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ANALYSIS OF THE IMPACT OF PHYSICOCHEMICAL PARAMETERS CHARACTERIZING COAL MINE WASTE ON THE INITIALIZATION OF SELF-IGNITION PROCESS WITH APPLICATION OF CLUSTER ANALYSIS

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ABSTRACT

Purpose	The subject of the research presented in this paper is the analysis of physicochemical parameters characterizing coal mine waste, in terms of their impact on the initialization of the self-ignition phenomenon.	
Methods	The model was constructed with the application of Hierarchical Cluster Analysis complemented with a colour data map enabling the tracing of similarities between the samples of coal mine waste in the space of parameters and between the examined physicochemical parameters in the space of samples. The data set analysed included parameters characterizing coal mine waste collected from 12 various coal mine waste dumps, either in operation or closed, and where thermal effects either took place or were not reported.	
Results	The HCA model constructed and complemented with a colour data map revealed that the tendency of coal mine waste to self-ignite is primarily affected by the contents of moisture, ash, volatile matter, C and S, values of heat of combustion, calorific value and contents of SiO ₂ , Al ₂ O ₃ , K ₂ O, SO ₃ , TiO ₂ , Co, Ni and Rb.	
Practical implications	One of the major environmental hazards associated with the storage of coal mine waste is the possibility of self-ignition. At present, there are no applicable methods of assessment of this risk. The application of Hierarchical Clustering Analysis complemented with a colour data map enabled the analysis of data structures organized in matrix $\mathbf{X}(m \times n)$ by tracing the similarities between the examined objects in the parameter space and between the measured parameters in the object space, and therefore contributed to the development of procedures of coal mine waste self-ignition risk assessment.	
Originality/ value	The originality of the study presented in this paper comes from finding the parameters affecting the tendency of coal mine waste to self-ignite.	

Keywords

coal mine waste, self-ignition, HCA

1. INTRODUCTION

Poland is the largest hard coal producer in the European Union. Documented resources of hard coal deposits in Poland amount to 48,226 million Mg (Państwowy Instytut Geologiczny, 2014). Global forecasts of energy and energy resources demand, clearly indicate that coal will continue to play an important role in securing energy demand in the coming decades. The world hard coal reserves are estimated to be sufficient for 109 years, while crude oil and natural gas enough for 53 and 56 years (BP, 2013), respectively. Currently, approximately 90% of electricity production in Poland and 40% worldwide is based on coal. In a report by the U.S. Department of Energy (U.S. DOE, 2013) an increase in energy consumption by about 56% from 13,100 Mtoe in 2010 to 20,500 Mtoe in 2040 is projected. An increase in coal consumption is also expected from 3,675 Mtoe in 2010 to 5,500 Mtoe in 2040, making an average increase of 1.3% per annum, while the growth of global CO₂ emission will rise from 31.2 bn tons in 2010 to 45.5 bn tons in 2040. In light of the above data, the development of Clean Coal Technologies could be considered as a means of strengthening the role of coal in the Polish and world economy. Clean Coal Technologies denote technologies developed to increase the efficiency of coal extraction, treatment, processing and use, while decreasing their negative environmental impact (OECD/IEA, 1993; Smoliński, 2010; Smoliński & Pichlak, 2009; Stańczyk et al., 2010; World Coal Institute, 2000). Clean Coal Technologies refer to "the entire coal chain" from extraction to waste disposal. This includes, primarily, the so-called sustainable management of resources, modern and efficient coal treatment technologies, transport and storage of coal, coal processing (including utilization for energy purposes), and management of waste formed at each stage of this chain (Smoliński, 2007). One of the most serious environmental hazard related to coal mine dumps is self-heating and self-ignition of waste (Cebulak, Gardocki, Miczajka, Szlosarek, & Tabor, 2010; Cairney, 1991; Cebulak, Miczajka, Tabor, Skręt, & Gardocki, 2009; Cebulak et al., 2005; Choudhry, 1977; Dulewski, Madej, & Uzarowicz, 2010; Gawor, 2010; Gogola, Bajerski, & Smoliński, 2012; Gogola, Iwaszenko, & Smoliński, 2012).

There are still no applicable procedures suitable for the evaluation of risk associated with the self-ignition of waste in coal mine waste dumps. The analysis of the physicochemical parameters characterizing coal mine waste and their impact on the initialization of the self-ignition process with the application of chemometric methods of data exploration and modelling may be useful in the assessment of such risks (Djaković-Sekulić, Smoliński, Perisić-Janjić, & Janicka, 2008; Smoliński, Falkowska, & Pryputniewicz, 2008; Smoliński & Hławiczka, 2007; Smoliński, Rompalski, Cybulski, Chećko & Howaniec, 2014; Zołotajkin, Smoliński, Ciba, Skwira, & Kluczka, 2014). In this paper, the application of Cluster Analysis in the exploration of data characterizing the waste collected from 12 mines' waste dumps, where the phenomenon of endogenous fires either occurred or was not observed is presented.

2. THE EXPERIMENTAL DATA AND CLUSTER ANALYSIS METHOD

The data on physical and chemical parameters, including the content of trace elements in 12 samples of coal mine waste from selected mine waste dumps, was organized in matrix $\mathbf{X}(12 \times 31)$, whose rows describe waste samples (1–12), and the columns represent measured parameters (see Table 1). Seven samples were collected from the area of postmining slag heaps, where waste from one coal mine was deposited, including three samples from areas of recently created storage sites and four samples from decades-old heaps formed by no longer existing mines. Three samples were collected from areas where coal mine waste from various mines is stored and two from slime separators, where fine-grained coal mine waste from water and sludge circulation systems was deposited.

The analysis of the profiles of mean and standard deviations for the data organized in matrix $\mathbf{X}(12 \times 31)$ proved that the data should be standardized (see Fig. 1). The data from matrix \mathbf{X} was subjected to centring, and divided by the standard deviation for each column:

$$xc_{ij} = \frac{(x_{ij} - x_j)}{d_j} \tag{1}$$

where x_{ij} and d_j denote the mean value of the *j*-th parameter and their standard deviation, respectively:

$$x_{j} = \frac{1}{m} \sum_{i=1}^{m} x_{ij}$$
(2)

$$d_{j} = \sqrt{\frac{1}{m} \sum_{i=1}^{m} (x_{ij} - x_{j})^{2}}$$
(3)

and *m* denotes the number of the objects in matrix **X**.

Table 1. Physicochemical parameters characterizing coal mine waste samples tested

No.	Parameter	Unit
1	moisture content W ^a	wt%
2	ash content Aª	wt%
3	content of volatile matter V ^a	wt%
4	content of C	wt%
5	content of H	wt%
6	content of S	wt%
7	heat of combustion Qs ^a	[kJ/kg]
8	calorific value Q _i	[kJ/kg]
9	content of SiO ₂	wt%
10	content of Al ₂ O ₃	wt%
11	content of Fe ₂ O ₃	wt%
12	content of CaO	wt%
13	content of MgO	wt%
14	content of Na ₂ O	wt%
15	content of K ₂ O	wt%
16	content of SO ₃	wt%
17	content of TiO ₂	wt%
18	content of P ₂ O ₅	wt%
19	content of As	ppm
20	content of Ba	ppm
21	content of Co	ppm
22	content of Cr	ppm
23	content of Cu	ppm
24	content of Mn	ppm
25	content of Ni	ppm
26	content of Pb	ppm
27	content of Rb	ppm
28	content of Sn	ppm
29	content of Sr	ppm
30	content of V	ppm
31	content of Zn	ppm

One of the classic methods of data structure exploration, the Cluster Analysis, was applied in the study of physicochemical parameters characterizing coal mine waste samples.

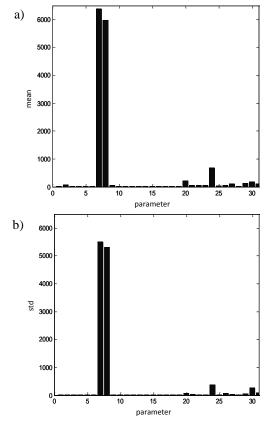


Fig. 1. A graphical representation of the profiles of: (a) mean and (b) standard deviations of the variables from matrix $X(12 \times 31)$

2.1. Cluster Analysis

Cluster Analysis, also called the Hierarchical Clustering method (Kauffman & Rousseeuw, 1990; Massart & Kauffman, 1983; Noworol, 1989; Romesburg, 1984; Vandeginste et al., 1998; Vogt, Nagel, & Sator, 1987; Ward, 1963), enables the analysis of data structures organized in matrix $\mathbf{X}(m \times n)$ by tracing the similarities between the examined objects in the parameter space, and between the measured parameters in the object space (Smoliński, 2008). Cluster Analysis methods differ in terms of the applied similarity measure between objects, as well as the way of connecting similar objects. In the case of continuous variables, the measures of similarity most commonly applied are the Euclidean distance or the Manhattan distance, which are special cases of the Minkowski distance (Vandeginste et al., 1998).

$$d_{ij} = \left[\sum_{k=1}^{l} \left| x_{i,m} - x_{j,m} \right|^{q} \right]^{1/q}$$
(4)

where I denotes the number of variables. If q = 2, then d_{ij} is the Euclidean distance, whereas if q = 1, d_{ij} is the Manhattan distance. Among the various ways of clustering similar objects one should mention single linkage, complete linkage, average linkage, centroid linkage and Ward's linkage methods (Hartigan, 1985; Ward, 1963).

The results of Cluster Analysis are presented in the form of dendrograms. The x-axis describes the order in which objects/parameters were combined and the y-axis defines their mutual similarity. Dendrograms allow for the tracing of data structure, such as clustering tendency. Cluster Analysis does not allow, however, for the simultaneous tracing of the relationship between objects and the measured parameters. This problem was solved by complementing the Cluster Analysis with a colour map of experimental data, enabling more in-depth interpretation of the data structure, tracing the similarities and differences between the clusters on the dendrogram, as well as an indication of the samples with the greatest values of the measured parameters (Smoliński, 2008; Smoliński, Walczak, & Einax, 2002).

3. RESULTS AND DISCUSSION

The dendrogram shown in Figure 2 was constructed for the analysed standardised data organized in matrix $X_c(12 \times 31)$, showing the examined coal mine waste samples in the space of 31 physiochemical parameters (listed in Table 1), obtained using Ward's linkage method, in which the distance between two clusters is defined as the sum of the squared distances of each of the components towards cluster centroids. Figure 3 shows the dendrogram for the 31 physiochemical parameters under consideration, characterizing the examined coal mine waste from various coal waste dumps in the space of 12 samples, obtained by Ward's linkage method.

The dendrogram showing the examined objects in the space of physiochemical parameters (see Fig. 2) enabled the grouping of the examined coal waste samples into two clusters. Cluster A consists of five samples (objects 2, 6, 7, 11 and 12), for which self-ignition was observed. Within cluster B there are four samples for which the phenomenon of self-ignition was not observed (objects 3, 5, 8 and 9) and three samples for which self-ignition was reported (objects 1, 4 and

10). Therefore, the Cluster Analysis did not enable the unambiguous division of coal waste, taking into account the physicochemical parameters determined, into those which undergo or do not undergo the phenomenon of self-ignition. In addition, certain subgroups were distinguished within each cluster. Within cluster A, a subgroup A₁ including samples 2, 6, 7 and 12 was identified, and within cluster B subgroups B₁ and B₂. Subgroup B₁ consists of three samples showing no tendency towards self-ignition (objects 3, 8 and 9) and two samples, for which such a phenomenon was observed (objects 1 and 4). Within subgroup B₂ there is a sample showing no tendency towards self-ignition (object 5), and the other one, for which self-ignition in laboratory conditions was confirmed (object 10).

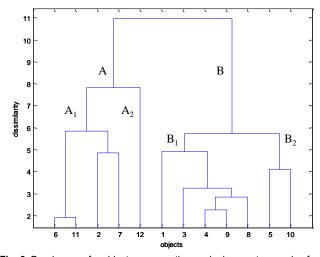


Fig. 2. Dendrogram for objects representing coal mine waste samples from 12 coal waste dumps in the space of 31 physicochemical parameters (described in Table 1), obtained by Ward's linkage method

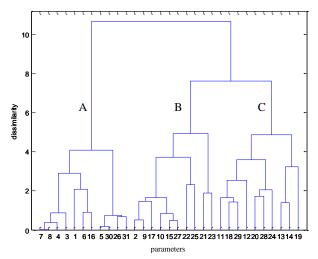


Fig. 3. Dendrogram for 31 physicochemical parameters characterizing the examined samples (as described in Table 1) in the space of 12 samples collected from various coal waste dumps, obtained by Ward's linkage method

In the dendrogram obtained for 31 of the measured physicochemical parameters characterizing the examined waste in the space of 12 objects, three groups of parameters were distinguished. The first one (group A) collected parameters 1, 3–8, 16, 26, 30 and 31, representing the contents of the total moisture, volatile matter, C, H, S, heat of combustion, the calorific value and trace elements: Pb, V and Zn, respectively. The second group (group B) encompassed parameters 9, 10, 15, 17, 21–23, 25 and 27, describing the contents of ash, SiO₂, K₂O, TiO₂, and trace elements: Co, Cr, Cu, Ni and Rb in a sample. The third group (cluster C) clustered parameters 11–14, 18–20, 24, 28 and 29, representing the contents of Fe₂O₃, CaO, MgO, Na₂O, P₂O₅ and trace elements: As, Ba, Mn, Sn and Sr. On this basis, it was found that a tendency of the self-ignition of coal mine waste, was largely related to the parameters included in cluster A, determining the amount of chemical energy accumulated in waste (the contents of volatiles, C, H, S, and the heat of combustion).

In order to trace the relationship between the examined samples of waste and the measured parameters, the dendrogram presenting objects in the space of parameters was complemented with a colour map of experimental data, proposed by Smoliński et al. (2002) and showing the values of the measured parameters, listed in the order of organization of the objects and parameters, obtained by means of the applied method of Hierarchical Clustering (see Fig. 4).

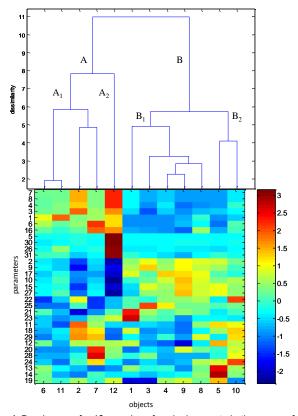


Fig. 4. Dendrogram for 12 samples of coal mine waste in the space of 31 measured physicochemical parameters (described in Table 1) with a colour map showing values of the measured parameters for individual samples of waste

Simultaneous interpretation of the dendrogram and the colour data map enabled a division of the waste samples into two clusters, mainly due to the difference in contents of moisture, ash, volatile matter, C and S, the heat of combustion, calorific value and the contents of SiO₂ and Al₂O₃, K₂O, SO₃, TiO₂, Co, Ni and Rb (parameters 1–4, 6–10, 15–17, 21, 25, and 27). Objects grouped in cluster A were characterized by relatively higher contents of moisture, volatiles, C and S, higher heat of combustion and calorific value as well as

higher content of SO₃ (parameters 1, 3, 4, 6-8 and 16) and relatively lower contents of ash, SiO₂, Al₂O₃, K₂O, TiO₂, Ni, and Rb (parameters 2, 9, 10, 15, 17, 25 and 27) than the objects grouped in cluster B. Samples 2, 6, 7 and 11, marked as subgroup A1, were characterized by relatively lower contents of Co and Ni (parameters 21 and 25). Furthermore, the specificity of samples 2 and 7 was found, due to the highest contents of Fe₂O₃, P₂O₅ and Sr (parameters 11, 18 and 29). Sample 7 was also characterized by the highest contents of S, SO₃, Ba and Sn (parameters 6, 16, 20 and 28) and the lowest contents of Cr and Ni (parameters 22 and 25). Sample 2 was characterized by relatively high values of C content, heat of combustion and the calorific value (parameter 4, 7 and 8), the highest contents of volatile matter, Fe₂O₃, P₂O₅ and Sr (parameter 3, 11, 18 and 29) and the lowest content of Co (parameter 21). Moreover, within subgroup A1, the specificity of sample 11 was observed resulting from the highest moisture content (parameter 1) of all the samples tested.

Sample 12 (not classified into subgroup A1) was unique and this resulted from the highest contents of C and H, the highest value of the heat of combustion, the highest calorific value and the highest contents of Pb, V, and Zn (parameters 4, 5, 7, 8, 26, 30 and 31), and also the lowest contents of ash, SiO₂, Al₂O₃, K₂O, TiO₂ and Rb (parameters 2, 9, 10, 15, 17 and 27) of all the samples tested.

Samples grouped within cluster B were characterized by relatively high contents of ash, SiO₂, Al₂O₃, K₂O, TiO₂, Co, Cr, Ni and Rb (parameters 2, 9, 10, 15, 17, 21, 22, 25 and 27). The maximum contents of ash, SiO₂, Al₂O₃ and TiO₂ (parameters 2, 9, 10 and 17) were observed for samples in subgroup B₁. In addition, this subgroup was characterized by the relatively low content of Sn (parameter 28). Within subgroup B₁ the specificity of samples 1 and 3 was also found, resulting from the lowest levels of As content (parameter 19) of all the samples tested. The specificity of sample 1 resulted from it having the highest contents of Co and Cu (parameters 21 and 23), and of sample 3, from having the highest content of Ni (parameter 25) of all the samples analysed.

Subgroup B_2 , consisting of two samples (objects 5 and 10), was distinguished from the remaining samples under analysis due to high contents of Fe₂O₃, CaO, MgO, Na₂O, P₂O₅ and Mn (parameters 11, 12, 13, 14, 18 and 24). Furthermore, sample 5 was characterized by the highest contents of MgO and Na₂O (parameters 13 and 14), and sample 10 by the highest content of Mn (parameter 24) of all the samples tested.

4. SUMMARY

Analysis of the experimental data with the application of the Hierarchical Clustering method showed that the tendency of coal mine waste to self-ignite is primarily affected by the contents of moisture, ash, volatile matter, C and S, values of heat of combustion, calorific value and contents of SiO₂, Al₂O₃, K₂O, SO₃, TiO₂, Co, Ni and Rb. The analysis did not show any impact from the other examined physiochemical parameters characterizing coal mine waste on their tendency towards self-ignition.

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