



Comparative mineralogical study of thermally-altered coal-dump waste, natural rocks and the products of laboratory heating experiments



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ABSTRACT

Research on rocks formed due to pyrometamorphism of waste in burning coal-mine dumps (BCMD) mainly in the Upper Silesian Coal Basin has enabled identification of a large number of different mineral species. These species are usually well-known minerals, e.g., olivines, plagioclases and clinopyroxenes. However, their crystal chemistry is often unique. Mineralogical- and chemical similarities between the BCMD and non-anthropogenic geological environments are outlined here. To better understand the crystallization processes of the minerals occurring in the BCMD, three types of heating experiments were performed. For these, ten protolith (thermally-unchanged) dump samples, mostly shales and carbonate rocks, were heated alone and mixed together and with a CaF₂ flux. Quantitative chemical analyses of the synthesized mixtures have shown that they are mineralogically similar to the rocks found in the BCMD. They are also similar in terms of their crystal chemistry, e.g., synthesized clinopyroxenes are rich in diopside and esseneite components and may capture phosphorus, plagioclase is rich in anorthite and contains iron and magnesium, and wüstite exists as a solid solution with periclase and is doped with calcium and other elements. Highly variable amounts of indialite–ferroindialite were formed in some samples due to solid-phase transformations or melt crystallization, depending on the experimental conditions and the protolith used.

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1. Introduction

Burning coal-mine dumps (BCMD) are known worldwide. They contain remnants of coal and coexisting sedimentary rocks discarded as mining waste. BCMD are sites at which significant mineralisation occurs due to fire-induced transformation of the waste sedimentary rocks and the coal itself. Of these transformations, high-temperature pyrometamorphism – contact metamorphism due to fossil fuel fires – is one of the most widely recognized phenomena. Although the BCMD are of anthropogenic origin, the mineral-forming processes occurring in them were caused naturally by the self-combustion of the coal remnants.

This paper describes the physicochemical features of the most important pyrometamorphic minerals found in five BCMD in the Upper Silesian Coal Basin (USCB; Fig. 1). Some samples for this research were part of the Ph.D. study of Kruszewski (2009). The choice of mineral species described was based on their chemical analogy to species known from geological environments. The work also involved heating experiments conducted to better understand the processes responsible for mineral formation. Preliminary results have been reported in abstract

form by Ciesielczuk et al. (2013) and Kruszewski and Ciesielczuk (2013). For comparison, some rock samples from the Lower Silesian Coal Basin (LSCB) were also investigated. The main goal of the study was to compare the BCMD environments with natural geological analogues in terms of their mineralogy and mineral-forming processes. Thermal experiments were conducted, under known conditions, to imitate mineral-forming processes to better understand them.

1.1. Coal fires in dumps

Spontaneous coal combustion is caused by oxidation of both coal and coal-disseminated sulphide minerals, the latter being of catalytic importance (Srebrodolskiy, 1989). Important internal factors influencing the initiation of coal combustion include coal petrography (maceral composition), coal structure and coal rank (Cebulak et al., 2005; Mastalerz et al., 2011; Sawicki, 2004; Wagner, 1980). The active surfaces of some microlithotypes allow them to accumulate heat induced due to initial coal oxidation, thus making them catalyzers, i.e., so-called fire-cores. Machnikowska et al. (2003) proved, that vitrinite is the maceral group which undergoes oxidation prior to other macerals. Iron sulphides, especially pyrite that is common in coal beds, also act as combustion catalysts. Their oxidation provides an additional heat source. The high porosity and granularity of waste rocks, the high

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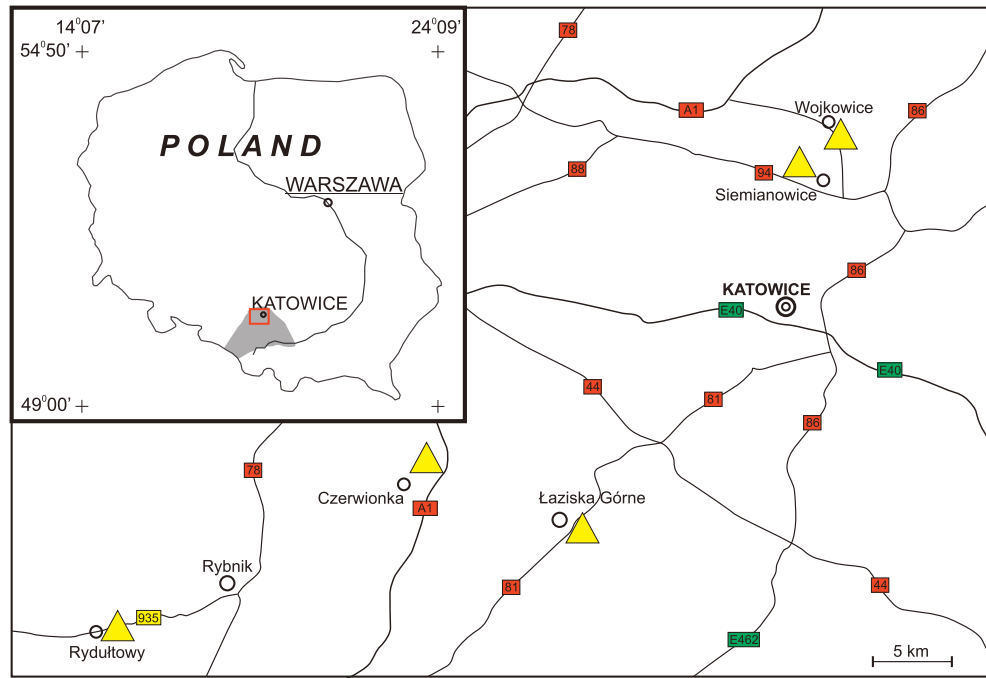


Fig. 1. Location of the coal waste dumps (yellow triangles) on a sketch map of the Upper Silesian Coal Basin (grey area). Red rectangle indicates the dump locations in Poland.

content of ash in fuel, and humidity, are additional internal factors. An important external factor is rainwater serving to transport oxygen. Rainwater also contributes to crack formation and, thus, facilitates oxygen penetration (Panov et al., 1999; Srebrodolskiy, 1989; Świętosławski, 1953; Wagner, 1980). As fire gases from degassing coal ascend, their interaction with air triggers secondary combustion above initial fire centres. Fire gas transfer is an important heat redistribution factor and is crucial for thermal transformation of waste. Combustion in dumps, a cyclic process, may be summarized as follows: combustion → gas explosion → rock crushing → cooling → protolith volume loss → development of cracks → O₂ flux → formation of fire → combustion (Sokol et al., 2005).

Self heating and fire-related processes, including mineralization, are seen in many dumps in the Upper and Lower Silesian Coal Basins. Research already performed on USCB material includes studies of the heating itself (e.g., Wagner, 1980), transformations of organic matter (Misz-Kennan and Fabiańska, 2011) and mineralogy (Ciesielczuk, 2008; Ciesielczuk et al., 2010; Kruszewski, 2006, 2009, 2013; Parafiniuk and Kruszewski, 2009, 2010) and gas-phase chemistry (Fabiańska et al., 2013). In contrast, data on the mineralogy of LSCB dumps is limited (Fabiańska et al., 2013; Kruszewski et al., 2012; Wielogórski et al., 1975).

1.2. Mineral-forming stages in burning coal-mining dumps

The formation of minerals in BCMD involves three stages: (1) a burnout stage – a high-temperature, metamorphic stage (ca. 330–1200 °C); (2) exhalative stage – a mid-temperature stage (typically 50–450 °C, rarely more), and (3) a low-temperature supergene stage (Kruszewski, 2009; Nasdala and Pekov, 1993; Sokol et al., 2005; Srebrodolskiy, 1989). The upper T limit of the last stage is sometimes given as ca. 50 °C (Srebrodolskiy, 1989). During the burnout stage, mineral phase oxidation, dehydration, mineral structure recrystallization, formation of metastable phases, formation of high-temperature minerals and, finally, melt crystallization may occur (Ciesielczuk et al., 2014). Exhalative-stage minerals may originate from condensing gases on dump surfaces, from acidic solutions formed as a result of gas liquifaction (a hydrothermal process) or, in sub-surface areas, be

associated with the extraction of metals from waste rocks by aggressive NH₃-bearing, S-rich gases (a pneumatolytic/hydrothermal process). At the supergene stage, minerals crystallize due to evaporation of acidic solutions formed at the surface due to oxidation of S-rich gases and pyrite (Kruszewski, 2009, 2013).

1.3. Fire-induced metamorphism of waste

The burnout stage is responsible for a marked transformation of the waste rocks, including the coal itself. The action of high temperatures at more or less atmospheric pressure allows this transformation to be classified as pyrometamorphism (e.g., Grapes, 2006; Sokol et al., 2005). Depending on the temperature and lithology of the protolith, the pyrometamorphism leads to partial, or total, melting and degassing of the waste rocks. Total melting, catalyzed by the presence of fluxes such as simple oxides, especially FeO in carbonate waste rocks, and fluorine derived from decomposing clay minerals of the coal-bearing shales, leads to the formation of melts which, on cooling, partial recrystallization and vitrification, give rise to so-called parabasalts (paralavas and scoria). However, due to the very dynamic conditions in the dump interior, parabasalts are relatively rare. Much more common among the newly-formed rocks are metapelites, especially clinkers formed from claystones and siltstones, and buchites derived from sandstone protoliths (Sokol et al., 2002, 2005). Mineral transformation processes taking place at the burnout stage include (1) dehydration and structural degradation at 500–650 °C of shale aluminosilicate minerals, mainly kaolinite, illite, and chlorites, followed by (2) thermal dissociation of carbonates at 600–750 °C, (3) local melting of some aluminosilicate minerals at 700 °C, (4) sanidinization of microcline at 800 °C and (5) intensive melting of the inner parts of the waste pile at temperatures < 1200 °C (Filippidis et al., 1996; Reifenstein et al., 1999; Sokol et al., 2005, 2002; Ward, 2002). At such high temperatures, carbonate decomposition products, e.g., lime (CaO) and periclase (MgO), may react with hot S-, Cl- and F-bearing gases to give oldhamite (CaS), anhydrite (CaSO₄), and F-S-Si-rich apatite-group minerals and fluorides. The latter serve to lower the temperatures of melt generation. Later, cooling melts give silicate and oxide minerals, and glass (Sokol

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