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MODELLING THE RELATIONSHIP BETWEEN SELF-IGNITION TEMPERATURE AND PHYSICOCHEMICAL PARAMETERS OF COAL MINE WASTE WITH THE APPLICATION OF THE PARTIAL LEAST SQUARES METHOD

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ABSTRACT

Purpose The aim of the work presented in this paper was the construction of a regression model describing the relationship between the experimentally determined value of the maximum temperature observed during the coal mine waste fire and physicochemical parameters characterizing the coal mine waste.

Methods The model was constructed with the application of the Partial Least Squares method. The experimental data analysed was acquired through the use of a laboratory test stand with a fixed bed reactor and analytical method of gas chromatography.

Results The constructed model was characterized by a good fit of the data used in its construction and the strong predictive ability for the new data. It illustrated the significant impact of the content of H and Fe_2O_3 and trace elements: Co, Cu, Pb, Sr, V and Zn in a sample on the value of the maximum temperature reached during the fire of coal mine waste.

Practical implications The practical importance of the work presented is clear in the light of the role of coal in the Polish economy and environmental aspects related to coal mining and the coal-based energy sector, in particularly to coal waste disposal and utilization. The model constructed contributes to the development of methods of self-ignition and fire risk assessment on coal waste dumps by determining the relationship between waste dump fire occurrence, the temperature observed during the fire and the physicochemical parameters characterizing the coal mine waste.

Originality/value The novelty of the study presented in the paper consists in both finding the relationships modelled and the data extraction methods applied in the research field concerned.

Keywords
PLS, self-ignition, coal mine waste

1. INTRODUCTION

The increase in energy consumption and increased demand for fossil fuels have created new opportunities for the mining industry, especially in conditions of limited global resources of oil and natural gas (BP, 2013; Smoliński & Howaniec, 2010). The environment protection regulations, especially in terms of greenhouse gas emissions reduction, represent a major challenge for the coal mining sector and coal-based energy sector (Smoliński & Howaniec, 2007). Cole mine waste from the extraction and processing of coal poses a serious threat to the environment and human health. Areas of mine waste dumps are often characterized by high investment potential (Gogola, Bajerski, & Smoliński, 2012; Hudecek, Cerna, & Adamec, 2012; Kosmaty, 2011; Ostręga & Ubermann, 2010). Issues related to the protection of the environment include the quality of land, excessive content of certain elements and compounds, leaching of pollutants that can migrate into the surface water and groundwater (POŚ, 2001; RMŚ, 2002) as well as the phenomenon of self-ignition of waste (Gogola, Iwaszenko, & Smoliński, 2012) and are of key importance in the management of these areas.

Mine waste dumps differ significantly in terms of waste composition. This results from the type of waste sources (one or several mines), the age of the dump and the differences in the operation of a mine (different technologies of coal processing). The oldest coal mine waste comes from the period when sorting stations operated in mines solely. Another type of waste is that from inactive slime separators. Slime has a specified market value: it is utilized in fuel blend production. However, self-ignition of slime stored in heaps often occurs during its extraction. Therefore, it is important to find the relationship between the waste dump fire, the temperature observed during the fire and the physicochemical parameters characterizing the waste to estimate the risk of fire incidents in the areas where coal mine waste is stored. The construction of a mathematical model illustrating the relationship between the maximum temperature observed during the mine waste fire and physicochemical parameters characterizing the wastes was the subject of the work presented in the paper.

2. DESCRIPTION OF THE EXPERIMENTAL DATA AND THE METHOD OF THE PARTIAL LEAST SQUARES

Data characterizing the physical and chemical parameters, including the content of trace elements in samples of coal mine waste, for which self-ignition was observed, were organized in the matrix $\mathbf{X}(8 \times 31)$. The matrix rows describe waste samples, and its columns represent the physicochemical parameters tested (see Table 1), whereas the dependent variable $\mathbf{y}(8 \times 1)$ contains the maximum temperatures observed during the tests of self-ignition carried out in a fixed-bed reactor installation. The data organized in the matrix $\mathbf{X}(8 \times 31)$ were standardized since the physical and chemical parameters characterizing the examined waste were in different measurement ranges.

Table 1. The physicochemical parameters characterizing the coal mine waste samples tested

No.	Parameter	Unit
1	moisture content W^a	wt%
2	ash content A^a	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 Q_s^a	kJ/kg
8	calorific value Q_i	kJ/kg
9	content of SiO_2	wt%
10	content of Al_2O_3	wt%
11	content of Fe_2O_3	wt%
12	content of CaO	wt%
13	content of MgO	wt%
14	content of Na_2O	wt%
15	content of K_2O	wt%
16	content of SO_3	wt%
17	content of TiO_2	wt%
18	content of P_2O_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

2.1. Partial Least Squares Method

It is often necessary to match a linear function in order to find the relationship between the independent variables and the dependent variable or variables in studies conducted in the field of coal processing. For this purpose, the Linear Regression Method is applied and its generalization for the case when more than one parameter is measured, i.e. the method of Multivariate Linear Regression (MLR) (Brandt, 1998; Martens & Naes, 1989; Mazerski 2000; Smoliński, 2012, 2014b). Data sets with correlated measured parameters are often found, which makes the application of the classical method of MLR impossible. In such cases, the method of the Principal Components Regression (PCR) (Joliffe, 1982; Martens & Naes, 1989; Smoliński, 2012, 2014b) or the Partial Least Squares (PLS) method (Djaković-Sekulić, Smolinski, Trišović & Ušćumlić, 2012; Martens & Naes, 1987, 1989; Massart et. al., 1997; Smoliński, Walczak & Einax, 2003; Smoliński, Zołotajkin, Ciba, Dydo, & Kluczka, 2009; Wold, 1981; Wold, Martens, & Wold, 1983) are most commonly used. The PLS method was applied in the study presented. In the PLS model, latent variables are established, which differ from the Principal Components applied in the PCR method. Namely, the principal components of the PCR method are constructed so as to maximize the description of the variation of the data, i.e., all the variables in block $\mathbf{X}$ are taken into consideration, yet in the case of PLS models, latent variables are constructed in such a way as to maximize the description of the covariance between $\mathbf{X}$ and $\mathbf{y}$.

The developed calibration models should not only be characterized by a good fit to the studied data, but especially by their strong prediction abilities for new objects. Cross Validation (CV) procedure was applied in order to determine the optimum complexity of the PLS model (Wold, 1978).

3. RESULTS AND DISCUSSION

An analysis of the physicochemical parameters for 12 coal mine waste materials collected from mine waste dumps located in the Silesian region has been presented (Smoliński, 2014a). The self-ignition temperature was determined experimentally for these samples (Gogola, Iwaszenko, & Smoliński, 2012). The self-ignition effect was observed for eight of them. Table 2 shows the maximum temperature values that were obtained under laboratory conditions during the test of self-ignition of 150 g waste in the fixed-bed reactor installation.

Table 2. The maximum temperature reported in tests of coal mine waste self-ignition in the fixed-bed reactor installation

Sample No.	Temperature, °C
1	434
2	292
3	364
4	275
5	251
6	293
7	360
8	462

A regression model was constructed, describing the relationship between the experimentally determined value of the maximum temperature observed during the fire of coal mine waste dumps, and physicochemical parameters characterizing waste samples. The maximum number of uncorrelated variables is indicated in the rank of a matrix, which is, by definition, equal to the smaller dimension of the matrix $\mathbf{X}(8 \times 31)$, in which physicochemical parameters characterizing the tested waste are arranged. In the case of data under consideration, the matrix rank is equal to eight. This means that the maximum number of uncorrelated parameters is eight. The parameters in the matrix analysed are correlated and it is not possible to construct a model describing the relationship between the maximum temperature observed during coal mine waste fire, and physicochemical parameters with application of the classical method of Multivariate Regression. Therefore, Partial Least Squares method was applied.

The designed PLS model should be characterized by a good fit for the data used for their construction and a good prediction abilities for new variables. The accuracy of matching the PLS model, based on the model of the dependent variable, to the appropriate experimental value for the objects from the model set, is determined by Root Mean Square Error (RMS)

$$RMS = \sqrt{\frac{\sum_{i=1}^{m}(y_i - \hat{y}_i)^2}{m}} \tag{1}$$

where y_i and $\hat{y}_i$ denote the experimental values of the dependent variable for a model set and their predicted values, respectively, while m stands for the number of objects in the model set.

The prediction accuracy is determined on the basis of the Cross Validation (CV) procedure consisting of the exclusion from the data, used for the construction of the model, one or more successive rows of the matrix **X** and the corresponding elements of the dependent variable **y** and treating them as a test set. Based on the model constructed for the other objects, the value of dependent variable from the test set is predicted and the Root Mean Square Error of Cross Validation (RMSCV) is calculated with a different number of components

$$RMSCV\,(A) = \sqrt{\frac{\sum_{i=1}^{m}\left(y_i^t - \hat{y}_i^t(A)\right)^2}{m}} \tag{2}$$

where y_i^t and $\hat{y}_i^t(A)$ denote the values from the test set and their values provided by the model of the complexity A, respectively.

In the case of the data concerned, the PLS model was developed in order to describe the relationship between the dependent variable **y**(8 × 1), describing the maximum temperature reached during the fire, and the standardized physicochemical parameters for the selected coal mine waste, organized in the matrix $\mathbf{X_c}$(8 × 31). Based on the analysis of the Root Mean Square Error, the correct complexity of the PLS model was determined. The constructed PLS model, describing the relationship between the maximum temperature of the simulated fire (**y**) and all other parameters organized in the matrix $\mathbf{X_c}$(8 × 31) is shown in figure 1. Values of the Root Mean Square Error and Root Mean Square Error of Cross Validation (*RMS* and *RMSCV*) determine how accurately the constructed model illustrates the data applied in its construction (matching the model to the data), and how precisely the model will predict for the new variables, respectively. The estimated root mean square error (*RMS*) for the constructed model with two components was 11.40%, and the root mean square error of cross validation (*RMSCV*): 18.71%.

Since the data applied in the construction of the PLS model were subjected to standardization, the weight of each of the model parameters can be determined based on the values of the regression coefficients. Table 3 shows the values of the coefficients b_0 and b for the constructed PLS model, describing the relationship between the maximum temperature reached during the fire of coal mine waste (**y**) and the physicochemical parameters characterizing the waste. If the weight (value of coefficient b) of a given parameter is positive, then with the increase in the value of this parameter the value of the dependent variable **y** will increase, whereas when the value of the weight is negative, then its growth will reduce the value of **y**. The model constructed demonstrated the significant impact of the content of H and Fe_2O_3 and trace elements Co, Cu, Pb, Sr, V and Zn in a sample (parameters 5, 11, 21, 23, 26, 29, 30 and 31) on the value of the maximum temperature reached during the fire of coal mine waste.

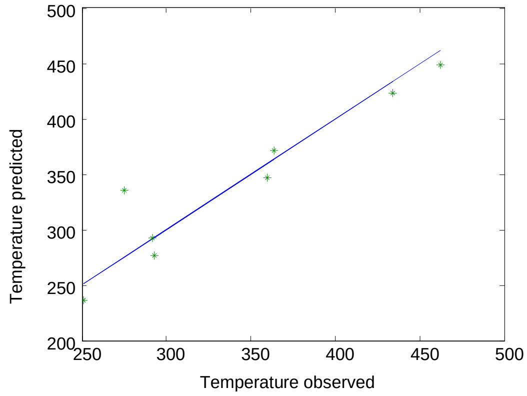

Fig. 1. The constructed PLS model illustrating the relationship between the maximum temperature observed during coal mine waste fire simulated in the fixed-bed reactor and the physicochemical parameters characterizing the waste, organized in the matrix $\mathbf{X_c}$(8 × 31)

Table 3. Values of regression coefficients for the constructed PLS model illustrating the relationship between the maximum temperature observed during the fire of coal mine waste simulated in the fixed-bed reactor installation, and the physicochemical parameters characterizing the waste, organized in the matrix $\mathbf{X_c}$(8 x 31)

Regression coefficient	Value
b_0	341.38
b_1	–2.95
b_2	–0.50
b_3	–1.95
b_4	2.12
b_5	8.00
b_6	–7.57
b_7	1.69
b_8	1.74
b_9	2.24
b_{10}	–2.75
b_{11}	–8.24
b_{12}	–1.80
b_{13}	0.91
b_{14}	–5.67
b_{15}	–3.80
b_{16}	–4.19
b_{17}	3.34
b_{18}	–5.68
b_{19}	–3.85
b_{20}	–5.25
b_{21}	10.50
b_{22}	1.98
b_{23}	10.52
b_{24}	–7.72
b_{25}	5.69
b_{26}	7.95
b_{27}	–5.38
b_{28}	–5.03
b_{29}	–8.29
b_{30}	8.27
b_{31}	8.50

4. CONCLUSIONS

The constructed PLS model, describing the relationship between the maximum temperature of coal mine waste fire simulated in the fixed-bed reactor installation, and the physicochemical parameters characterizing the tested waste was characterized by both, a good fit and strong predictive ability. The analysis of regression coefficients for the constructed PLS model for the standardized data organized in the matrix $\mathbf{X}(8 \times 31)$ allowed for the determination of the importance of each of the parameters under consideration. It was found that the content of H and Fe_2O_3 in the sample and the content of trace elements Co, Cu, Pb, Sr, V and Zn have a significant influence on the value of the maximum temperature reached during the coal mine waste fire.

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References

BP. (2013). *Statistical review of World Energy. Retrieved July 16, 2014* from http://www.bp.com/content/dam/bp/pdf/statistical-review/statistical_review_of_world_energy_2013.pdf.

Brandt, S. (1998). *Analiza danych* [Data analysis]. Warszawa: Wydawnictwo Naukowe PWN.

Djaković-Sekulić, T.LJ., Smolinski, A., Trišović, N. & Ušćumlić, G. (2012). Multivariate evaluation of the correlation between retention data and molecular descriptors of antiepileptic hydantoin analogs. *Journal of Chemometrics*, *26*, 95–107.

Gogola, K., Bajerski, A. & Smoliński, A. (2012). Modyfikacja metody oceny zagrożenia pożarowego na terenach lokowania odpadów powęglowych [Modification of the method of assesment of fire hazards in coal mine waste dumps]. *Prace Naukowe GIG. Górnictwo i Środowisko*, *11*(2), 13–32.

Gogola, K., Iwaszenko, S. & Smoliński, A. (2012). Próba zastosowania reaktora ze złożem stałym do oceny punktu inicjacji zapłonu obiektów formowanych z odpadów powęglowych [Application of a fixed-bed reactor for assessment of the ignition initiation point of objects formed from coal wastes]. *Prace Naukowe GIG. Górnictwo i Środowisko*, *11*(1), 35–46.

Hudecek, V., Cerna, K. & Adamec, Z. (2012). Rekultywacja Hałdy Centralnej w kopalni Jana Švermy w Žacléř [Rehabilitation of Central Tailing Heap of Jan Šverma in Žacléř]. *Inżynieria Mineralna*, *13*(1), 41–53.

Joliffe, I.T. (1982). A Note on the Use of Principal Components in Regression. *Applied Statistics*, *31*(3), 300–303.

Kosmaty, J. (2011). Wałbrzyskie tereny pogórnicze po 15 latach od zakończenia eksploatacji węgla [Wałbrzych post-mining land 15 years after coal extraction]. *Górnictwo i Geologia*, *6*(1), 134–148.

Martens, H. & Naes, T. (1987). Multivariate calibration by data compression. In P.C. Williams, K. Norris (Eds.), *Near-infrared technology in agricultural and food industries* (pp. 57–87). St. Paul, MN: American Association of Cereal Chemists.

Martens, H. & Naes, T. (1989). *Multivariate Calibration*. New York: John Wiley & Sons.

Massart, D.L., Vandeginste, B.G.M., Buydens L.M.C., De Jong S., Lewi P.J. & Smeyers-Verbeke, J. (1997). *Handbook of Chemometrics and Qualimetrics: Part A*. Amsterdam: Elsevier.

Mazerski, J. (2000). *Podstawy chemometrii* [Fundamentals of chemometrics]. Gdańsk: Wydawnictwo Politechniki Gdańskiej.

Ostręga, A. & Uberman, R. (2010). Kierunki rekultywacji i zagospodarowania – sposób wyboru, klasyfikacja i przykłady [Modes of reclamation and redevelopment – selection, classification and examples]. *Górnictwo i Geoinżynieria*, *34*(4), 445–461.

POŚ. (2001). *Obwieszczenie Marszałka Sejmu Rzeczypospolitej Polskiej z dnia 26 sierpnia 2013 r. w sprawie ogłoszenia jednolitego tekstu ustawy – Prawo ochrony środowiska* [Notice of the Marshall of Parliament of the Republic of Poland of the 26th August, 2013 on the publication of the consolidated text of the Act – Environmental Protection Law]. Dz. U. poz. 1232 z dnia 23 października 2013.

RMŚ. (2002). *Rozporządzenie Ministra Środowiska z dnia 9 września 2002 r. w sprawie standardów jakości gleby oraz standardów jakości ziemi* [Regulation of the Ministry of Environment of the 9th September, 2002 on soil quality and earth quality standards]. Dz.U.02.165.1359 z dnia 4 października 2002.

Smoliński, A. (2012). *Modelowanie ilościowych zależności w danych chemicznych* [Modeling of quantitative relationships in chemical data]. Katowice: Główny Instytut Górnictwa.

Smoliński, A. (2014a). Analiza wpływu parametrów fizykochemicznych charakteryzujących odpady powęglowe na inicjalizację procesu samozapłonu z zastosowaniem metody analizy skupień [Analysis of the impact of physicochemical parameters characterizing coal mine waste on the initialization of self-ignition process with application of Cluster Analysis]. *Journal of Sustainable Mining*, in press.

Smoliński, A. (2014b). *Metody opracowania danych wielowymiarowych i planowania eksperymentów w badaniach naukowych* [Methods for the development of multivariate data and planning experiments in scientific research]. Katowice: Główny Instytut Górnictwa.

Smoliński, A. & Howaniec, N. (2007). Możliwości redukcji stężenia dwutlenku węgla w atmosferze w procesie sekwestracji z wykorzystaniem formacji geologicznych [Reduction of carbon dioxide concentration in the atmosphere in sequestration process with utilization of geological formations]. *Ochrona Powietrza i Problemy Odpadów*, *44*(1), 14–22.

Smoliński, A. & Howaniec, N. (2010). Environment Friendly Coal Processing Technologies for Sustainable Development of Polish Energy Sector. *Ecological Chemistry and Engineering S*, *17*(3), 297–307.

Smoliński, A., Walczak, B. & Einax, J.W. (2003). Robust Multivariate Calibration in Environmental Studies. *Analytical Letters*, *36*(10), 2317–2336.

Smoliński, A., Zołotajkin, M., Ciba, J., Dydo, P. & Kluczka, J. (2009). Robust PLS Regression Models to Predict Aluminum Content in Soils of Beskid Mountains Region. *Chemosphere*, *76*(4), 565–571.

Wold, H. (1981). *Soft Modeling: The Basic Design and Some Extensions, Systems Under Indirect Observation, Causality-Structure-Prediction*. Amsterdam: North Holland.

Wold, S. (1978). Cross-validatory estimation of the number of components in factor and principal components models. *Technometrics*, *20*(4), 397–406.

Wold, S., Martens, H. & Wold, H. (1983). The Multivariate Calibration Problem in Chemistry Solved by the PLS Method. In *Lecture Notes in Mathematics Vol. 973* (pp. 286–293). Heidelberg: Springer Verlag.