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Leaching behaviour of metals from post-underground coal gasification cavity residues in water differing in mineralization

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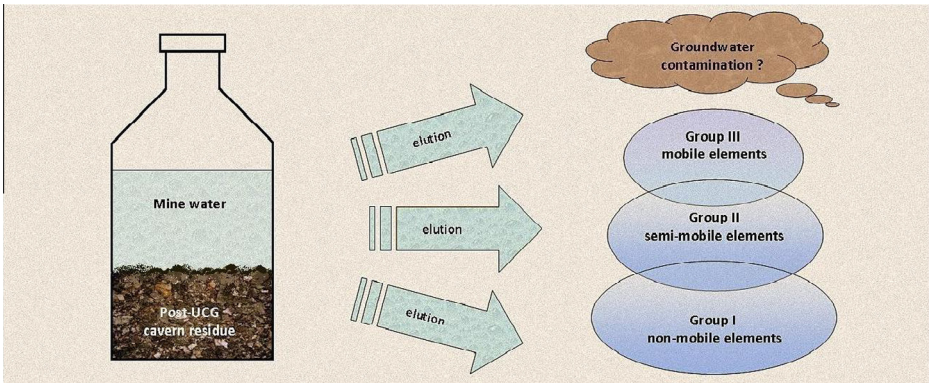
HIGHLIGHTS

- Ashes and chars from quenched reactors after three UCG experiments were tested.
- Waters with diverse mineralization may have different leaching abilities.
- Eluted metals were divided into groups based on their mobility to water phase.
- Some toxic elements (Cr, Pb, Cd) may migrate to water from UCG hard coal residues.

GRAPHICAL ABSTRACT



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ABSTRACT

The aim of this study was to examine whether water entering the post-reaction area after underground coal gasification (UCG) leaches metals from the post-processing waste in the cavity to such a degree that it can pose a threat to the environment.

In the studies, ashes and chars arising from hard coal and lignite gasification have been used. Leaching tests have been performed with the usage of deionized water and two types of mine water that greatly differ in mineralization. The physicochemical composition of the obtained eluates has been analysed. The coefficients of dispersion (distribution), k_d, have been estimated, which allowed determination of the mobility of metals under test and thus their ability to leach to the water phase. Metals present in the process of UCG have been described in terms of volatility and divided into three groups. In addition, the concentrations of metals in the eluates, depending on the leaching medium used, have been investigated.

It has been found that the majority of tested elements tend to be leached in a stronger degree from ash residues than from chars. In comparison with the residues from hard coal, the residues of gasified lignite show the higher relation to the water phase. The presence of such elements as Cr, Pb or Cd in eluates may suggest that toxic metals are able to leach to water from the residues of the gasification process and a non-negligible risk of environment contamination might occur.

1. Introduction

The technology of underground coal gasification (UCG) allows energy to be obtained in the form of gas discharged to the surface

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as a result of the in situ coal seam gasification. This process is initiated by inflammation of the part of deposit to which the adequate gasification agent is supplied (oxygen, air, steam). The obtained gas is a product of industrial value that can be used in the production of electricity and heat or as a synthesis gas. Underground coal gasification is an important process not only from business and economic points of view [1,2] (it also assumes the usage of those coal deposits that, because of their deep position or residual nature, are unsuitable for operation with conventional methods) but also from the perspective of the environment (technology minimally impacts the environment) [3]. The last issue is not entirely clear and therefore needs further research.

Every oxidation process, including hard coal or lignite gasification, generates the formation of solid, liquid and gas products containing significant concentrations of organic and inorganic substances, including substances hazardous to the environment [4–9]. Moreover, as a result of coal pyrolysis and the series of homo- and heterogeneous reactions occurring between the gasification products, number of further hazardous environmental contaminants can be produced during the UCG process. The major organic ground water pollutants typical for UCG are phenols, benzene and its derivatives (BTEX), polycyclic aromatic compounds (PAHs) and heterocyclic compounds containing nitrogen, sulphur and oxygen heteroatoms. The inorganic matrix includes ionic compounds, such as ammonia, cyanides, sulphates, chlorides and wide range of metal and nonmetal elements [10–13]. Therefore the threat of ground water pollution is the dominant environmental concern during and after UCG. The results of the extensive studies on the potential ground water pollution resulting from the UCG operation based on Hanna and Hoe Creek experience (Wyoming, USA) have been reported in the 1980s and 1990s [10,14–16]. The potential environmental problem is also announced in study performed previously in Europe, Thulin project [17] and in Australia, Chinchilla project [18]. The report issued by Queensland Government, Australia also proves that the environmental aspects of the UCG process should not be neglected [19]. Generally it may be concluded that more is known about organic contaminants from UCG but less is learned about inorganic pollutants and research on metal elution from the cavity residues have been not addressed yet.

Because of the location of the geo-reactor, which is surrounded by a natural geological stratum of diverse structure and properties, the unique conditions of coal seam operation and the process of underground gasification are very complex. There is a risk that the thermal and mechanical factors associated with UCG will meaningfully change the parameters of the rock mass, allowing water penetration of the rock through the newly created crevices, cracks or post-exploitation voids [20]. That is why it seems necessary to study the post-reaction space formed as a result of the underground coal gasification, the cavity, in which, after process termination, a part of the solid products such as ash residue or carbon chars lies. No published test results have determined whether a cavity is a neutral object and whether the UCG post-process residues do not present a danger to the aquatic environment surrounding the geo-reactor. Each stored waste may pose a potential pollution, affecting the local ground water. It is known that combustion waste, among others, is a category of waste that is important from the perspective of the metal hazard to the ground water [21]. Because the UCG process is accompanied by the generation of waste with properties similar to those of ashes, it can be assumed that there are such risks as leaching of metals from pollutants lying in the cavity.

All waste deposited in underground workings are exposed to contact with the natural water flowing to the workings. From waste, water may leach substances that, after dissolution, contribute to the growth of the total mineralization or the increase of pollutant content in the waters. The risk of important changes in ground water quality (also mine ground water) occurs mainly at shallow levels, where most ground water runs from the infiltration of meteoric waters from the surface or waters from aquifer layers of the overburden (usually poorly mineralized). Generally, it is assumed that along with the degree of water mineralization that occurs with increasing depth, the participation of the water resources in relict reserves (static) becomes greater as well.

Long-term mining work contributes to the transformation of the original hydro-geological conditions of the Upper Silesian Coal Basin over a substantial area, not only in the rock mass (drainage of the aquifer layers) but also at the surface (mine waters with a significant contamination carried to surface waters). The coal exploitation provokes the creation of cracks and a continuity break in the isolation layers dividing the particular aquifer layers [20]. The natural and mining technical factors cause significant changes in volume quality and huge variation of physicochemical composition of water flowing to mining workings. It can be generally assumed that in the Upper Silesian Coal Basin, mineralization increases with depth (hydro-geochemical zoning); however, this increase is dependent on the nature of the overburden. When the Carboniferous rock mass is located under the isolating complex of the Miocene clay, the highly mineralized waters are located at a definitely shallower level compared to Carboniferous formations covered by water-saturated Triassic formations. With an increase in depth, the water supply to the mining workings decreases because the permeability of layers decreases.

Waters flowing to mining workings in the Upper Silesian Coal Basin mines can be divided into three groups based on the degree of mineralization: waters containing dissolved substances up to 1.5 g/l, from 1.5 g/l up to 10 g/l and above 10 g/l [22].

Tests carried out within the framework of this study have been proposed primarily in terms of the safety of the natural aquatic environment, particularly ground water.

The subjects of tests were the residues obtained from three different experiments of UCG (one in situ experiment and two experimental simulations of UCG in an ex situ laboratory installations). Two hard coal samples and one sample of lignite were investigated. To estimate the quantity of substances leaching to the aquatic environment, leaching tests have been carried out for the ash residues and chars taken from the shut-down reactors after the above mentioned experiments. Leaching was carried out using deionized water and also two types of mine water, significantly differing in their general degree of mineralization. The analysis of the physicochemical composition of the obtained eluates has been carried out, and the concentrations of metals leached from individual samples to various leaching media have been investigated.

The knowledge about inorganic contaminations and its leaching by different types of water will be incorporated into reactive transport model of the pollutants migration so it would be useful for environmental assessment of UCG technology.

2. Experimental

2.1. Sample preparation and analyses

Two hard coal samples and one sample of lignite constituted the raw feed. The first one was taken from experiment conducted at the experimental mine ‘Barbara’. The georeactor was placed in coal seam at a depth of 30 m, and oxygen and water were applied as gasification reagents. During this experiment 5364 kg of hard coal was gasified, with an average consumption rate of 37.8 kg/h [23]. The second sample of hard coal was taken from the pressurized reactor used for the experimental simulations of UCG process in an artificial coal seam. The total length of the simulated coal