeding time. Details for the procedure is found in Deliverable 4.2.

The bed was calcined in batch mode in order to regenerate the bed material before the subsequent gasification test. Calcination was performed in batch mode and although the average temperature during calcination was 890°C, short temperature highs of 920°C occurred which may have led to sintering. However, the results agree with cyclic experiments at atmospheric pressure in which the bed was calcined at controlled temperature of 850°C.

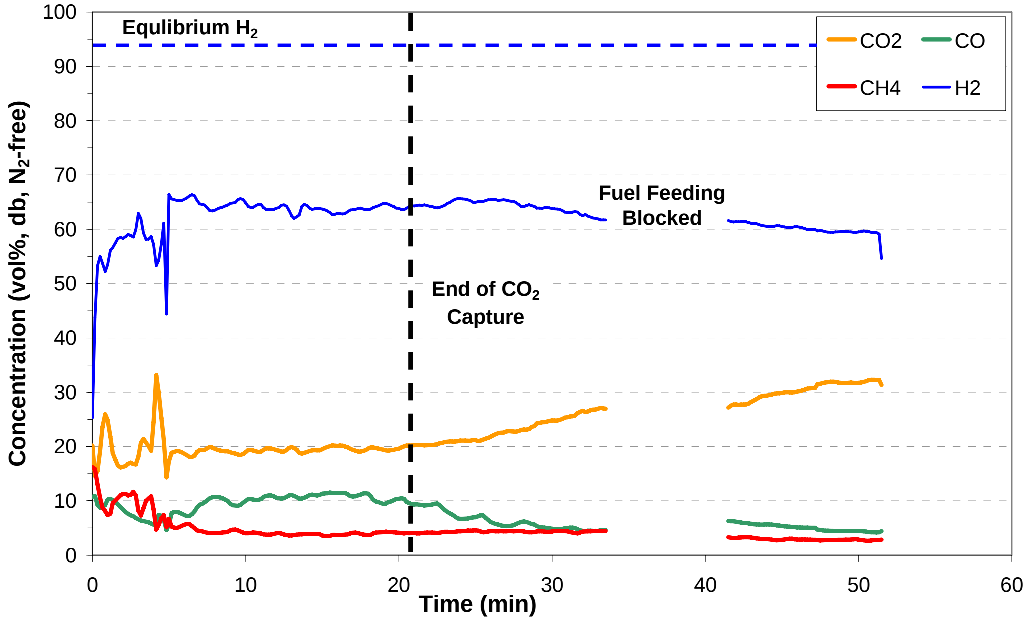

Figure 14: Run IVD-P#11.Cycle 3, $P_{ave} = 3.5$ bar, $T_b = 670°C$ WHVS = 0.65 kg daf/h-kg CaO_o , S/F= 5 kg / kg

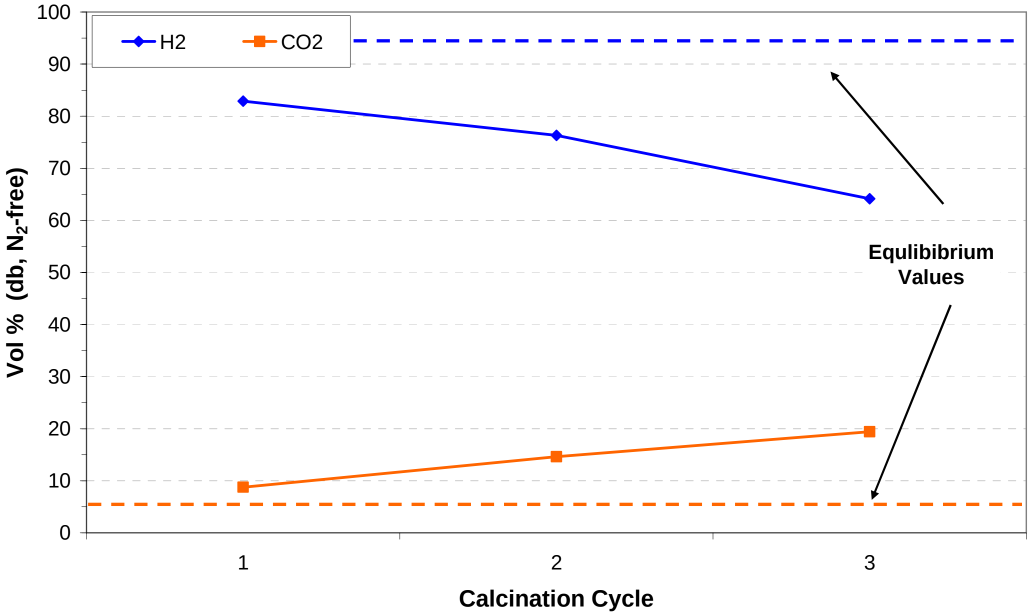

Figure 15: H_2 and CO_2 Concentration as a function of Calcination Cycle

P = 3-3.5 bar, T=670-715°C, S/F = 5, WHSV = 0.65 , kg daf/h-kg CaO_o calcined Rübeland limestone

Table 4: Approach to hydrogen equilibrium concentration

	IVD-P#9	IVD-P#10	IVD-P#11	VTT-P#4
Cycle	1	2	3	1
Approach to H_2 Equilibrium, %	88.2	81.2	68.3	86.3

Figure 16 demonstrates the influence of various parameters on tar concentration and producer gas yield. It is well known that temperature has a large influence on tar density. This also holds true for the C2H gasifier, where for the same weight-hourly space velocity, the gravimetric tar concentration decreases from 9.8 g/Nm^3 to 1.7 g/Nm^3 when the fluid bed temperature increases from 655°C to 722°C. The fluid bed also shows that CO_2 absorption process increases the rate of gravimetric tar destruction (1.7 g/Nm^3 compared to 3.3 g/Nm^3 at 712-720°C and 9.8 g/Nm^3 compared to 13.2 g/Nm^3 at 655°C).
This mechanism is not well understand and is currently being investigated. The effect of calcine number is minimal at both atmospheric and pressurised operation.

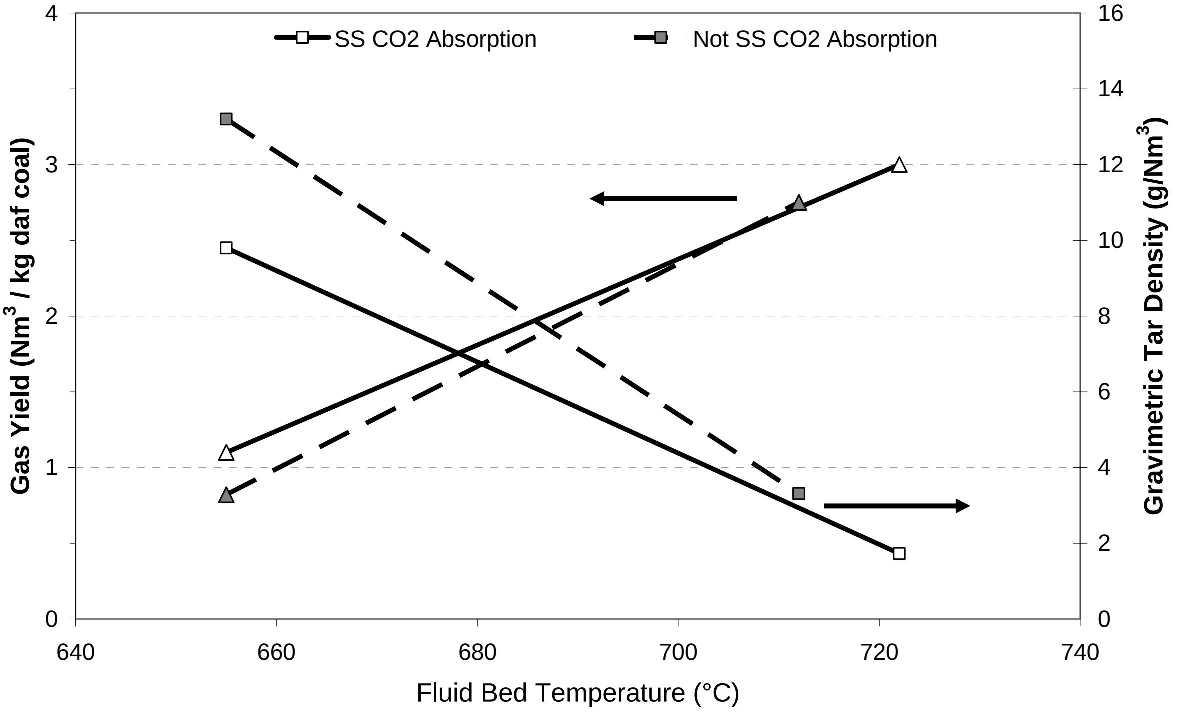

Figure 16: Influence of bed temperature and steady-state[†] CO_2 adsorption on tar concentration and gas yield. ([†] Steady-state CO_2 absorption is characterised by constant CO_2 and H_2 concentrations in the product gas)

The following observations are made from the previous sets of experiments:

- Hydrogen gas concentrations close to equilibrium (> 82 vol.%) were achieved. Improved CO_2 capture can be achieved by increasing the fluidised bed height thereby increasing gas contact time with CaO.
- Gravimetric tar density under 2 g/Nm^3 is achieved for temperatures at 720°C during CO_2 adsorption. The tar content is low enough so as not to interfere with downstream gas-cleaning unit operations.
- Deactivation of CaO with respect to calcine number was observed. The degree of deactivation was more severe under pressurised conditions due to uncontrolled calcination conditions (to be discussed in Task 4.4)
- Continuous circulating dual fluidised bed system required in order to properly characterise and validate the ISCC process.

Conclusions:

The feasibility of the process has been demonstrated. The semi batch tests showed good and promising results and the deactivation of the sorbents was corresponding to the TGA results which confirmed the design rules used in the following work packages. The future works should concentrate on testing the coupled process in an interconnected fluidized bed system.

In a first step atmospheric testing is suggested before a pressurized plant with its huge investment requirements is planed.

1.3.5 Process design and technical assessment (WP 5)

The objectives of WP5, according to the Work Programme, were the following:

- Determine the plant design with all required components
- Simulation of the overall process and evaluation of the efficiency
- Comparison with alternative processes for very low emission power generation from coal
- Design of a semi-technical plant to demonstrate the technical feasibility of the process and industrial plant design

By the end of the project, the original goals of the WP have been achieved. A description of the work conducted within each Task of WP5 is provided below.

1.3.5.1 Thermodynamic integration of coal drying system

BTU Cottbus was integrated in this Work package to perform the basic engineering of a drying system for the high moisture low rank coal to be adapted to the developed gasification/reforming and regeneration processes. Initially, different existing pre-drying concepts for brown coal, considering the boundary conditions and the basic requirements of the connected processes, were elaborated. For keeping the boundary conditions and the basic requirements of the ISCC process, a thermal drying process was selected for the ISCC process. The highest water vaporisation and mass throughput can be realised by the fluidised bed dryer. Especially the pressurised fluidised bed contact drying using steam leads to a process with compact low cost design. The selected drying system was adapted principally to the basis and boundary conditions of the ISCC process. Based on the defined ISCC process plant sizes (1 MW and 500 MW electrical output), dryer units process flow sheets were created and all main components were designed. The thermodynamic results (pressures, temperatures, enthalpies etc.) of the dryer units were used for the thermodynamic analysis and optimisation of the overall process. Many variants for the thermodynamic integration of the dryer unit into the overall process were worked out. The variants were simulated by NTUA to have basics for rating, resulting into a final variant, which is optimal for the dryer unit and for the entire process.

However, it was finally decided to exclude the pre-drying of the lignite from the ISCC cycle as it thermodynamically does not make any sense. Instead of this finally evaluated concepts use the moist coal which is directly injected into the boot of the gasifier and recirculated product gas is used for the fluidization of the reactor.

1.3.5.2 Thermodynamic analysis and optimisation of the overall process

NTUA/LSB, has devoted its efforts during the first 18 months of the project on the creation of the basic thermodynamic model of the ISCC process, covering fuel preparation, gasification/reforming, regeneration and the products, using the thermodynamic cycle simulation software ENBIPRO (modified with new routines) and the software code ASPEN, especially for the modelling of the gasifier/regenerator module. Following discussions on the size of the industrial power plant to be modelled, a ca. 500 MW Combined Cycle power plant was selected and the basic model was created. The reference power plant used for the modelling of the process is a 500 MW natural-gas fired single-shaft Combined Cycle plant with a net efficiency of 56.5%, consisting of one gas turbine, one steam turbine and a triple-pressure HRSG. For the simulation of the Combined Cycle fired with the carbon-free fuel gas produced from lignite gasification, it is assumed that the water/steam cycle and the heat input provided by fuel remain the same as in the original case. In addition, the flue gas flow entering the turbine is kept at the same level, whereas a coal drying system was integrated in the process based on the input from BTU.

Following this study, NTUA/LSB proceeded in the second half of the project period in the optimisation of the process. In order to determine an optimised power plant that utilises a carbon-free fuel, which is produced through the carbonation/ calcination process, several power plant configurations were evaluated, taking into account different reactor (gasifier and regenerator) operating conditions as well as different power plant outlays. The simulation of the gasifier and the regenerator was based on the equilibrium of the carbonation and calcination reactions respectively in specified conditions. On the other hand, the power plants' performance was defined through heat and mass balance solving of the thermodynamic circuits. In all cases examined, the lignite input is no longer pre-dried, as was the case in the configurations studied for the basic thermodynamic model. Eight power plant configurations in total were investigated in order to determine the optimised scenarios. The differences between the examined cases concern:

- The gasifier and the regenerator operating pressure
- The regenerator oxidation medium (pure oxygen or air). In the case of air, the CO_2 is captured from the flue gases by means of amine scrubbing
- The use or not of an expander to produce power from the exploitation of the regenerator flue gases
- The use of a Humid air turbine to produce power from the exploitation of the high-moisture regenerator flue gases
- The use of a membrane ASU instead of a cryogenic ASU

In all cases, the GT power output was retained at the same level (288.5 MW), reflecting the choice of a certain GT for all configurations. In cases 1, 2, 4, 5, 6, 7 and 8 the flue gas that exits the regenerator is expanded to produce electric power. Consequently, in the cases where the char and the lignite that enters the regenerator are combusted with oxygen, which is produced by a cryogenic ASU, the exhaust gas heats the nitrogen up to 150 ºC for the regeneration of the Air Separation Unit molecular sieves. In all cases, except case 4 (air-blown regenerator) the flue gas is used in a water-gas heat exchanger to produce steam. The high moisture content of the lignite eliminates the need of extra steam that would otherwise be required for gasification.

The final product of the regenerator (rich CO_2stream) is compressed up to 110 bar in a 5 stage compression process with intermediate cooling. These steps are: 1.0 - 3.2 bara, 3.2 - 10.4 bara, 10.4 - 33.5 bara, 33.5 - 71 bara and 71 - 110 bara. The flue gas condensation occurs before the CO_2 compression and most of the water content is removed. The TEG dehydration system is placed after the 33.5 bara step and the removal of non-condensables at 71 bara. On the other hand, in the oxygen-fired regenerator cases with the cryogenic ASU, the air compression for the ASU occurs at 2 steps with intermediate cooling. These steps are: 1.013 - 2.37 bara and 2.37 - 5.52 bara. The power consumption for the air separation has been estimated to 203 kWh/ t of pure O_2. The oxygen purity is 95 v% with the remaining 5 v% mainly Ar. The oxygen that exits the ASU is compressed up to the regenerator pressure for the char and lignite combustion. In all the examined cases the Calcium/ Carbon ratio is 3- 3.5, whereas the purge rate is 10%.

The above-mentioned cases can be grouped in two categories: a) Power plants that are based on commercially available or state of the art components b) Power plants representing more advanced solutions. The first category includes the cases from 1 to 5, while the second the cases 6 to 8 (high-efficiency advanced HAT concept). In summary, in the second category (cases 6, 7 and 8) the power plants' efficiency has been assessed to 37.4%, 37.6 % and 40.2 % respectively. Among the cases of the first category, case 2 and 4 have demonstrated the best performance with 37.1 % and 37.5 % net efficiency, while the poorest performance is demonstrated for the case 1 power plant outlay, with a calculated net efficiency of 31.2 %.

1.3.5.3 Comparison with alternative processes and very low emission CO_2 concepts

Sorption enhanced coal gasification to hydrogen or synthesis gas arrives to be one of the most popular areas in the applied energy industry research of last few years. The state of the art of different versions of the process has been reviewed and discussed during the special ISCC conference in Stuttgart in 2005. The concept seems to be the one of the most popular technology explored pathway in the frame of Zero Emission Fossil Fuel Power Plant European Technology Platform. However this new ideas strongly compete with more mature classic coal gasification technologies (IGCC). The advantages of the sorption enhanced processes are based on reduced consumption of pure oxygen in the full technology scheme, on the anaerobic character of coal gasification of LEGS what radically decreases NO_x emission and on possibility of work with high sulphur content fuels. In this last case the hydrogen rich gaseous product generated in the process could be used for on site reduction of gypsum during the sorbent regeneration, as described in the most recent patent application by Anthony (2007). The following trends could be identified in the sorption enhanced gasification process development:

1. Searching for the specially prepared sorbents with the high carbon capture capacity and long term stability of the sorption properties in the calcination-carbonation cycles.
2. Modification of the coal gasification process parameters looking for working conditions warranting the extended service of the sorbent in c-c cycle with it's acceptable capture efficiency
3. Exploitation of synergy effects through combining the gasification process with other industrial technologies able to consume the by-products. Integration of LEGs with cement production is extensively explored in the frame of this project. The low temperature, sorption enhanced biomass – lignite co-gasification process carried out in the multiphase, pressurized sub-critical region of water is the other example of this type of development.

In the following a preliminary techno-economic evaluation of the investigated cycles is summarized. A deeper economic analysis of the finally decided plant scenarios can be found in section 1.3.6. Because the economic data of LEGS power stations have not been available at the end of ISCC project, the comparison of LEGS based CO_2 emission free power stations with Oxyfuel and IGCC concepts is based on the simulations of ISCC project partner NTUA. It is assumed that a sufficient S removal can be achieved in the gasifier and that high temperature gas cleaning in reducing atmosphere is not a crucial issue for the investigated LEGS concepts. Then the following conclusions are derived:

- The performance of LEGS based power stations is significantly improved if the solid purge is used for cement clinkering with related energy savings and if the outstanding high CO_2 capture of 115 % related to C input with coal is taken into account (from 37 – 38 % to 42%)
- Compared to Oxyfuel LEGS based power stations show a better performance in terms of efficiency for power production (37 -38 % compared to 34 %) and CO_2 capture (115 % compared to 100 %) even without any efficiency corrections related to cement industry
- To compete with Oxyfuel concept a specific investment in the same range (1500 €/kW electricity) should be achieved for LEGS based power stations with integrated cement production.
- Advanced IGCC concepts show a better efficiency for power production compared to LEGS power stations (42 % compared to 37 – 38 %) if energy savings and CO_2 emission reduction for the cement process are not taken into account for LEGS power stations

- A LEGS power plant with integrated cement production has comparable efficiency with 50 % higher CO_2 capture rate
- It is assumed that LEGS power stations are less expensive compared to advanced IGCC concepts with investment of about 1800 €/kW
- If Ion Transport Membranes are available for O_2 separation from air the efficiency can be improved especially for Oxyfuel and LEGS based power plants because of their higher O_2 demand.

LEGS based power stations are a promising alternative for CO_2 free power production from coal if the following remaining technical and economical assumptions become true:

- Integration of cement production leading to outstanding ecological advantages
- Sufficient S removal in the gasifier
- Hot gas cleaning to GT quality at moderate cost
- Total investment for combined power and cement production < 1500 €/kW electricity

It was decided to use the cases 2 and 7 of the NTUA/CERTH simulations for comparison with other processes. The simplified flow sheet for the case 2 power station is illustrated in the figure below.

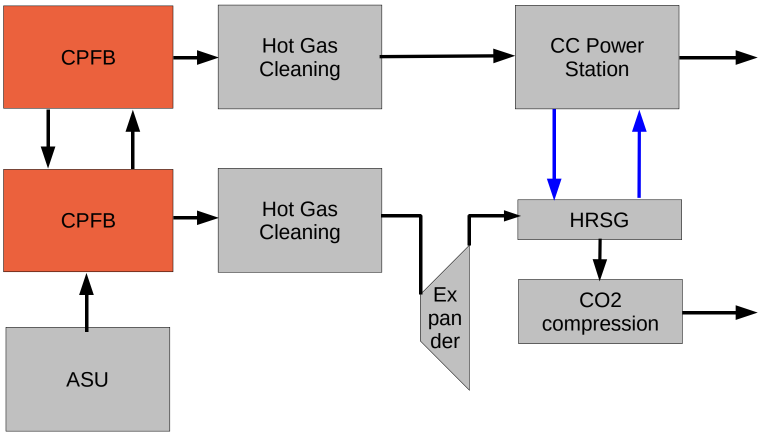

Figure 17: Simplified flow sheet for case 2 ISCC station

In Case 7 the Expander was replaced with a HAT Turbine and generated steam is injected to regeneration in order to dilute the CO_2.

1.3.5.4 Comparison with Oxyfuel Concepts

The Oxyfuel concept was studied in detail by Andersson and Maksinen [1]. O_2/CO_2 combustion combines a conventional combustion process with a cryogenic air separation process. The fuel is burnt in oxygen and recycled flue gas, yielding a high concentration of CO_2 in the flue gas which reduces the cost of CO_2 capture. Their work applies an O_2/CO_2 concept to commercial data from an 865 MWe lignite fired reference power plant and large air separation units (ASU). A detailed design of the flue gas treatment before transportation of the separated carbon dioxide is also proposed. It is assumed that the sulphur dioxide can be sequestered together with the carbon dioxide, provided that the gas is dry, and, consequently,

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[1] Andersson,K., Maksinen, P.: Process evaluation of CO2 free combustion in an O2/CO2 power plant; master thesis, Chalmers University of Technology, Sweden; www.entek.chalmers.se/~klon/msc

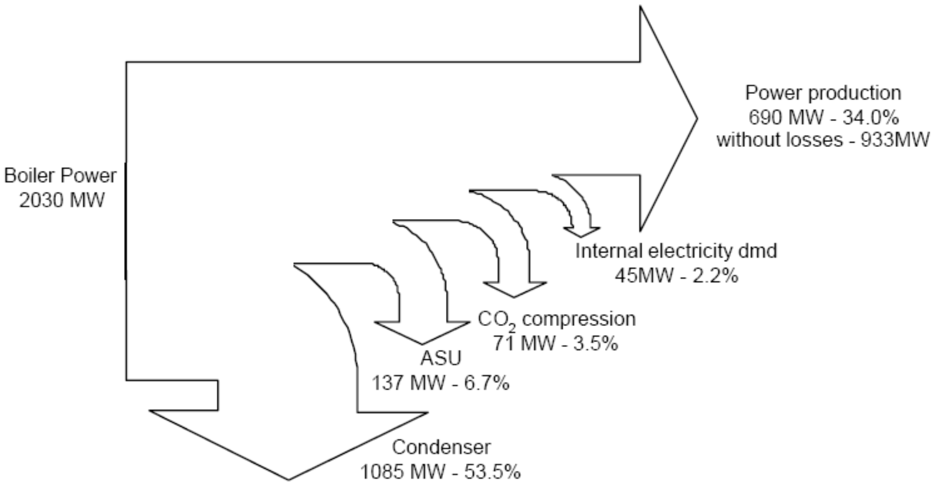

Figure 18: Energy flow in Oxyfuel power station

there is no need for a desulfurising unit. Since the investment cost of an ASU is slightly lower than for the desulfurising unit, the investment cost of the O_2/CO_2 plant will be slightly lower than for the reference plant. With all identified integration possibilities the net electrical efficiency becomes approximately 34%, which should be compared to 42.6% in the reference plant. The basic results can be summarised in a Sankey diagram.

The basic data of the LEGS and Oxyfuel power station are summarised in the following table:

	Case2	Case7	Oxyfuel
Total Heat Input, MW	2030	2030	2030
ASU power incl. O_2 compression, MW	115.6	128.6	137
CO_2 compression 110 bar, MW	94.9	97.84	71
CO_2 recycle, MW	0.00	39.24	0
Consumption, MW	210.7	266.0	253
Power, MW	963.6	1029.0	933
Net Power Produced, MW	752.9	763	680
Net Efficiency LHV based	0.37	0.38	0.34
Fuel saving cement (150 kJ/molCaO), MW	201.1	179.3	0
Corrected efficiency	0.41	0.41	0.34
CO_2 capture rate, %	118	117	100

1.3.5.5 Comparison with Advanced IGCC Concepts

The most advanced IGCC concept with integrated CO_2 capture is described by Ogriseck from Bergakademie Freiberg [2]. A power plant efficiency of 41% is achieved with a CO_2 capture rate of 78.5 %. In the following table, the basic data of the LEGS and IGCC plant are compared based on the same fuel input. Regarding the data of the LEGS and IGCC plant, it can be seen that the efficiencies are comparable if the higher CO_2 capture and cement energy saving is taken into account for the LEGS concepts.

Regarding the process complexity, both concepts use CC power plant technology. The LEGS requires two CPFB systems (gasifier and calciner) whereas the IGCC concept requires additional lignite drying, and a number of process steps for CO_2 and S removal (see red box in Figure 1.3.5.1). Because in the IGCC design the lignite drying and gasifier are fluidised bed

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[2] Ogriseck, K.: Untersuchung von IGCC-Kraftwerkskonzepten mit Polygeneration und CO2-Abtrennung, VDI Verlag, Düsseldorf 2006

reactors as well, a rough estimation gives the same investment for the IGCC drying + gasification as for the two CPFB reactors in the LEGS concept. With this assumption, the LEGS process has significant lower investment costs compared to the IGCC concept with an investment of about 1800 €/kW electricity, because CO_2 removal is an in-situ feature of LEGS and sulphur is as well already captured to a large extend in the bed.

	Case2	Case7	IGCC
Total Heat Input, MW	1620	1620	1620
ASU power incl. O_2 compression, MW	92	103	66
CO_2 compression 110 bar, MW	76	78	59
CO_2 recycle, MW	0.00	31	0
Consumption, MW	168	212	202
Power, MW	769	821	885
Net Power Produced, MW	601	609	673
Net Efficiency LHV based	0.37	0.38	0.42
CO_2 capture rate, %	118	117	79
Efficiency 79 % capture	0.39	0,39	0.42
Fuel saving cement (150 kJ/molCaO), MW	160	143	0
Double Corrected efficiency	0.43	0.43	0.42

This is as well a first indicator that investment for LEGS based power plants might be in the area of 1500 €/kW investment costs as for Oxyfuel plants.

1.3.5.6 CO_2 storage options

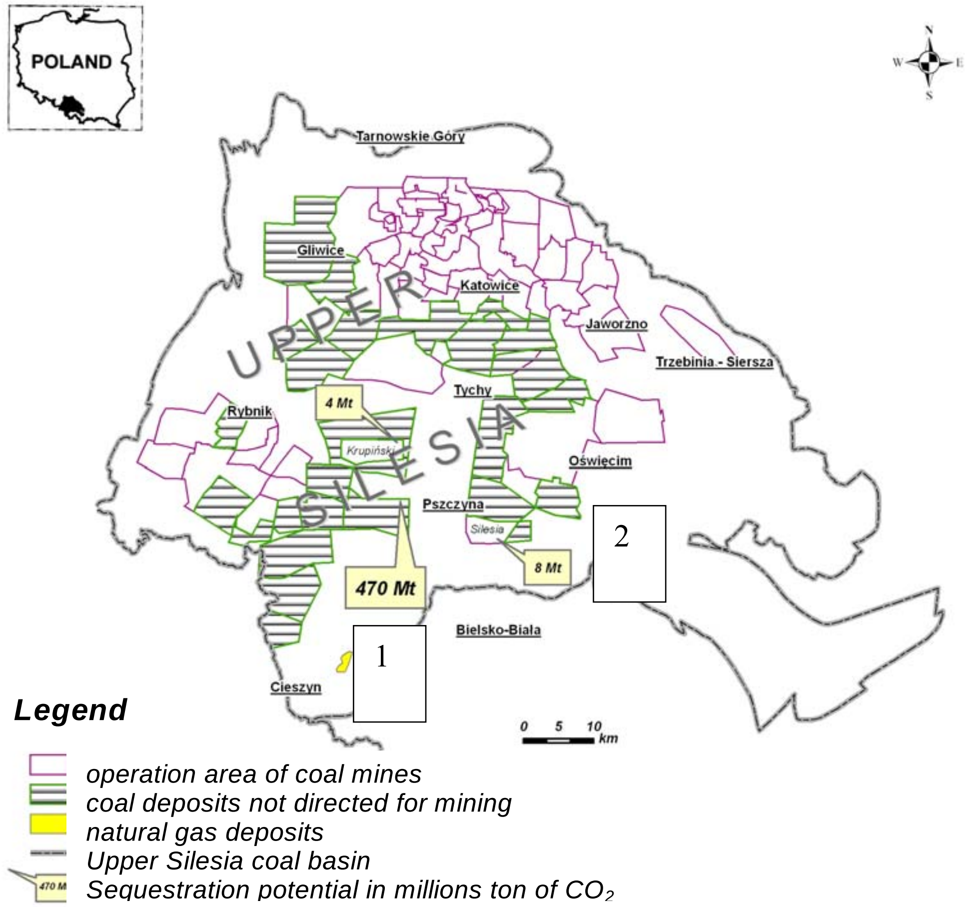
Work on the CO_2 storage options for the ISCC process was also conducted. The objective of the work was to present the potential of CO_2 sequestration in the region of Upper Silesia. In order to assess the potential, certain rock traps and rock formations suitable for sequestration have been selected for the analysis. Research concerning the possibilities of injecting compressed gases into strata has been carried out in numerous European countries as well as in Poland. However, for such a heavily industrialized region as the Upper Silesia, the research activities had to be focused only on certain coal seams. Therefore the conditions of CO_2 storage in other formations such as the aquifers or rocks have not been subject to the research. The most frequently proposed reservoirs for underground gas storage are the following:

- Deep aquifers and rock formations
- Coal seams
- Depleted oil and gas bearing formations
- Rock Salt formations
- Mineral sequestration (carbonatization of rocks)

The carbon dioxide storage potential in the Silesian region, as assessed in this study, is presented in Figure 19. The requirements for the reviewed sequestration options combined with the LEGS technology should be rated according to the business attractiveness as follows:

1. H_2 production through adsorption enhanced gasification of bituminous coal combined with CO_2 storage in unmined, deep coal deposits integrated with a parallel coal bed methane recovery as the additional source of fuel, or the gasification raw material
2. Lignite gasification in the plants located close to the coal deposits with storage of CO_2 in the deep aquifers and rock formations, with pipeline transport of carbon dioxide from generation to the storage site.
3. Adsorption enhanced gasification of coal, biomass or refinery waste to hydrogen in locations determined according to the availability of sources of carbon fuels and recognized local market for hydrogen consumption. The case is addressed to the low

and medium scale gasification plants with carbon dioxide sequestration options employing the local storage opportunities (mineral sequestration, depleted oil and gas bearing formations, rock salt formations)



Map of the Silesian region with deposits of hard coal shown and the carbon dioxide storage potential visualized. Location of CO_2 in deep coal deposits with CBM recovery seems to be the most feasible option for further technology development. A more ambitious but difficult idea aimed on the development of the LEGS type process assumes the integration in-situ of coal conversion with carbon dioxide storage and CBM utilization performed underground. The concept, derived from the ISCC and RECOPOL results will be explored further in the frames of the HUGE project just accepted by CSRF. The optimal locations of the plant are the area in the vincinity of the Polish – Czech border – the Morcinek coal mine, or the Nadwislanska coal deposits close to the Dwory – Oswiecim Chemical Plant

Figure 19: Carbon dioxide storage potential in the Silesian region

Taking into account the proven data on carbon dioxide storage potential in the Silesian region, it is rational to state that, as yet, the technically feasible option of CO_2 sequestration is the storage of carbon dioxide in deep unmined coal beds combined with a parallel technology of ECBM recovery and utilization. In the most feasible combined approach, this option will produce the opportunity of total coal to hydrogen conversion up to 100 millions of ton of hard coal during the whole time of the project. The idea is the main business-aimed result of the ISCC project and was partly exploited in the preparation of the HUGE project proposals just accepted by CSRF.

1.3.5.7 Plant design (semi-technical and commercial)

NTUA/LSB based on the findings of the thermodynamic optimisation work, completed the study of synthesizing a complete plant design for a 1 MW semi-technical plant and a 500 MW (coal) commercial design. The proposed plant design for a semi-technical 1 MW plant aims at the proof of the concept and therefore the proposal aims at simplicity and low cost of all equipment other than the gasifier/regenerator module, this being the basic module of a future ISCC commercial power plant. Both gasifier and regenerator operate at 10 bar and the regenerator is oxygen-blown. The power generation block is based on an Allied-Signal ASE8-1000 Gas Turbine, with a nominal output of 556 kW on natural gas. No CO_2 sequestration is envisaged for the plant and a high temperature inlet expander (not readily available in the market) is not included in the concept, so the flue gas steam exiting the gas turbine is expanded and mixed with the regenerator stream and then passes through a single-pressure HRSG producing steam that is expanded in a condensing steam turbine.

Four power plant configurations were identified as having the greatest potential for a 500 MW ISCC commercial power plant design with CO_2 capture, based either on commercially available/state of the art components or representing more advanced solutions.

State of the art 500 MW ISCC Power Plant designs:

Case 2: Oxygen-blown regenerator, expander producing extra power from regenerator flue gas. Efficiency: 37.1%

Case 4: Air-blown regenerator, expander producing extra power from regenerator flue gas, CO_2 removal from regenerator flue gas flow with amine scrubbing. Efficiency: 37.5%

Advanced 500 MW ISCC Power Plant designs:

Case 7: Oxygen-blown regenerator, HAT (Humid Air Turbine) for power production, H2O and CO2 recirculation in the regenerator, expander producing extra power from regenerator flue gas. Efficiency: 37.6%

Case 8: Oxygen-blown regenerator with membrane air separation unit, HAT (Humid Air Turbine) for power production, H_2O and CO_2 recirculation in the regenerator, expander producing extra power from regenerator flue gas. Efficiency: 40.2%

For all four cases, design data were provided, for each basic power plant unit, as follows:

- Gasifier/regenerator module (including coal handling and solids/solids heat exchanger)
- ASU or air compressor
- Power generation module (including all water/steam heat exchangers)
- Flue gas cooling and CO_2 compression train

1.3.6 Socio-economic assessment (WP 6)

1.3.6.1 Life Cycle Analysis of the ISCC process

Details of the work done in this work package can be seen in the deliverable D6.1. The final activity report just summarizes the work done and the results of this reporting period.

The technical, economic and environmental performance of six IGCC plant designs, including four ISCC plant concepts with CO_2 capture and two conventional Shell type IGCC plants, one with and one without CO_2 capture, fuelled by lignite coal were studied through ECLIPSE simulations. ECLIPSE mass and energy (M&E) simulation results were validated with NTUA results from WP5.2 and 5.4. The main aims and objectives of the techno-economic study were to evaluate the impacts of new ISCC CO_2 concepts on the plant output, efficiency, CO2 capture efficiency, environmental emissions, investment costs, cost of electricity and CO2 mitigation costs including sensitivity studies. The following cases were reviewed:

A-ISCC Case 2: Oxygen-blown regenerator; expander to produce extra power from regenerator flue gas; gasifier and regenerator temperatures: 750°C and 1100°C, respectively; operating pressure 20 bar; cryogenic Air Separation Unit (ASU) for O_2 supply to the regenerator.

B-ISCC Case 4: Air-blown regenerator expander to produce extra power from regenerator flue gas; gasifier and regenerator temperatures: 750°C and 1100°C, respectively; operating pressure 20 bar; CO_2; amine scrubber to remove CO_2 from regenerator flue gas; cryogenic Air Separation Unit (ASU) for O_2 supply to the regenerator.

C-ISCC Case 7: Oxygen-blown regenerator, expander to produce extra power from regenerator flue gas and Humid Air Turbine (HAT) for power production; gasifier and regenerator temperatures: 750°C and 1050°C, respectively; operating pressure 20 bar; cryogenic ASU for O_2 supply to the regenerator.

D-ISCC Case 8: Oxygen-blown regenerator, expander to produce extra power from regenerator flue gas and Humid Air Turbine (HAT) for power production; gasifier and regenerator temperatures: 750°C and 1100°C, respectively; operating pressure 20 bar; membrane ASU for O_2 supply to the regenerator; CO_2 and CO_2 recirculation in the regenerator.

E-Conventional (With CO_2 capture): Lignite fuelled oxygen blown shell gasification, cryogenic air separation unit, three pressure level reheat steam cycle, gas scrubbing and low temperature cooling, sulphur recovery unit and COS hydrolysis, shift reactor, CO_2 compression to 110 bar.

F- Conventional (W/O CO_2 capture): Lignite fuelled oxygen blown shell gasification, cryogenic air separation unit, three pressure level reheat steam cycle, gas scrubbing and low temperature cooling, sulphur recovery unit and COS hydrolysis.

The results of a detailed mass and energy balance of these cases can be seen in Table 5:

Table 5: Plant operational input and output data for the six cases

Plant Operating Input and Outputs Data							
PARAMETERS		ISCC				CONVENTIONAL	
		A	B	C	D	E	F
Lignite (tonne/day)		12917	14990	15467	18751	10282	10282
Lime Stone (tonne/day)		7513	8638	8023	8023	0	0
Fuel Heat rate		8800					
Gross electric output		624	827	799	985	417	509
Net electric output		488	567	592	760	329	448
Plant Efficiency (%)		37.1	37.2	37.6	39.8	31.4	42.7
By Product (tonne/day)	Ash	504	585	603	731	264	393
	CaO	4036	4639	3793	4247	0	0
	$CaSO_4$	421	493	503	606	0	0
Total (tonne/day)		4961	5717	4898	5585	264	393
CO_2 emission (kg/s)		7.9	8.1	8.7	7.7	14.33	116.27
CO_2 capture (kg/s)		178.5	208.0	205.6	248.7	101.9	
CO_2 Capture (%)		95.8	96.3	96.0	97.0	87.7	
CO_2 emission (kg/MWh-		57.9	51.4	52.6	36.6	156.8	934.3
CO_2 capture (kg/MWh-		1317.0	1320.3	1250.3	1178.1	1114.7	0.0

Furthermore the plant investment and operational costs were estimated, based on known data from ISCC plants (see deliverable D6.1). With these costs and the results of the mass and energy balance the specific investment costs in €/kWel (Figure 20) and the CO_2 capture costs (Figure 21) were calculated.

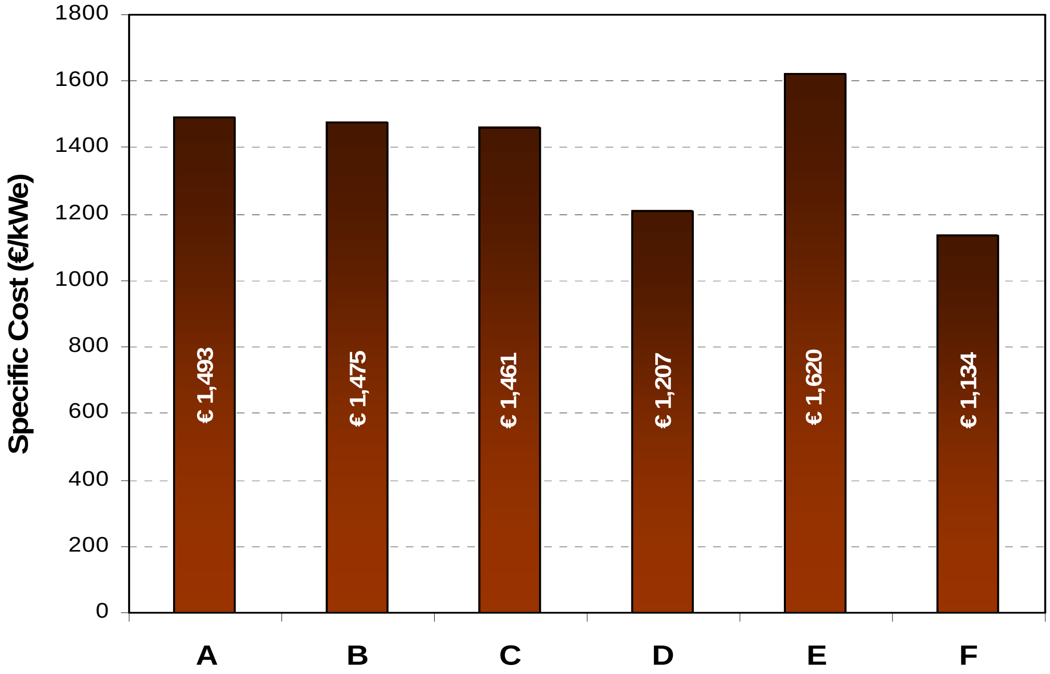


Figure 20 Specific investment costs (€/kWe) for six cases

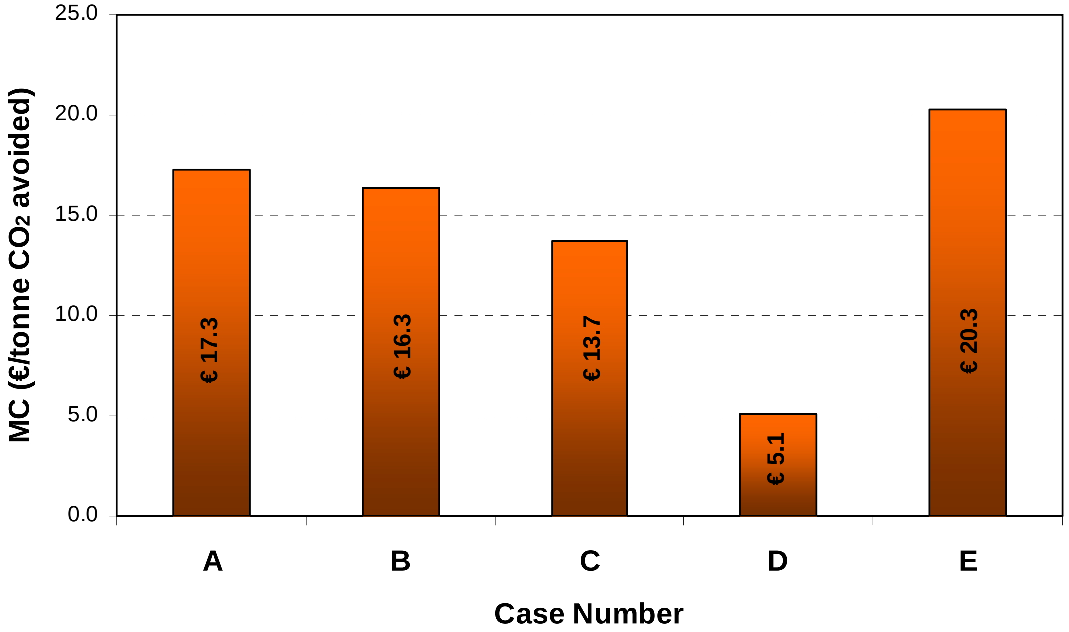


Figure 21: CO_2 mitigation cost for the six cases

The following conclusions can be drawn from the study:

- The new ISCC concepts, which combine gasification and regeneration processes, offer significant techno-economic advantages due to their CO_2 and H_2S co-capturing.
- The efficiencies of the ISCC CO_2 capture plants were 18.5 to 21% higher than the conventional IGCC CO_2 capture plant.

- The new ISCC processes are capable of capturing CO_2 more efficiently, with an efficiency penalty lower than those of the conventional IGCC plants.
- The CO_2 capture efficiencies of ISCC plants were estimated to be 95.8% to 97%, whereas for the conventional IGCC plants the CO_2 capture efficiency was 87.7%.
- The CO_2 emissions of ISCC plants ranged between 0.04 to 0.06 $kg/kW_{e,}$ while it was 0.16 kg/kW_e for the conventional plant.
- The specific investment costs of the ISCC plants ranged from 1207 to 1479 €/kW_e and for the conventional IGCC plants with without CO_2 capture plant the Sis were 1620 €/kW_e and 1134 €/kW_e, respectively
- The investment cost of the advanced ISCC plants is significantly lower due to the application of the advanced HAT turbine, transport membrane ASU unit and single pressure HRSG units.
- The levelised COE for the ISCC plants ranged between 46.84 and 57.43 €/MWh, resulting in 1% to 19% lower cost than the conventional IGCC CO_2 capture plant.
- The CO_2 mitigation costs for the ISCC plants were found to be within 5 to 17.3 €/tonne CO_2 avoided, whereas for the conventional IGCC plants the mitigation cost was 20.3 €/tonne CO_2 avoided.
- Sensitivity analysis shows that the highest impact on the cost of electricity was due to the variation of plant occupancy followed by the discounted cash flow. Variation of operating, maintenance, insurance and variable cost have the least impact on the cost of electricity

1.3.6.2 Assessment of the Ash quality

The ISCC process (Innovative In Situ CO_2 Capture Technology for Solid Fuel Gasification) is a new technology in support of European Union policy objectives.These include reduction of greenhouse gas emissions, a safe and cost effective energy supply, and decreased dependency on energy imports. Europe has large, economically available reserves of brown coal for which the ISCC process is designed.

The project aims to develop a new process for upgrading high moisture low rank brown coals yielding into three valuable products:

- Fuel gas consisting of mainly hydrogen
- Purge gas stream consisting of mainly CO_2, ready for transportation to sequestration (CO_2 capture > 90 %) or chemical fixation
- Pre-calcined feed for a cement kiln consisting of CaO, $CaSO_4$ and coal ash

This document shows the feasibility of utilization of the pre-calcined feed material, the so-called purge, in the production of cement.

After a basic description of cement production process, the specific problems of using purge as a raw material substitute like influence of sulfur from brown coal and re-carbonation of CaO by kiln exhaust gas are made evident.

Process simulations for both conventional and ISCC based cement production show the significant savings of fuel and raw material consumption and consequently of CO_2-emissions.

The utilization of purge from ISCC process as raw material substitute in the cement production allows remarkable savings of fuel and CO_2-emissions because of the already pre-calcined raw material and consequently because of the absence of the strong endothermic reaction of calcite de-carbonation. The amount of savings is determined by the required quantity of additives to be mixed with purge with a given chemical analysis to achieve the rather limited figures of clinker chemistry. The substantial difficulties in operation of a cement kiln using purge as main raw material component are re-carbonation of raw meal by CO_2 containing kiln gas and the impact of increased sulfur input, which causes circulating effects and higher SO_2-emissions. In order to fulfill the strict emission limits for cement plants, a waste gas desulphurization by e.g. wet scrubber is absolutely necessary. Transport and handling of purge, which as a calcined material should be kept dry is a further major issue of the proposed industrial application. In general, the evaluation in chapter 4 shows a high quantity of cement related to the amount of electric energy produced by a proposed ISCC power plant. In order to place this by-product on the European cement market, a close cooperation with the cement industry is recommendable.

1.3.6.3 Comparison with other pre-combustion CO_2 capture processes

The pre-combustion techniques of fossil fuel decarbonisation, assume transferring the carbon contained in raw fuel into sequestration ready carbon dioxide and the recovery of this gas during processing. The CO_2 free conversion products (hydrogen, synthesis gas, substitute natural gas, liquid fuels) are next consumed in the final combustion or in chemical processes. This ambitious target has been chosen as a priority of the energy research in the first energy call of 7Th RTD FP. Chemical looping processes, membrane assisted coal gasification, coal gasification in pure oxygen are examples of the technology options competitive to LEGS.

The non-questionable advantage of LEGS is the ability of direct coal conversion to one valuable product - hydrogen with a separate stream of sequestration ready carbon dioxide as the second major product from the plant. The high sulphur content, low quality coal up to the wasted coal residues could be gasified in the process. This makes the LEGS to be an attractive offer for the potential utilization of coal fines produced in large amounts by long wall mining techniques. Contrary to the classic IGCC techniques, low quality fuels with high sulphur content could be processed in the LEGS system without any extra modification of the future plant.

1.3.6.4 Development of dissemination strategy and business plan

The direct use of wet, highly contaminated by sulphur and mineral matters coal for energy or chemicals production has been announced by Polish coal and energy industry as the country priority technology goal. The vigorous discussions on implementing in Poland the first large scale coal gasification plant are carried out by Southern Energy Concern (PKE S.A.) and Polish Coal Company (K.W. S.A.) and KGHM S.A.. The mature coal gasification technology combined with liquid fuel synthesis based on Sasol technology and the gasification technologies exploited in the Schwarze Pumpe chemical complex are taken into account in the recently carried out pre feasibility studies of potential investors. The scale of the potential investment is 3 billion Euro and the estimated conversion capacity of the plant equals 6 million ton of hard coal a year.

The Polish coal mining and energy industry are also interested in renewing the research on underground coal gasification. This is due to the expected shortage of hard coal available for large scale long wall extraction technology. The majority of Polish coal reserves (also lignite) available for mining is located deeply underground. The industrial interest in underground gasification has resulted in the preparation the HUGE (Hydrogen Oriented Underground Coal Gasification for Europe) project proposal which was accepted in 2007 for execution by Coal and Steel Research Fund. The HUGE project is aimed at dynamic operation of in-situ sorption enhanced coal gasification to hydrogen and exploits in part the scientific results of ISCC research.

The integration of the coal energy industry with nuclear and hydrogen economy development has been announced recently as the priority by the Polish Government. It is aimed on combining the processes of coal gasification to hydrogen and synthesis gas using the heat generated in a nuclear plant. In this technology concept, the high temperature stream of heat produced will be consumed in the process of water decomposition to hydrogen and oxygen at temperatures of 800-1000 ^{o}C. The generated hydrogen will be consumed in coal hydrogenation to liquid fuels or substitute of natural gas (energy carriers substantially de-carbonated in comparison with raw coal). The oxygen stream from the nuclear plant will be consumed directly in the coal gasification with steam to hydrogen giving sequestration ready stream of CO_2. The results of ISCC project could find interesting innovative application in the frame of this technology concept. The research is assumed to be presented as a joint, Central European initiative to the Hydrogen and Fuel Cell JTI. It could be treated as an important initiative serving the European energy security needs.

2 Dissemination and use

The consortium publishes the work in several journals and conferences. Furthermore this report will be available for download on the project web site for interested parties.

Furthermore USTUTT was did host the In-Situ CO2 Removal Workshop 2005 (ISCR 2005). This is the annual scientific event where scientists from North America, Japan and Europe meet to discuss processes like the LEGS gasification. This workshop will be further used to disseminate the results and to install new global co-operations.

Based on the results of the ISCC project USTUTT will erect a larger pilot size test plant of a interconnected fluidized bed system to test and optimize the LEGS process under atmospheric pressure. This further research work is funded by the regional government of Baden Württemberg and the companies ENBW (utility) and Alstom (Fluidized bed boilers). This is a direct continuation of the ISCC work towards an industrial application of the LEGS process. However, it is only a first step and further work in pressurized systems must follow.

GIG intends to further develop a dissemination strategy which shall be integrated in the Polish coal to hydrogen roadmap. Poland seems to be the most promising location for a future ISCC power plant.