

SUMMARY

The doctoral thesis represents a set of theoretical and practical achievements on the processes of energy recovery of plant biomass through gasification technology.

Gasification has been studied to the limit of industrial achievements, initially for coal and later for woody biomass. The application of the technology to plant biomass (of agricultural origin) is in an initiation phase, with laboratory or pilot scale achievements for low thermal outputs.

World research on gasification of plant biomass has only gone through the phase of choosing the technically agreed technology in correlation with the energy characteristics of primary fuel. Unanimously, the technology of gasification in fixed layer, with descending flow, or otherwise formulated with equicurrent flow, is accepted for the current phase.

Research on the gasification of plant biomass will have to go through the whole cycle related to specific thermo-chemical processes previously developed for coal and wood. There is the problem of the temperature field in the areas of drying and devolatilization of the vegetal biomass and especially for the embers bed. As a rule, plant biomass has a significant moisture content, but especially a high content of volatiles (the emission of H_2 and CH_4 from volatiles will positively influence the content of fuel elements in the gas of gasifier produced). Regarding the embers bed, depending on the energy characteristics of plant biomass, the problem arises of achieving a temperature by partial combustion of fixed carbon controlled by excess air that ultimately controls the conversion CO_2 to CO . In parallel, conversion to form gaseous combustible components CH_4 and H_2

The developed analytical model responds to these aspects imposed by the characteristics of plant biomass. The developed model comprises autothermal gasification (without external heat contribution) for a fixed-bed and downdraft gasification system.

The analytical calculation model was validated by experiments on a specially constructed gasification plant. The installation comprises a fixed-bed gasogen with vertical flow (Lurgi type), an installation equipped with a temperature sensor located in the layer and with a flue-gas analyzer for samples taken from the exhaust gas.

The paper includes 7 chapters, of which the last one represents the general conclusions, the contributions made and the perspectives for the future.

Going through the chapters of the paper leads to the following aspects on the achievements in the work:

Chapter 1

Entitled "Basics of the gasification process" makes a summary presentation of the processes followed by the primary fuel in the gasification reactor, basically shown in figure 1.1. For a fixed-layer gasogen with descending flow, the main processes were positioned, comprising drying, pyrolysis, combustion and reduction of gaseous components in the hot carbon layer. These processes exist and develop in all types of gasogens, presented in Table 1.1., with reference to the fuel-oxidant circulation and the required temperature level.

The gaseous components emitted during the pyrolysis phase, as well as the influence of reduction reactions in the fixed bed, were presented. Reference has also been made to the resulting tar as a by-product with totally negative effects.

The chapter also made bibliographical references to oxygen oxidation and steam effect as technological principles for a particular class of gasogens and primary fuels. Data in Figure 1.2. link the nature of fuels and gasification technologies to the level of industrial technological applications.

The chapter also includes significant achievements of gasification plants in the European Union, specifying the primary fuels used, the gasification technology and the energy use of the gasogen gas produced.

Chapter 3 deals with "numerical modelling of gasification processes". This approach has emerged necessary to compare and translate long-running coal data to renewable primary fuels. To begin with, a review of the main achievements of modeling gasification processes was carried out. Next comes a classification of numerical simulation models, classified into zero-dimensional, one-dimensional, bidimensional and three-dimensional models, based on the principle of thermodynamic equilibrium.

The importance of the combustion regime, which can be stoichiometric or substoichiometric, as well as the Gibbs total energy factor, was highlighted. Table 2.3 presented a synthesis of stoichiometric modelling principles, followed by clarifications on the transition to non-stoichiometric models. Table 3.3. includes a systematization of the results of various numerical simulation methods for all gasification technologies.

The chapter concludes with a detailed presentation of TEM thermodynamic equilibrium models, models considered relatively simple to apply. These models are characterised by an appropriate level of simplicity and are very flexible to the quality of the primary fuel. The structure of the models is shown in Figure 3.1. and comprises three units, namely drying, pyrolysis and gasification.

The model contains a complete set of experimentally determined computational relationships for pyrolysis and gasification units, depending on the temperature in the respective unit, including steam reforming and methane-steam reforming reaction in the embers bed.

These models allowed the development of further applications, highlighting the influence of excess air, air preheating, steam injection or enrichment of air with oxygen.

In Figure 3.3. A summary of the influence of excess air on the composition of the gasogen gas and thus on its calorific value shall be provided.

At the end of the chapter for the development of the doctoral thesis objectives, it is specified that starting from the data presented for the gasification of plant biomass, the theoretical and experimental development of a simple installation, easily applicable in agriculture, with autothermal gasification with fixed bed air and descending flow is envisaged.

Chapter 4, Process modelling of a low-power fixed-layer gas for biomass, contains the data that led to the development of an original analytical calculation model, finally validated by the values obtained for plant biomass.

In the initial part, in subchapter 4.1. the biomass produced in Romania is presented, in terms of energy characteristics determined for the gasification process.

The basis for defining the energy characteristics of primary fuels was given to technical analysis covering moisture, volatiles, fixed carbon and ash. The volatile and fixed carbon content determines the processes in the pyrolysis zone, when the gaseous components CO , CO_2 , CH_4 and H_2 occur, and combustion respectively with increasing temperature in the fixed layer and reducing and reforming to form the final components of the gas of gasifier.

The energy characteristics of vegetal biomass represented by agricultural waste (cereal straw, corn and sunflower stalks, cobs) and waste represented by vine cords, apple branches, etc. were presented. These characteristics have been compared to those of wood of woody crops (energy willow) and even coal, a fuel with many applications previously for gasification.

The pretreatment of biomass was also taken into account, which leads to increased energy performance, namely:

- Obtaining optimal dimensions for primary fuel;
- Densification of biomass
- Increase in calorific value

To achieve these goals, operations occur:

- Chopping and sorting
- Hot pressing of chopped biomass to a certain size
- Forced drying of biomass

The possible amount of use of plant biomass was also presented, which is a definite application for the future.

Subchapter 4.2. includes the presentation of the original analytical model developed by the author and successfully applied in the continuation of the doctoral thesis. In Figure 4.2. The areas of the gasification process shall be presented within the model carried out, specifying as originality the uniform distribution of air along the length of the combustion zone.

The analytical model developed is particular to a fixed-bed down-flow gasogen without the use of auxiliary oxidizing agent (steam and/or oxygen). The model belongs to the category of those based on thermobalance, gasification being autothermic.

The process of analytical calculation starts from the anhydrous mass obtained by drying (dehydration) in the first phase. Next comes pyrolysis where the release relationships of volatiles (CO , CO_2 , H_2 , CH_4) depend on temperature. Heat for drying and pyrolysis is obtained by combustion of fixed carbon. With air comes a new gas, namely nitrogen that becomes dominant.

The heat released by combustion will be controlled by excess air, through two ways, namely: λ

- Pyrolysis zone temperature
- The temperature in the fixed layer, referred to in the paper as the "reference temperature"

Excess air in the literature for biomass gasification has recommended values in the domain $\lambda = 0.25 \div 0.45$.

The reference temperature controls the final chemical processes, especially the conversion of CO_2 to CO by reduction reaction (Boudouard reaction) as well as reforming with production of H_2 and CH_4

This makes it possible to determine the components of the gas, their mass from the initial mass of the fuel and the calorific value of the gaseous gas (gasification efficiency becomes an easily calculating element). The analytical calculation model was validated by a numerical application for woody biomass for an excess air value in the range of $\lambda = 0.4 \div 0.6$. In Table 4.8. the results of the calculation shall be provided, showing the variation in the final gaseous components. There was a slight decrease of components H_2 and CH_4 in gas of gasifier as excess air increased, and of course the proportion of N_2 . The calorific value varied in the range of $8000 - 7000 \text{ kJ}/m_N^3$ (the lower value being at the maximum excess air – figure 4.9.)

The transition to the calculation of vegetal (agricultural) biomass gasification for control required the use of data from specialized literature. The graph in figures 4.11 and 4.13 summaries the quality of the gasogen gas as a function of operating temperature (fixed layer temperature). These values will be another criterion for validating the original analytical calculation model for the primary fuel domain of agricultural (vegetable) biomass. The gasification of plant biomass covered three areas of plant biomass quality:

- Domain I, characterized by $C_f = 10\%$ and $V = 65\%$
- Domain II, characterized by $C_f = 15\%$ and $V = 60\%$
- Domain III, characterized by $C_f = 20\%$ and $V = 65\%$

The composition of plant biomass with a very high volatile content ($V = 60 - 65\%$) and low fixed carbon content ($C_f = 10 - 20\%$) shall be considered. This covers the quality of vegetable biomass represented by cereal straw, corn and sunflower stalks and vines, sunflower husks, or walnut shells, respectively.

The excess air had the values required by the autothermia of the process, to ensure heat for drying and pyrolysis (devolatilization) and the operating temperature of the fixed layer, with optimal values of $800 - 950^\circ\text{C}$. The result was the need to reduce excess air when increasing the value of fixed carbon from values $\lambda = 0.4 \div 0.5$ for $C_f = 10\%$, to $\lambda = 0.2 \div 0.3$ for $C_f = 20\%$.

Calorific value increased from a value around $3\,500 \text{ kJ}/m_N^3$ a value around $8\,800 \text{ kJ}/m_N^3$ with the increase of fixed carbon within the computational limits.

The results obtained are validated by the literature data presented above.

In conclusion, the gasification of plant biomass allows efficient operation for strict operation, especially for excess air. The strict control of excess air also allowed to obtain a gasogen gas with a relatively high content of H_2 and CH_4 , resulting in a calorific value at high values.

Chapter 5 addresses "Experimental results of plant biomass gasification at fixed layer pilot plant" The chapter contains in the first part a synthesis of the experimental results in the European Union regarding the gasification of plant biomass. However, research is still at an early stage, at pilot

plant level, even for a primary fuel consisting of a mixture of wood and plant biomass. Experimental results were presented, including for a mixture of avian manure with woody biomass in certain proportions. The results demonstrated low calorific value gas and efficiencies in the range 72 – 77%. Only small amounts of tar were noted, below $1g/m_N^3$.

Data on the basic dimensions of the reactor and gasification efficiency were the basis for the sizing of the gasifier made for experimental research. It has been designed for a quantity of $1kg/h$, the primary fuel having the construction characteristics shown in figure 5.3. Thus, gasogen represents:

- A cylindrical reactor;
- Superior primary fuel supply,
- Downward flow
- Uniform air distributed through 4 pipes over the height of the combustion zone;
- The existence of a grill at the base of the embers bed.

Sizing calculations indicated a diameter of 80 mm and a length of 400 mm, of which 230 is the active length.

The pilot plant is with batch supply. For starting, the embers bed is made outside the installation (solution used in many installations of the Lurgi generation). Next comes the introduction of primary fuel and air intake with an overpressure of 1.1-1.2 bar. The air flow is admitted by a compressor, it is measured using a rotameter, observing the imposed excess. The temperature of the embers layer is measured using a high-temperature thermocouple. In Figure 5.6. It is a photo of the plant assembly. The gasogen gas components are measured with a TESTO 350 analyzer when the gasogen gas is discharged into the atmosphere. As shown in photo 5.9, samples of ignition of gasogen gas in the atmosphere were also carried out.

The experiments included 3 qualities of vegetable biomass, consisting of cereal straw, pellets of cereal straw and wood (30%) and pellets of cereal straw with the results presented in Table 5.4.

Experimental research also included the successful gasification of walnut shells, a fairly common local fuel.

Experiments on the most difficult primary fuel tested represented by cereal straw confirmed the effectiveness of the proposed technology, namely:

- Constant-diameter, down-flowing cylindrical body reactor;
- Use of a uniform intake distributed over the height of the combustion zone;
- Maintaining excess air within optimal limits, $\lambda = 0.3 \div 0.5$

The experimental data were validated by the concordance of the degree of conversion CO_2 to CO by the reduction reaction in the layer, by observing Boudouard's law (represented by the correspondence of the temperature layer-degree of conversion CO_2 in CO).

The experimental data were also supplemented with those obtained when gasifying walnut shells. The chapter concluded with general recommendations.

Chapter 6 "Decarbonisation of flue gases by gasification" is another possible application of the results obtained. The conversion of the component CO_2 from the flue-gas to CO the embers layer (bed) shall be considered. Studies were conducted for the gasogen variant developed for

gasification of plant biomass. For efficient operation, the degree of CO_2 conversion to CO , was required to be a minimum 95%, so that the temperature was greater than 850°C.

This temperature will be obtained by burning part of the fixed carbon bed formed by charcoal through a certain amount of oxygen or oxygen introduced into the reactor (the burnt carbon mass will have to be completed, so that a balance, temperature, content of O_2 and introduced carbon mass were presented in the paper).

The chapter concludes with an analysis of the possibilities of using the gas resulting from conversion CO_2 into CO .

Key words: gasification, vegetal biomass, decarbonization, modelling

Table of content

List of figures	Error! Bookmark not defined.
List of tables.....	Error! Bookmark not defined.
1. Basics of the gasification process	Error! Bookmark not defined.
1.1. The objectives of the research work.....	Error! Bookmark not defined.
1.2. Basic processes in gasification technology	Error! Bookmark not defined.
2. Gasification installations.....	Error! Bookmark not defined.
2.1. The physico-chemical characteristics of the gasification process.....	Error! Bookmark not defined.
2.2. Chemical and energetic characteristics of gas of gasifier ...	Error! Bookmark not defined.
2.3. Types of gasification reactors and respective technologies	Error! Bookmark not defined.
2.4. Achievements in the European Union at the pilot and industrial level of energy use of gas of gasifier.....	Error! Bookmark not defined.
3. Numerical modeling of gasification processes	Error! Bookmark not defined.
3.1. Modeling objectives	Error! Bookmark not defined.
3.2. Classification of numerical simulation models	Error! Bookmark not defined.
4. Process modeling of a fixed-bed low-power gasifier for biomass.....	Error! Bookmark not defined.
4.1. Biomass produced in Romania – possibility of gasification	Error! Bookmark not defined.
4.2. Analytical Model for Fixed Bed Biomass Gasifier for Small Consumers.....	Error! Bookmark not defined.
4.2.1. Modeling of thermochemical processes	Error! Bookmark not defined.
4.2.2. Determination of the thermal field	Error! Bookmark not defined.
4.2.3. Validation of the model by numerical applications.....	Error! Bookmark not defined.

Conclusions	Error! Bookmark not defined.
4.3. Analytical modeling of fixed bed air gasification performances of agricultural biomass	Error! Bookmark not defined.
4.3.1. General aspects related to air gasification of agricultural biomass in fixed bed ..	Error! Bookmark not defined.
4.3.2. Particularities of air gasification of agricultural biomass in fixed bed	Error! Bookmark not defined.
4.3.3. Modeling of agricultural biomass gasification and analysis of results	Error! Bookmark not defined.
I. Gasification of biomass with the lowest fixed carbon content	Error! Bookmark not defined.
II. Gasification of agricultural biomass with technical analysis	Error! Bookmark not defined.
III. Gasification of agricultural biomass with technical analysis	Error! Bookmark not defined.
Conclusions	Error! Bookmark not defined.
5. Experimental results of the gasification of plant biomass at the pilot plant with fixed bed	Error! Bookmark not defined.
5.1. Research objective	Error! Bookmark not defined.
5.2. Achievements in the field of gasification of plant biomass	Error! Bookmark not defined.
5.3. Dimensioning of pilot gasifier	Error! Bookmark not defined.
5.4. Results of the gasification process of agricultural biomass	Error! Bookmark not defined.
5.4.1. Results obtained by analytical modeling	Error! Bookmark not defined.
5.4.2. Experimental results	Error! Bookmark not defined.
Conclusions	Error! Bookmark not defined.
6. Decarbonization of combustion gases by gasification	Error! Bookmark not defined.
6.1. General problem	Error! Bookmark not defined.
6.2. Thermochemical processes of CO_2 capture from combustion gases by gasification ..	Error! Bookmark not defined.
6.3. Use of gasification gas	Error! Bookmark not defined.
Conclusions	Error! Bookmark not defined.
7. General conclusions, personal contributions and future perspectives	Error! Bookmark not defined.
7.1. Conclusions	Error! Bookmark not defined.
7.2. Contributions	Error! Bookmark not defined.

7.3. Future perspectives.....**Error! Bookmark not defined.**
Bibliography **Error! Bookmark not defined.**