



## Full Length Article

## Study of properties of tar obtained from underground coal gasification trials

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## ARTICLE INFO

## Keywords:

Tar  
Hard coal  
Underground coal gasification  
UCG

## ABSTRACT

This work presents the results of an experimental study on the properties of tar that originated from underground coal gasification (UCG) trials. Two in situ experiments of UCG were performed using oxygen as the gasification agent under the HUGE and HUGE 2 projects. Trials were conducted using different configurations of the fire channel, gasification time and method of raw gas collection and purification. The obtained tars were analysed over a wide range of parameters that are typical for the assessment of the quality of conventional coke oven tar. The results of the analysis showed that in terms of physical–chemical properties, both tars are highly different from each other, as well as from typical coal tar from a coking plant. One characteristic observation of the UCG-derived tars is a higher content of ash as well as toluene and quinoline insolubles compared with a typical coke oven tar. Both tars are susceptible to thermal destruction and degradation to lighter products, and the characteristics of the coking distillation residue after heating to high temperatures is different from coke oven tar. The tested tars vary in boiling range and fractional composition. The analysis also showed that tars from UCG contain more heterocyclic compounds than does coke oven tar, and the tar from HUGE is heavier than tar from HUGE 2. The content of PAHs in the tar from HUGE is many times higher than in tar from HUGE 2 and at a similar level as coke oven tar; however, the naphthalene content in both UCG tars is many times lower compared to typical coke oven tar. The probable reasons for the differences described in this report include the tar residence time in the sampling point, as well as different extents of tar fractionation phenomena, which resulted from the distinct configurations of the gas transportation and purification systems.

## 1. Introduction

Underground coal gasification is the process of coal conversion into combustible gas which main components are hydrogen, methane and carbon monoxide [1–4]. This process is a combination of several unit processes, such as drying, pyrolysis, gasification and combustion. In addition to UCG gas, liquid tar products are also obtained, which are undesirable but unavoidable contamination products of the process gas. Tar is generated the most intensively at the beginning of the gasification process, and as the gasification progresses, the amount of tar is reduced; however, depending on the process conditions, the raw gas always contains a certain amounts of tar. The results of American and Russian studies [5,6] show that the contents of the tar in raw gas may be in the range of 0.5 to 15.5 g/m<sup>3</sup>, depending on the process conditions. For this reason, the gasification conditions are chosen such that the tar content in the resulting gas is not too high [7]. The generation of tar starts at a temperature of approximately 350 °C and ends at approximately 1000 °C. Because tar is formed primarily from the thermal transformations of volatile compounds, the higher the content of

volatile matter in coal is, the more tar is produced [8]. With an increase in temperature and residence time, the resulting tar undergoes secondary reactions (cracking, partial combustion) that change its properties and yield [9–11]. The gasification temperature depends on the oxygen concentration and its supply rates in the gasified medium; hence, when air is used, lower temperatures are achieved compared to pure oxygen. An important factor influencing the tar quantity and composition is also the configuration of the UCG gas collection system. As a result of the tar fractionation phenomena, the longer the output pipeline is, and the lower the temperature inside is, the more tar is condensed [12,13]. This phenomenon may cause problems with collection of the process gas because condensed tar mixed with water and dust may reduce the cross-section of the output pipeline, and in extreme cases, it can completely block the production well [10,14]. To prevent this outcome, the process gas temperature should be maintained at the appropriate level. In studies on UCG, depending on the type of installation used, various process gas temperatures have been reported. A Russian study [10] shows that the best operating temperature range for syngas cooling is between 100 and 200 °C. During American trials,

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Hanna [15,16], Hanna II, IVa, and b [17] the process gas temperature was significantly higher and amounted to approximately 140–370 °C. In a Chinchilla project, the temperature of the process gas was approximately 300 °C [18], and during the gasification of coal in Swan Hills, Alberta (Canada), the syngas temperature leaving the production well was 50 °C or less [19].

For further downstream gas applications, tar should be effectively removed [10,20,21] using standard physico-chemical methods. The installation of tar and water separation should include the following elements: coolers, decanters, electrostatic precipitators, absorbers, cyclones, and centrifuges. The elements should provide a maximum level of tar and water removal from the gas, as well as removal of solid particles that consist mainly of dust and ash. The separated tar can be subjected to further processing for economically valuable products, e.g., by hydrocracking [6] or reforming [22]. An alternative method of utilizing UCG tar may incorporate it into a stream of other processed products with similar properties. From mass market products, the most available are coke oven tar and heavy fuel oils. However, the high content of ash may be a major obstacle. If tar from UCG contains more than 0.1% ash, adding it to coke oven tar can significantly degrade its quality because the ash content is one of the most crucial parameters that determines the quality of the coke oven tar. When the level of the ash content in coke oven tar is well below 0.1%, adding tar with increased ash content even in relatively small quantities is unacceptable because it negatively affects the quality of coal tar, which is the primary coke oven tar distillation product. When trying to use these tars as heavy fuel oil components, in addition to the high ash content, which can cause clogging of the nozzles in the burners, mixing with the fuel oil can also be a problem and can create a homogeneous mixture. The most favourable option would be to minimize the amount of tar produced in the underground coal gasification process; therefore, a small amount of tar could be treated as waste and utilized, e.g., in a waste incineration plant. This approach would avoid problems resulting from the REACH system concerning the registration, assessment and authorization of chemical substances in the European Union [23].

This paper describes the physicochemical properties of tars obtained from two underground coal gasification tests. The gasifications of hard coal were performed using oxygen as the gasification agent. Both UCG trials were conducted with different configuration of the fire channel, gasification times and methods of raw gas collection. The tar samples were analysed using standard methods for the assessment of typical coke oven tar. The obtained results were compared with literature data on the properties of typical coke oven tar.

## 2. Experimental

### 2.1. Description of the installation for coal gasification

The tested tars were obtained during two in-situ underground coal gasification experiments during the projects of HUGE and HUGE 2 [24,25]. These experiments were conducted in the experimental “Barbara” mine in southern Poland. The UCG reactors were located in coal seam No. 310 at a depth of 30 m. The thickness of the coal seam was 1.5 m. The results of the coal analysis used for gasification are presented in Table 1. The analyses were performed in a certified laboratory in the Central Mining Institute according to Polish Standards. In both experiments, the gasified coal was characterized using the same physico-chemical properties.

The schemes of the used installations are shown in Figs. 1a,b and 2a,b and the most significant results of these trials are shown in Tables 2 and 3.

The HUGE trial was performed in the reactor equipped with a straight fire channel (Table 2) with a length of 15 m, whereas for the trial HUGE 2, the fire channel was the V-shape type with a length of 2 × 17.3 m. The duration of the HUGE and HUGE 2 trials were 355 and 142 h, respectively. In both trials, the main gasification agent was

**Table 1**  
Proximate and ultimate characteristics of the coal from the “Barbara” mine.

|                     |               | Raw coal from the<br>“Barbara” mine | Standard                            |
|---------------------|---------------|-------------------------------------|-------------------------------------|
| <i>As received:</i> |               |                                     |                                     |
| Moisture            | $W^r$ (%)     | 11.81                               | PN-G-04511:1980                     |
| Ash                 | $A^r$ (%)     | 15.56                               | PN-G-04560:1998<br>PN-ISO 1171:2002 |
| Sulphur total       | $S^r$ (%)     | 0.51                                | PN-G-04584:2001<br>PN-ISO 334:1997  |
| Lower heating value | $Q^r$ (kJ/kg) | 21,708                              | PN-G-04513:1981                     |
| <i>Analytical:</i>  |               |                                     |                                     |
| Moisture            | $W^a$ (%)     | 6.39                                | PN-G-04511:1980                     |
| Ash                 | $A^a$ (%)     | 16.52                               | PN-G-04560:1998<br>PN-ISO 1171:2002 |
| Volatile matter     | $V^a$ (%)     | 29.84                               | PN-G-04516:1998<br>PN ISO-562:2000  |
| Lower heating value | $Q^a$ (kJ/kg) | 23,192                              | PN-G-04513:1981                     |
| Carbon              | $C^a$ (%)     | 57.95                               | PN-G-04571:1998                     |
| Hydrogen            | $H^a$ (%)     | 3.70                                | PN-G-04571:1998                     |
| Nitrogen            | $N^a$ (%)     | 0.87                                | PN-G-04571:1998                     |
| Sulphur total       | $S^a$ (%)     | 0.54                                | PN-G-04584:2001<br>PN-ISO 334:1997  |
| Sulphur combustible | $S_c^a$ (%)   | 0.54                                | PN-G-04584:2001                     |
| Oxygen              | $O^a$ (%)     | 14.03                               | PN-ISO 334:1997                     |

oxygen. Due to an uncontrolled inflow of air into the reactor, the average calorific value of the process gas for HUGE trial was approximately two times lower.

The data presented in Table 3 show that the conditions under which the tars remained before sampling until analysis were considerably different from each other, which could have significantly affected their properties.

The HUGE tar was sampled from the flame breaker (Fig. 1b), whereas the tar from HUGE 2 was sampled from the tar separator (Fig. 2b). The residence time of the HUGE tar sample in the flame breaker was two days, whereas for the tar sample from HUGE 2 in the tar separator, it was only several hours. Significant differences also existed between the temperature in the places of tar sampling, lengths of pipelines and gas cooling system used. The temperature in the flame breaker (HUGE tar) was approximately three times higher than in the tar separator (HUGE 2 tar). The output pipeline with the process gases for the HUGE trial was not cooled in the underground part of installation, whereas the pipeline for the HUGE 2 trial was equipped with a 15-meter-long water cooler. In addition, the length of the output pipeline for the HUGE trial was two times less compared with HUGE 2.

### 2.2. Preparation of tars for analysis

The tars obtained from the UCG trials visually differed from each other, and their properties were different from conventional coke oven tar. The tar from HUGE at room temperature was a black, messy and smelly plastic mass similar to glue. The tar from experiment HUGE 2 was a black viscous liquid with a faint odour of carbochemical products and visible inclusions of dust and high-molecular-weight agglomerates. Before performing the analyses, both tars were pre-homogenized and averaged by gentle heating in a drier and by thoroughly mixing.

The tar from HUGE at 50 °C had no tendency to melt or increase liquidity, which enabled its averaging. To avoid evaporation of the light fractions and changes in the other properties, the sample was not heated to a higher temperature. This tar was treated similarly as a typical coal tar from coking plant and was prepared for analysis by applying methods of homogenization and averaging according to standards pertaining to coal tar. The HUGE 2 tar did not pose such problems as the HUGE tar, and was homogenized and averaged by carefully mixing

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