The efficiency of the process of coal gasification in the presence of hydrogen

Gracjana Woźniak[1,*], *Rafał* Longwic[1], *Kamil* Szydło[1], *Adam* Kryłowicz[2], *Jarosław* Kryłowicz[2], and *Robert* Juszczak[2]

[1]Lublin University of Technology, Department of Vehilces, Nadbystrzycka 36, 20-618 Lublin, Poland
[2]Bicarbo Sp. z o.o., Grzybowska 2/29, 00-131 Warsaw, Poland

Abstract. The results of the research on the process of gasification of coal mixture in a closed system was evaluated in this article. The method presented includes oxygen-free gasification of coal at high pressure and its thermal degradation. Hydrogen is a gasifying factor. Gas containing, among others, methane, carbon dioxide, carbon monoxide, hydrogen is obtained as a result of reaction. Fossil coal as a chemical and energetic raw material is very significant in the process of demand for energy. Due to more and more difficult access to petroleum and natural gas, as well as growing prices, fossil fuels are becoming economically attractive. However, the works on more effective and environmentally friendly methods of energetic use of coal - including its gassing - should be continued. The use of hydrogen during coal gasification allows to get emission-free fuel gas, having better energetic properties than primal raw material.The laboratory site was built and consisted of: 4,5dm^3 reactor with a heating system, system of supply reactor with hydrogen and rinsing with argon and cooling system. The results of hydrogasification of coal with specific parameters, for selected time intervals were presented in this article. The efficiency of hydrogasification depending on the type of applied coal was assessed. Powdered charcoal, meeting the norm of German Committee for Standardization DIN EN1860 2 with a number 3H020, hard coal and lignite coal, as well as their mixtures were applied during the research.

1 Introduction

Limited resources of petroleum and natural gas, instability of the prices of petroleum-derived fuels, the necessity of controlling the emission of greenhouse gasses, development of new technologies are the factors affecting changes in global structure of the use of primary fuels. According to the forecasts of International Energy Agency (IEA), coal will remain basic chemical raw material and energetic fuel (Fig. 1.1). Therefore, coal gasification as a technology of energy production is becoming more popular due to global availability of this raw material and economic conditions. Global coal resources are estimated to last for about 200 years. Polish resources of hard coal amount to about 43,3 billion tons, whereas,

* Corresponding author: g.wozniak@pollub.pl

lignite coal – 13,7 billion tons. Global petroleum resources are estimated to last for about 45 years, and natural gas for about 60 years.

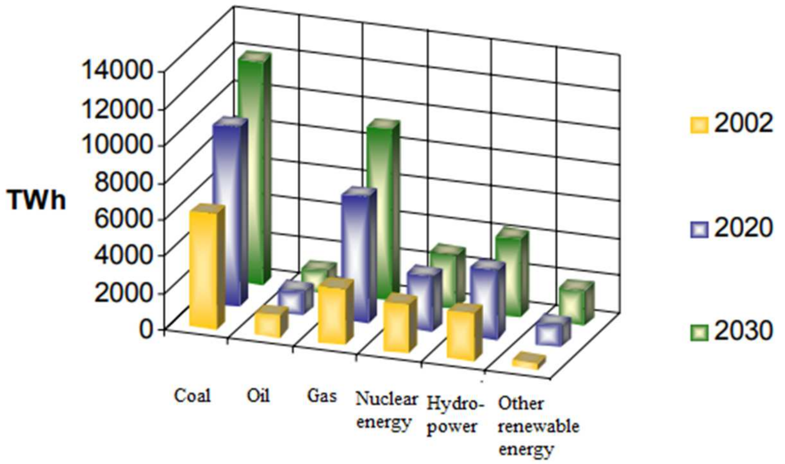

Fig. 1. IEA forecasts – perspective within the scope of amount of energy that can be obtained from various primal sources

Coal gasification is a complex of multidirectional changes, both chemical and thermal, occurring in high temperature between coal and gasifying factor. Air, oxygen, steam, carbon dioxide, rarely hydrogen, are usually applied as a gasifying factors. Gasifying factor is usually transmitted through heated coal to ensure close molecular contact for chemical reaction. The goal of the process of gasification is a complete conversion of coal to combustible gas that can be used as fuel or synthesis gas. The efficiency of gasification depends on the type of coal and gasifier. Technology of coal gasification allows to use generated gas in many ways [26]. The majority of the process of gasification is endothermic reactions, part of the element of coal is oxidized to carbon dioxide and applied in the process. Unfortunately, raw material such as coal is not only hydrogen and coal as the element, it is also the so-called heteroatoms, which generate such harmful substances in the process of coal gasification as: hydrogen sulphide, ammonia, carbon oxysulfide in the amount not exceeding 1% of generated gases.

Technology of coal gasification has been applied around the world for many years. It dates back to the 19th century, when Lurgi gasifier with slidable deposit of coal (1887) was patented. There are many processes of gasification. They can be divided due to the way of flow of raw material inside the reactor, therefore, the reactors can be divided into three categories: with permanent or slidable deposit, with fluidal deposit and dispersive reactors. The process of coal gasification can be conducted in two ways. Firstly, in the gasification reactors, the so-called gas generators, located on the surface, secondly as the so-called ground-based gasification or in the georeactor located in the coal, as underground gasification (gasification in situ). The results of research on coal gasification in a prototype of ground-based reactor were presented in the article. Hydrogen was applied as a gasifying factor Hydrogen is a coal carrier affecting improvement of efficiency of the process. The research results showed the positive impact of application of H_2 as a gasifying factor. Technologies based on hydrogen may use energy in a more efficient way, provide ultra clean fuels, eliminate emission of pollutants and considerably limit emission of greenhouse gasses to atmosphere, particularly carbon dioxide.

2 Empirical research

2.1 Research site

Figure 2 shows a scheme of the research site used for coal gasification. The system consists of the following main elements: about 4,5 l reactor with a heating system, system of supply reactor with hydrogen and rinsing with argon and cooling system.

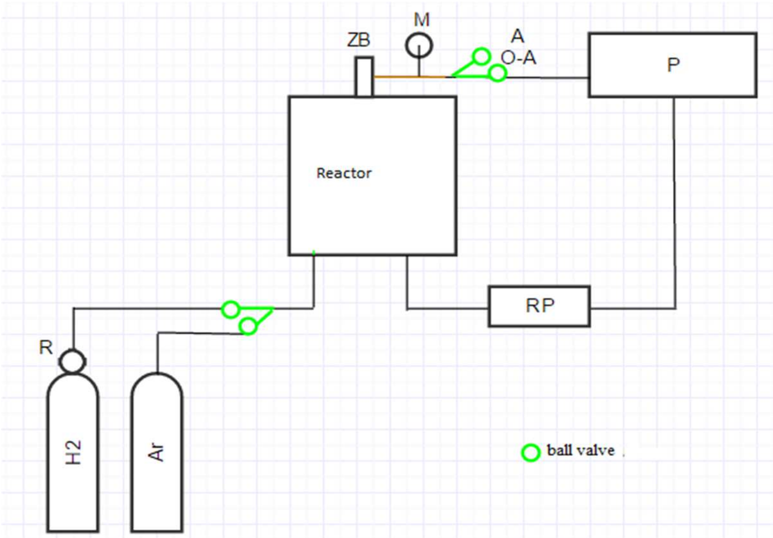

Fig. 2. Scheme of the measuring site used for coal gasification: R- reducer, H2 - hydrogen cylinder, Ar- argon cylinder, ZB – cylinder valve, M – manometer, A O-A- receiving-analysing apparatus, P – pump, RP – flow regulator

The casing of the reactor (Fig.2, Fig. 3) consisted of a cover made of heat resisting steel with the stub pipes charging and discharging gas and coolant (water). Double walls of the cylinder form an external chamber. Temperature of the charge was measured with the use of a sensor containing thermoelement PTTN TKb-30.

Fig. 3. Reactor

Fig. 4. Thermal shield

Fig. 5. Diffuser

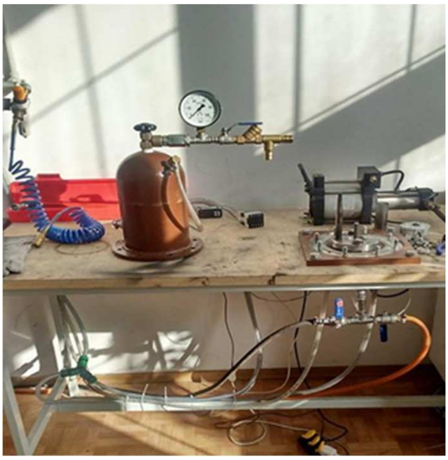

Fig. 6. Site used for coal gasification

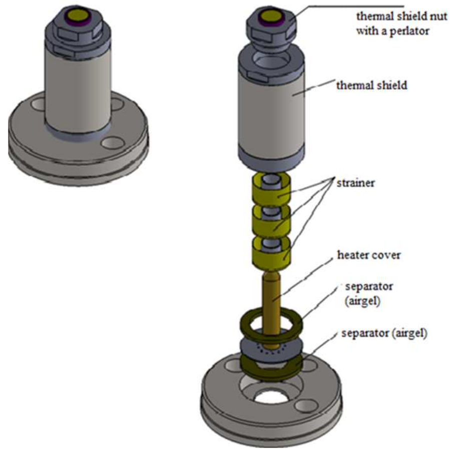

Fig. 7. Mody1 site with the heater cover

Hydrogen supply system – technical compressed hydrogen 5,0 and rinsing with argon – compressed technical argon 5,0 consists of a pressure cylinder, manometer of reducing and discharge valves. Post-reaction gas flows to the radiator. The main element of the reactor is a furnace. The research was conducted on MODY1 site (Fig. 4). The furnace consisted of a thermal shield with a nut containing perlator (Fig. 5), distance and diffuser (Fig. 6). Figure 7 shows a site used for coal gasification.

2.2 Methodology of the research

The process of gasification was conducted in repeatable initial conditions. Sample of coal in the ground form of the weight about 9-19 g was heated. At first, the samples of coal for the research were prepared and weighed, then charge in the form of two filling sieves with coal in the furnace, was placed in the bottom part of the reactor, and thermal and cylindrical shield placed. When tightness was checked, the reactor was screwed together. Argon under the pressure of 1,2 Mpa was raised and dropped three times to rinse out every sample. The same action was repeated when the samples were rinsed out two times with hydrogen under the pressure of 1 MPa. Upon rinsing with gases, the reactor was filled with hydrogen up to 1,2 MPa. Heater and cooling of the bottom cover of the reactor were turned on. The measurements at every attempt were started from 800°C. Temperature in the oxidation zone should be maintained at the level of 700-900°C (this temperature would not exceed 1000°C due to the structure of the reactor). When temperature in the oxidation zone is lower than recommended, carbon dioxide is not completely reduced to oxide. If the temperature is above recommended value, ash is melting and slag is forming, causing suspension of the charge and, as a result, limitation of flow of gas [6]. The pressure inside the reactor was controlled by elastic pressure gauge on the outlet of the reactor and initially set with the use of a pressure regulator. In order to increase fluidization in the reactor, constant blow through was performed with the use of a piston pump forced pressure 0,1 MPa. Aerogel was applied as insulation.

3 The results of the research

Coal gasification for its various types, at constant parameters specified in point 2.2 were analysed on the laboratory scale during experimental research. The research was conducted for various times of hydrogen gasification. Three measurements for various time intervals were taken, that is, 15 minutes, 30 minutes and 60 minutes. The tests were performed for charcoal, hard coal and lignite coal and their mixtures, with constant blow through, temperature was between 800 °C and 950 °C, pressure was 1,2-1,3 MPa. The results of the research are presented in Tab. 3 and Fig. 8. Relative amount of gasified coal was calculated using dependencies 1 and 2.

$$\Delta mk = mp - mk \tag{1}$$

$$G\% = \frac{\Delta mk}{mp} \tag{2}$$

where :

Δmk	–	absolute amount of gasified coal, kg,
mp	–	initial mass of coal (before gasification), kg,
mk	–	final mass of coal (after gasification), kg,
G%	–	relative amount of gasified coal.

Charcoal, meeting the norm of German Committee for Standardization DIN EN1860 2 with a number 3H020, as well as hard coal and lignite coal were applied during the research. Description and content of elements in charcoal are presented in table 1 and table 2. The coals for research gasification were dried and grinded. Technical hydrogen produced by Linde of 5.0 (99,999%) purity were used as a gasifying factor.

Table 1. Technical analysis of the sample of charcoal

Element	Contents [% by weight]
Mg	0.018 ± 0.002
Al	0.0026 ±0.0006
Si	0.0167 ± 0.0020
P	0.0136 ± 0.0015
S	0.0139 ± 0..0007
K	0.176 ± 0.005
Ca	0.133 ± 0.004
Ti	0.016 ± 0.002
Mn	0.0083 ± 0.0015
Fe	0.0197 ± 0.0025
Cu	0.00064 ± 0.00020
Zn	0.00057 ± 0.00015
Rb	0.00042 ± 0.00013
Sr	0.0011 ± 0.0003

Table 2. Elemental analysis CHN

Sample name	%C	%H	%N
P-LA-79/2016/Z – charcoal - A	average: 77.435	average: 3.45	average: 0.49

During gasification of hard coal, slight relation between gasification and process duration has been observed. The difference between the measurements for various times was about 2%. After 15 minutes of the process, gasification process at the average level of about 48% was obtained. The lowest standard deviation was obtained for the process lasting 60 minutes, the process was repeatable for this value. In the event of mixture of hard coal and charcoal in the proportion 50% to 50%, moderate relation between gasification and process duration has been observed. Between 15-minute and 30-minute and 60-minute measurements, there was an absolute difference at the level of about 8-9%. In this case, the lowest standard deviation was obtained for the process duration of 15 minutes, which proves that the process was most repeatable then. The relation between gasification and process is small in the case of lignite coal. After 15 minutes of the process, gassing process at the average level of about 59% was obtained. Standard deviation for all three times was constant and slight, which may show high repeatability of the process. In the case of mixture of lignite coal and hard coal, gassing process at the average level of about 55% was obtained after 15 minutes. The highest results for carbon mixture were achieved for 15 minutes, and then for 30 minutes, however, the most repeatable process with the lowest standard deviation was achieved for the process that lasted 60 minutes. In the case of charcoal and its mixture with lignite coal, slight relation between gasification and process duration has been observed. After 15 minutes of the process, gassing process at the average level of about 39,18% was obtained. The lowest standard deviation was obtained for the process that lasted 30 minutes.

Table 3. Averaged percentage values of gasified samples, minimum, maximum and standard deviation for all types of coal

Hard coal	**MIN**	**MAX**	**AVERAGE**	**STANDARD DEVIATION**
15 minutes	42.92 %	53.92 %	47.89 %	0.055760829
30 minutes	43.46 %	53.14 %	48.23 %	0.048416664
60 minutes	44.64 %	53. 04%	50.06 %	0.046990247
			48.72 %	
Hard coal and charcoal	**MIN**	**MAX**	**AVERAGE**	**STANDARD DEVIATION**
15 minutes	34.01 %	39.13 %	35.76 %	0.029165
30 minutes	42.02 %	49.47 %	44.81 %	0.040618
60 minutes	39.84 %	46.75 %	43.00 %	0.034917
			41.19 %	
Lignite coal	**MIN**	**MAX**	**AVERAGE**	**STANDARD DEVIATION**
15 minutes	58.19%	60.90 %	59.26%	0.0144411
30 minutes	56.57 %	60.83%	58.91%	0.0216083
60 minutes	60.81 %	62.85 %	61.52%	0.0115539
			59.89 %	
Lignite coal and hard coal	**MIN**	**MAX**	**AVERAGE**	**STANDARD DEVIATION**
15 minutes	51.69 %	58.45 %	55.03%	0.0338083
30 minutes	50.49 %	56.97 %	52.75%	0.0365503
60 minutes	47.30 %	53.70%	50.65%	0.0299377
			52.81%	
Lignite coal and charcoal	**MIN**	**MAX**	**AVERAGE**	**STANDARD DEVIATION**
15 minutes	47.62%	52.43%	50.03 %	0.0340118
30 minutes	48.65 %	52.26%	50.46%	0.0255266
60 minutes	58.99%	58.99%	58.99%	0.0210526
			53.16%	
Charcoal	**MIN**	**MAX**	**AVERAGE**	**STANDARD DEVIATION**
15 minutes	35.83 %	49.27 %	39.81 %	0.064108
30 minutes	38.93 %	41.70 %	40.10 %	0.012858
60 minutes	37.91 %	49.60%	41.91 %	0.052563
			40.61 %	

The research presented above shows that the coal that is gasified to the largest extent is lignite coal, which may result from its properties and chemical composition. This type of coal has the highest repeatability of the measurements for each time interval. The mixtures with lignite coal also achieved high degree of gasification above 50%. Hard coal is also a good raw material for gasification. It has been observed that coke was formed in the filling sieves after every measurement, which may have impact on low losses during the research.

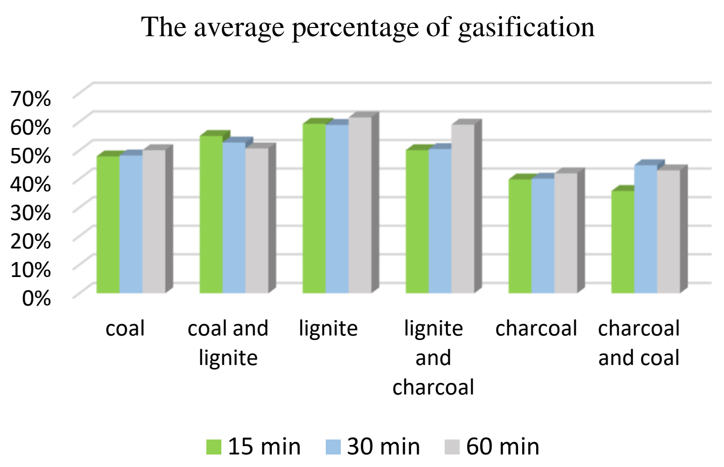

Fig. 8. Relative amount of gasified coal for various types of coal, process duration 15 min, 30 min, 60 min

The results were repeatable at constant level of about 48%. Apart from repeatability of the results, the mixture of hard coal and lignite coal was characterized by high temperature during the measurement, which results from fusion of coals of similar physical properties (temperature exceeded 950 degrees two times). Charcoal is characterized by the lowest loss of initial mass of coal at the level of about 40%. During the research on charcoal and its mixtures, it was difficult to achieve the temperature of 800 degrees. Time of conducting the research only slightly affects the value of gasification mixtures - at the level of a few per cent. Gas formed during coal gasification contains H_2, CO, CH_4, NO, NO_2, CO_2, CH_4, as well as side tarry substances.

The average value of gasification of carbon mixtures was obtained in conducted research, understood as relative loss of initial mass of coal (Fig. 9):

- lignite coal 59,89 %,
- the mixtures of lignite coal and hard coal 52,81 %,
- the mixtures of lignite coal and charcoal 53,16%,
- hard coal 48,72%,
- the mixtures of hard coal and charcoal 41,19 %,
- charcoal 40,61%.

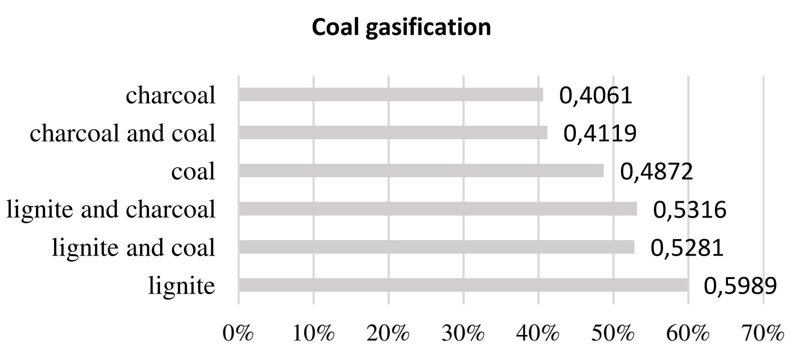

Fig. 9. Relative loss of initial mass of coal during gasification

4 Conclusion

Coal gasification is a controlled process, characterized by high efficiency. Its advantage is the fact that it reduces emission of pollutants from combustion process, which makes this process environmentally friendly. One of the products of coal gasification in the presence of hydrogen is methane, depending on the source of origin, it can be biomethane. It can be applied for production of electric and thermal energy, but also as a renewable motor fuel, which is characterized by high efficiency. Conducted research showed that temperature slightly affects the process of gasification, however, in order to increase the share of burnable components in the reactions, we must accept minimal temperature of 800 degrees. Lignite coal is characterized by the highest reactivity in comparison with hard coal or charcoal, achieving coal gasification at the level of nearly 60% after 15 minutes of experimental research. The use of hydrogen as a gasifying factor seems to be a prospective direction of development of the process of coal gasification. The works on coal gasification in the presence of hydrogen and the works on higher deposits of coal should be continued. It will cause many problems, mainly rinsing the deposit with hydrogen.

References

1. Bigda J., Burchart-Korol D., Porada S., Chart of technology solutions for coal gasification processes. Przegląd górniczy, pp. 86 – 96, (2014)
2. Borkiewicz M. Zgazowanie węgla wodór nośnikiem energii. Biuletyn Górniczy, **5-6**, pp. 155-156, (2008)
3. Chmielniak T., Sobolewski A., Tomaszewicz G., Zgazowanie węgla przy wykorzystaniu CO2 jako czynnika zgazowującego. Doświadczenia IChPW. Przemysł Chemiczny **94,** 4, pp. 442-448, (2015)
4. Chmielniak T., Bigda J., Czardybon A., Popowicz J., Tomaszewicz G., Technologie oczyszczania gazu ze zgazowania węgla. Przemysł Chemiczny, **97**, 2, pp. 1000÷1010, (2014)
5. Chmielniak T., Stelmach S., Współczesne technologie zgazowania węgla, **13**, 2 p. 69-76, (2009)
6. Gawlik L., Mokrzycki W., Paliwa kopalne w krajowej energetyce – problemy i wyzwania. Energy Policy Journal, **20**, 4, pp. 6-26, (2017)
7. IEA Publications 2014. World Outlook EIA
8. Kamble AD., Saxenaa VD., Chavan PD., Mendhe VA., Co-gasification of coal and biomass an emerging clean energy technology: Status and prospects of development in Indian context. International Journal of Mining Science and Technology, (2018)
9. Kezhong L., Zhangand R., B and J., Experimental study on syngas production by co-gasification of coal and biomass in a fluidized bed. Int J Hydrogen Energy, **35**, 7, pp. 2722–2726, (2010)
10. Kumabe K., Hanaoka T., Fujimoto S., Minowa T., Sakanish K., Co-gasification of woody biomass and coal with air and steam. Fuel, **86**, 5-6, pp. 684–689, (2007)
11. Minchener A. J., Coal gasification for advanced power generation. Fuel, **84**, pp. 2222-2235, (2005)
12. Porada S., Otrzymywanie substytutu gazu ziemnego na drodze hydrozgazowania węgla. Karbo Energochemia Ekologia, **5**, 186-188, (1996)
13. Smoliński A., Howaniec N., Badania procesu zgazowania węgla kamiennego w obecności K_2CO_3 Research reports mining and environment, **1**, pp. 81- 92, (2012)