Mineralogy of thermally altered coal mining residues in self-burning coal waste piles in Portugal and Spain

Mineralogia de resíduos da exploração mineira de carvão termicamente alterados em escombreiras em auto-combustão em Portugal e Espanha

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Abstract: The environmental risks associated with coal mining activities include, among others, the production of solid wastes. Self-burning coal waste piles may present serious potential environmental and human health hazards. The geological characterization of waste materials in self-burning waste piles is essential in order to identify the related negative impacts and to further contribute to their mitigation. The main goal of this study is to identify the mineralogical composition of burning or already burnt and unburned coal waste material from self-burning anthracite waste piles and the changes attributed to the combustion process. X-ray powder diffraction was used for the determination of the mineralogical composition of relevant coal waste materials. The results show that illite+mica and quartz are the main constituents of the unburned material, together with minor proportions of muscovite, kaolinite, chlorite, pyrophyllite, anatase, gypsum, and rutile. However, other minerals, such as mullite, cristobalite, hematite, jarosite, anorthite, were identified in burning/burnt waste samples. The formation of these minerals is attributed to the combustion process, and suggests that they may have reached temperatures of at least 1000°C.

Keywords: Mineralogy, Anthracite, Coal mining residues, Self-combustion, Thermal changes.

Introduction and objectives

Despite the benefits to the economic and social sectors of many countries, the activities associated with coal mining and coal consumption may cause significant negative impacts to the environment and human health (Orem & Finkelman, 2004; Finkelman, 2004; Suárez-Ruiz & Crelling, 2008; Suárez-Ruiz et al., 2012). The environmental risks associated with coal mining include the production of solid wastes, essentially composed of overburden and other discarded materials resulting from mining activities. The disposal of such wastes may lead to: atmospheric dispersion of particles, self-heating, spontaneous combustion, self-burning, landslides and mass movements, leaching of toxic elements, and formation of acid drainage caused by weathering and/or oxidation processes. The uncontrolled release of pollutants (such as greenhouse gases, particulate matter, organic compounds, toxic trace elements) from self-burning coal wastes present serious potential environmental and human health hazards, especially if the coal waste piles are located close to urban centers (Finkelman, 2004; Pone et al., 2007; Finkelman & Stracher, 2011). Comprehensive geological characterization of materials in self-burning waste piles is essential in order to identify the related environmental and human health impacts, and to contribute where possible to their mitigation (Ribeiro et al., 2010a, 2010b; Misz-Kennan & Fabiánska, 2011; Ribeiro et al., 2012).

Lower rank coals, such as lignites and subbituminous coals are, generally, much more prone to self-heating than higher rank coals (Taylor et al., 1998). However, self-burning coal waste piles resulting from mining of...
high rank coals (anthracite) have been identified in Portugal (Douro Coalfield) and in Spain (El Bierzo Coalfield). The S. Pedro da Cova, Lomba, and Midões self-burning waste piles resulted from past anthracite mining in the Douro Coalfield (Portugal), with the original ignition caused by forest fires. The Fabero and Arroyo Galladas coal waste piles result from past anthracite mining in the El Bierzo Coalfield (Spain); the combustion process was initiated by oxidation of the organic matter, and also of the pyrite in the coal, with the latter producing acid waters as well. Mining operations have completely ceased in both cases.

Previous research on self-burning of the Douro Coalfield waste piles addressed the petrographic and geochemical characterization of the waste materials (Ribeiro et al., 2010a; Ribeiro et al., 2013). The aim of this study is to add data on the self-burning of high rank coals (anthracites) and the changes taking place during the combustion process, since this phenomenon is more frequent in lower rank coals. The specific objectives of this work are: (i) to determine the mineralogical composition of burning or already burnt and unburned coal waste materials from anthracite self-burning coal waste piles in Portugal and Spain; (ii) to identify the mineralogical changes and influences attributed to the combustion process.

2. Materials and methods

The coal waste piles sampled were the S. Pedro da Cova, Lomba and Midões waste piles in the Douro Coalfield (Portugal) and the Fabero and Arroyo Galladas waste piles in the El Bierzo Coalfield (Spain). In addition to burning or already burnt materials in each waste pile, samples of unburned material were also collected.

The samples were collected close to the surface (up to 20 cm depth). They were homogenized, dried and quartered in the laboratory to obtain representative sub-samples and were then crushed (< 212 µm).

The methodologies applied to this study were optical microscopy and X-ray powder diffraction (XRD). The microscopic observations were performed in whole-rock polished blocks prepared according to standard procedures (ISO 7404-2, 2009) and were done using a Leitz Orthoplan microscope equipped with a Discus-Fossil system under standard conditions. For the determination of the mineralogical composition, the samples were analyzed by XRD using a Philips PW-1830 diffractometer with Cu K-alpha radiation. Quantitative analyses of mineral phases in each sample were made from the X-ray diffractograms using Siroquant™, commercial interpretation software based on the Rietveld XRD analysis technique. The proportions of amorphous material in the burnt samples were determined from the XRD patterns (Ward & French, 2006).

3. Results and discussion

Mineralogical analyses were performed on 3 samples of unburned coal waste material and on 11 samples of burning or already burnt coal waste material. The results obtained are presented in Table 1.

The results show that illite+mica and quartz are the main constituents of the unburned material. Significant proportions of muscovite, kaolinite, chloride, and pyrophyllite are also present in these samples, along with minor proportions of anatase, gypsum, and rutile. Such a mineralogical composition is as expected, considering that: (i) the waste studied is a mixture of coal and the surrounding lithologies of the coal seams (essentially lithic arenites and carbonaceous shales in both Coalfields); and (ii) these phases are among the most common minerals found in coals (Ward, 2002 and references therein).

The minerals found in the samples from the unburned zones are also expected to have been originally present in the burning or burnt samples, as well. However, a range of other minerals were also identified in the burning/burnt samples, mainly anhydrite, anorthite, coquimbite, cordierite, cristobalite, hematite, jarosite, mullite, rozenite, sanidine, sulfur, and tschermigite+boussingaultite, and also amorphous material, as indicated in Table 1. The formation of these minerals is attributed to the combustion process since most of these new minerals, such as hematite, jarosite, mullite, cristobalite, as well as the amorphous material, are recognized as mineral phases commonly formed from the thermal decomposition of minerals during coal combustion (Vassilev & Vassileva, 1996; Saxby, 2000; French et al., 2001; Ward, 2002). Figure 1 provides a general view of the mineralogical constituents of the burning/burnt waste samples from petrographic observations.

French et al. (2001) present results from thermomechanical analysis combined with dynamic high-temperature X-ray diffraction that show the relative abundance of the different mineral and amorphous phases developed when coal mineral matter is heated progressively in an oxidizing atmosphere to more than 1500°C. These authors have shown that kaolinite begins to disappear at around 600°C, being replaced by amorphous material. Mullite and cristobalite begin to form at around 1000°C, accompanied by a decrease in the amount of amorphous material that resulted from the thermal decomposition of the kaolinite. It was also observed that anorthite may form in Ca-bearing materials at around 1200°C.

Considering the observations above in conjunction with the results obtained in the present study, especially the general decrease in the abundance of kaolinite, the presence of amorphous material, and the occurrence of mullite, cristobalite, and anorthite only in the burning/burnt material, it can be concluded that the combustion temperature in the coal waste piles reached temperatures of around 1000°C or even more. These temperatures were reached only in some areas of the waste piles, as the self-burning process is very heterogeneous within the coal waste materials.
Table 1. Mineralogical composition of coal waste piles material from unburned and burning/burnt zones (values in wt. %).

<table>
<thead>
<tr>
<th>Mineral</th>
<th>Unburned waste material</th>
<th>Burning/burnt waste material</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>DC  S 34, L 68, M 11</td>
<td>EB  F 1, G 1, G 2, G 3, G 4, G 5, G 6</td>
</tr>
<tr>
<td>Anatase</td>
<td>- 0.6</td>
<td>- 2.7</td>
</tr>
<tr>
<td>Chlorite</td>
<td>- 1.0</td>
<td>- 1.8</td>
</tr>
<tr>
<td>Gypsum</td>
<td>- 4.5</td>
<td>- 2.1</td>
</tr>
<tr>
<td>Illite +</td>
<td>- 12.9</td>
<td>- 17.8</td>
</tr>
<tr>
<td>Kaolinite</td>
<td>- 17.8</td>
<td>- 25.5</td>
</tr>
<tr>
<td>Muscovite</td>
<td>- 21.3</td>
<td>- 31.3</td>
</tr>
<tr>
<td>Pyrophyllite</td>
<td>- 6.6</td>
<td>- 2.7</td>
</tr>
<tr>
<td>Quartz</td>
<td>- 13.7</td>
<td>- 18.7</td>
</tr>
<tr>
<td>Rutile</td>
<td>- 0.5</td>
<td>- 0.4</td>
</tr>
</tbody>
</table>

DC - Douro Coalfield (Portugal); EB - El Bierzo Coalfield (Spain); S - S. Pedro da Cova waste pile; L - Lomba waste pile; F - Fabero; M - Midões waste pile; G - Arroyo Galladas waste pile.

Fig. 1. Petrographic images of the mineralogical constituents of burning/burnt material from coal waste piles in the Douro Coalfield (A and B) and the El Bierzo Coalfield (C and D), showing the presence of iron oxides (Fe) and other lithic fragments (LF) made up of quartz and clays associated with organic matter (OM).

4. Conclusions

The assessment of the mineralogical composition of material from self-burning coal waste pile was the main goal of this research. This study provides comprehensive evidence of the mineralogical changes that have occurred as a result of combustion processes in the self-burning coal waste piles of the Douro and El Bierzo Coalfields. The results indicate the formation of amorphous material, along with cristobalite, mullite, anorthite, hematite, and jarosite, among other phases, due to combustion in the coal waste piles, suggesting that temperatures of 1000°C were attained.

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