"BABES–BOLYAI" UNIVERSITY
CLUJ-NAPOCA

Faculty of Chemistry and Chemical Engineering

Victoria Goia (Maxim)

Energy conversion processes of coal and biomass through gasification with CO_2 capture

PhD THESIS ABSTRACT

PhD Supervisor:
Prof. Univ. Paul Şerban Agachi,
Reviewers:
Prof. Dr. Ing Teodor Todincă, Polytechnic University of Timişoara
Prof. Dr. Ing Grigore Bozga, Polytechnic University Bucharest
Conf. Dr. Ing. Călin Cristian Cormoş, Universitatea Babeş-Bolyai, Cluj-Napoca

Date of public support: December 16, 2011

CONTENTS

MOTIVATION AND OBJECTIVES OF THE THESIS

1. INTRODUCTION

1.1. History of gasification process and IGCC technology

1.2. The current state of knowledge

1.3. The importance of CO_2 captures

2. FEEDSTOCK

2.1. Solid fossil fuels

2.2. Renewable energy resources

2.3. Composition and properties of fuels

2.3.1. Fuel analysis

2.3.2. Calorific value

2.3.3. Ash properties

3. COAL GASIFICATION. GASIFICATION REACTORS

3.1. Gasification

3.1.1. Chemical reactions

3.1.2. Thermodynamics of Gasifications

3.1.3. Kinetics of Gasifications

3.2. Gasification reactors

3.2.1. Moving-bed reactors

3.2.1.1. Lurgi reactor

3.2.1.2. British Gas Lurgi reactor (BGL)

3.2.2. Fluidized-bed reactors

3.2.2.1. Winkler reactor

3.2.2.2. High Temperature Winkler - reactor (HTW)

3.2.3. Entrained-flow reactors

3.2.3.1. Siemens reactor

3.2.3.2. Shell and Prenflo reactors

3.2.3.3. ConocoPhillips E-Gas reactor

3.2.3.4. GE-Texaco reactor

3.3. Evaluation criteria of gasification reactors

4. IGCC TECHNOLOGY

4.1. IGCC technology presentation

4.2. TSyngas treatment and purification

4.2.1. Water–gas shift conversion

4.2.2. Acid gas removal

4.2.3. CO_2 conditioning, compression and storage

4.2.4. Hydrogen Purification

4.3. Electricity generation island

4.3.1. Combined cycle

4.3.2. Environmental impact

5. PYROLYSIS PRETREATMENT OF BIOMASS

5.1. Introduction

5.2. Pyrolysis processes

5.2.1 The composition and use of pyrolysis products

5.2.2. Kinetics of Pyrolysis

6. IGCC PLANT ASSESSMENT

6.1. Multi-criteria analysis of gasification reactors

6.2. General presentation of igcc plant

6.3. Case Study: IGCC plant performance analysis using different gasification technologies

6.4. Case Study: IGCC plant performance analysis with and without CO_2 capture

6.5. Case Study: IGCC plant performance analysis for electricity and hydrogen co-generation with CCS

6.6. Case Study: IGCC plant performance analysis for co-gasification of coal with biomass and waste

6.7. Conclusions

7. PYROLYSIS PRETREATMENT OF BIOMASS FOR AN IGCC PLANT

7.1. Equipment and materials

7.2. Influence of temperature and heating rate on pyrolysis process

7.2.1. Influence of temperature on pyrolysis process

7.2.2. Influence of heating rate on pyrolysis process

7.2.3. Influence of pyrolysis temperature on energy efficiency

7.3. Case study: the use of pyrolysis products in an IGCC plant

7.4. Conclusions

8. CONCLUSIONS

9. PERSONAL APPROACH

REFERENCES

LIST OF PUBLICATIONS

LIST OF ABBREVIATIONS

LIST OF FIGURES

LIST OF TABLES

APPENDIXES

APPENDICS I. CHARACTERISTICS OF FEEDSTOCKS

APPENDICS II. EXPERIMENTAL DATA FOR PYROLYSIS PRODUCTS AT 250-300 ° C

Keywords:

Gasification

Energy

Carbon Capture

Renewable energy resources

Motivation and objectives of the thesis

In order to limit the climate change, carbon dioxide emissions must be reduced by capturing and storing it. This is possible for electricity generation through gasification of solid fossil fuels, using an IGCC plant with CO_2 capture.

The continuous increase in the price of fossil fuels and also the increased interest in global environmental protection, make biofuel production to grow rapidly. Currently, an estimated global potential of biomass energy is large enough to meet global energy demand. Although the European Union wants a swift transition from coal to biomass, for short and medium term, coal will remain the main source of electricity generation.

Biomass gasification using existing reactors in IGCC plants it is difficult because of biomass properties. Therefore direct gasification of biomass is not the best option, taking into account existing commercial reactors. Worldwide, energy generation from biomass is growing and gasification reactors are developed for biomass conversion.

This thesis presents an IGCC plant for electricity and hydrogen co-generation with carbon capture, which can process both coal (with or without the addition of biomass or waste) and biomass pyrolysis products. This concept is very promising, since the plant can run on *coal* with or without addition of renewable energy resources in this transition period from coal to biomass and on *biomass pyrolysis products*, with no further investment in research and development of novel gasification reactors. In this context this thesis is aligned with the highest level of energy research and utilization of renewable energy resources.

The main objective of this thesis is to investigate innovative ways of converting coal, waste and biomass into energy vectors (electricity and hydrogen), through gasification with carbon capture.

The thesis aims at achieving the following objectives:

- Establishing of technical characteristics of IGCC plant for electricity and hydrogen co-generation with carbon dioxide capture;

- Gasification technology assessment and draw up a multi-criteria analysis, in order to narrow the range of gasification reactors that will be simulated in an IGCC plant with CO_2 capture. The choice of the four most promising options for electricity and hydrogen co-generation with carbon dioxide capture;
- Mathematical modeling and simulation of IGCC scheme using the four chosen gasification technologies, using coal as feedstock. Results evaluation results and choosing the right options for the studied installation;
- Mathematical modeling and simulation of IGCC scheme without carbon capture and comparison with the case when the carbon dioxide is captured
- Evaluation of IGCC plant flexibility to co-generate electricity and hydrogen while capturing carbon dioxide, depending on the electricity demand;
- Investigation of co-gasification processes of coal with biomass or waste. Mathematical modeling and simulation of co-gasification systems, performance evaluation and comparison with the case when is used only coal as feedstock;
- Proposal of an innovative and efficient method for biomass conversion into electricity using biomass pyrolysis products in an IGCC plant.

1. INTRODUCTION

Gasification is a process by which solid fossil fuels are converted into a fuel gas, synthesis gas (mainly a mixture of carbon monoxide and hydrogen), and is one of the oldest industrial processes for energy conversion. Generally gasification process involves the reaction of solid fuel with an oxidizing agent (air or oxygen) in the presence of moderator (steam) at an elevated temperature from 1200 to 1500 ° C resulting syngas which is used for power generation or as raw material for other substances synthesis such as methanol, urea, ammonia, etc. [1].

Fundamental principles of electricity generation were discovered in the years 1820 - 1830 by british scientist Michael Faraday. His method consists in generating energy by moving a wire loop or copper disc between the poles of a magnet, this method being still used today [2].

Centralized energy production became possible when it was found that AC power lines can transport electricity at very low cost on large distances. Since 1881 began generating centralized electricity. The first power plants were based on water or coal. For power generation are used as fuels: coal (44.9%), gas (23.4%), nuclear fuel (20.3%), water (6.9%), oil (1%) and other energy sources (wind, solar, geothermal) [3,4].

In order to limit the climate change, carbon dioxide emissions must be reduced by capturing and storing it. This is possible for electricity generation through gasification of solid fossil fuels, using an IGCC plant with CO_2 capture.

IGCC technology is very important in coal power generation and environmental protection because it has many advantages compared to classical technology used in steam power plants based on coal or lignite to generate steam which is then expanded in a steam turbine to produce electricity. The first advantage concerns the significantly lower environmental impact of IGCC technology. Another advantage is related to the flexibility of IGCC technology to produce various energy vectors according to the demand at a time, leading to higher energy and economic efficiency. Another important factor is that IGCC technology allows the capture of carbon dioxide (pre-combustion capture) at lower costs and higher efficiency than for capture from flue gas (post-combustion capture).

IGCC technology is becoming more widespread, and in recent years more and more gas turbines manufactured by the largest manufacturers in the field (Alstom, Siemens, General Electric, Mitsubishi) have been adapted to be used with syngas.

2. FEEDSTOCK

Coal is the oldest known fossil fuel use. Coal can be defined as a sedimentary brown-black rock with combustible properties, formed by the slow degradation of vegetation. Over millions of years vegetation remnants have suffered a slow process of carbonization, resulting in different sorts of coal, peat is the youngest and oldest anthracite coal [1.4 to 6].

Biomass is the first form of energy used by humans, with the fire discovery. Biomass is the most abundant renewable resource on the planet, including all organic matter produced by the metabolic processes of living organisms. Biomass is not a commonly used industrial fuel; a rate of 15-20% of the total fuel is represented by biomass and is being used mainly for heating and domestic use. Biomass as a fuel has a major advantage over other renewable energy resources: can be used as liquid, gaseous and solid for power generation [1].

Waste as raw material for gasification cover a wide range of materials, both solids and liquids. The European Union has grown increasingly in recent years and with it the amount of waste produced. According to European Environment Agency, the European Union annually produces 1.3 billion tonnes of waste, of which about 40 million tonnes are hazardous waste and for every man about 3.5 tonnes of waste annually. At these quantities are added 700 million tonnes of agricultural waste. Treatment and disposal of all such waste without harming the environment becomes a major problem [7].

European Commission encourages the use of renewable resources for electricity generation both to reduce dependence on oil and coal and to reduce emissions of greenhouse gases. Biomass is a renewable resource with almost zero CO_2 emissions because it absorbs CO_2 from the atmosphere is formed, so when burned does not contribute to global CO_2 emissions. However when biomass is used as fuel, some CO_2 emissions are correlated with cultivation and its processing [7].

3. COAL GASIFICATION. GASIFICATION REACTORS

3.1. Gasification

During the gasification process a series of chemical reactions take place [1, 8,10]:

- Combustion reactions

$$C + \frac{1}{2}O_2 \rightarrow CO \qquad \Delta H_1 = -111\ MJ/Kmol \tag{3.1}$$

$$CO + \frac{1}{2}O_2 \rightarrow CO_2 \qquad \Delta H_2 = -283\ MJ/Kmol \tag{3.2}$$

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \qquad \Delta H_3 = -242\ MJ/Kmol \tag{3.3}$$

- Boudouard reaction

$$C + CO_2 \leftrightarrow 2CO \qquad \Delta H_4 = +172\ MJ/Kmol \tag{3.4}$$

- Water-gas reaction

$$C + H_2O \leftrightarrow CO + H_2 \qquad \Delta H_5 = +131\ MJ/Kmol \tag{3.5}$$

- Methanation reaction

$$C + 2H_2 \leftrightarrow CH_4 \qquad \Delta H_6 = -75\ MJ/Kmol \tag{3.6}$$

$$CO + 3H_2 \leftrightarrow CH_4 + H_2O \qquad \Delta H_7 = -206.3\ MJ/Kmol \tag{3.7}$$

- CO shift reaction

$$CO + H_2O \leftrightarrow CO_2 + H_2 \qquad \Delta\, H_8 = -41\ MJ/Kmol \tag{3.8}$$

- Pyrolysis reactions

$$4C_nH_m \rightarrow mCH_4 + (4n - m)C \qquad \Delta\, H < 0 \tag{3.9}$$

Fossil fuels used in gasification contain in addition to carbon, oxygen and hydrogen and other elements such as sulfur, nitrogen or halogens (mainly chlorine). These components also changes during the reactions, so that nitrogen turns into NH_3 and HCN and sulfur into H_2S and COS (carbonyl sulphide). If not removed, sulfur compounds will be emitted into the atmosphere as sulfur oxides (SOx). To avoid air pollution with SOx, IGCC technology provides a purification step of the syngas when

COS is converted into H_2S according to one of the following chemical reactions [1.10, 11]:

$$COS + H_2 \leftrightarrow H_2S + CO \qquad \Delta H_9 = +7\ MJ/Kmol \qquad (3.10)$$

$$COS + H_2O \leftrightarrow CO_2 + H_2S \qquad \Delta H_{10} = -34\ MJ/Kmol \qquad (3.11)$$

3.2. Gasification reactors

Worldwide there are more than 140 gasification plants, of which 90 are located in the U.S. and it is estimated that by 2020 their number will increase by 70%. These plants are based on a wide range of reactors that can be classified into three categories [1, 5, 9, 11]:

- Moving-bed gasifiers were the first modern type of solid fuels gasification reactors. Moving-bed reactor, illustrated in Figure 1, has a feeding system at the top and at the bottom in countercurrent with fuel is the feeding system for gas phase (oxidation agent and moderator) [1,5].

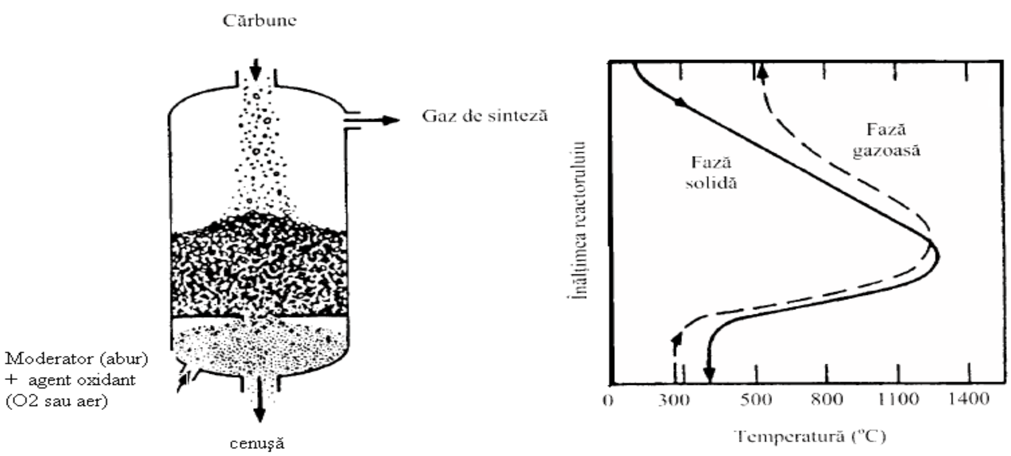

Figure 1. Moving-bed gasifier

- Fluidized-bed gasifiers - this type of reactor provides a very good mixing between fuel and oxidizing agent. Oxidizing agent, oxygen or air is blown through a bed of solid fuel with a certain speed so that the fluidization of solid matter occurs. This type of reactor is suitable for reactive materials such as coal or biomass. Figure 2 illustrates a fluidized bed reactor and its temperature profile.

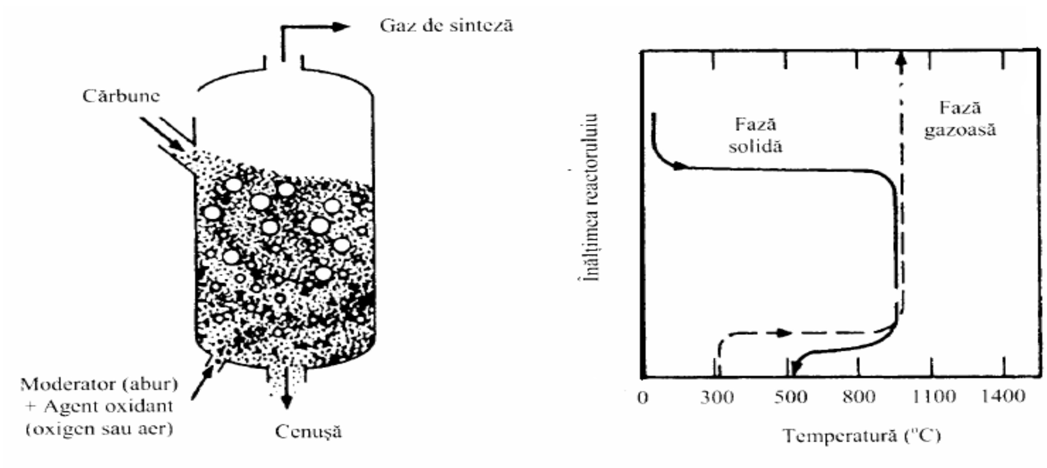

Figure 2. Fluidized bed gasifier

- Entrained-flow gasifiers - the solid phase and gas phase are moving in the same direction. Entrained-flow gasifiers can be used for less reactive raw materials like coal. This type of reactor is shown in Figure 3. together with associated temperature profile.

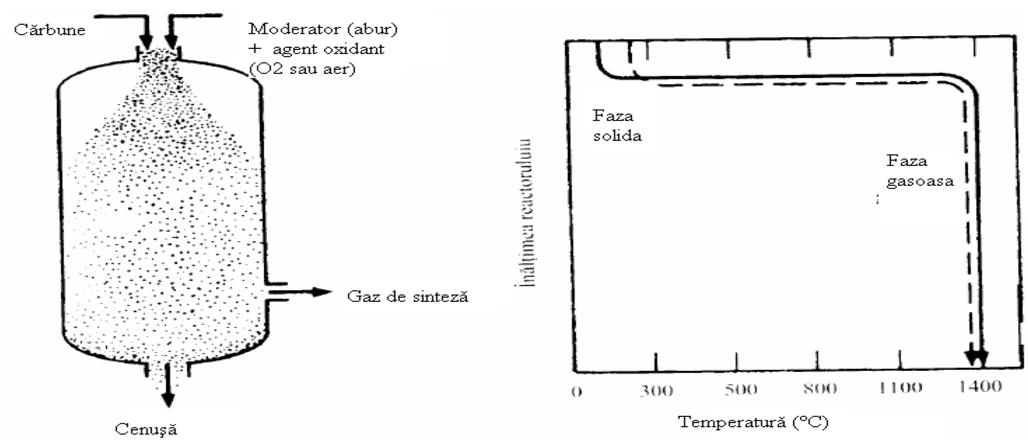

Figure 3. Entrained-flow gasifier

4. *IGCC TECHNOLOGY*

IGCC technology is very important in coal power generation and environmental protection because its advantages against to classical technology used in power plants based on coal or lignite to generate steam which is then expanded in a steam turbine to produce electricity.

- The first advantage concerns the significantly lower impact on environment of IGCC technology than coal-based technologies.
- Another advantage of IGCC plants is plant flexibility to produce electricity or hydrogen depending on the demand. In periods when electricity demand is low the plant can produce more hydrogen which can be stored and used for other applications. Therefore on account of the installation flexibility, full load operation leads to lower operating and maintenance costs.
- Another important factor is that IGCC technology allows the capture of carbon dioxide (pre-combustion capture) at lower costs and higher efficiency than for capture from flue gas (post-combustion capture).

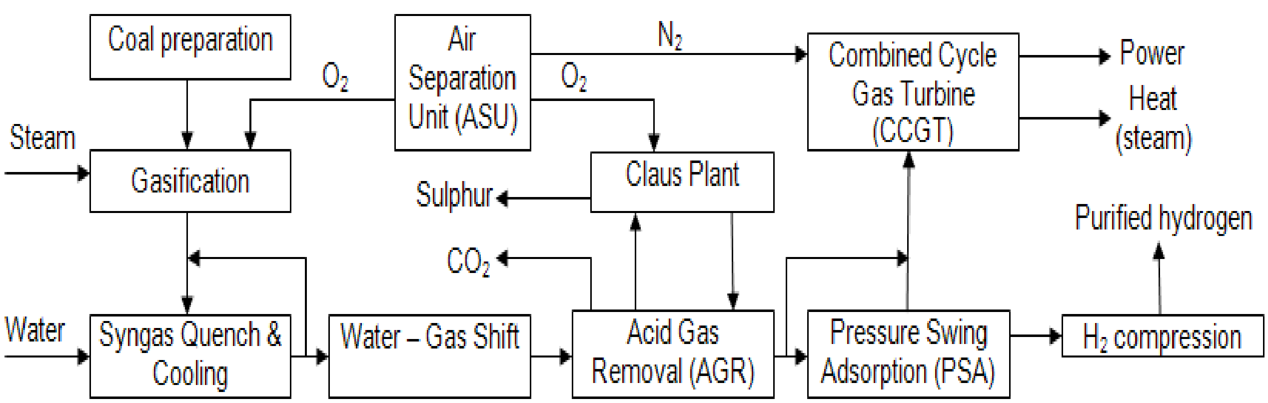

Figure 4. Block diagram of electricity and H_2 co-generation ICGG plant with CO2 capture

Figure 4. illustrates a block diagram of IGCC plant with CO_2 capture and storage. Unlike a conventional plant, an IGCC plant with CO_2 capture has in addition a catalytic conversion of carbon monoxide CO with water vapor into hydrogen and carbon dioxide. This step is designed to increase the H_2 concentration in syngas and to transform chemical species containing carbon into carbon dioxide which can be captured [1, 8, 14].

Another difference of this scheme is that the acid gas separation unit separates both H_2S and CO_2. Now the syngas contains mostly hydrogen which is divided: one part going to Pressure Swing Adsorbtion - PSA to obtain high purity hydrogen (> 99.9% vol) able to be used not only in chemical and petrochemical processes or as fuel for fuel cells but also for the transport sector and the other part, together with gas coming from acid gas separation unit is used in combined cycle for power generation.

5. PYROLYSIS PRETREATMENT OF BIOMASS

Biomass is a renewable energy resource, which includes organic matter formed by photosynthesis. The most important renewable fuel is wood, but trees are too valuable to be burned, but residues from wood processing industry (e.g. sawdust), could be a very valuable feedstock. Other sorts of biomass that can be used as fuels are agricultural residues such as wheat straw, corn stalks, rice husks, coconut etc. Fossil fuels (e.g. oil, coal, lignite) are also derived from plant species with the difference that was formed during millions of years. Worldwide biomass has always been a major source of energy since the beginning of civilization. In under development countries and rural areas woody biomass and agriculture residues still represent a significant proportion of feedstock for thermal energy supply [16-18].

Gasification of biomass in existent gasification reactors is difficult, because of the properties of biomass. It is known that to have high efficiency gasification process is necessary that the ratio O / C of the fuel to be as small as possible, like in the case of coal, but biomass is a fuel that has high O/C ratio. Another problem is the feeding of existing gasification reactors with biomass, which should be shredded at 100 mm, which means an energy penalty of about 20%. Thus the direct gasification of biomass is not the best option, taking into account existing commercial reactors at this time. But an attractive alternative is the pretreatment of biomass through pyrolysis at low temperature before being gasified.

Pyrolysis is the thermo chemical decomposition of solid fuels (biomass, waste, fossil fuels) in the absence of oxygen with production of chemicals, heat or energy. Pyrolysis is the first step in all other thermo conversion technologies such as combustion and gasification. The process takes place at relatively low temperatures (300-800 ° C) compared to 900-1500 ° C for gasification [11, 19-21].

6. IGCC PLANT ASSESSMENT

Multi-criteria analysis of gasification reactors

The aim of this multi-criteria analysis is to narrow the range of gasification reactors that will be simulated in an IGCC plant with CO_2 capture. Using the data obtained from simulations a selection regarding the gasification reactor can be made, which will be used in a co-generation of electricity and hydrogen IGCC plant with CO_2 capture and which can process a wide range of feedstocks (e.g. coal, coal in addition to various renewable energy resources, biomass pyrolysis products).

Due to the large variety of gasification rectors, a multi-criteria analysis is needed to evaluate these reactors. Table 6.1 shows the multi-criteria analysis for 7 gasification reactors.

Table 6.1. Multi-criteria analysis of gasification reactors

Parameters	Case 1 Lurgi	Case 2 BGL	Case 3 HTW	Case 4 Siemens	Case 5 Shell	6 E-Gas Case	GE-Texaco Case 7
Feedstock coal	Yes	Yes	Lignite	Yes	Yes	Yes	Yes
Maximum pressure (bar)	100	60	30	40	40	40	100
Temperature (° C)	450-650	450-600	900-1050	1400-1600	1400-1600	950-1400	1200-1450
Carbon conversion (%)	> 92	> 95	90-95	> 99	> 99	> 98	> 98
Steam / oxygen requirement	High	Low	Medium	High	High	High	High
Syngas clean up issues	Yes	Medium	Yes	Not	Not	Medium	Not
H_2 production potential HPP	Low	High	Medium	High	High	Medium	High
Cold Gas Efficiency CGE (%)	85-87	82-87	80-85	75-79	75-79	78-80	65-75

CO_2 capture capacity	Low	Low	Medium	High	High	Medium	High

Excellent	Satisfactory	Unsatisfactory

Table 6.1. presents an analysis of the gasification reactors in order to choose the most feasible gasifier for an IGCC plant with carbon capture. Such a reactor used in IGCC plant with CO_2 capture and energy vectors co-generation (electricity and hydrogen), using coal as feedstock should meet the following conditions: high pressure (60 - 100bar), high temperature (1400 -1600 ° C), carbon conversion> 99%, low requirement for steam / oxygen, easy syngas clean up, high hydrogen production potential (HPP), high Cold Gas Efficiency (CGE) and high CO_2 capture capacity.

Assessing the technical level of equipment shall be based on comparison of their technical characteristics. Some features it is best to be higher, while others should be smaller, each feature with a certain degree of influence on the overall technical level indicator. To determine the technical level a relationship has been developed which combines the principle of Von Neumann-Morgenstern utility and Cobb-Douglas production function [23, 24].

Technical level calculated using this relationship has to be as high as possible. Technical levels calculated for the 7 reactors, taking into account the evaluation criteria presented in Table 6.1, are:

Reactor						
Lurgi	BGL	HTW	Siemens	Shell	E-GAS	GE
506.28	705.71	634.07	817.55	817.55	690.29	812.77

Based calculated technical levels, but also because of their advantages, it appears that the entrained-flow reactors are the most appropriate choice for IGCC plant CO_2 capture and storage (cases 4-7).

The purpose of this analysis was to narrow the range of reactors to be simulated in an IGCC plant with CO_2 capture. Based on multi-criteria analysis performed for energy vectors poly-generation IGCC plant with CO_2 capture and storage were chosen as most suitable reactors the entrained flow gasifiers.

Case Study: IGCC plant performance using different gasification technologies

Among the commercially available entrained-flow technologies four have been chosen:

- ❖ Case 1: Siemens reactor
- ❖ Case 2: Shell reactor
- ❖ Case 3: E-Gas ConocoPhillips reactor
- ❖ Case 4: GE-Texaco reactor

Table 6. 2. IGCC plant performance indicators for four types of reactors

	UM	**Case 1**	**Case 2**	**Case 3**	**Case 4**
Solid fuel flowrate (a.r.)	t / h	168.1	169.1	167.0	180.5
Feedstock tthermal energy	MWt	1183.7	1190.74	1175.95	1271.02
Syngas flowrate	kmol / h	29116.3	15483.47	18898.0	34943.23
CO	% Vol	29.56	55.77	34.24	23.78
H_2	% Vol	13.83	25.78	30.75	12.68
CH_4	% Vol	0	0	0.6	0
H_2S	% Vol	0.09	0.18	0.15	0.08
Thermal energy of raw syngas	MWt	949.92	950.04	927.04	953.85
Cold Gas Efficiency (CGE)	%	80.25	79.79	78.83	75.05
Thermal energy of CO an H_2	MWt	946.16	946.03	897.69	949.83
H_2 production potential (HPP)	%	79.93	79.45	76.34	74.73
Thermal Energy of clean syngas	MWt	845.82	846.79	844.65	843.27

(from AGR)					
Syngas treatment efficiency	%	89.04	89.13	91.11	89.44
Gas turbine output	MWe	334.00	334.00	334.00	334.00
Steam turbine output	MWe	200.9	209.30	203.33	194.13
Gross electric power output	MWe	534.9	543.30	537.33	528.13
ASU power consumption + O_2 compr.	MWe	45.78	46.56	44.17	56.03
Gasification island power consumption	MWe	7.68	8.6	7.01	6.23
AGR & CO_2 drying & compression	MWe	39.18	39.18	38.75	39.00
Power island power consumption	MWe	19.06	19.00	19.03	18.71
Total ancillary power consumption	MWe	111.7	113.34	108.96	121.48
Net electric power output	MWe	423.23	429.36	428.37	413.21
Gross electrical efficiency	%	45.19	45.62	45.69	37.78
Net electrical efficiency	%	35.75	36.1	36.42	29.20
CO_2 specific emissions	kg CO2/MWh	82.25	100.6	344.05	88.39

Technical levels were calculated for each reactor:

Reactor			
Siemens	Shell	E-GAS	GE
313.40	297.86	216.68	282.96

Based on calculated technical levels, but also on performance indicators from Table 6.2 it appears that Siemens reactors (case 1) and Shell (case 2) are the best options.

When a Shell reactor is used in an IGCC plant, the net plant efficiency increases by 0.98% comparing with the case when a Siemens reactor is used. But the main advantage of Siemens gasification reactor is the cooling system of the syngas "water-quench" which provides optimal conditions for the conversion of carbon monoxide with water vapor, a precondition to capture carbon dioxide. Another advantage is due to lower CO_2 emissions by 22% than in the Shell reactor, which means 137 tCO2/an.

Based on these considerations was chosen as best option for the considered plant a Siemens reactor. So case studies to be further conducted in this paper will be based on Siemens gasification technology.

Case Study: IGCC plant performance assessment with and without CO_2 capture

To highlight the benefits of capturing carbon dioxide emitted in an IGCC plant a case study was set involving the following cases:

- ❖ Case 1 - IGCC plant without CO2 capture, entrained-flow gasification reactor Siemens
- ❖ Case 2 - IGCC plant with CO2 capture, entrained-flow gasification reactor Siemens
- ❖ Case 3 - IGCC plant without CO2 capture, entrained-flow gasification reactor Shell
- ❖ Case 4 - IGCC plant with CO2 capture, entrained-flow gasification reactor Shell

Table 6. 3. IGCC plant performance indicators with and without CO2 capture

	UM	**Case 1**	**Case 2**	**Case 3**	**Case 4**
Solid fuel flowrate (a.r.)	t / h	151.0	168.1	152.0	169.1
Feedstock thermal energy	MWt	1063.2	1183.7	1070.3	1190.7
Syngas flowrate	kmol / h	26088.3	29116.3	14082.3	15483.47
CO	% Vol	29.47	29.56	54.83	55.77
H_2	% Vol	14.04	13.83	25.83	25.78
CH_4	% Vol	0	0	0	0
H_2S	% Vol	0.09	0.09	0.18	0.18

Thermal energy of raw syngas	MWt	853.22	949.92	854.13	950.04
Cold Gas Efficiency (CGE)	%	80.24	80.25	79.80	79.79
Thermal Energy of clean syngas (from AGR)	MWt	849.41	845.82	850.51	843.27
Syngas treatment efficiency	%	99.55	89.04	99.57	89.44
Gas turbine output	MWe	334	334	334	334.00
Steam turbine output	MWe	186.65	200.9	200.89	209.30
Gross electric power output	MWe	520.65	534.9	534.89	543.30
ASU power consumption + O_2 compr.	MWe	41.12	45.78	40.19	46.56
Gasification island power consumption	MWe	6.81	7.68	7.87	8.6
AGR & CO_2 drying & compression	MWe	6.01	39.18	6.04	39.18
Power island power consumption	MWe	19.17	19.06	19.24	19.00
Total ancillary power consumption	MWe	73.11	111.7	73.34	113.34
Net electric power output	MWe	447.54	423.23	461.55	429.96
Gross electrical efficiency	%	48.96	45.19	49.97	45.62
Net electrical efficiency	%	42.09	35.75	43.12	36.1
CO_2 specific emissions	kg CO2/MWh	853.44	82.25	843.78	100.6

Calculated technical levels for each case are:

Reactor			
Siemens without capture	Siemens with capture	Shell without capture	Shell with capture
176.59	313.40	177.68	297.86

It may be noted that in cases without CO_2 capture the net efficiency is higher by 12.52% for Siemens reactor and by 13.38% for the Shell reactor. Decrease in net efficiency of the plant in cases where CO_2 was captured is due to significant increase in energy consumption of AGR and CO_2 compression. CO_2 emissions were drastically reduced in cases where the carbon dioxide was captured.

IGCC technology has other advantages in terms of environmental impact: lower emissions of SOx and NOx, but also the possibility of using inferior coal as feedstock, but also biomass or waste. [25-27].

Case Study: IGCC plant performance assessment for electricity and hydrogen co-generation with CO_2 capture

This section evaluates the performance of IGCC plant that produces electricity and hydrogen with CO_2 capture, based on Siemens gasification reactor. The data obtained from simulations are presented in Table 6.4.

Table 6. 4. IGCC plant performance indicators for electricity and hydrogen co-generation

	UM	**Electricity**	**Electricity + H_2**			
Solid fuel flowrate (a.r.)	t / h	168.1				
Feedstock thermal energy	MWt	1183.7				
Syngas flowrate	kmol / h	29116.3				
Thermal energy of raw syngas	MWt	949.92				
Cold Gas Efficiency (CGE)	%	80.25				
Thermal energy of CO an H_2	MWt	946.16				
H_2 production potential (HPP)	%	79.93				
Thermal Energy of clean syngas (from AGR)	MWt	845.82				
Syngas treatment efficiency	%	89.04				
Gas turbine output	MWe	334.0	313.47	292.89	272.23	251.65
Steam turbine output	MWe	200.9	190.14	178.64	166.74	155.86
Gross electric power output	MWe	534.9	503.60	471.52	438.97	407.51
Hydrogen output – LHV	MWt	0	50	100	150	200

Total ancillary power consumption	MWe	111.7	111.59	111.57	111.55	111.51
Net electric power output	MWe	423.23	392.01	359.95	327.42	296.00
Gross electrical efficiency	%	45.19	42.54	39.83	37.08	34.42
Net electrical efficiency	%	35.75	33.11	30.40	27.66	25.00
Hydrogen efficiency	%	0	4.22	8.44	12.67	16.89
Cumulative efficiency	%	35.75	37.27	38.83	40.32	41.84
CO_2 specific emissions	kg CO2/MWh	82.25	78.20	75.15	72.00	69.12

From this study it can be seen that the combined efficiency of the process increases with increasing the amount of hydrogen generated. Carbon dioxide emissions decreases with increasing the amount of hydrogen generated. Because of the flexibility facility to produce electricity and hydrogen according to the request at a time, the cumulative efficiency is greater for co-generation and the quantity of carbon dioxide is reduced, the electricity and hydrogen co-generation IGCC plant is a very attractive option.

Case Study: IGCC plant performance assessment for co-gasification of coal with biomass and waste

This study evaluates the use of coal with or without addition of biomass / waste in gasification process in order to produce syngas for electricity generation.

- Case 1: coal
- Case 2: coal in addition with Sawdust SWD
- Case 3: coal in addition with sewage sludge SWG
- Case 4: coal in addition with meat and bone meal MBM

Table 6. 5. IGCC plant performance indicators for co-gasification of coal with biomass / waste

	UM	Case 1	Case 2	Case 3	Case 4
Solid fuel flowrate (a.r.)	t / h	168.1	181.5	192.5	168.6
Mixing ratio (coal / biomass)	% Wt.	100/0.0	80/20	80/20	80/20
Feedstock thermal energy	MWt	1169.7	1184.85	1219.24	1130.18
Syngas flowrate	kmol / h	29116.3	31074.39	32294.97	26458.26
Thermal energy of raw syngas	MWt	949.92	946.82	950.86	942.48
Cold Gas Efficiency (CGE)	%	80.25	79.94	77.99	83.39
Thermal energy of CO an H_2	MWt	946.16	943.95	945.75	938.08
H_2 production potential (HPP)	%	79.93	79.65	77.57	83.00
Thermal Energy of clean syngas (from AGR)	MWt	845.82	845.03	847.08	845.62
Syngas treatment efficiency	%	89.04	89.11	88.85	89.72
Gas turbine output	MWe	334.00	334.00	334.00	334.00
Steam turbine output	MWe	200.9	200.9	205.7	196.28
Gross electric power output	MWe	534.9	534.9	539.7	530.28
ASU power consumption + O_2 compr.	MWe	45.78	45.4	50.5	41.16
Gasification island power consumption	MWe	7.68	7.75	7.87	7.74
AGR & CO_2 drying & compression	MWe	39.18	39.87	40.06	41.73
Power island power consumption	MWe	19.06	19.06	19.06	19.06
Total ancillary power consumption	MWe	111.7	112.08	117.04	109.69
Net electric power output	MWe	423.23	422.82	422.66	420.59
Gross electrical efficiency	%	45.19	45.16	44.26	46.91
Net electrical efficiency	%	35.75	35.70	34.66	37.21
CO_2 specific emissions	kg CO2/MWh	82.25	62.4	58.91	60.16

As can be noticed in Table 6.5. all four cases evaluated have a net electric power output of about 420 MW and plant net efficiency between 34% and 37%. For the same power output of about 420 MW is required a different amount of feedstock, according to its calorific value.

Calculated technical levels for each case are:

Feedstock			
Coal	Coal with SWD	Coal with SWG	Coal with MBM
313.40	335.07	333.13	348.87

Based on key performance indicators and calculated technical levels it appears that the best mixtures are coal in addition with sawdust and coal in addition with meat and bone meal.

Conclusively IGCC plant developed in this thesis is flexible in terms of feedstock supply can be fed with both coal and coal in addition with renewable energy resources (biomass) or solid wastes. In this context, this flexible plant is a solution that can ensure the transition from an economy based almost entirely on coal to an economy based on renewable energy resources.

7. PYROLYSIS PRETREATMENT OF BIOMASS FOR AN IGCC PLANT

Coal gasification technology is a mature technology without issues, but biomass gasification due to its proprieties significant large problems. IGCC demonstrative plants which used biomass feedstock have been set up, such as the one in Värnamo, Sweden (1991 - 1993). These plants ran for a short period of time, aiming to demonstrate the technical possibility of biomass gasification, but biomass fueled IGCC plants are not yet efficient and optimized, being under continuous research and development.

This study aims to demonstrate the advantages pyrolysis pretreatment of biomass before gasification, and investigates the possibility to generate electricity in an IGCC plant using as feedstock biomass pyrolysis products.

Pyrolysis products were analyzed by different methods:

- Solid analysis by proximate/ultimate analysis
- Permanent gases removed by argon purge gas, collected in gasbag and analyzed by (GC),
- Liquid products analysis by HPLC.

178 experiments were performed at temperatures between 250 ° C and 700 ° C and heating rates between 5-80 ° C / min. As feedstocks were used 14 types of biomass with different humidity, characteristics of the feedstocks used are presented in Appendix I.

Product composition and yield are affected by pyrolysis temperature. Figure 7.2. shows the effect of temperature on pyrolysis products distribution for the four types of biomass selected for example, at temperatures between 250 ° C and 700 ° C in a fixed bed reactor.

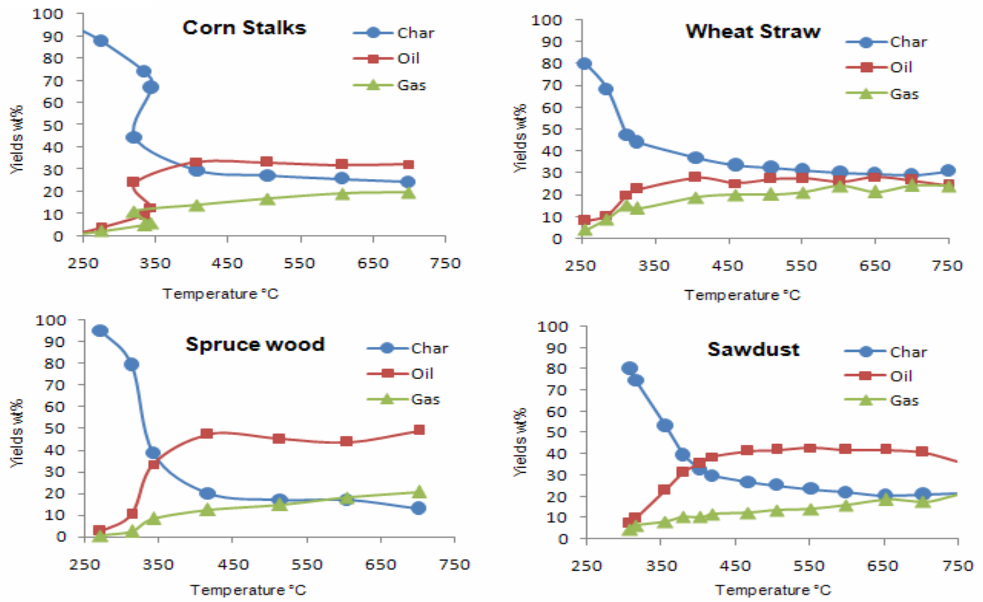

Figure 7.1. Influence of pyrolysis temperature on the yield of pyrolysis products

If the desired product is char is recommended that pyrolysis temperature to be low (250-300 ° C), but if tar or gas is the desired product, it is recommended that the temperature to be higher (700-750 ° C).

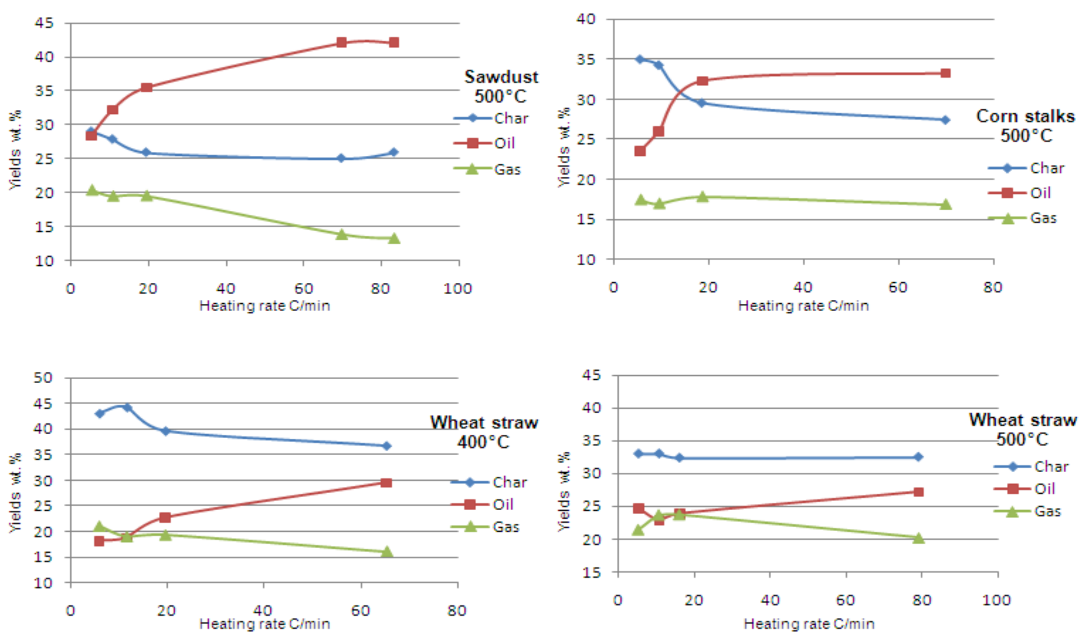

Figure 7. 1. The influence of heating rate on the yield of pyrolysis products

Figure 7.3. illustrates the effect of heating rate on the distribution of pyrolysis products for the four biomass types selected, at heating rates between 5-83 ° C / min in a fixed bed reactor.

If the desired product is char is recommended that the heating rate and final temperature to be low (10-20 ° C / min, respectively 250-300 ° C). To maximize the production of tar is recommended that both temperature and heating rate to have higher values (70-80 ° C / min, respectively 650-750 ° C).

Case study: the use of pyrolysis products in an IGCC plant

Pretreatment of biomass through pyrolysis at low temperatures (~ 300 ° C) increase its calorific value and the energy is concentrated in solid and liquid product. This case study examines the possibility of using biomass pyrolysis products in an IGCC plant. IGCC plant described in Chapter 6 can be adapted to run on biomass pyrolysis products by introducing a fluidized bed reactor prior gasification reactor.

Table 7.1. IGCC plant performance indicators for the four types of biomass

	UM	Corn stalks (CST)	Spruce wood (SPW)	Sawdust (SWD)	Wheat straw (WST)
Biomass flowrate	t / h	249.32	259.22	237.24	290.10
Pyrolysis products flowrate	t / h	240.00	255.13	217.32	255.00
Thermal energy of pyrolysis products	MWt	1157.36	1180.97	1147.38	1205.43
Syngas flowrate	moles / h	32649.38	31215.28	26573.82	15688.83
CO	% Vol	23.80	24.20	29.54	51.04
H_2	% Vol	14.58	16.08	17.93	27.46

CH_4	% Vol	0.00	0.03	0.09	0.00
H_2S	% Vol	0.06	0.06	0.00	0.25
Thermal energy of raw syngas	MWt	932.34	934.64	941.33	923.54
Cold Gas Efficiency (CGE)	%	80.70	79.22	82.53	78.25
Thermal energy of CO an H_2	MWt	929.53	929.86	936.06	917.89
H_2 production potential (HPP)	%	80.45	78.82	82.07	77.77
Thermal Energy of clean syngas (from AGR)	MWt	840.17	847.55	854.96	841.69
Syngas treatment efficiency	%	90.11	90.68	90.82	91.14
Gas turbine output	MWe	334.00	334.00	334.00	334.00
Steam turbine output	MWe	194.95	198.77	194.65	199.82
Gross electric power output	MWe	528.95	532.77	528.65	533.82
ASU power consumption + O_2 compr.	MWe	37.09	41.53	40.42	40.05
Gasification island power consumption	MWe	7.76	7.72	8.08	9.27
AGR & CO_2 drying & compression	MWe	39.72	41.53	39.77	41.07
Power island power consumption	MWe	19.05	19.06	19.07	19.07
Total ancillary power consumption	MWe	103.62	109.84	107.34	109.46
Net electric power output	MWe	425.33	422.93	421.31	424.36
Gross electrical efficiency	%	45.70	45.11	46.07	44.28
Net electrical efficiency	%	36.75	35.81	36.73	35.20

Calculated technical levels for each case are:

Feedstock			
CST	PWS	SWD	WST
953.21	937.87	963.23	927.87

Based on calculated technical levels but also as can be seen in Table 7.2., for an installed capacity of about 420 MW it can be said that from biomass sorts used in simulations, the most efficient, in terms of energy efficiency and Cold Gas Efficiency (CGE), are cases where sawdust (SWD) or corn stalks (CST) are used as feedstock.

As shown, direct use of biomass as fuel in an entrained flow gasifier has certain technological constrains, but pretreatment of biomass through pyrolysis before being gasified is a very attractive option. Biomass pretreatment through pyrolysis improves its properties enabling the feeding in an entrained-flow reactor.

The IGCC plant proposed in this section provides the possibility to generate a large amount of electricity in IGCC plant using biomass, as pyrolysis products, using a pyrolysis reactor before the gasification reactor. Energy produced from biomass is considered a green energy because as is burned and emits a certain amount of carbon dioxide in the atmosphere, the same amount of carbon dioxide is absorbed by other plants during growth.

8. Conclusions and personal approach

The IGCC plant proposed in this thesis is flexible in terms of electricity generation and hydrogen according to the requirement at a time, has a very low environmental impact compared with conventional technology and is flexible in terms of supply raw material.

For the evaluation of the innovative energy vectors co-generation schemes with carbon dioxide capture mathematical modeling and simulations were carried out using specific software ChemCAD. Data obtained from the simulations led to the preparation of case studies analyzed.

The main objective of this thesis is to investigate innovative ways of converting coal, waste and biomass into energy vectors (electricity and hydrogen), through gasification with carbon capture.

The IGCC plant scheme proposed in this thesis is very promising because of its advantages:

- Plant flexibility to produce electricity and hydrogen: depending on the demand the system has the ability to produce one of the two energy vectors;
- Plant flexibility to be fed with different feedstocks: coal, coal in addition with biomass or waste or biomass pyrolysis products;
- No metter the feedstock used, the plant has a very low environmental impact;
- This plant is a solution that can ensure the transition from an economy based almost entirely on coal by an economy based on renewable energy resources.

The results of this thesis contribute to the research field of energy conversion systems and use of renewable energy resources with carbon dioxide capture by the following additional contributions:

- Detailed analysis of coal gasification processes with or without the addition of biomass or solid waste, in order to be transformed into energy vectors (electricity or hydrogen) with carbon dioxide capture.
- Evaluation of IGCC plant flexibility to co-generate electricity and hydrogen with carbon dioxide capture, depending on the electricity demand at a time. Thus in

this period of continuous development of hydrogen based applications the IGCC plant can produce more electricity than hydrogen, and gradually as the demand for hydrogen will increase the plant will produce more hydrogen according to demand.

- Assessment of environmental impact of the proposed IGCC schemes with carbon capture compared with the current conventional IGCC plants without capture of carbon dioxide.
- Proofing, based on experimental data, the advantages of biomass pretreatment through pyrolysis before being gasified.
- The proposal of an efficient solution of using biomass as feedstock for electricity generation using biomass pyrolysis products in an IGCC plant. Biomass pyrolysis products can be used either in an IGCC plant with CO_2 capture such as the one proposed in this paper either in a conventional IGCC plant without CO_2 capture. This is possible by including a fluidized bed pyrolysis reactor before the gasification reactor.

The IGCC concept proposed in this thesis, due to its advantages, is a promising solution not only for short and medium term (until depletion of coal), but also for long term it will ease the transition from an economy based almost entirely on coal to an economy based entirely on renewable energy resources.

SELECTED REFERENCES

1. Higman, C., Van Der Burgt, M., 2008, *Gasification Second edition*, Elsevier Science.
2. The Institution of Engineering & Technology: Michael Faraday , www.theiet.org.
3. US Environmental Protection Agency, www.epa.gov.
4. Fanchi, J.R., 2004, *Energy: Tecnology and directions for the future*, Elsevier.
5. Demirbas, A., 2009, *Biofuels: Securing the Planet's Future Energy Needs*, Springer-Verlag London Limited.
6. Miller, B.G., 2005, *Coal Energy Systems*, Elsevier Academic Press.
7. Comisia Europeană, www. ec.europa.eu.
8. Cormoş, C.C., 2008, *Decarbonizarea combustibililor fosili solizi prin gazeificare*, Cluj University Press.
9. Gasification Technologies Council, 2011, www.gasification.org.
10. Basu, P., 2006, *Combustion and gasification in fluidized beds*, Taylor&Francis, New York.
11. De Souza-Santos, M.L., 2004, *Solid fuels combustion and gasification. Modelling, simulation and equipment operation*, Marcel Dekker, New-York.
12. Shoko, E., McLellan, B., Dicks, A.L., Diniz da Costa, J.C., 2006, Hydrogen from coal: Production and utilisation technologies, *International Journal of Coal Geology*, **65**, 213– 222.
13. Emun, F., Gadalla, M., Jimenez, L., 2008, Integrated Gasification Combined Cycle (IGCC) process simulation and optimization, *Computer Aided Chemical Engineering*, **25**, 1059-1064.
14. Kunze, C., Spliethoff, H., 2010, Modelling of an IGCC plant with carbon capture for 2020, *Fuel Processing Technology*, **91**, 934-941.
15. Massachusetts Institute of Technology Laboratory for Energy and the Environment, Report MIT LFEE 2005-002 WP, September 2005

16. Ladanai, S., Winterbäch, J., 2009, *Report: Global potential of sustainable biomass for energy,* Swedish University of Agricultural Sciences Department of Energy and Technology.
17. Jaccard, M., 2005, *Sustainable Fossil Fuels*, Cambridge University Press.
18. Quaak, P., Knoef, H., Stassen, H., 1999, *Energy from biomass*, World Bank technical paper; 422, Energy series, Library of Congress Cataloging-in-Publication Data.
19. I.W. Smith, The combustion rates of coal chars: a review, Proceedings of the Combustion Institute, 1045 – 1065, 1982
20. Lurgi GmbH, www.lurgi.com
21. Erasmus, H.B., van Nierop, P., 2002, Sasol: fifty years of growth, *IChemE 5'th European Gasification Conference*, Noordwijk, The Netherlands.
22. Wampler, T.P., 2007, *Applied pyrolysis handbook 2nd edition*, Taylor&Francis Group New York.
23. Douglas, P.H., 1976, The Cobb-Douglas Production Function Once Again: Its History, Its Testing, and Some New Empirical Values, *Journal of Political Economy*, **84**, 903-916
24. Fishburn, P.C.,Kochenberger, G.A., 1979, Two-piece von Neumann-Morgenstern utility functions, *Decision Sciences*, **10**, 503-18
25. Raja, A.K., Srivastava, A.P., Dwivedi, M., 2006, *Power plant engineering*, New Age Interantional Limited Publishers.
26. Cormos, C.C., 2010, Evaluation of iron based chemical looping for hydrogen and electricity co-production by gasification processwith carbon capture and storage, *International Journal of Hydrogen and Energy,* **35**, 2278 - 2289
27. Bhattacharya, A., Manna, D., Paul, B., Datta, A., 2011, Biomass integrated gasification combined cycle power generation with supplementary biomass firing: Energy and exergy based performance analysis, *Energy*, **36**, 2599-2610.