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Comparison of metal elution from cavern residue after underground coal gasification and from ash obtained during coal combustion

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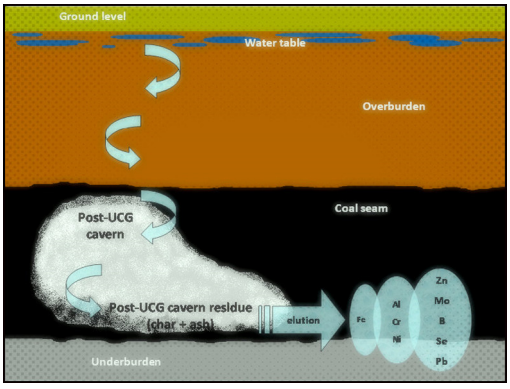
HIGHLIGHTS

- The first research on metal elution from residues after in situ UCG were conducted.
- Hydro-geochemical background typical for groundwater was exceeded in eluates.
- Highly toxic elements (Sb, Cd, Hg, As) were not detected in any of water extracts.
- Metals were classified into groups according to their mobility to water phase.
- Ashes from four UCG processes were compared with ashes from coal combustion.

GRAPHICAL ABSTRACT



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ABSTRACT

The aim of this paper was to determine the capability for metal elution that may constitute potential contamination to the water environment from post-process cavern residue after underground coal gasification (UCG).

Samples of raw coals that had been subjected to gasification (three samples of hard coal, the coal mine "Bobrek", the experimental mine "Barbara" and the coal mine "Piast", and a sample of brown coal from the coal mine "Bełchatów") were analysed for their physicochemical properties. Similar tests were performed with samples of ash and char that remained in the cavern after experiments associated with underground coal gasification. For comparison, samples of raw coals were also burned under controlled conditions in air, in a specially designed furnace, until their complete incineration. Each of these materials was subjected to an elution test using deionised water.

An analysis of the physicochemical composition of the water extracts for the selected parameters was carried out. The qualitative–quantitative composition of the water extracts was related to the hydro-geochemical background characteristics of groundwater. The effect of the degree of coal conversion on the ability of metals to elute into the water environment was discussed.

1. Introduction

The origins of studies on the technology of underground coal gasification (UCG) date back to the 1930s, and their results have been presented in several review papers [1–4]. This process

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acquires energy through in situ coal bed gasification and consists in bringing the appropriate gasifying agent (air, oxygen, water vapour) to the ignited bed and then collecting the generated gas on the surface. Gas, a by-product of industrial value, can be used as synthesis gas or can be used to produce electricity and heat [5]. The process of underground coal gasification involves the use of this material with the lowest possible burden on the natural environment. One of the main advantages of UCG is the possibility of using deeply deposited coal beds or residual beds, which for economic or technological reasons are not suitable for operation using traditional methods [6].

Due to unique geological conditions, the diversified deposition of coal resources and the variability of the process itself, UCG is such a complex process that its implementation requires a number of detailed research studies and analyses, including studies on both the bed intended for gasification as well as its immediate surroundings before, during and after exploitation of the deposit. Particularly interesting and still poorly investigated is the determination of the potential risk of contamination of the natural water environment near quenched post-process caverns. This risk results directly from the coal gasification process, during which solid, liquid and gas products are generated. These products contain significant concentrations of organic and inorganic substances, including substances that are toxic and hazardous to the environment [7–12].

Any oxidation process (including gasification) involving hard coal or brown coal results in the generation of by-products in the form of ash, slag and char. The quality and quantity of the by-products generated in the gasification process depends on the quality and quantity of the mineral components of the combusted fuel, the gasifying agent used and the conditions under which the gasification process occurs. The main source of inorganic impurities is the eluate formed after contact with the residues of coal gasification, whereas the tar formed during the process is mainly responsible for the formation of organic pollutants and ammonia [4].

In general, the elements present in coal, depending on the weight of the dry mass of fuel, can be divided into major elements (forming the organic component of fuel with contents on the order of a few % by weight); ash-forming elements such as Al, Ca, Fe, K, Mg, Na, and Si (at concentrations ranging from approximately 1000 ppm to several weight%); and trace elements such as As, B, Cd, Cr, Hg, Pb, Se, Zn, Cl, and F (at concentrations below 1000 ppm) [13]. The organic matter in coal consists mainly of elements such as carbon, hydrogen, nitrogen, sulphur and oxygen in proportions that depend on the type and degree of coalification of coal.

The mineral content of solid fuels can be classified into two groups based on origin: syngenetic (internal, which accompanies the formation of coal beds from the beginning) and epigenetic (external, non-homogeneous mixture containing organic coal). During thermal processing, coal mineral matter undergoes profound physicochemical changes, resulting in the formation of, e.g., slags and ashes composed mainly of the oxides of Si, Al, Ti, Fe, Ca, Mg, Na, K, and P.

The trace elements present in coal have been studied by a number of authors to examine their behaviour at the different temperatures of pyrolysis, gasification and combustion; to determine their affinity for organic and inorganic substances; or to classify them with respect to their volatility or importance to the environment [14–19].

After the completion of the process of underground coal gasification, a portion of solid products such as char, ash and slag remains in the cavern where the quenching occurred, and the complete removal of these solid products is not possible. The reference literature presents studies whose aim was to determine how different metals behave under the conditions of underground coal gasification or pyrolysis [20–22]. Such studies, among others, have attempted to determine the feasibility of exploiting coal seams by means of underground coal gasification for commercial purposes [23]. However, there is a lack of research that provides a definite answer to the question regarding whether, after the UCG process, a cavern is a neutral place and does not pose a threat to the natural environment.

A report published by the EPA in 2009 [24] documents that from the residue stored after the thermal treatment of coal (from ash or sludge), harmful substances can be eluted at concentrations that pose a serious threat to human health. An elution test of 73 samples of coal ash conducted by the EPA showed that from some of the ashes under certain conditions, harmful substances can be released, whereby hundreds or even thousands of times the adopted standards for drinking water are exceeded, and some of these substances, such as arsenic or selenium, elute at the level classifying water as a dangerous waste substance. Water flowing into the post-reaction space in contact with post-process cavern residue may also be assumed, under certain circumstances, to elute significant amounts of toxic substances. Therefore, it is extremely important to monitor the surroundings of geo-reactors for the elution of pollutants into groundwater, either directly before or after gasification, as well as during a sufficiently long period after the termination of the process.

Safe storage of waste, including the residues of the underground coal gasification process, is a serious problem from a technical point of view and requires knowledge regarding not only the chemical composition of the pollutants themselves but also the extent of their elution. Because geo-reactors are situated in the vicinity of natural geological formations of varying structure and properties, there is always a risk that, despite proper prior hydrogeological examination, the thermal and mechanical factors accompanying the process of UCG significantly alter rock mass parameters and thus enable the migration of eluted substances into the environment (for example, with newly formed cracks, crevices or sinks). Because each waste remaining in the environment represents a potential threat, the possibility of quantifying the waste is even more important.

During UCG risk of water pollution in the vicinity of the site of operation is negligible, because the pressure of the process is lower than the pressure in the overburden and contaminants are retained inside the cavern. After completion of the process, groundwater may flow into the post-UCG area and have contact with the post-process residues, eluting impurities from waste and carrying them into the surrounding aquifers (on the principle of circulation of groundwater).

Due to the physicochemical transformations that waste is subject to with the passage of time in the above-described environment, it is important to find a proper method that would not only allow us to assess pollutants' susceptibility to elution and degree of toxicity but also determine the anticipated effects of long-term storage. A number of elution tests for solid waste have been developed. These include, among others, the "EP toxicity test" used in the United States; the Toxicity Characteristic Leaching Procedure (TCLP) method, based on US EPA Method 1311; the Synthetic Precipitation Leaching Procedure (SPLP) method, based on US EPA Method 1312 or the method recommended by the Swiss Office for the Protection of the Environment (BUS); and the elution test based on TVA AS.1991 for the determination of hazardous substances in waste. The last test enables, in a relatively objective way, the effects of the long-term storage of solid waste to be predicted.

In the case of UCG post-process waste, it is important to adopt the proper method for assessing elution. In many of the methods used to assess solid waste, the degree of elution and the type of chemicals eluted from UCG post-process cavern residues are crucial factors in determining the quality and safety of natural soil