

## Tar related issues in underground coal gasification

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*Technology UCG is worldwide presented as the technology with the lower adverse effect on the environment in comparison to all until now applied techniques of the underground and surface coal mining. The present paper deals with the pollution from the underground coal gasification in situ taking into the consideration the amounts of the gasified coal based on the samples from the simulated gasification and chemical analysis of tar. Pollution of rock surrounding can be caused by evasive gas, aromatic hydrocarbon, extractable non-polar substances and solid residues. During individual gasification, there are raising polluting gases as hydrocarbon, hydrosulphide, carbon disulfide, oxide nitrogen, mercaptans. Such gas compounds have high toxicity and significant stink. Some of the less qualitative sorts of coal obtain till 6 % of sulphur, that is oxidised during gasification and burning to the sulphur dioxide, and hydrosulfide is also rising. By not perfect coal burning, there are also raising polyaromatic hydrocarbons (PAH), many of them are toxic, or they have mutagenic effects. The question is how these tars will affect the underground and surface water in the area which has been encumbered with the mining and chemical industry for a longer period.*

**Key word:** underground coal gasification, surface coal mining, gasification, coal deposits

### Introduction

Gasification is the chemical process of the solid or liquid fuels transformation in gaseous fuels, which take place in the gasifiers (generators, reactors). In the course of the gasification of coal, the decomposition of the organic mass occurs due to the effect of the gasification media under high temperatures. The resulting are gaseous products, tars and solid residues, as clinker or ash. The gaseous products are after their cleaning used for the electricity production or as the raw material for the chemical products production. Tar related issues represent one of more severe problems in coal gasification since it is an unwanted product, which hinders the produced gas exploitation and increases the cost of the whole facility operation. Tar is the complex mixture of the organic substances. Underground coal gasification is based on the same principle as the classical gasification with the unique feature that the location of the gasification is in the coal deposit (Bhutto et al., 2013; Blišťanová and Blišťan, 2012; Boyd et al., 1981; Lamb, 1977; Uppal et al., 2014; Straka et al., 2014).

The contribution shows the problems of underground coal gasification. Currently, this topic is timely, seeing that many coal deposits can not mine through traditional mining methods. Through underground gasification, the so-called synthetic gas - syngas can be obtained from these deposits, which can be converted into electricity or used in industry and households as fuel. Underground gasification technology has been verified in laboratory conditions for several experiments. Experiments differed from each other by methods of coal bedding, using different oxidants and methods of management. Experimental coal gasification in experimental conditions, which took place in the solutions of applied research projects, has enabled us to gain knowledge about this process. We analysed the useful, but even harmful products of this so-called "clean technology" (Durdán et al., 2014; Laciak et al., 2011).

The contribution is divided into two parts. By comparison of two experiments, the contribution investigates the process of coal gasification concerning produced gas - syngas. Analysis of the gasification process regarding syngas was focused on the quality of the produced gas, which can be characterised by its calorific value. From this perspective, the primary goal of the coal gasification process is to generate the gas with the highest calorific value. In the second part of the contribution, the coal gasification process concerning by-product is analysed (a mixture of tar and water) and the generation of hazardous substances that may endanger the environment (Sasvári et al., 2007).

### Underground coal gasification

More than 95 % of the world energy sources from fossil fuels are bound in coal. Despite vast world coal reserves, only a small proportion of coal can be mined applying the recent technology of mining. Approximately

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85 % of known coal reserves are called non-extractable coal sources, which, according to many experts, may be made available using the new technology of the underground coal (UCG), (Blišťan and Blišťanová, 2009; Sasvári et al., 2007; Yang, 2004).

This method is based on the underground coal "combustion" right in the deposit. For the gasification in the industrial scale, it is necessary to build the system of drills – as a minimum one injection and one production drill and also the system for the produced gas cleaning and storing, shown in Figure 1. By exploiting the injection drill, the deposit is ignited, and gasification medium is blown inside. Produced gas is passed on the surface via the production drill. The goal of this process is to generate as much gas as possible from the coal with the maximum calorific value. To achieve that purpose the methodology based on the algorithms of controlling the input oxidising agents and exhaust of the produced gas must be applied. The process is safe and from the point of the economy is efficient, which is its highest advantage.

The described method enables to react flexibly to the energy market, because the transformation of the primary product, the so-called syngas, in electricity is recently executable without serious problems. The further possibility of the syngas as the gaseous fuel exploitation is in the industry and homes. The objective of the coal gasification is to produce gas - syngas with the as high calorific value as possible, in works (Kačur et al., 2014; Kostúr et al., 2015; Pástor, 2003).

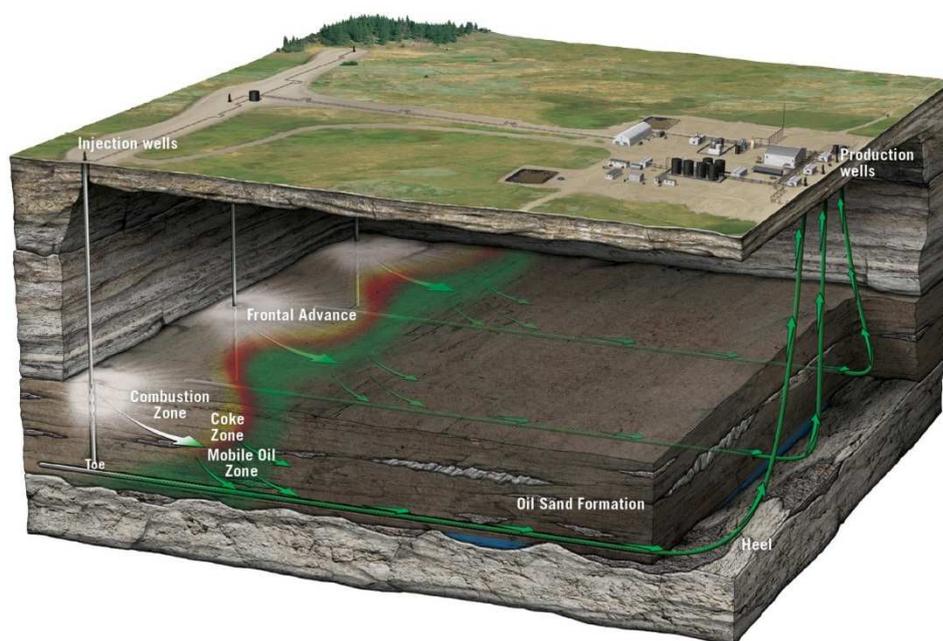


Fig. 1. Principle of UCG technology (www1).

### Production of tar in the underground coal gasification

Processes and chemical reactions that originated during gasification of brown coal are described in literature (Sasvári et al., 2007), chapter 3.

Individual possible negative influences to the living environment on the surface and underground can be divided into:

1. the change of environment morphology
2. pollution by gas,
3. pollution by aromatic hydrocarbon,
4. pollution by VOCs,
5. pollution by solid remains after gasification and trace elements.

In the 1st point - Terrain declines are a single influence of underground gasification that can be observed at the surface. During gasification process, there are volume changes at the rock. Only remains and tars that are not gasified remain in originated hole from the former coal stuff. Volume change has a decrease of the top layer of rock as a consequence that leads to the creation of declined hollow basin at the surface according to the rock type. Deformation of the surface does not occur immediately during gasification, but only after 2,5-3 months. Declined hollow basin is regularly created in the frame of geometric space that is limited by marginal

(tangential) drill holes of the generator, and they are outstanding around the drill holes. It is possible to avoid terrain decline by choice of proper locality for gasification and by the evaluation of geological factors, mainly when a discontinuity occurs in the chosen locality.

In the 2nd point - Rising of pollutive gases ( $\text{CO}_2$ ,  $\text{H}_2\text{S}$ ,  $\text{C}_2\text{S}$ ,  $\text{NO}$ ,  $\text{NO}_2$  and mercaptans) is influencing by high toxicity and expressive odour.

In the 3rd point - Polycyclic aromatic hydrocarbon (PAH) are organic compositions consisted of 3 and more aromatic cycles that contain only carbon and hydrogen. They are created during heat decomposition and not-perfect burning of coke, black coal, asphalt and naphtha. Some PAH elements are easily vaporised to the atmosphere from the soil or surface water. The majority of PAH is not soluble in the water, and they are joining to the soil elements and sediment on the river and lake basin, and consequently they contaminate underground water. Their content characterises loading of the environment. Due to the activity of microorganisms, PAH are decomposed in soil or water element during several weeks or months. They are accumulated in plants and animals, many of them are toxic, or they have mutagenic effects.

DHHS (The Department of Health and Human Services) classifies PAH as a potential carcinogen for the health of people. Environmental Protection Agency USA (US EPA) mentioned them in the list of priority pollutants with the necessity to monitor them in terrain as well as water elements of the living environment. US EPA suggested to state 16 PAH as standards for risk PAH that contaminate mostly soil and water: naphthalene, acetanaphthalene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b,k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene, indeno(1,2,3-c,d)pyrene.

In the 4th point - Volatile Organic Compounds (VOCs). VOCs is a collective term given to organic compounds, which have high vapour pressure and become gases at ambient pressure and temperatures. Many are hazardous to human health, with several classified as carcinogenic. The key sources of VOCs are industrial processes. A group of VOCs, collectively known as BTEX, comprising benzene, toluene, ethylbenzene and xylene (often expressed as total xylenes (m+p), can be very dangerous for living environment and they can occur during coal gasification.

In the 5th point - black and brown coal contains a certain concentration of trace elements, including heavy metals. The majority of heavy metals evaporates during burning. Lately, during the process, it is condensed on the surface of solid elements (flue ash). There can be pollution by fluorine, bromine, manganese, iron, calcium, etc.

Table 2 illustrates the volume of pollutive elements in detracted samples of tar and water mixture for analysis in accredited laboratory (extractable non-polar substances - ENP, total organic carbon -TOC, BTEX, PAH), (Laciak et al., 2011).

Gasification process, heating with the air absence, is conducted as follows. At the temperatures about  $100\text{ }^\circ\text{C}$ , the part of water (free) and absorbed gases are released. By  $300\text{ }^\circ\text{C}$ , a substation proportion of water and a certain amount of gases, mainly carbon dioxide, is released from brown coal due to which in particular oxygen is released, then the small amount of nitrogen and carbon oxide. Further heating within the temperatures range of  $300 - 350\text{ }^\circ\text{C}$  causes the continuation of the bound water and oxygen splitting off, and combustible gases begin to release (methane) as well. Up to this temperature level, only the ballast substances are released. Above the temperature of  $350\text{ }^\circ\text{C}$ , the decomposition of the carbon components is processed, the combustible gases, vapours of the hydrocarbons and tars which exist in the gaseous phase are released. However, the most intense production of the hydrocarbon gases and tars, which turn in the liquid fraction after their cooling, is at the temperatures about  $600\text{ }^\circ\text{C}$ . At high temperatures ( $1\ 000\text{ }^\circ\text{C}$ ), the majority of the product is released in the form of the gas, and their amount is quickly reduced when the temperature drops below  $800\text{ }^\circ\text{C}$ . As the solid fraction is considered, the porous residuum is produced in connection with the gas and vapours release; the temperatures should not exceed  $600\text{ }^\circ\text{C}$ . This is marked as the low-temperature coke (char), in the case of the higher temperatures (up to  $1\ 000\text{ }^\circ\text{C}$  and more) as mentioned in the works (Zelenák, Škvareková, 2012; Friedmann et al., 2009; Durdán, Kostúr, 2015).

The higher temperatures cause the coal decomposition, the product of which is the gaseous fraction and substantial residuum. By cooling these gases and vapours the condensate is produced, consisting of the hydrocarbon and water fraction. Their chemical character is identical with the hydrocarbon fractions as it is with crude oil: petrol, motor naphtha, paraffin, light and heavy oils and asphalt matters. Next, there are water fractions and soluble compounds which are formed during the coal thermal decomposition. That is the first of all the ammonia, the particular amount of the sulphur containing matters and the broad spectrum of the organic compounds of the phenol type, ketones, and other polarised cases.

Table 1 provides the physical and chemical parameters of the brown coal mined in Slovakia (Škvareková, Kozáková, 2012).

Tab. 1. Characteristic of physicochemical parameters of the Slovak brown coal (Škvareková, Kozáková, 2012).

Item	Fuel	Slovak brown coal		
		assorted	boiler	average
Elemental fraction in the fuel		/ wt. %		
C <sup>r</sup>	fraction of carbon	47.29	28.35	33.80
H <sup>r</sup>	fraction of hydrogen	3.62	2.49	2.86
O <sup>r</sup>	fraction of oxygen	14.36	12.42	13.52
N <sup>r</sup>	fraction of nitrogen	0.78	0.45	0.54
S <sup>r</sup>	fraction of combustible sulphur	1.37	1.76	1.78
Amount of combustibles "V <sup>r</sup> " in the fuel		67.41	45.48	52.50
W <sup>r</sup>	total humidity	24.26	36.04	32.00
Sv (Dm)	total dry matter	75.74	63.96	68.00
A <sup>r</sup>	ash in the fuel	8.33	18.48	15.50
A <sup>d</sup>	ash in dry matter	11.00	28.90	22.80
S <sub>t</sub> <sup>r</sup>	total S in the fuel	1.51	2.58	2.38
S <sub>t</sub> <sup>d</sup>	total S in dry matter	1.99	4.04	3.50
Q <sub>i</sub> <sup>r</sup> [MJ.kg <sup>-1</sup> ]	fuel efficiency	15.27	10.76	12.04
Q <sup>daf</sup> [MJ.kg <sup>-1</sup> ]	caloric value of combustible	22.65	23.66	22.93
Q <sub>s</sub> <sup>r</sup> [MJ.kg <sup>-1</sup> ]	caloric value of fuel	16.03	11.30	12.64

Tab. 2. Composition of tar from the coal gasification, mentioned in work (Laciak et al., 2012).

Experiment	1	2	Limit values for the synthetically produced polluting substances [19] [µg/l]
	Amount [µg/l]	Amount [µg/l]	
ENP	1 324 000	144 900	
TOC	2 824 000	22 656 000	
Benzene (BTEX)	3.80	393.90	0.75
o – xylene ( BTEX)	3.00	41.00	312.5
m, p – xylene ( BTEX)	4.75	76.20	
Toluene ( BTEX)	3.20	199.90	437.5
Acenaphftene (PAH)	3.08	1 022.19	
Acenaphthylene (PAH)	2.36	3 766.27	
Anthracene (PAH)	2.61	880.49	
Benzo(b)fluoranthene (PAH)	0.00	29.67	
Benzo(a)anthracene (PAH)	0.34	358.28	
Benzo(k) fluoranthene (PAH)	0.00	15.64	
Benzo(g,h,i)perylene (PAH)	0.00	11.58	
Benzo(a)pyrene (PAH)	0.00	44.35	0.00625
Dibenzo(a,h) anthracene (PAH)	0.00	5.48	
Phenanthrene (PAH)	16.72	2 299.92	
Fluoranthene (PAH)	3.20	1 036.90	
Fluorene (PAH)	4.92	1 162.52	
Chrysene (PAH)	0.38	363.43	
Indeno(1,2,3-c,d)pyrene (PAH)	0.00	20.83	
Naphthalene (PAH)	1.77	2 894.26	
Pyrene (PAH)	2.26	660.04	
∑PAH	41.102	14 784	0.0625
Tar amount [litre]	21.8	10	

## Analysis and discussion

For the analysis of the underground coal gasification from the view of syngas and secondary product (a mixture of tar and water) that had been retracted immediately after finishing of gasification, two experiments were done in laboratory conditions. Tar samples from the experiments in the two generators were taken and analysed in the accredited laboratory (Laciak et al., 2011). In Table 2, the chemical analysis of the individual components of the tar (31.8 litres) is given, of which about 650 kg of brown coal was used in the laboratory. They are compared with the limit values with the synthetically produced polluting matters (Zelenák, Škvareková, 2012). Figure 2 shows the scheme of one of the generators used for the gasification of coal in laboratory conditions.

The fuel, including charge, consisted of the coal, coal powder and goudron (residual substance after the oil distillation), which together with the coal powder served to bind the coal pieces into one monolithic layer (Kačur, Durdán, 2014). According to Table 2, tar is prevalingly formed of polycyclic aromatic hydrocarbons (PAH).

PAH are the organic compounds composed of two or more aromatic cycles containing only carbon and hydrogen. They are created in the course of the thermal decomposition and incomplete combustion of coke, black and brown coal, asphalt and naphtha. Some PAH particles easily evaporate into the atmosphere from the soil or surface water. The majority of PAH is not solvable in water and are bound to the particles of soil and the sediment on the bottom of rivers and lakes and following they contaminate underground water. Their content characterises the state of the environment. Due to the activity of the microorganisms, PAH is decomposed into the soil or water component of the environment in the course of several weeks or months. They are accumulated in the bodies of the plants and animals, and many of them are toxic or have the mutagenic effects. These are the matters which are subject to the monitoring in the frame of the Slovak Republic with the objective to meet the limit values of the pollution of the industrial waste waters released into the surface waters containing the hazardous matters. The recommended value for the surface water is 1.0 g/l and for water determined for the irrigation 0.05 µg/l (Laciak et al., 2012).

In the environment, a degradation of PAH can occur, during which various reactions arise: electrophilic and nucleophilic substitutions, 1,2 and 1,4 cyclic reactions, oxidation, hydrogenation, condensation reaction, biodegradation.

In the atmosphere, environmental factors (intensity of radiation, concentration of gas reactants, physical and chemical characteristics of the elements) influence PAH decomposition due to the solar radiation and oxidation, or by substitution reactions.

In the water environment, photo-oxidation, chemical oxidation and biodegradation by water organism contribute to the PAH decomposition.

Also, microbial degradation is a single way of PAH transformation in soil due to the absence of radiation and limitation of oxygen presence. The following belongs among factors that influence the speed and extent of PAH degradation in the ground: composition of the microbial population, soil composition, temperature, pH, oxygen and nutritive content, physical and chemical characteristics of PAH.

The initial reaction is running in bacteria, fungus and advance organism by which oxygen loading to the PAH molecule is ensured. In the environment, several types of such initiation reactions are appearing, and they provide oxygen intrinsic to the PAH molecule:

1. Initial reaction by the help of bacteria and the green alga (for example naphthalene oxidation, toluene, dibenzothiophene, anthracene, phenanthrene, fluoranthene, pyrene, chrysene, benzo anthracene and benzopyrene during *Pseudomonas*).
2. Initial reaction with the help of methanotrophs bacteria (for example bioremediation of the soil contaminated by trichloroethylene, it changes naphthalene to 1- and 2-naftoly).
3. Initial reaction by the help of fungus and bacteria, whereby enzyme that turns unsaturated compound to the arene oxide and consequently to the trans-dihydro diol is raising. Various organisms are changing activated molecules to the different products, for example, sulphate, xyloside, glucoside. Such compounds are substrates for other microorganisms, and by this way, the removing of polycyclic compounds from the biosphere exists.
4. Initial reaction by the help of the so-called wood-rotting fungus. Such fungus is also able to make PAH degradation during rising of Chino that are afterwards mineralized backwardly by fungus and bacteria, mentioned in work (Škvareková et al., 2011).

For evaluation of PAH toxicity, it is necessary to know not only the mechanism of such elements, the relationship of toxicity and structure, but also results of synergic or antagonistic acting of individual polyaromatic and results of their interaction with other compounds.

Sulphur dioxide can create in the water together with PAH dissoluble sulphur acids that show carcinogenic effects. The presence of sulphur dioxide and nitrogen increase even more potential carcinogenic effects of PAH. Through water colorization, PAH can be changed to the related derivatives of chlorine, where many of them have the biggest mutagenic and carcinogenic effects.

Scanning of interactions among not carcinogenic PAH show the following: slightly carcinogenic or not carcinogenic PAH, for example, benzo(e)pyrene, benzo(ghi)perylene, fluoranthene or pyrene - they increase the significant occurrence of tumours together with benzo(a)pyrene. However, others are not carcinogenic PAH, for example, benzo(a)fluoranthene, benzo(k)fluoranthene, chrysene or mixture of anthracene, phenanthrene, and pyrene. Some experiments show that PAH mixture is less efficient as individual PAH.

Due to the carcinogenic effects, PAH demands metabolic activation of monooxygenation.

Sequel of pollutants in the environment that can induce monooxygenation can act with PAH synergic. For example 2,3,7,8 - tetrachloro dibenzo-p-dioxin can increase the activity of microsomal enzymes, and by this way increase toxicity, mentioned in work (Škvareková, Kozáková, 2012).

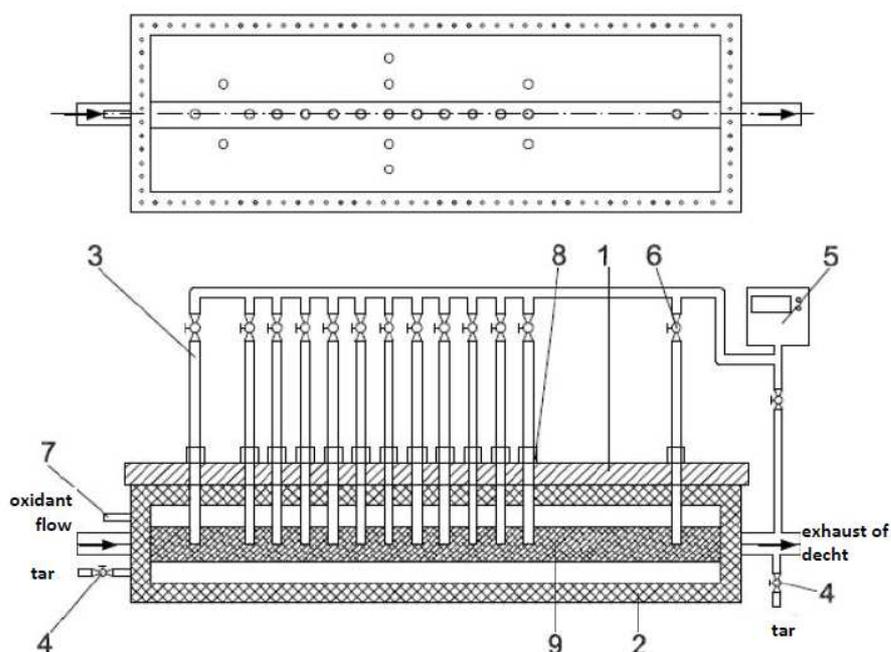


Fig. 2. The scheme of the generator (1- cover of generator, 2- container of generator, 3- the sampling probe of syngas, 4- , 5- gas analyser, 6- the valve, 7- the ignition opening, 8- nut for positioning of the sample probe) (Laciak et al., 2012).

## Conclusion

In the strategy of energy security of the Slovak Republic, the end of the use of the Slovak coal deposits is expected by 2030. The obtaining of all energy sources that coal offers is, therefore, obvious. In addition to traditional coal, natural methane, or gas generated by underground gasification, can also be used. Technologies for obtaining gas have already been ranked among the clean coal technologies (Clean Coal Technology- CCT) and are considered to have advantages, but also disadvantages (Škvareková, Kozáková, 2011; Lin et al., 2014; Taušová, 2007).

In assessing the activity of technology UCG and its effects on the elements of the environment, an individual approach is important. It is necessary to evaluate impacts in a given rock environment with its specific properties.

Coal production has been increasing over the past 10 years, despite calls for lower emissions and continued research into the development of alternative energy sources.

The International Energy Association predicts an increase in coal usage of 55 % to 2030 as emerging nations develop industrial infrastructure, and the world moves from over-reliance on depleting supplies of Oil and gas.

UCG technology has a less detrimental environmental impact, as all coal stays underground, there are less emissions, fewer surface footprint as no surface gasifier is required and the gas is processed to remove harmful particulates, including CO<sub>2</sub> capture.

The process is safe and economical, being of great importance to all - but especially the emerging markets that are currently building large scale coal fired power stations at an unprecedented rate. The requirement for secure supplies of gas for both domestic and industrial use has never been greater (Laciak et al., 2011).

Analysis of coal gasification in laboratory condition has been made from two points of view - from the view of produced gas – syngas as the main product of gasification and from the view of secondary product – mixture of tar and water. Both products are the result of coal gasification. The review of amounts of the polluting substances occurring in the samples of tar provided by the accredited laboratory (extractable non-polar substances - ENP, total organic carbon - TOC, BTEX, PAH) is given in Table 2. The values of the polluting substances are compared with the limit values for the synthetically produced polluting substances.

The most significant impact has the underground coal gasification on the underground water. The operation of the underground generator may violate the chemical balance and overall mineralisation and thereby also the hardness of the underground water. Due to the generator operation, the water may be polluted by, for example, hydrogen sulphide, carbon dioxide, and tar. Also, the effect of the temperature in the underground generator on the surrounding area is significant. Concerning high temperatures, it is evident that surrounding rocks and the underground water will be heated, mentioned in work (Laciak et al., 2012).

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