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RESEARCH

Use of coals and wastes in a co-gasification process aimed at producing hydrogen rich gas

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Abstract

The use of low-quality coals and flotoconcentrates is currently severely limited, and the problem of managing municipal waste from anthropogenic activities is currently a challenge. The problems of reducing carbon dioxide emissions, utilizing the energy potential of waste and increasing its recycling have an impact on the costs of electricity production. Considering the abundant streams of unused fuels, they can be considered as attractive energy materials, so environmentally-friendly and cost-effective options for their utilization should be developed. A study was conducted using steam co-gasification technology on selected coals, flotation concentrates and Refuse Derived Fuel (RDF) alternative fuel. Selected low-quality coals were combined with RDF alternative fuel in a process aimed at hydrogen production. The experiments produced gas with hydrogen concentrations ranging from 67% (vol.) to 68% (vol.) with low methane concentrations. It was observed that the addition of alternative fuels helped to increase the hydrogen concentration in syngas. Attention was paid to the catalytic ability of the metal oxides contained in the fuel blend, with particular reference to K_2O and Al_2O_3 and TiO_2.

Keywords Coal · Hydrogen · Flotation concentrate · Refuse derived fuel · Catalytic capacity · Catalysts

1 Introduction

The economic attractiveness of low-quality coals and flotoconcentrates is a challenge, with the problem of managing municipal waste from the wide range of human activities also currently posing a clear challenge (Jingchao et al. 2019; Gregson and Crang 2015). The production of alternative fuels is dictated by environmental considerations, but also by economics (Yang et al. 2021; Sarquah et al. 2022). One should bear in mind the reduction of carbon dioxide emissions, the use of the energy potential of waste and increasing its recovery in circulation, and the reduction of energy production costs (lower cost of obtaining alternative fuel in relation to the cost of conventional fuels). Considering the above components as an abundant fuel stream, they can be considered as attractive energy raw materials. Therefore, environmentally-friendly and economically-viable options for their use should be developed.

In this paper, selected coals, flotoconcentrates and Refuse Derived Fuel (RDF) alternative fuels were examined for their steam gasification. The commercialization of coal gasification plants is one of the methods for the proper utilization and processing of low-quality fuels.

Gasification technology is currently being developed into three main technologies ConocoPhillips (E-Gas), SES technologies and Shell technologies. The basic element of a gasification plant is the reactor in which thermochemical transformations take place, with a distinction being made between fixed bed, fluidized bed and flow bed reactors (Mahinpey and Gomez 2016). Gasification plants are used in biomass processing (Howaniec and Smolinski 2017), polyethylene processing (Jeong et al. 2020) or municipal waste (Singh et al. 2020).

This technology enables proper utilization of the energy potential of fuels. The requirements of commercial coal gasification technology solutions are mainly the appropriate determination of the parameters of the feedstock fuel, as well as the determination of the parameters of the fuel processing (gasification), which must be determined in order

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to direct the processing of the feedstock to obtain the desired output product.

In the research presented in this paper, analyses of low-quality coals including flotation concentrates were performed, taking into account their purpose in steam co-gasification installations with RDF energy waste. Models have been developed to study the similarities and differences between the examined fuels, as well as the relationship between physical parameters and the catalytic abilities of the oxides contained in the fuel that can be managed in the steam gasification process.

Low-quality coals were selected, blended with RDF alternative fuel at 5% (m/m). and subjected to experimental co-gasification. The study focused on the production of hydrogen-rich gas, which is a clean, environmentally-friendly energy carrier that can be used in power generation and transportation. The experiments produced gas with hydrogen concentrations ranging from 67% (vol.) to 68% (vol.). Analysis of the results was performed using chemometric tools with MATLAB and MS Excel software. The obtained results were traced using the hierarchical clustering method. The correlation of oxide composition in the production process of hydrogen obtained in syngas was considered. It was observed that the addition of alternative fuels contributed to the increase in hydrogen concentrations. The catalytic ability of the metal oxides contained in the fuel was observed. The catalytic properties are exhibited by the individual oxides in the coal fuel and the alternative fuel, with particular emphasis on K_2O, Al_2O_3 and TiO_2 supplied to the gasified fuel mixture along with RDF. The desired hydrogen production yields were obtained with 5% (m/m). RDF at a gasification temperature of 900 °C. An increase in temperature caused a decrease in carbon dioxide concentrations, while methane concentrations were low regardless of the temperature in the reactor, which may indicate the disappearance of the hydrogen gasification and methanation reaction due to the influence of K_2O and Al_2O_3, TiO_2 catalysts, through more efficient processing of tar and soot.

A systematic approach to the processing of hard-to-recycle raw materials confirms the direction of waste management and utilization in an energy-efficient manner, while maintaining the principles of minimizing the impact of human activity on the surrounding environment with the use of steam gasification technology. The selection of the fuel mixture requires a thorough analysis, which indicates a number of relationships between fuel quality and physicochemical parameters. Further research directions require the determination of an appropriate proportion of RDF alternative fuel, taking into account the correlation of oxide composition in the studied samples, to obtain the most beneficial effect on hydrogen production when co-gasifying with low-quality fuel and flotoconcentrates.

The basic premise of coal gasification technology is to conduct the process in such a way as to produce a synthesis gas suitable for a specific application, such as one with the highest possible composition of methane or hydrogen (Speight 2014). Hydrogen as a clean environmentally-friendly energy carrier obtained from coal and waste processing technology can become an attractive solution for the management of these products, realizing the assumptions of the circular economy cycle, which has been addressed in a number of scientific studies (Miandad et al. 2017; Rehan et al. 2017; Howaniec and Smolinski 2017; Moghadam et al. 2013; Mazloomi and Gomes 2012; Conte et al. 2009). The papers discussed biological and chemical means of hydrogen production or, as addressed in the study by Rehan et al., the energy potential of waste generated in Medina located in Saudi Arabia (the amount of waste generated is an energy potential of 74.45 MW of electricity equivalent). Howaniec et al. recommended the utilization of bio-waste by co-gasification with coal and lignite at 700 °C and 900 °C. They showed the dependence of the increase in the coal conversion rate in co-gasification increased with the increase in biomass content in the fuel. Moghadam et al. studied the gasification in a fluidized bed reactor at 650 °C to 1000 °C of a mixture of waste polyethylene with biomass, which promoted an increase in hydrogen production. The mixture of 30% polyethylene with biomass feedstock increased the hydrogen content of syngas to 84.71%. Mazloomi et al. demonstrated the potential for hydrogen as a key energy carrier and fuel source. They also touched on hydrogen as energy storage, indicating that, by the year 2050, the demand for this carrier will be over 42 million tonnes in the United States alone. Conte et al. (2009) touched on aspects related to the role of hydrogen in the functioning of society in the future. They pointed out that hydrogen can be used in the future, considering the radical changes in the way energy is perceived in human life.

Adaptation of Texaco technology targeting the production of hydrogen-rich gas from waste materials was studied by Wallman et al. (1998) and Kim (2003). In work focused on the production of hydrogen-rich gas, Kim (2003) dealt with the topic of utilizing waste materials such as used tyres and oils. The production of hydrogen-rich syngas was also the topic of He et al. (He et al. 2009a, b, c). The work presented the possibility of using municipal waste in the steam gasification process. The problem of waste management was also the subject of the works of Wang et al. (2019) and Zheng et al. (2018). The topic of co-gasification of low-quality coals with wastes from coal enrichment processes (silts, flotoconcentrates), and also low-quality coals with RDF alternative fuels in a process aimed at obtaining synthesis gas with a significant proportion of hydrogen, requires continued research.

2 Results and discussion

Co-gasification studies were carried out in a laboratory installation with a fixed bed reactor at the Central Mining Institute. The fixed bed reactor with a working volume 800 cm^3 could work with maximum temperature 900 °C. The reactor was heated by a double-sphere resistance furnace. The gasifying medium was injected at the bottom of the reactor. The scheme of the installation is presented in Fig. 1.

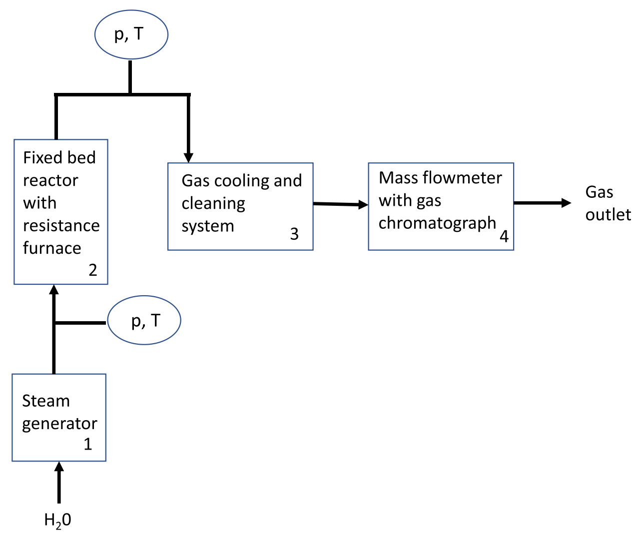

Fig.1 Scheme of the gasification installation: (1) gasification agent supply with steam generator, (2) fixed bed reactor with resistance furnace, (3) gas cooling and cleaning system, (4) gas chromatograph (GC) with a flow rate measurement system

Co-gasification of coals, flotoconcentrates and RDF was performed on 3 g samples in an analytical state. Fourteen coal samples from the Upper Silesian Coal Basin were dried and ground to grains below 0.2 mm and heated in an inert nitrogen atmosphere at a heating rate of 10 °C/min. until temperatures of 800 °C and 900 °C were reached. Steam gasification of the samples was carried out at a flow rate of 3.2 mL/min.

In order to characterise the research samples of coal materials from fourteen mines in the Upper Silesian Coal Basin and one RDF sample (sample No. 15), physical and chemical analyses of the fuel were carried out in the Solid Waste Analyses Laboratory of the Department of Environmental Monitoring GIG, in accordance with Polish standards including: transient moisture content (PN-G-04511:1980), total moisture content (IC-29.1 edition 16 25.05. 18), ash content (PN-G-04560:1998), volatile matter content (PN-G-04516:1998), combustion heat (PN-G-04513:1981), calorific value (PN-G-04513:1981), sintering capacity according to Roga (PN-G-04518: 1981), free smoke index (PN-ISO-502:2007), sulphur content (PN-G-04584:2001), carbon content (PN-G-04571:1998), hydrogen content (PN-G-04571:1998), nitrogen content (PN-G-04571:1998), and oxygen content (IC-29. 1st edition 16 25.05.18). The results are shown in Tables 1 and 2.

Analysing the results of the physical parameters of the fuels in Table 1 in relation to the estimation of coal quality, i.e. its energy parameters in the working state, the moisture content is significant, ranging from 1.17% to 9.70%. Based on the moisture content, it can be concluded that samples 5 to 9 come from coal mines that are failing. Another

Table 1 Physical parameters

No.	Moisture (%)	Ash (%)	Volatile parts (%)	Combustion heat (kJ/kg)	Calorific value (kJ/kg)
1	1.32	2.66	32.18	33,697	32,506
2	1.39	2.81	30.23	33,604	32,531
3	1.74	4.47	27.38	32,745	31,663
4	2.25	15.49	26.50	27,705	26,744
5	9.70	6.66	31.69	26,264	25,115
6	9.40	10.82	29.33	24,938	23,820
7	7.77	9.52	30.91	26,076	24,984
8	7.50	7.85	31.67	27,170	26,066
9	5.79	5.48	35.10	28,914	27,744
10	1.65	4.64	27.73	33,166	32,037
11	1.60	1.86	33.54	33,652	32,476
12	1.85	2.05	28.42	33,484	32,369
13	1.53	18.24	32.15	27,907	26,881
14	1.17	1.66	31.31	34,923	33,770
15	1.99	15.69	75.07	25,730	24,267

Table 2 Elemental content parameters

No.	S (%)	C (%)	H (%)	N (%)	O (%)
1	0.29	82.19	5.31	1.52	6.89
2	0.31	83.62	4.76	1.35	5.94
3	0.42	80.16	4.76	1.35	7.28
4	0.51	69.71	4.15	1.10	6.92
5	1.59	67.61	4.18	0.99	9.70
6	1.83	64.27	4.07	0.95	9.06
7	1.67	67.32	4.13	0.99	8.72
8	0.90	68.14	4.22	1.02	11.09
9	0.77	71.52	4.71	1.08	11.17
10	0.51	80.20	4.99	1.43	6.73
11	0.31	82.53	5.21	1.50	7.17
12	0.30	82.44	4.90	1.48	7.18
13	0.84	69.97	4.53	1.27	3.92
14	0.27	85.59	5.15	1.48	4.74
15	0.26	56.18	6.88	0.88	N.A

Note: N.A. refers to not applicable

important parameter is the ash content which, in the samples tested, ranged from 1.66% to 18.24%.

The selection of fuel (coal, flotation concentrate) for the co-gasification process with RDF alternative fuel was preceded by the analysis of physical parameters of ashes and their oxide composition. The analysis of ash fusibility temperature was performed in the Solid Waste Analysis Laboratory of the Department of Environmental Monitoring GIG, in accordance with the PN-G-04535:1982 standard; with the results presented in Table 3.

Then, a correlation study of eight ash melt temperature parameters was also performed using MATLAB Simulink software, as shown in Table 4.

Table 4 Matrix of physical parameters of tested samples

No.	Parametr	Jednostka
1	Ash sintering temperature (oxid.)	°C
2	Ash sintering temperature (red.)	°C
3	Ash softening temperature (oxl.)	°C
4	Ash softening temperature (red.)	°C
5	Ash melting point (oxl.)	°C
6	Ash melting point (red.)	°C
7	Ash melt temperature (oxl.)	°C
8	Ash melt temperature (red.)	°C

The results of the correlation are shown in Fig. 2. In our study the Pearson correlation was used to evaluate whether there is a statistical evidence for a linear relationship among the studied parameters. The absolute value of Pearson correlation coefficients are between -1 and 1. Values -1 and 1 mean that the negative or positive correlation between studied parameters is observed, respectively. Based on the analysis performed, it was found that there is a correlation between the physical parameters of ash melting temperature 3–4, 4–6, 5–6, 5–7, 5–8, 6–7, 6–8, 7–8.

Based on the analysis of physical parameters of fourteen coal samples and finding the correlation between these parameters, the analysis of the oxide composition of ash of coal samples and one sample of RDF alternative fuel was undertaken. The analysis was performed according to ISO/TS 13605:2012 using Wavelength Dispersive X-ray Fluorescence Spectrometry. The metal oxide composition of SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, Na_2O, K_2O, SO_3, TiO_2, and P_2O_5 was determined, and the results are shown in Table 5.

Table 3 Ash melting points (°C)

No.	Sintering		Softening		Melting		Melt	
	Oxid	Red.	Oxid	Red.	Oxid	Red.	Oxid	Red.
1	1040	960	1340	1280	1350	1320	1400	1360
2	1020	950	1250	1180	1300	1200	1360	1230
3	960	920	1220	1160	1280	1230	1320	1300
4	970	890	1260	1220	1490	1430	1500	1480
5	930	880	1300	1130	1360	1160	1400	1180
6	940	910	1270	1170	1300	1220	1330	1280
7	970	900	1330	1280	1460	1420	1500	1450
8	950	1140	1350	1330	1360	1360	1380	1360
9	950	1140	1370	1350	1380	1370	1400	1380
10	1050	1000	1270	1230	1320	1310	1360	1340
11	1020	960	1340	1300	1380	1360	1420	1370
12	1100	1050	1350	1330	1370	1360	1420	1370
13	990	940	1380	1350	1460	1440	1480	1460
14	1070	980	1290	1240	1350	1300	1430	1340

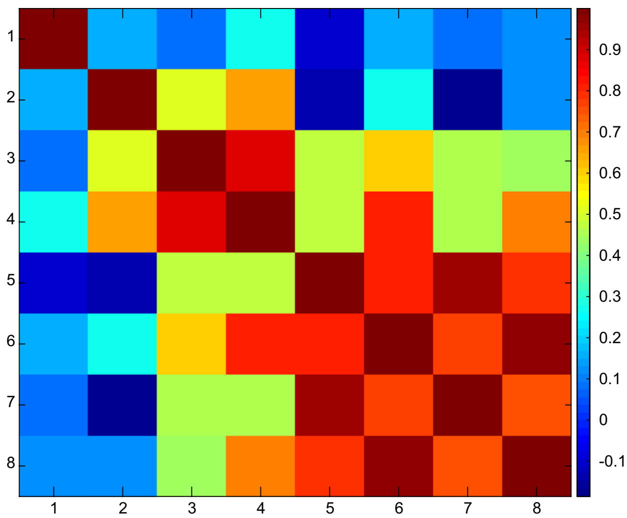

Fig. 2 Correlation of ash fusibility temperature

In order to fully interpret the obtained results of the analysed samples, the obtained data was organized in an X matrix (15 × 10) according to Table 3, then cluster analysis, also called Hierarchical Clustering Analysis (HCA), was applied. As a result of the HCA method, similarities between the studied objects (samples) in the parameter space and parameters in the object space were tracked (Smoliński 2011). In the organized matrix, rows were formed by the fifteen studied objects (research samples) and columns were formed by the ten studied parameters of the oxide composition of ashes. Then, hierarchical grouping of the studied samples (objects) in the space of the measured parameters and hierarchical grouping of parameters in the space of objects were performed. Further analysis was extended with a colour similarity map, which enabled the interpretation of the data to be broadened by indicating similarities and differences of the grouped data represented in the form of dendrograms.

Graphical representation in the form of a dendrogram (Fig. 3) shows on the abscissa axis the order of grouping of fifteen particular objects. While on the ordinate axis it defines the degree of their similarity. Graphical representation of hierarchical cluster analysis simplifies the analysis of clustering ability of individual objects. However, it is not sufficient to simultaneously determine the relationships between a given object (studied sample) and its ten parameters of oxide composition.

To facilitate further analyses of object similarities in the space of the measured parameters. a colour map of the studied data was made using MATLAB Simulink software (Fig. 4). The analysis focused on sample 4—flotation concentrate. sample 7—coal from a mine with increased unreliability. and sample 15—alternative fuel.

Samples 4, 7 and 15 with respect to the percentage of aerobic compounds in their ash showed a tendency to cluster within one cluster B (Fig. 2). These samples show low levels of parameters 5, 8 and 10 (MgO, SO_3, P_2O_5). which can be observed in Fig. 3. Analysing separately only objects 4 and 7 which are samples produced in a coal mining company, parameters 1, 2 and 4 (SiO_2, Al_2O_3, CaO) are similar. For object 4, there is a significantly higher level of parameter 7 (K_2O) while for object 7 it is parameter 2 (Al_2O_3). Considering object 15 (RDF) separately, the alternative fuel shows high contents of parameters 1, 4 and 9 (SiO_2, CaO. TiO_2) with parameter 9 (TiO_2) reaching the highest value among all the tested samples. The correlation of the oxide composition of ashes in the co-gasification process was shown

Table 5 Oxide composition of ashes in Sample No. 15- RDF

No.	Parameter (%, m/m)									
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3	TiO_2	P_2O_5
1	16.60	17.47	13.38	18.54	10.29	2.65	1.28	15.38	0.71	1.48
2	22.93	19.53	13.62	15.55	6.88	2.86	0.84	13.58	0.61	1.71
3	36.06	21.13	9.28	9.89	4.91	3.69	2.01	9.29	1.72	0.54
4	49.56	29.82	4.50	2.87	2.07	2.11	3.69	1.53	1.99	0.83
5	24.75	18.56	23.05	10.72	3.95	4.14	1.17	12.00	0.61	0.08
6	32.55	24.77	16.43	6.10	3.52	5.56	1.21	7.05	1.80	0.06
7	40.18	32.26	13.18	2.69	1.23	5.02	0.81	1.97	1.75	0.07
8	30.91	23.28	16.47	8.57	4.95	4.34	0.61	8.58	1.01	0.06
9	11.65	10.90	13.21	22.90	11.10	2.91	1.72	22.78	0.57	0.77
10	22.94	25.14	8.83	14.06	4.32	3.38	0.45	9.06	0.51	8.60
11	11.62	9.56	20.07	18.90	9.97	3.24	0.70	23.85	0.30	0.16
12	15.50	10.01	17.36	18.30	8.69	2.80	2.62	22.65	0.27	0.19
13	45.81	30.90	4.95	4.29	3.17	1.04	3.08	3.74	1.18	0.77
14	26.35	27.10	15.71	9.02	4.47	2.53	3.06	8.50	0.74	0.29
15	45.22	13.41	6.06	17.15	2.01	3.02	2.05	6.79	2.15	1.23

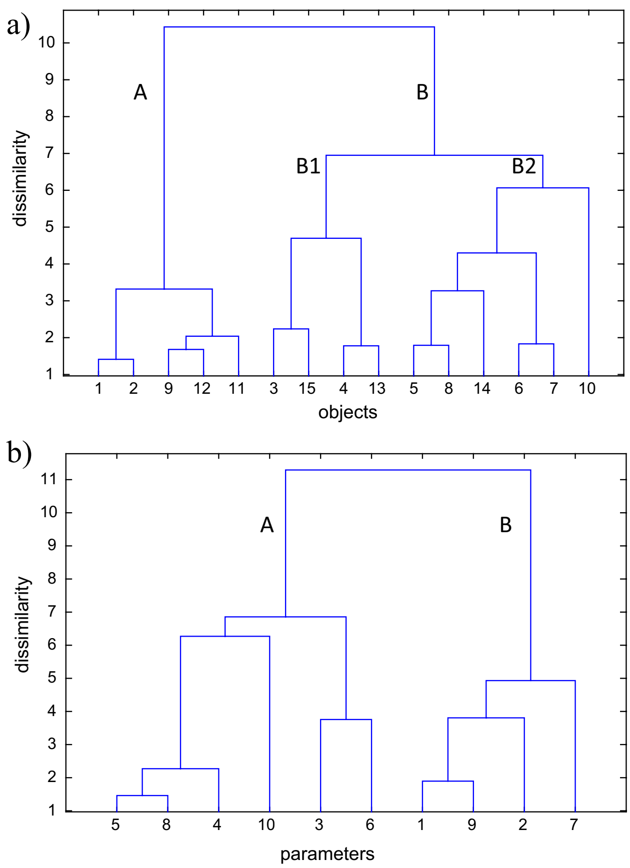

Fig. 3 Dendrograms of HCA cluster analysis. **a** Clustering of objects in the parameter space; **b** Clustering of parameters in the feature space

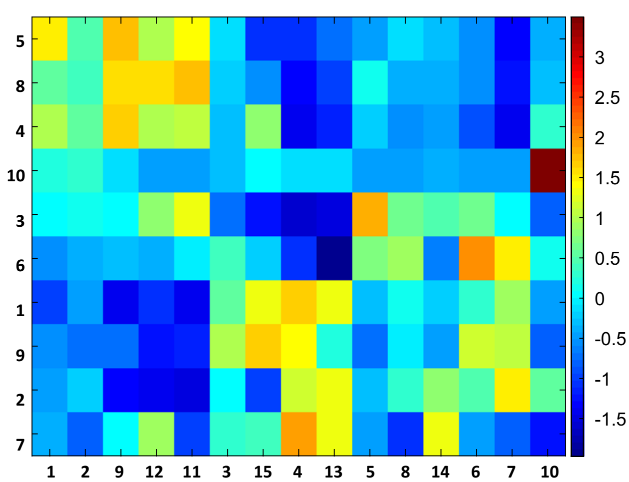

Fig. 4 Colour interpretive map of the oxide composition of ash

in the work of He et al. (He et al. 2009a, b, c). It was also confirmed by studies in the work of Howaniec and Smoliński (2014a; b) that particular oxides with catalytic properties are the metal oxides Al_2O_3, Fe_2O_3, Na_2O, K_2O and TiO_2.

Based on the analyses carried out tests were conducted on co-gasification with water vapour of samples 4 (flotoconcentrate) and 7 (coal from the coal mine with increased unreliability) with RDF alternative fuel. The tests were carried out in a laboratory installation with a solid bed located at the Department of Energy Saving and Air Protection of GIG. Co-gasification was performed at a temperature of 900 °C of a sample in an analytical state containing 5% (m/m) RDF alternative fuel. was heated in an inert nitrogen atmosphere and then exposed to the gasification agent at a flow rate of 3.2 mL/min. The results of the steam co-gasification are illustrated in a graph (Fig. 5).

In the synthesis gas produced, the concentration of hydrogen reached a maximum of 68% vol. while the proportion of methane reached a minimum of 1% vol., which confirms the catalytic influence of the metal oxides present in the fuel. Considering the examined fuel blends in terms of the catalytic properties of the metal oxides present in them. significant catalytic effects of individual oxides in a given fuel group were observed with particular emphasis on K_2O and Al_2O_3. Also important is the contribution of TiO_2, which is supplied to the gasified fuel mixture in the form of added RDF. The contribution of individual oxides contributes to the interaction in the co-gasification of fuel blends. confirming the synergy of catalytic properties in the co-gasification process.

3 Conclusions

Investigations of the co-gasification of coals and flotoconcentrates with the participation of 5% (m/m) RDF at 900 °C confirmed the possibility of obtaining a gas with hydrogen content above 60% vol.

The in-depth analysis of the physical parameters of fourteen coal samples and finding the correlation between these parameters, the analysis of the oxide composition of ash of coal samples and one sample of RDF alternative fuel was undertaken.

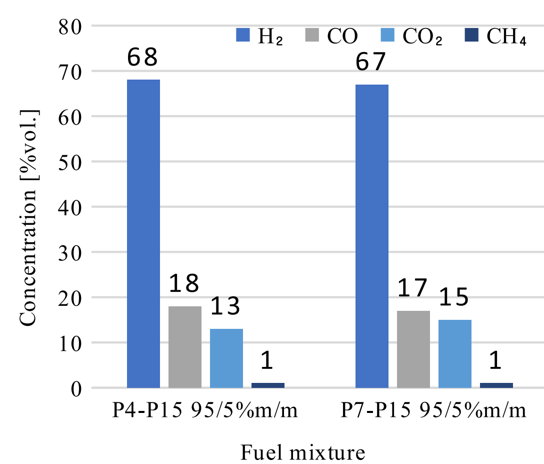

Fig. 5 Gas concentrations in the co-gasification process

The catalytic potential of metal oxides contained in the fuel mixture was observed.

Particular catalytic properties are indicated by the oxides contained in the fuel group with particular emphasis on K_2O and Al_2O_3 as well as TiO_2. which is supplied to the gasified fuel mixture in the form of added RDF.

As a result of the catalytic ability of K_2O, Al_2O_3 and TiO_2. there is an increase in hydrogen generation. which may indicate the disappearance of the hydrogasification and methanation reactions in the steam co-gasification process. through more efficient processing of tar and soot.

Declarations

Conflict of interest The authors declare no conflict of interest. This research received no external funding.

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