Hungarian Materials and Chemical Sciences and Engineering, Volume 47, No. 1 (2023), pp. 5–15.
http://doi.org/10.32974/mse.2022.001

COAL CHAR STEAM GASIFICATION OF THREE DIFFERENT HUNGARIAN COAL TYPES

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Abstract: In this study, there were two brown coal sample and a black coal sample gasified in a down draft fixed bed reactor at 800 and 900 °C, within 10 g/min of steam flow rate in all experiments. The aim of this research is the investigation of gasification process using Hungarian brown coal char and back coal char samples from pyrolysis process in the distribution of dry gas yields, as well as the synthesis gas composition. Based on the gasification experiments, the brown coal samples showed a potential utilisation for the gasification process. In the case of brown coal samples, the gasification time was shorter at the higher gasification temperature. Using steam as reactant in the gasification process generated a significant quantity of gas with a high hydrogen concentration, which may be suitable to produce secondary raw materials, as methanol.

Keywords: thermochemical processes, gasification process, coal gasification, synthesis gas

INTRODUCTION

Presently, coal gasification is the most interesting process among thermochemical process (combustion, pyrolysis and gasification process) because it allows the produced synthesis gas (mainly including H_2, CO, CO_2 and CH_4) to use in various fields as electricity and heat production, chemical production, or transportation fuel [1]–[3]. Coal gasification process is used to convert C-content into synthesis gas at elevated temperature with the presence of gasifying reactants.

In general, gasifiers can be classified into three configurations, entrained flow, fluidized bed, and fixed bed (moving bed) type. The fixed bed gasifier seems to be more suitable for the small decentralized combined heat and power plants due to its simple construction, low operation cost, low syngas exit temperature, and high flexibility of feedstock materials [4].

In the gasification process, the air, steam, oxygen, and their combination can be used as gasifying agents. The selectivity of the gasification reactions varies with different gasifying agents, thus affecting the composition and low heating value of produced gas [5]. Air is the most popular media used in large scale gasification systems. It is the cheapest option, although the produced gas will have a high concentration of nitrogen, thus significantly decreases the heating value of the gas as well as limits the option of further use in chemical processes. Using oxygen leads to high operational cost and safety risks. Using steam as a gasifying agent requires separate heat introduction in the system, as the main reactions within the process are endothermic. Therefore, the utilisation of external heat source or partial combustion process is necessary. In general, the mixture of steam and air/oxygen is the most commonly used.

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In the case of Hungarian low-rank coal gasification, the relative works are quite rare. A. Pettinau et al. [6] studied the gasification process of Hungarian brown coal in both lab-scale and pilot-scale gasifiers. L. Bokányi and Á. Pintér-Móricz [7] researched the potential methanol-ethanol synthesis process from the plasma gasification of the sub-bituminous Hungarian coal sample. There is a clear gap in the description of the gasification process for coal samples from the different coal-mining areas in Hungary.

The main aim of this research is the investigation of gasification process using Hungarian brown coal and back coal samples as starting material in the distribution of dry gas yields, as well as the synthesis gas composition. In addition, the effect of gasification temperature is considered in the coal samples.

1. MATERIAL AND EXPERIMENTAL APPARATUS

1.1. Material

In this study, there were three different coal samples used for the steam gasification process. They were collected from different coal basins in Hungary, including brown coal from Felsőnyárád (FC) and Bükkábrány (BC) basin, and black coal from Pécs (PC) basin. The results of elemental analysis and proximate analysis is illustrated in *Table 1*.

The elemental composition of coal was examined under the standard ISO 29541:2010 Solid mineral fuels – Determination of total carbon, hydrogen, and nitrogen content – Instrumental method with a Carlo Erba EA 1108 equipment analyser. The results of proximate analysis were collected from the thermal analysis of coal samples. Thermal analysis was performed in a MOM Derivatograph-C type, with nitrogen ambient.

Table 1

Elemental and proximate analysis of coal samples

Elemental analysis in dry samples [wt%]					
	N-nitrogen	C-carbon	H-hydrogen	S-sulphur	O-oxygen
BC	0.42	45.61	3.46	3.31	22.20
FC	0.37	48.06	3.64	5.54	16.08
PC	0.23	29.37	2.13	1.15	5.71
Proximate analysis in air-dry samples [wt%]					
	M-moisture	V-volatile	FC-fixed carbon	A-ash	
BC	9.5	28.6	36.9	25.0	
FC	2.2	31.0	41.4	25.4	
PC	0.2	11.5	26.9	61.4	

1.2. Experiment apparatus

The schematic of lab-scale single stage gasification process is depicted in *Figure 1*. In every experiment 3 kg of coal sample placed in the downdraft gasifier except in the case of the coal sample from Pécs as its high density allowed a 5 kg charge for the experiment. The particle size was from 10 to 20 mm in all experiments. When the pyrolysis process was finished at the initial set-up temperature of electrical heater, the steam was introduced for reduction process at ambient pressure. A venturi scrubber following the gasifier was used to remove the tar and particle matters from the synthesis gas, and to cool down the synthesis gas. The cotton filter was set at the synthesis outlet of venturi scrubber to capture the residual tar matter in the synthesis gas before entering the gas meter and analysing the gas sample. Finally, the synthesis gas was combusted in a gas torch. The synthesis gas composition was analysed in every 4 minutes using micro thermal conductivity sensor by an Agilent 490 micro-GC. There were two different columns used in the gas analyser, 1 m Cox column for the analysis of hydrogen, carbon monoxide, carbon dioxide, oxygen, and methane contents, and the second one is 10 m PPU column for hydrocarbons content analysis. The gasifier is made of heat resistant steel with 1200 mm of length, 100 mm of outer diameter, and 10 mm of thickness. There are three blind holes along the gasifier to indicate the inside gasification temperature *(Figure 2)*. The gasifier was externally heated by a Carbolite 12/900 electrical furnace *(Figure 3)*.

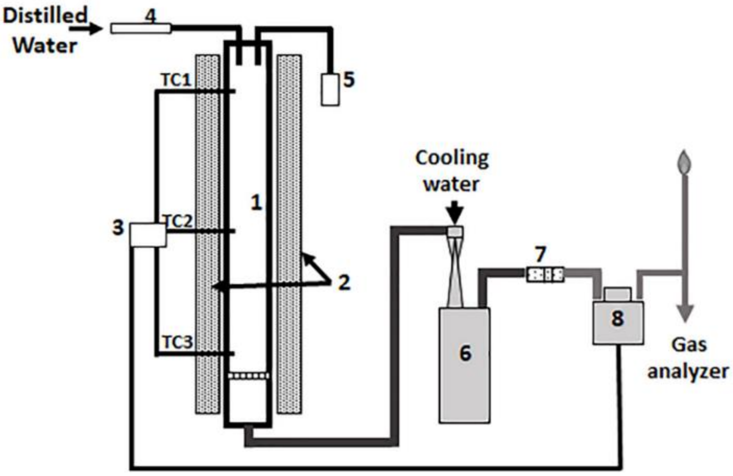

Figure 1

The schematic of single stage gasification process (1. Reactor, 2. Electrical heater, 3. Data logger, 4. Steam generator, 5. Manometer, 6. Venturi scrubber, 7. Cotton wool filter, 8. Gas meter)

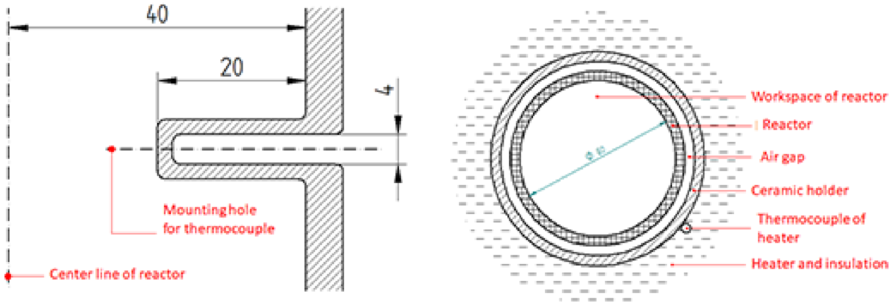

Figure 2

The cross-sections of gasifier

There was a pressure gauge placed in the top of gasifier to measure the actual pressure during the experiment, as well as to detect the possible blockages in the gasification system. In this case, the pressure was measured by an MRU-DM9100 type with a range of 0–100 mbar *(Figure 4)*. The steam was provided by a Maxi Vapor steam generator, with 3.5 L of built-in tank, 1300 W of power, and 2.8 bar of vapour pressure inside the tank *(Figure 4)*. The steam flow rate was manually adjusted according to the initial requirements in each experiment. There were two separated steam generators used during the gasification process, which allowed a continuous flow of steam at the entire length of the experiment.

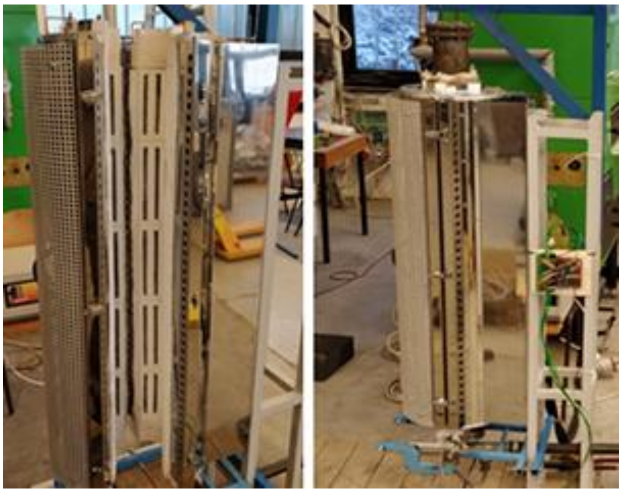

Figure 3
The electrical heater

Figure 4
The manometer and steam generator

The venturi scrubber is presented in *Figure 5*. The scrubbing water was injected through a fuel injector, which places on the top of the yellow-painted unit above the throat section of venturi scrubber. The scrubbing water was supplied at 2.5 bar and 15 °C from the district water system. During the gasification process, the wastewater was collected and treated before discharge.

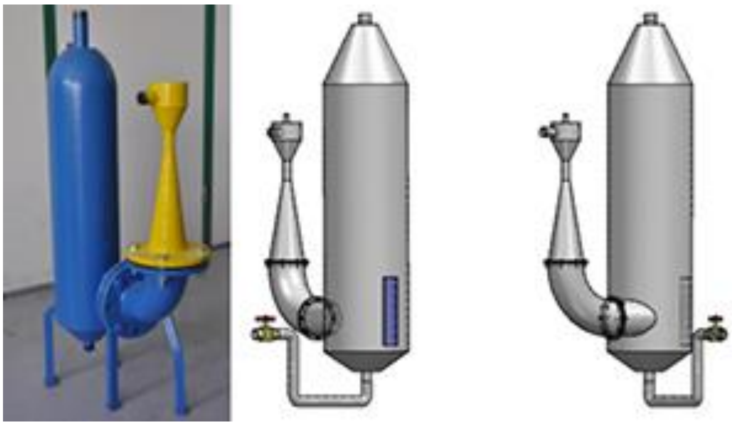

Figure 5
The venturi scrubber

2. RESULTS AND DISCUSSION

In this research, the experiments were conducted at 800 and 900 °C of gasification temperature and 10 g/min of steam flow rate within FC and BC samples. The PC sample was only gasified at 900 °C of gasification temperature and 10 g/min of steam flow rate as a result of the very low syngas production at this temperature. The pyrolysis process was carried out before the gasification process in each experiment.

2.1. Pyrolysis process

The temperature profile and syngas flow rate profile during pyrolysis period are shown in *Figure 6*. In all experiments, the temperature was higher at the top and middle of gasifier due to the heat transfer of matter inside the gasifier. The pyrolysis experiment started to produce syngas around 350-400 °C at the middle of gasifier in the case of BC and FC samples. While that was around 500 °C in the case of PC sample, which could be the result of the higher density and lower reactance of the coal sample. The flow rate of pyrolysis gas was quite similar in the experiments of BC and FC sample. While that was significantly lower in the experiment of PC sample.

The total volume of produced gas during pyrolysis period is described in *Figure 7*. At 900 °C of pyrolysis temperature, the total volume of pyrolysis gas was higher in the case of BC and FC coal than that of PC coal. There were 266.67 and 250 Nm^3/ton_{coal} for BC and FC coal sample, respectively. While there was approximately 3 times lower in the case of PC sample. It can be explained by the volatile content of PC sample which was lower than that of BC and FC samples. At the same temperature conditions, the pyrolysis gas in the experiment of BC sample was higher than that in the experiment of FC sample. In the experiments of BC and FC sample, the total volume of pyrolysis gas was higher at the higher pyrolysis temperature. It indicates that the higher temperature promoted the thermal cracking of the volatile matter during the pyrolysis period [8]–[10].

2.2. Gasification process

After pyrolysis process, the steam was introduced into the gasifier at 10 g/min mass flow rate for all experiments. The gasification process was terminated when the produced gas decreased to around 70 Nm^3/h for 1 ton of coal. The synthesis gas composition of the steam gasification process at ambient pressure is the result of the main reactions as follows:

Boudouard reaction:

$$C + CO_2 \rightarrow 2CO\ (\Delta H_{298} = 172\ kJ/mol) \tag{1}$$

Water gas reaction:

$$C + H_2O \rightarrow H_2 + CO\ (\Delta H_{298} = 131\ kJ/mol) \tag{2}$$

Water gas shift reaction:

$$CO + H_2O \rightarrow H_2 + CO_2\ (\Delta H_{298} = -41\ kJ/mol) \tag{3}$$

Methane reforming reaction:

$$CH_4 + H_2O \rightarrow CO + 3H_2\ (\Delta H_{298} = 206\ kJ/mol) \tag{4}$$

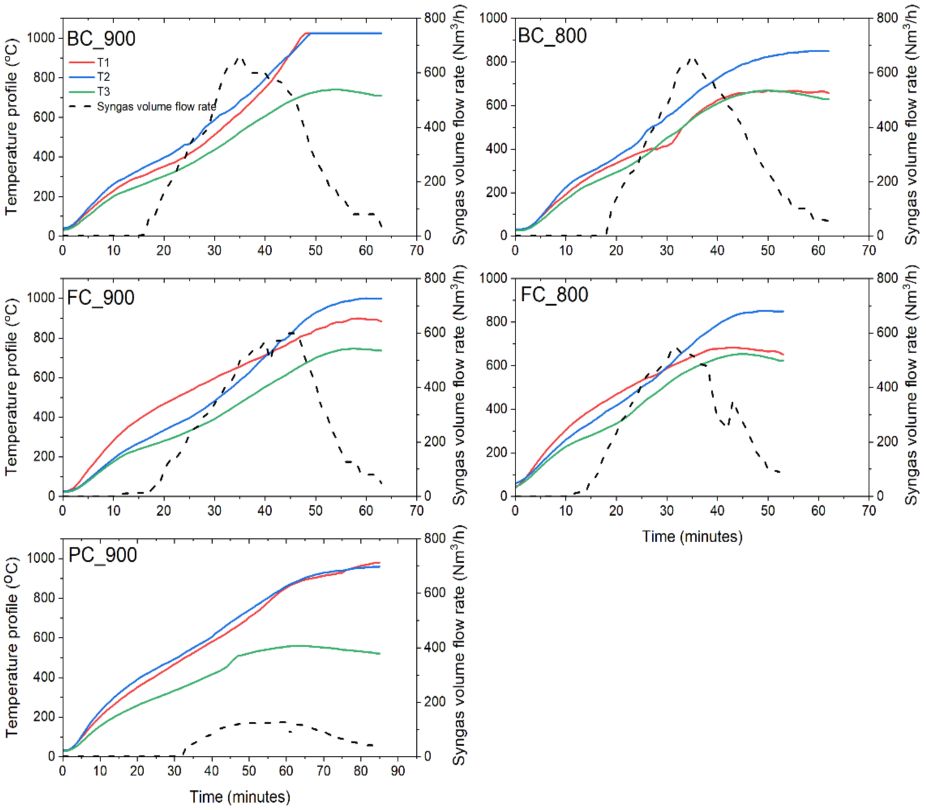

Figure 6

The temperature profile and syngas flow rate during pyrolysis period

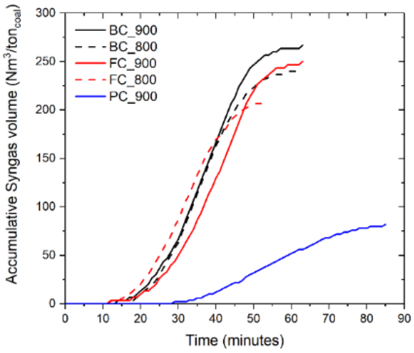

Figure 7
The accumulative syngas volume during pyrolysis period

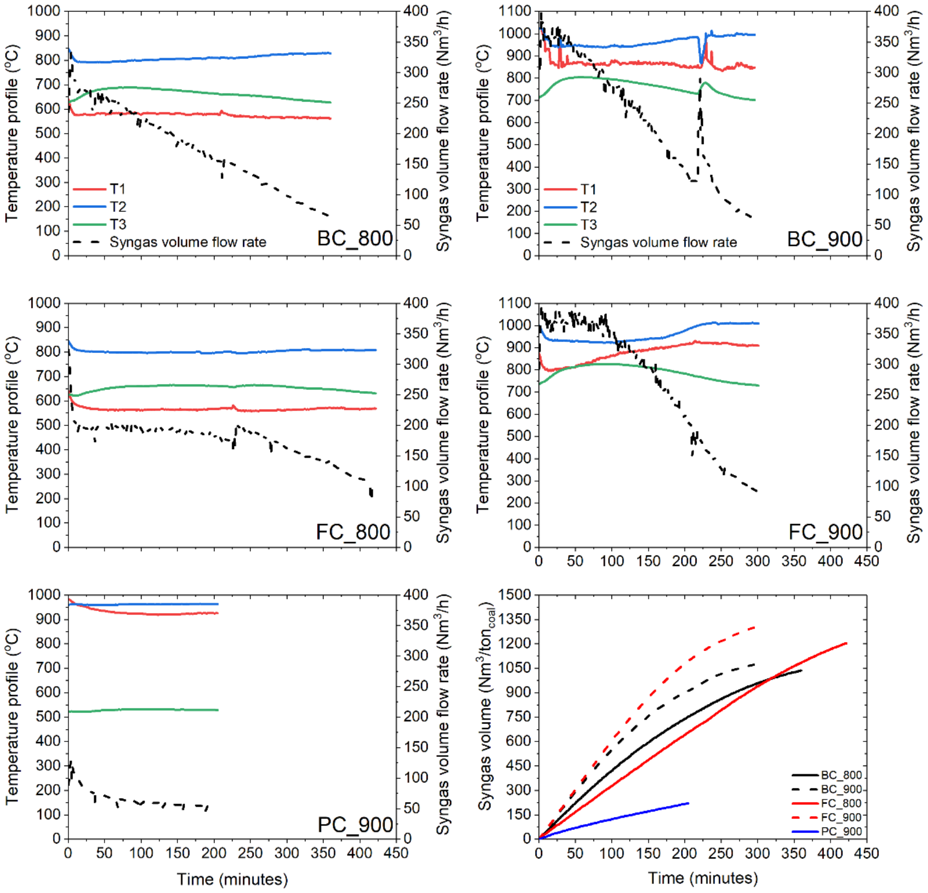

Figure 8
The temperature profile, syngas volume flow rate and accumulative syngas volume during the gasification process of BC, FC, and PC samples

The temperature profile, syngas volume flow rate, and accumulative syngas volume are exhibited in *Figure 8*. In which, the syngas volume flow rate and accumulative syngas volume was calculated for 1 ton of coal. At all experiments, the temperature at top and middle of gasifier were decreased due to the introduction of steam and endothermic reactions between the coal and steam at the beginning of gasification period. The total time of gasification process was longer in the case of 800 °C of gasification temperature for BC and FC samples. It indicates that the higher temperature promoted the gasification rate. At 900 °C of gasification temperature, the gasification process of BC and FC samples took a longer time than that of PC sample. This mainly could be the result of particularly high ash content in black coal sample from Pécs. In each experiment, the syngas flow rate was high at beginning of the experiment and decreased throughout the experiment. This is mainly the result of the decreased reaction rate as the carbon content in the surface of coal consumed in the reaction and these reactions are kinetically limited.

In the case of BC and FC samples only, the higher temperature resulted in a higher gas flow rate as a result of the increased rection rates of endothermic reactions at a higher temperature [11]–[14]. The dry gas yields were higher at 900 °C of gasification temperature. They were 1073 and 1310 Nm^3/ton_{coal} at 900 °C of gasification temperature for BC and FC samples, respectively. While the dry gas yields were only 1036 and 1203 Nm^3/ton_{coal} at 800 °C of gasification temperature for BC and FC samples, respectively.

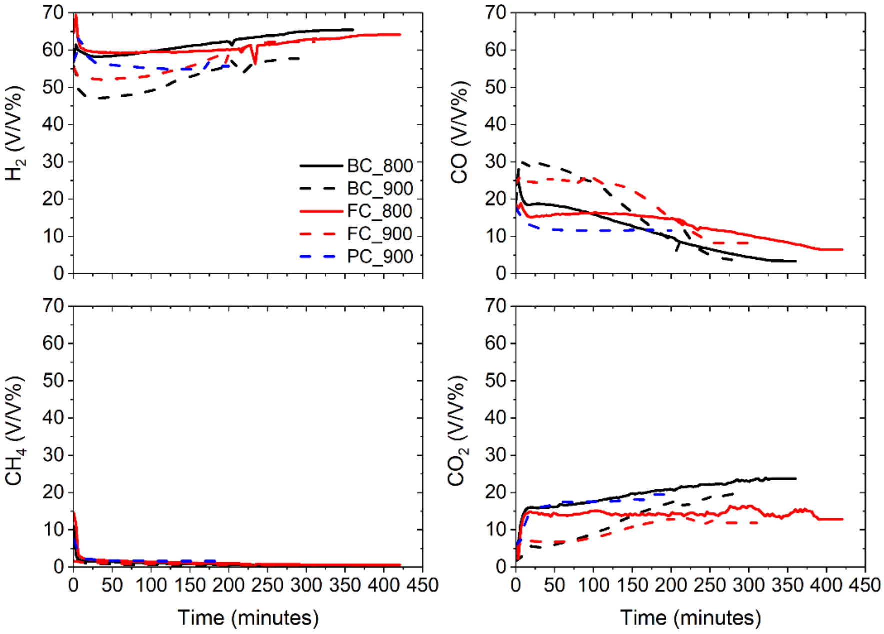

Figure 9
The syngas composition during the gasification process

The synthesis gas composition is illustrated in *Figure 9*. The main components of synthesis gas were hydrogen (H_2), carbon monoxide (CO), carbon dioxide (CO_2), and a small quantity of methane (CH_4). There were no detectable longer chain hydrocarbons in the gas samples. Typically, the H_2 concentration remained roughly above 60 V/V% during the gasification process at 800 °C of gasification temperature. The H_2 concentration was lower at 900 °C of gasification temperature for BC and FC samples, caused by the decreased reaction rate of WGS reaction *[Equation (3)]* at the higher temperature, and the reverse WGS reaction when temperature exceed the equilibrium temperature [15]–[17]. There was a reverse trend in the CO and CO_2 concentrations along the experiments. The CO concentration showed a decrease trend during the whole gasification period. In addition, the CO concentrations were higher at 900 °C of gasification temperature for BC and FC samples. While the gasification temperature at 800 °C produced a higher CO_2 concentration than that at 900 °C for both BC and FC samples.

During the gasification process, there was a small amount of $CH_{4,}$ and the presence of longer chain hydrocarbons were under detectable level. The CH_4 concentration decreased below 2 V/V% in the gasification process. The obtained trends could be explained by the higher temperature favouring the endothermic reactions *[Equations (1), (2), (4)]* in the gasification process [14], [18], [19].

3. CONCLUSION

The above approaches mainly focused on the multi-stage gasification (pyrolysis and steam gasification) to determine possibility of gasification as a source of synthesis gas that can be further utilised as secondary raw material. Both BC and FC samples show a good potential in the field of synthesis gas production for the chemical industry as a raw material. Both have a high H_2 and CO concentration and a low CO_2 concentration. With an increasing gasification temperature, they also showed a higher reaction rate, which resulted in shorter gasification time. However, the coal sample from Pécs has a very high ash content and very low carbon content compared to the other two coal samples. During the gasification the syngas flow rate and accumulative syngas production were three times lower compared to the other two samples. Therefore, this coal samples are not a good candidate for further analysis and optimisation of the gasification process for syngas production.

ACKNOWLEDGEMENTS

The cross-cutting research was conducted at the University of Miskolc as part of the *More efficient exploitation and use of subsurface resources project* implemented in the framework of the Thematic Excellence Program funded by the Ministry of Innovation and Technology of Hungary (Grant Contract reg. nr.: NKFIH-846-8/2019), and within the subsequent *Developments aimed at increasing social benefits deriving from more efficient exploitation and utilization of domestic subsurface natural resources project* supported by the Ministry of Innovation and Technology from the National Research, Development and Innovation Fund according to the Grant Contract issued by the National Research, Development and Innovation Office (Grant Contract reg. nr.: TKP-17-1/PALY-2020).

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