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Research Article

Contaminant Formation and Release During Underground Coal Gasification Process at High Pressure Regime

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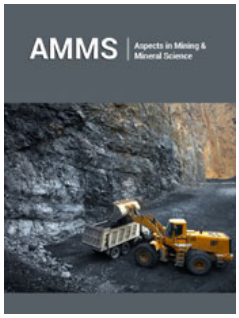

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Abstract

The use of underground coal gasification process for the recovery of deep underground seams for the production of methane-rich gas is now becoming a topical issue again. The protection of water resources, which could be contaminated with organic or inorganic pollutants during the washing of emerging products, is closely related to this issue. The article brings data on the elution of organic as well inorganic pollutants from samples obtained after the UCG process performed in ex-situ unit.

Keywords: Underground coal gasification; Groundwater contamination; Leaching tests; Emerging pollutants

Abbreviations: UCG: Underground Coal Gasification; DR: Dry Matter; MC: Moisture Content; GC: Gas Chromatography; MS: Mass Spectrometry

Introduction

At present, given the situation regarding energy resources, it can be assumed that some countries will be forced to continue using fossil fuels, including coal. This includes the possibility of using Underground Coal Gasification Process (UCG) [1,2] as a partial replacement for natural gas [3]. One of the most serious environmental problems related to UCG is contamination of groundwater, which can occur both at the production stage and after termination of the process. Therefore, the protection of water resources, which could be contaminated with organic or inorganic pollutants [4] by washing products after UCG, is also closely related to this issue. In the framework of the Study [5] ash and carbonaceous substances from the gasification of hard coal and lignite from an experimental simulation of the UCG process at 5 bars were used for leaching tests. The authors found that that most tested elements tend to be leached in a stronger degree from ash residues than from chars. From that reason the article is focused on testing the elution of individual pollutants from real patterns after the UCG process performed in ex-situ unit (Figure 1), however at higher pressures. The solid products forming during ex situ UCG process such as char and ash, which can be the secondary sources of contamination, were used for tests. Ash and char samples obtained after termination of the ex-situ UCG experiments accomplished in the Central Mining Institute (GIG), Poland, where the block of hard coal "Wesoła" was gasified at defined reaction conditions, were used for leaching tests.

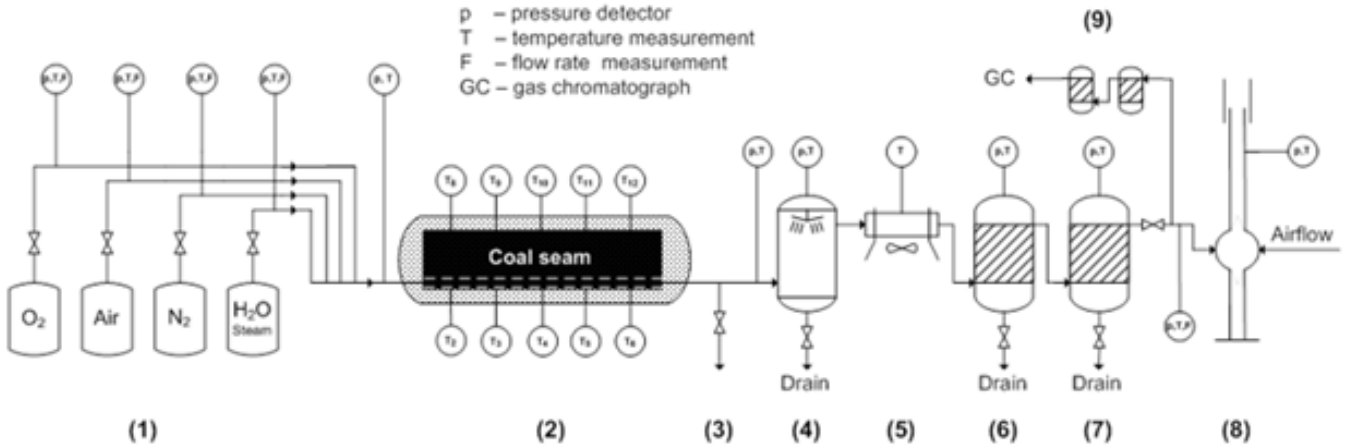

Figure 1: Scheme of the high-pressure UCG reactor: (1) gasification reagents, (2) large-scale gasification reactor, (3) sampling valve, (4) water scrubber, (5) air cooler, (6,7) separators, (8) thermal combustor, (9) gas treatment module prior to GC analysis.

Experimental

UCG experiments were proceed under two different pressure levels corresponding to 20 and 40 bars, respectively. Then, post-gasification solid residues were sampled along the reactor length – 1m, 2m and 3m from the reactor inlet. The ash and char set taken from the reactor at 3m was utilized for the selected contaminant leaching tests. The elemental analysis of delivered ash and char samples was accomplished by the method of AAS at GIG, Poland. The leaching tests were performed according to the Czech version of the European Standard [5] was utilized for making of leaching tests [6]. For determination of dry matter, the samples were weighted before and after drying. The drying took place at 105±5 °C.

Results and Discussion

The elemental content of the selected metal in coal sample is summarized in Table 1. The Dry Matter (DR) and Moisture Content (MC), both parameters are expressed in %, were then calculated. The statistical error of the determination was assessed as ±0.1% (Table 2). The number of selected metals in ash or char samples can be considered as independent of used reaction pressure. The content of Pb or Ni in ash samples is relatively high for the sample proceed at pressure 20 bar, the concentration of other elements is comparable. The high amount of carbon remains in both ash samples after burning, more than 50%.

Table 1: The elemental content of the ash and char samples produced through ex situ UCG of hard coal "Wesoła" at pressure 20 or 40 bars, sampled 3m from the reactor inlet.

Microelements mg/kg (ppm)	Ash		Char	
	20 bar	40 bar	20 bar	40 bar
As	8	8	2	1.8
B	143	118	19	14
Cd	<1	<1	<1	<1
Co	4	3	<1	<1
Cr	37	46	<1	<1
Cu	120	89	19	8.5
Hg	<0.01	<0.01	<0.01	<0.01
Mn	1150	1178	75	14.3
Mo	30	19	<1	<1
Ni	21	23	1	<1
Pb	41	36	<1	<1
Sb	203	129	<1	<1
Se	9	<2	<2	<2
Zn	229	153	5	1.9
Macroelements (wt. %)	**Ash**		**Char**	

Al	2.24	2.29	0.08	0.05
Fe	4.37	2.80	0.26	0.11
K	0.170	0.140	0.002	0.001
Ti	0.030	0.040	0.003	0.001
Macroelements (wt. %)	**Ash**		**Char**	
Al	7.90	5.00	0.51	0.48
Fe	1.63	0.48	0.04	0.11
K	0.120	0.120	0.013	0.008
Ti	0.130	0.090	0.006	0.003

Table 2: Dry matter (DR) and moisture content (MC) of the ash and char samples, hard coal 'Wesoła'.

Sample	Pressure (bar)	DR (%)	MC (%)
Ash	20	96.9	3.1
	40	96.8	3.2
Char	20	95.7	4.3
	40	95.9	4.1

Realization of leaching tests

One stage batch test at a liquid to solid ratio of 10L/kg±2% for both dried ash and char was accomplished. The char samples were crushed and sieved before leaching (particles <10mm), the ash samples were used without crushing and sieving. The sealed tubes filled by tested powder material were placed on the shaker. The leachability verification tests were carried out during 24 hours by continual mixing at 22±5 °C on the shaker. Then, the suspended solid phase was allowed to settle. After ~15 minutes, the liquid phase was separated from solid one by filtration through 0.45μm membrane filter (Whatmann membrane). The filtration was performed using the water pump which typically operates under pressure < 50kPa. All samples were filtered within 35 minutes. The obtained leachate containing metal and organic contaminants was analysed with aid of the AAS, mercury analyzation, and advanced capillary Gas Chromatography (GC) coupled with Mass Spectrometry (MS). The results for both tested post-gasification residua are summarized in Tables 3 & 4. Analyses of the filtrate obtained by the leaching test gave the concentration c of the individual contaminant in the filtrate (mg/L). Final results were expressed as the amount of leached component relative to the total weight of the sampled dry matter A in mg/kg calculated after A=c(V/MD+MC/(100), where V is the volume of leaching liquid used, MD is the mass of the dried analytical sample (kg), and MC is the moisture content (%).

Table 3: The concentration of selected substances leached from the ash or char samples after ex situ UCG process taken at 3m from reactor input (mg/L filtrate). – stands for concentration under the detection limit.

Hard Coal, 'Wesoła" Mine, Poland								
Sample	**Cl^-**	**SO_4^{2-}**	**Hg^{2+}**	**Cd^{2+}**	**As^{3+}**	**Zn^{2+}**	**Naph**	**Phen**
Ash, 20 bar	1000	3730	0.017	–	–	0.050	–	–
Ash, 40 bar	607	2250	0.022	–	–	0.008	–	–
Char, 20 bar	122	–	–	–	–	0.005	–	–
	111	–	–	–	–	0.017	–	–
Char, 40 bar	156	–	–	–	–	0.023	–	–
	116	11.5	–	–	–	0.067	–	–

Table 4: The concentration of selected substances leached from the ash or char samples after ex situ UCG process taken at 3m from reactor input (mg/kg dry matter). – stands for concentration under the detection limit.

Hard Coal, 'Wesoła" Mine, Poland								
Sample	**Cl^-**	**SO_4^{2-}**	**Hg^{2+}**	**Cd^{2+}**	**As^{3+}**	**Zn^{2+}**	**Naph**	**Phen**
Ash, 20 bar	9918	36993	0.169	–	–	0.500	–	–
Ash, 40 bar	6124	22699	0.222	–	–	0.076	–	–
Char, 20 bar	1228	–	–	–	–	0.050	–	–
	1115	–	–	–	–	0.171	–	–
Char, 40 bar	1572	–	–	–	–	0.229	–	–
	1162	115	–	–	–	0.670	–	–

Conclusion

The leaching studies revealed that the dominant inorganic compounds involve chlorides as well as sulphates. Both ash samples were characterized by higher concentrations of leachable zinc-and mercury-based compounds. On the contrary, cadmium and arsenic species were found in ash samples in the trace amounts approaching the limits of analytical methods used. Neither Cd nor As were found in the leachates obtained both from the tested ash and char. Because arsenic was previously determined in all tested parent ash and char in quantities significantly exceeding its detection limit, it confirms that the leaching capacity of As to the aqueous phase is very low. The concentrations of organic aromatic contaminants, represented by naphthalene (Naph) and phenanthrene (Phen) were found to be below detection limits for all tested leached samples. This can be explained by very high volatility of naphthalene and phenanthrene during UCG due to sublimation process. Tests have shown that in addition to chloride and sulphate ions, the extracts also contained heavy metals, namely mercury and zinc, but in small concentrations. For these reasons, it would be appropriate to consider in the future the possible installation of underground reaction barriers.

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