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The use of alternating conditional expectation to predict methane sorption capacity on coal

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ABSTRACT

Conventional approaches to determine methane sorption capacity, including manometric, volumetric and gravimetric methods, require complex procedure of preparation of coal samples and long-term sorption measurement. This article proposed the use of alternating conditional expectation (ACE) algorithm to relate methane sorption capacity (V_L) to coal composition, vitrinite reflectance and temperature without conducting sorption tests, which minimizes the work volume and time required in conventional measurement method.

The basic idea behind the ACE is to estimate a suit of optimal transforms of a dependent and a set of independent variables that result in a linear correlation between the transformed independent and dependent variables with minimum error. Underlying effect can be uncovered of the control of each independent variable on dependent variable through the transform. 139 sets of proximate analysis, maceral analysis and methane sorption data from previous studies were acquired. Ash, fixed carbon, moisture, vitrinite content, vitrinite reflectance and temperature were selected as independent variables to predict V_L. The resulted ACE transforms have a correlation coefficient R^2 of 0.91, indicating an excellent match between the predicted and measured V_L values. Normality and homoscedasticity were verified by Lilliefors- and F-test, which further confirmed the capability of ACE as being a correlation tool. The effects of independent variables on V_L observed from the transforms show an agreement with previous studies.

Based on the ACE transform results, an explicit V_L model was proposed bearing a polynomial correlation with the independent variables. The validity of the proposed model was proved by fitting it to another 43 data sets. Additionally, outlier diagnose was conducted through standardized residuals and their effect on prediction accuracy was investigated.

1. Introduction

Sorption of gases (CH_4, CO_2, N_2, or their binary or tertiary mixtures) on coals acts as a crucial role in the interactions of multiple processes within a coal–gas system (Liu et al., 2011). Modeling of CBM (Feng et al., 2012; Karacan, 2008, 2013) and enhanced coalbed methane (ECBM) (Kronimus et al., 2008; Masaji et al., 2010) production with injection of gases, as well as controls of coal or gas outburst (Basil and Crosdale Peter, 1998; Lama and Bodziony, 1998; Noack, 1998) to ensure mining safety require reliable information of the sorption behavior.

To date, numerous studies have been conducted on the sorption of gases on coals (e.g., Bell and Rakop, 1986; Ceglarska-Stefańska and Zarębska, 2005; Czerw, 2011; Dutta et al., 2011; Faiz et al., 2007; Gensterblum et al., 2013; Han et al., 2012; Macuda et al., 2011; Laxminarayana and Crosdale, 1999, 2002; Mazumder et al., 2006; Pan et al., 2012; Sakurovs et al., 2012; Siemons and Busch, 2007; Weniger et al., 2012). Three typical approaches to determine the sorption capacities of gases on coals include: 1) manometric, 2) volumetric and 3) gravimetric methods. Despite their differences in devices and some operation detail, those three methods bear similarities in, e.g., the need to determining void volume and long time required to reach equilibrium at each pressure step (Busch and Gensterblum, 2011).

Generally, measurement of a sorption isotherm calls for several to dozens of days, making itself time consuming work. More recently, the molecular simulation is applied in the prediction of sorption of methane and carbon dioxide in coals (Hu et al., 2010; Liu and Wilcox, 2012; Mosher et al., 2013). Though the use of molecular simulation eliminates the procedure of conducting laboratory test on sorption capacity, the accuracy depends highly on the constructed electronic structure of the coal sample and the cost is relatively high due to the use of specific software and structure-construction-related measurement (e.g., the use of MRI to obtain coal structure).

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Many sorption models have been developed and introduced to quantitatively represent the gas sorption isotherm on coals, including Langmuir model, Langmuir–Freundlich model, Toth model, UNILAN model, Brunauer–Emmett–Teller (BET) model, Dubinin–Astakhov (D–A) model and Simplified Local Density (SLD) model. The readers are encouraged to refer to Clarkson (1998), Fitzgerald (1999) and Ozdemir (2004) for more details of the models. Of the mentioned models, the two-parameter (V_L and P_L) Langmuir model is most widely used in gas–coal sorption modeling, especially for methane and nitrogen while the SLD model is gaining more attention especially for CO_2. Though the Langmuir model is derived based on thermodynamics and intends originally to represent sorption on homogeneous solid surface, it is frequently used as a simple method and provides a reasonable fit to most experimental data (Busch and Gensterblum, 2011) for gas sorption on coals (which are known to exhibit high heterogeneity). In this paper, the Langmuir volume (V_L) was specifically assigned to represent the methane sorption capacity on coals.

The dependence of coal sorption capacity in methane on coal composition and temperature (Clarkson and Bustin, 1996; Dutta et al., 2011; Faiz et al., 2007; Hildenbrand et al., 2006; Joubert et al., 1973; Krooss et al., 2002; Levy et al., 1997; Ozdemir, 2004; Pini et al., 2010; Sakurovs et al., 2008) has been investigated extensively. However, majorities are focused on single element analysis and there is rare comprehensive model for V_L that takes into account of the major constituents of proximate and maceral analyses and temperature. Hildenbrand et al. (2006) developed a V_L model using the multiple regression method. The model incorporates the vitrinite reflectance, temperature and moisture content as independent variables and produces an averaging R^2 of 0.73. However, the model does not include, e.g., ash or vitrinite content that are known to exert important effects on sorption capacity.

As gas sorption capacity is influenced by the joint action of multiple factors, their respective effect may be masked or reinforced by one another, which increases the difficulty in discovering the underlying relationship between them. One solution to quantitatively analyze the effects of multiple factors on sorption capacity is through conventional multivariate parametric regressions. However, the conventional method requires a priori assumption of the function forms relating dependent and independent variables, which may yield erroneous or misleading results once they are assigned improperly (Xue et al., 1997; Jassim et al., 2012). Artificial neural networks (ANNs) provide with an alternative to conventional multivariate parametric regressions, which have been demonstrated to be a powerful correlation tool in varying fields. Unfortunately, explicit function forms cannot be obtained using ANNs, and thus the effect of each independent variable on the dependent variable cannot be quantitatively represented.

This article proposed the use of the ACE (Breiman and Friedman, 1985) algorithm to relate methane sorption capacity to coal composition, vitrinite reflectance and temperature and gave an explicit comprehensive correlation between them. The ACE is in essence a nonparametric method for multivariate nonparametric regression that was first proposed by Breiman and Friedman (1985), and later refined by Xue et al. (1997). Different from conventional parametric regression methods, the ACE is totally data driven and does not require a priori assumption of the functional forms (Jassim et al., 2012). This enables the analyst to have a closer insight into the combined effect of varying factors on methane sorption capacity such that relationship between them can be best described and non-linear relationships can be uncovered (Wang and Murphy, 2004).

2. Methodology

2.1. ACE methodology

The basic idea behind ACE is to estimate the transformations of a dependent and a set of independent variables that produce the maximum linear effect between the transformed independent and dependent variables (Wang and Murphy, 2004). The ACE regression model can be written as:

$$\theta(y) = \alpha + \sum_{i=1}^{n} \phi_i(x_i) + \varepsilon \tag{1}$$

where ϕ_i and θ are the transform functions of the input independent variable x_i and dependent variable y, respectively. Thus the problem of correlating dependent variable y with independent variable x_i is replaced with estimating n separate one-dimensional functions ϕ_i and θ (Wang and Murphy, 2004). The objective of ACE is to minimize the error variance (ε^2) of the transformed dependent variable on the sum of transformed independent variables by holding $E[\theta^2(y)] = 1$ (Jassim et al., 2012). ε^2 is defined as:

$$\varepsilon^2(\theta, \phi_1, \ldots, \phi_n) = E\left\{\left[\theta(y) - \sum_{i=1}^{n} \phi_i(x_i)\right]^2\right\} \tag{2}$$

The ε^2 is minimized through a series of single function minimizations, resulting in (Breiman and Friedman, 1985):

$$\phi_i(x_i) = E\left[\theta(y) - \sum_{j \neq i}^{n} \phi_j\left(x_j\right) \middle| x_i\right] \tag{3}$$

$$\theta(y) = E\left[\sum_{i=1}^{n} \phi_i(x_i) \middle| y\right] \Big/ \left\| E\left[\sum_{i=1}^{n} \phi_i(x_i) \middle| y\right] \right\| \tag{4}$$

the resulted $\phi_i(x_i)$ and $\theta(y)$ in Eqs. (3) and (4) after an iterative process are referred to as optimal transformations $\phi_i^*(x_i)$ and $\theta^*(y)$, which are related as:

$$\theta^*(y) = \sum_{i=1}^{n} \phi_i^*(x_i) + \varepsilon^* \tag{5}$$

where ε^* is the error not captured by the use of the ACE transformations and is assumed to have a normal distribution with zero mean.

2.2. Data acquirement and normalization

Proximate analysis, maceral analysis and methane sorption data from previous literatures were acquired and inputted to develop a model for predicting V_L in this study (Table 1). Coals with rank in the range of subbituminous to anthracite were included. The samples were prepared in either as-received, or air dried, or dried state for sorption test. Test temperature varied from 296.75 K high up to 338 K. It is noted that though numerous sorption tests have been conducted on coals in a worldwide range and provide with a valuable data base, large variations exist in integrality of the reported data (e.g., some lack the information on proximate or maceral analysis), which therefore were not included in this study.

Due to differences in the report bases from literature to literature, the data were normalized to ensure accuracy and comparability prior to being inputted into the ACE transforming. Ash content, fixed carbon and vitrinite content were respectively corrected to dry, dry-ash-free (DAF) and mineral-matter-free (MMF) basis. Moisture content was invoked to characterize the sample preparation process. The moisture content was assigned to zero for dried samples assuming that the free water was totally removed during sample drying. For as-received or air-dry samples, the moisture content determined from proximate analysis was used. For moisture equilibrated samples, the moisture was assigned to be the sum of proximate-analysis-determined and equilibrated moisture.