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Selected aspects of coal gasification for application in low-emission energy technologies

Solid fuel electricity generation has been known and used for many years. The combustion of solid fuels is a complex process that requires proper preparation of the fuel, carrying out the combustion process, as well as the removal of harmful substances in the form of dust and gaseous pollutants (NOx, SOx, CO) from exhaust gases emitted into the environment. For decades, the gaseous form has been considered the noblest form of fuel. Gaseous fuels can be easily transported over long distances, are immediately ready for combustion and the composition of the fuel mixture can be freely adjusted. The constant pursuit to reduce anthropogenic greenhouse gas emissions require the use of low-emission and zero-emission energy generation technologies. In the case of coal, this will mean a shift from direct combustion to more advanced systems powered by gaseous fuel. The paper presents an overview of the available techniques and technologies of solid fuel gasification aimed at the production of gaseous fuels, which can be used in low-emission energy technologies. The computational methods of the gasification process are also presented, which allow the selection of the best technology and operating parameters of individual reactors.

Key words: *gasification, gas fuel, syngas, gas technology*

1. INTRODUCTION

The process of fossil-derived solid fuels gasification significantly increases their applicability while reducing their negative environmental impact [1]. Produced by thermal decomposition, in a small quantity of air\oxygen or water vapor, the basic product of gasification, called syngas (synthetic gas), can be used both for heat generation in the energy sector and for the production of synthetic hydrocarbons in the petroleum and chemical sector [2]. Such a wide range of applications is, on the one hand, an opportunity for the development of this technology and, on the other, a risk that the products obtained will not be used where the benefits of their use will be the greatest. Given the current problems of ensuring clean air in many regions of the world, and the need to reduce greenhouse gas emissions [3], the use of syngas in energy technologies seems appropriate (Fig. 1). The combustion of solid fuels causes relatively greater emissions than the combustion of gaseous fuels (emission of both solid and gaseous pollutants), especially in the case of low-power boilers that are not equipped with flue gas cleaning systems, and where the combustion process conditions are often significantly different from optimal [4]. There are many indications that the greater use of syngas can be expected in the chemical industry in the future [5].

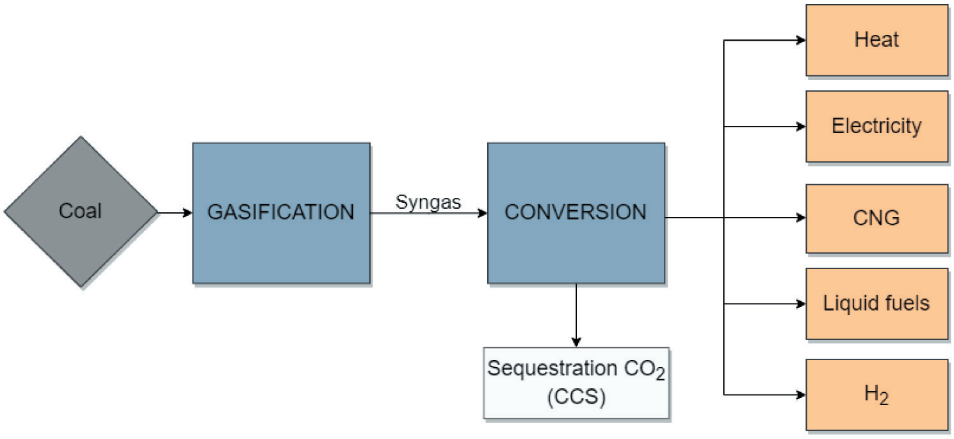

Fig. 1. Syngas as universal energy resource

2. GASIFICATION TECHNOLOGIES – SOLUTION OVERVIEW

Solid fuel gasification technologies are categorized according to the design solutions of the system used for the gasification process (including the reactor itself) and the process parameters: temperature, properties of the feedstock, gasifying agent and its concentration, pressure. The achievable process parameters parameters are determined by the choice of the gasification system.

The gasification systems are distinguished by the method of heat supply to the process (with direct heating – autothermal, with indirect heating – allothermal) and the movement of the reactants in the reactor. The basic types include moving bed gasifiers (downdraft and updraft), fluidized bed gasifiers (bubbling fluidized-bed or circulating fluidized-bed), and entrained-flow gasifiers. Moving bed gasifiers are referred to in the literature and hereinafter interchangeably as fixed bed reactors. Diagrams of most common gasifier types are shown in Figure 2.

The type of reactor determines the temperature range in which the gasification process is carried out and affects the temperature profile along the length of the reactor. An increase in temperature usually increases the proportion of hydrogen and carbon monoxide in the produced syngas, while an increase in pressure increases the proportion of methane [6]. The main differences between reactor types are presented in Table 1.

In commercial applications, reactors of various designs are used, operating with different sets of the process parameters – at present it is difficult to define the optimal variant, although development trends can already be indicated. Updraft reactors are suitable for raw materials where moisture accounts for up to 50 wt%. Feedstock with a high inert content, such as coal dust, sewage sludge or municipal waste, which are not suitable for fixed bed reactors, have been successfully gasified in bubbling fluidized bed reactors. Wood biomass is not suitable for entrained-flow reactors, unless it is pretreated in torrefaction and grinding processes [7]. In practice, bio-coal suspensions with bio-oil are used, as well as wood or bio-coal dispersed/dissolved in glycerol, ethylene glycol, phenolic oil or bioliquids [8]. Fixed bed reactors are currently being used less and less. Fluidized bed reactors, despite many advantages, have gained relatively little commercial interest. Currently, the most commonly developed and used reactors are entrained-flow ones [9].

2.1. Coal

Coal gasification technologies have been in operation since the beginning of the 19th century. The popularity of this raw material as a gasification feedstock is high due to the stability of supplies, relatively stable quality parameters and low price. Coal is commercially gasified in fixed bed gasifiers (Lurgi [10] and British Gas Lurgi [11, 12]), fluidized bed gasifiers (Great Point Energy [13], Winkler [14], TRIG [15]) and entrained-flow gasifiers (GE Energy [16], Texo, Koppers-Tozek, Shell [1]).

In the past, the synthesis gas obtained from coal was used for various purposes. During World War II, processing into transport fuels in the Fischer-Tropsch synthesis process predominated [17], while in the last 60 to 70 years a significant amount of it was converted into hydrogen for the production of ammonia [18]. In the last 25 years it has been intensively used in commercial power plants with the use of technologies based on an integrated gasification combined cycle (IGCC) [19].

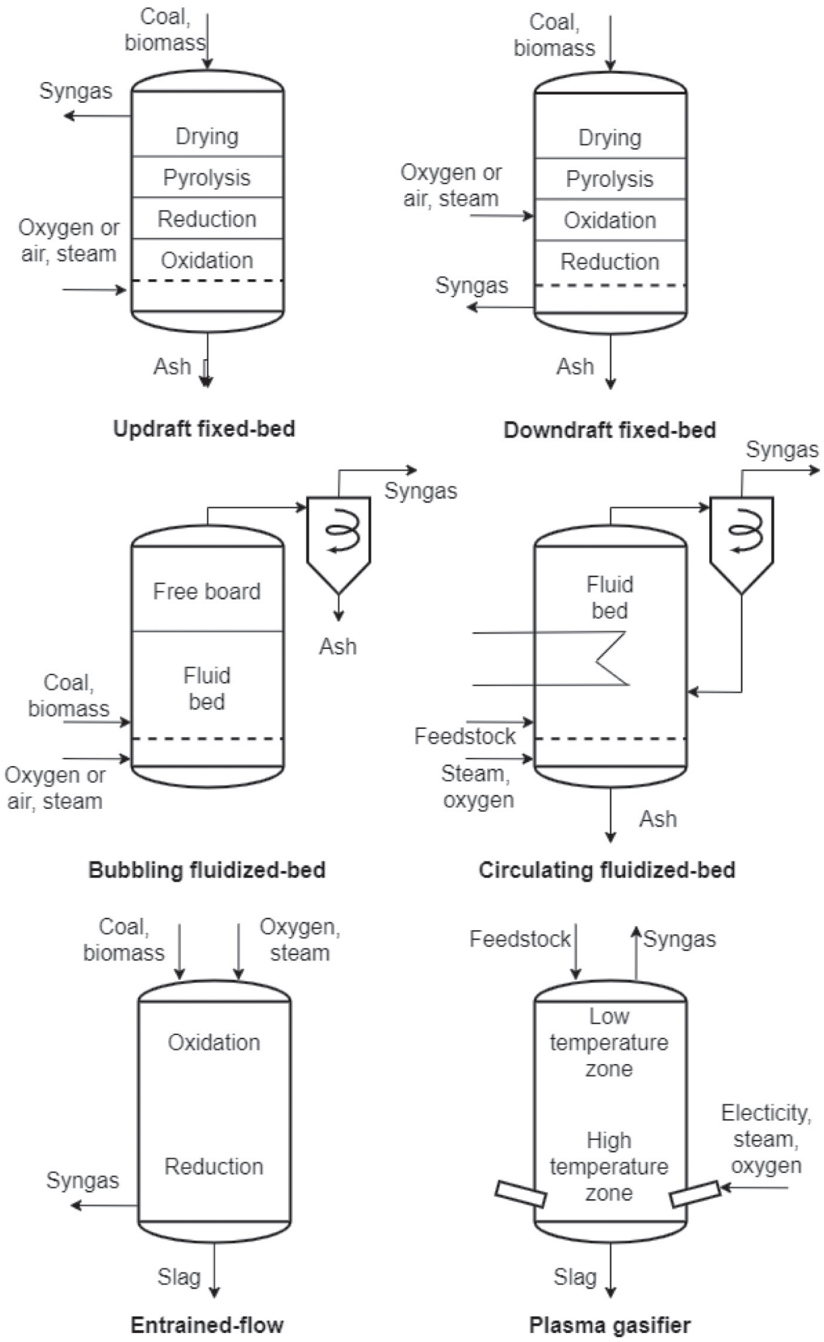

Fig. 2. General scheme of synthesis gas reactors (based on [3, 9, 10])

Table 1

Comparison of the primary types of gasification reactors [6]

Property	Fixed-bed	Fluidized-bed	Entrained-flow
Particle size [mm]	<51	<6	<0,15
Gas outlet temperature [°C]	450–600	800–1000	>1260
Temperature in the reaction zone [°C]	1090	800–1000	1990
Demand for an oxidizing agent	low	moderate	high
Characteristics of ash	dry	dry	slag
Scale	small	medium	large
Operational problems	tars blocking equipment and pipelines	low carbon conversion	material problems due to high temp.
Installed power [GW]	18.7 (42%)	0.9 (2%)	25.4 (56%)

2.2. Cogasification with biomass

The level of greenhouse gas emissions has become one of the benchmarks for the application of technological solutions in both environmental and economic terms. For this reason, mainly those gasification technologies that allow the use of biomass or waste not suitable for further processing are being developed. As a rule, the gasification process of such materials is analogous to that of coal, but the differences in their structure and heterogeneity constitute a major technical challenge. This means that the design of the reactors must be modified [20], to increase their flexibility in working with particles of greater differentiation in size and shape, moisture, calorific value and content of the elements: carbon, hydrogen, oxygen, sulfur and nitrogen, as well as ash and trace elements (which, however, can significantly affect the course of the process and the quality of the products received) [21]. Taking into account the features of individual types of reactors, a fluidized bed reactor is often used for gasification of biomass (HoSt [22], ANDRITZ AG [23], Valmet Corporation [24], Eqtec [25]), but also in this case other reactors can be used: fixed bed (Shangqiu Haiqi Machinery Equipment [26], Chanderpur Works [27], Infinite Energy [28]), entrained-flow (Siemens AG [29]). Despite many technical and logistical problems [30], which often ended with the cessation of operation of biomass gasification plants, there is still considerable interest in these technologies [12]. It is related to the higher exergy efficiency of gasification compared to the combustion process [31].

The reduction in the number of operational problems typical for biomass gasification can be achieved by co-gasification with coal [32]. Adapting the technology to gasification of these fuels in different mass proportions greatly facilitates the management of the continuity of energy production. In cogasification, the efficiency of the process is also improved by ensuring a sufficiently high calorific value of the feed.

3. CHEMICAL MECHANISM OF THE GASIFICATION PROCESS

From a chemical point of view, a number of redox reactions occur during the gasification process. Depending on the process temperature, carbon in the form of either pre-decomposed organic compounds or in the form of radicals is oxidized by the so-called gasifying agent. Oxygen, hydrogen, water vapor, or even carbon dioxide may be the gasifying agent. Depending on the gasification agent used, chemical reactions during gasification can be as follows:

– Gasification with oxygen:

$$2C + O_2 \rightarrow 2CO \qquad \Delta H = -246\ \text{KJ/mol}$$

Oxygen as a gasifying agent may be in the form of air, but due to the formation of nitrogen oxides at high temperatures, it is better to use pure oxygen for gasification. The reaction is exothermic and with volume increases, so the shifting of the equilibrium toward the products is achieved by cooling and reducing the pressure, i.e. taking away heat and removing the CO produced.

– Gasification with hydrogen:

$$C + 2H_2 \rightarrow CH_4 \qquad \Delta H = -75\ \text{KJ/mol}$$

Here, the reaction is also exothermic, but proceeds with a reduction in volume, so to speed up the process, heat should be taken and pressure increased.

– Gasification with steam:

$$C + H_2O \rightarrow CO + H_2 \qquad \Delta H = +119\ \text{KJ/mol}$$

The reaction is endothermic and is proceeding with an increase in volume.

– Gasification with carbon dioxide:

$$C + CO_2 \rightarrow 2CO \qquad \Delta H = +172\ \text{KJ/mol}$$

The reaction is also endothermic and is proceeding with an increase in volume.

Apart from the reaction of coal with the gasifying agents in the gasification process, there are a number of reactions of gasification products. Here are some:

$$CO + 3H_2 \rightarrow CH_4 + H_2O \qquad \Delta H = -206\ \text{KJ/mol}$$

$$CO + H_2O \rightarrow CO_2 + H_2 \qquad \Delta H = -41\ \text{KJ/mol}$$

$$2CO + O_2 \rightarrow 2CO_2 \qquad \Delta H = -676\ \text{KJ/mol}$$

$$CO_2 + 4H_2 \rightarrow CH_4 + H_2O \qquad \Delta H = +119\ \text{KJ/mol}$$

$$CH_4 + H_2O \rightarrow CO + 3H_2 \qquad \Delta H = +206\ \text{KJ/mol}$$

In the real gasification process, not one reaction takes place, but several at once, therefore controlling these processes requires a lot of effort. It is performed by controlling the pressure (delivering or receiving reagents) and controlling the temperature.

Moreover, when the process is carried out at very high temperatures, reagents in the radical form can participate in the reactions, in which take place the degradation of O_2 to radical oxygen O*, degradation of water molecules into O* radical, and hydrogen molecule or directly into hydrogen radicals 2H*. These radicals are very reactive and easily react with carbon as well as other compounds present in the gasifying mixture.

The use of carbon dioxide as a gasifying agent can be a very attractive method of its management – the product of such a process is carbon monoxide (CO), which is an energy carrier. The most promising gasification option is the use of steam as a gassing agent – the product is not only combustible carbon monoxide, but also gaseous hydrogen, which after separation is the desired energy carrier. It is gaining more and more popularity due to the possibility of its use in conventional combustion engines, gas turbines and fuel cells.

4. CALCULATION METHODS FOR THE GASIFICATION PROCESS

The accuracy of the results of the modeling gasification processes depends on the assessment of technical, economic, and ecological efficiency, which is the basis for deciding on the legitimacy of the project that may increase the efficiency of using solid fuels. As a result of modeling, process parameters are determined: the amount of individual media and the resulting gas composition [33]. Modeling also allows to examine the influence of the composition of the feedstock, the type of gasifying agent, and the parameters of the gasification process on the obtained results [31]. The gasification process calculations are laborious. Therefore, they are performed almost exclusively in software. Their further development in terms of the accuracy of results is supported by the so-called machine learning and the use of neural networks [34].

4.1. Engineering methods

Conversion of solid fuel in the gasification process can be modeled using the equilibrium approach alone, using only the kinetic approach, and also using these two approaches simultaneously [35]. The use of engineering computational methods is associated with making many assumptions, depending on the selected method. These are the assumptions related to the temperature distribution in the reactor, heat exchange with the environment, the individual reactions rate. These methods usually don't take into account the formation of tars, replace coal fuel with pure carbon (graphite) and ignore the impact of the ash present in the raw material on the process. Nevertheless, practice has shown that the use of these methods leads to an approximation sufficient for engineering purposes [33].

Equilibrium models are among the oldest, most well documented and frequently used. The results obtained from these models are characterized by high compliance, especially in the analyzes of gasification at high temperatures (>850° C). Wang et al. showed that they are very useful, especially in the modeling of IGCC installations [36]. In one of the modeled cases, they obtained the consistency of the syngas composition modeling results with the experiment results at the level above 99% (relative error below 1%). Based on this model, they demonstrated how to maximize Cold Gas Efficiency (CGE) by optimizing the gasification temperature, oxygen supply and steam supply. The achieved improvement of CGE was 10% [37].

Kinetic models provide more information on the gasification mechanisms useful for process design and development but require more computational effort and are partly based on complex empirical kinetic equations [3]. Nowadays, a convenient method of modeling is the use of Aspen Plus software [38–41], in which there is no need to determine many chemical reactions occurring during gasification, but a process flow diagram should be created, and chemical compounds that may be present in the process, as well as the conditions of the process (flow, temperature, pressure), should be defined.

4.2. CFD modeling of the solid fuel conversion process

Many multidirectional changes between the substrates and the gasifying agent occur in the gasifier, which may also be affected by gasification products. The final effect of the process is significantly influenced, apart from the process parameters, by the shape and dimensions of the reactor, which determine the time and conditions in which the reactants stay. For this reason, the engineering methods that have proven successful in balancing the mass and energy of the gasification process are not sufficient to improve the design of reactors and minimize the emission of undesirable compounds [42]. The significant progress made in recent years in the field of numerical modeling techniques and the efficiency of computing machines allows for the modeling of the behavior of raw materials and products in the 3D model of the reactor [43, 44]. The model can take into account the fluid dynamics, the size and structure of the raw material particles, the kinetics of chemical reactions, mass and energy exchange between the solid and gas phases [45]. Modeling methods are further developed to reduce calculation time while maintaining or improving the consistency of results. For example, Mularski and Modlinski have demonstrated the significant impact of the adopted kinetic parameters on the composition of the producer gas and proposed a new procedure for optimizing input data [46]. The result of CFD modeling is, inter alia, distribution of velocity, temperature, unreacted raw material and gasification products.

5. STRATEGIES FOR COAL GASIFICATION

The energy sector is a key element of the economy, as its competitiveness largely depends on it. For this reason, it is present in the policies and strategies of countries around the world, and these, in turn, largely determine the feasibility and cost-effectiveness of projects in specific technologies. As an opportunity to convert raw materials into useful products (including fuels and energy), gasification is included in the strategies of all developed countries. Selected information, which presents a political attitude and reflects the state of knowledge, is presented in the following subsections.

5.1. European Union

The European Green Deal, together with Fit for 55, aims at the reduction of at least 55% of the European Union's greenhouse gas emissions by 2030 compared to 1990. Increased use of hydrogen is meant to help achieve this goal. The EU strategy for the development of hydrogen production advocates obtaining hydrogen from fossil fuels with carbon dioxide capture. According to EU estimates, in order for it to be able to compete with hydrogen from fossil fuels without CCS, carbon-emission prices between 55 and 90 Euro per ton of carbon dioxide equivalent (CO_2e) would be necessary. This level was already reached in mid-2021, and at the time of writing this paper, the price was above its upper limit.

5.2. Poland

The government document "Energy policy of Poland until 2040" states that the global effects of research and development activities (R&D) indicate the existence of a potential for low-emission or zero-emission use of coal. In practice, this is to allow for partial further use of coal generating units. For this reason, the policy recommends searching, testing and implementing new methods of coal use and processing, i.e. gasification, oxy-combustion and other clean coal technologies.

At present, LW Bogdanka is discussing the cost-effectiveness of IGCC using Mitsubishi Hitachi Power Systems technology with a capacity of 500 Mwe [47].

5.3. United States of America

The Fossil Fuels Office of the US Department of Energy (DOE) supports the development of modular gasification technologies of various types and qualities of coal, as well as blends of coal with biomass and other waste, aimed at the production of clean syngas suitable for the production of electricity, chemicals, hydrogen, transport fuels and other products depending on the needs of the internal market. In 2021, the DOE selected four projects for which it awarded $2 million in research and development (R&D) support to refine the technology for the gasification of coal-biomass mixtures and plastic waste. According to assumptions, these technologies are designed to produce hydrogen while ensuring a negative carbon balance.

5.4. Japan

In the strategy of the Japanese Agency for Natural Resources and Energy of July 2018 (The 5th Strategic Energy Plan) it was stated, inter alia, that in order to further reduce greenhouse gas emissions, the development and practical applications of a new generation of highly efficient coal-based energy production technologies will be promoted, such as the integrated coal gasification combined cycle (IGCC) and the integrated coal gasification fuel cell combined cycle (IGFC). As a result, the world's largest 525 MWe IGCC installation was launched in Iwaki in 2021, with 48% efficiency.

5.5. Australia

Australia, rich in coal seams, in the published strategy „First Low Emissions Technology Statement – 2020" indicates coal gasification combined with carbon dioxide sequestration as one of the cheapest methods of producing pure hydrogen in the short-term perspective. For this reason, the Australian government is expected to allocate $50 million to research and development (R&D) projects aimed at achieving commercialization.

6. CONCLUSIONS

The discussion on the purposefulness and efficiency of coal gasification in the era of decarbonization, pursuing for climate neutrality and implementing new and more stringent EU regulations in the field of climate protection is of great importance. However, it requires the development and implementation of advanced and dedicated technologies that follow these regulations, ensuring highly efficient production of fuels for low-emission energy generation.

Coal has tremendous potential for other energy-chemical applications, the basis of which will be the processes of its conversion: gasification or hydrogenation. The scientific community is facing a number of challenges, the realization of which will create the possibility of developing effective, climate-friendly technologies of the 21st century for the generation of energy and raw materials from coal deposits. There are many arguments in favor of coal gasification, including:

- improving the efficiency of low-emission electricity generation from hard coal, aiming at high-efficiency zero-emission coal-fired units integrated with the capture of CO_2 from the flue gas,
- increasing the raw material independence for the petrochemical industry,
- facilitating the process of biomass gasification (co-gasification).

The review of solid fuel gasification technologies has shown that there are advanced technologies available that can be applied to many raw materials. The type of reactor has a significant influence on the physicochemical properties of the obtained products, apart from the parameters of the raw materials. It determines the feasible process conditions and the way in which the raw material should be prepared. The selection of an appropriate solution strongly depends on the intended method of further use of the products and the possibility of waste management (including waste heat). Proper correlation of the gasification technology with the technology of energy use of its products is necessary from the point of view of maximizing the total energy efficiency and contributes to the reduction of the emission of greenhouse gases and pollutants: tars, dust, ammonia, hydrogen sulfide, hydrogen chloride, etc. [48]. For this purpose, available modeling methods can be used, which have repeatedly shown a high correlation with the experimental results. Optimization of the design of the selected gasification reactor is possible thanks to the numerical modeling methods, which are being constantly developed, but already enable the achievement of reliable (verified and validated) results with the use of CFD (Computational Fluid Dynamics).

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