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Comparison of hydrocarbon gases (C_1–C_5) production from Carboniferous Donets (Ukraine) and Cretaceous Sabinas (Mexico) coals

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Abstract

The main purpose of this contribution is to compare the ability of Carboniferous coals from the Donets Basin of the Ukraine and Cretaceous coal from the Sabinas Basin of the Mexico to generate hydrocarbon gases (C_1–C_5). Two bituminous coals from the Donets Basin (2c10YD and 111Dim; 0.55 and 0.65%R_r respectively) and one bituminous coal from the Sabinas Basin (Olmos, 0.92%R_r) were studied using heating experiments in a confined-pyrolysis system. The highest rank reached during the heating experiments corresponds to the anthracite stage (2.78 and 2.57%R_r) for the 2c10YD and 111Dim coals and (2.65%R_r) for the Olmos coal. The composition of the generated (C_1–C_5) gases was evaluated using a thermodesorption-multidimensional gas chromatography. The results show that the Carboniferous Donets coals produced more wet gas and methane during pyrolysis than the Cretaceous Olmos coal. This is probably due to their higher liptinite (6–20%) and collodetrinite content and to the loss of a major part of the petroleum potential of the Olmos coal during natural coalification. C_2–C_5 compounds are mainly derived from the cracking of liquid hydrocarbons. Ethane is the most stable compound and formed from the cracking of higher hydrocarbon component.

Large amounts of methane (up to 81 mg/g coal for the Donets coals and 50 mg/g coal for the Sabinas coal) were formed at high temperatures by cracking of previously formed heavier hydrocarbons and by dealkylation of the coal matrix. A linear relationship was observed between methane generation and the maturity level of both coal types.

Keywords: Artificial maturation; Confined pyrolysis; Coal; Methane; Donets Basin; Sabinas Basin

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1. Introduction

The Donets Basin in the SE part of Ukraine contains one of the major late Paleozoic coal basins in the world with proven reserves in the order of 60 Gt. Total coal thickness in Carboniferous formations is about 60 m. The Donets coal mines are among the gasiest and the most dangerous in the world (Privalov et al., 1998, 2003; Privalov, 2002). Thermal maturation of coals and dispersed organic matter in the Donets Basin resulted in the formation of an enormous methane resource of about 278 Tm^3 (Uziyuk et al., 2001; Triplett et al. 2001) estimated the total methane resource in the Donets Basin from coals to be 117 Tm^3 and the methane content in recoverable coal seams at 1400 to be 2500 Gm^3. The high methane content in coal seams in the Donets Basin presents a high potential for coal bed methane recovery (Privalov et al., 2004a; Alsaab et al 2007a). On the other hand it represents a severe mine safety problem, where coal and gas outbursts constitute a major mining hazard and account for many fatalities (Privalov et al., 2004a,b). As well, as an energy resource, CH_4 is a greenhouse gas whose atmospheric concentration has been increasing at a rate of about 1% annually (Tyler, 1991). For instance in the Donets Basin only 13% of the total generated methane from coal beds is detected in the degasification system

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during coal mining activity. Only 30% of that captured methane is used as an energy resource. Thus the emissions of large amounts of methane from Donets coal beds contribute to the gas greenhouse effect (Triplett et al., 2001).

The Sabinas Basin in Mexico contains Upper Cretaceous (Maastrichtian Age) very gassy coals (the Double Seam) in the Olmos Formation, of high to medium volatile bituminous rank (Gentzis et al., 2005). Brunner and Ponce (1999) estimated the presence of 12.2 Gt of coal in the Maastrichtian coals in Coahuila State. Olmos coal is considered as a potential resource of natural gas, with coal seams ranging in thickness from 1.5 to 4 m (Eguiluz de Antuñano, 2001; Gentzis et al., 2005). The average of gas content of Olmos coals is of 8–15 m^3/ton (Eguiluz de Antuñano, 2001). Coal bed methane is mainly methane (98%) (Eguiluz de Antuñano, 2001; Gentzis et al., 2006). The amount of adsorbed methane in Olmos coal formation is still under investigation. Gentzis et al. (2006) estimated this amount to be more than 9 cm^3/g, and that in some mines such as Esmeralda Mine the adsorption capacity at a depth of 300 m is as high as 15 cm^3/g. A report by Minerales Monclova (Santillan-Gonzalez, 2004) indicated an average in-situ content of gas in the coals of Sabinas Basin of 10–14 m^3/ton (98.5% methane) with gas total resource in Coahuila estimated between 1.22×10^{11} m^3 and 2.2×10^{11} m^3.

Previous studies by Law and Rice (1993), Béhar et al. (1995), Taylor et al. (1998), and Petersen (2006) presented coals as being both gas-prone source rocks and reservoir rocks. In accordance with the work of Alsaab et al. (2007b), the Donets and Olmos coals have different abilities to oil generation during heating experiments in a confined system; the Donets coals, which are less mature and richer in liptinite and collodetrinite produce more bitumen than the Olmos coals. Moreover, Alsaab et al. (2007b) observed an increase in the porosity with increasing coal rank for both the Donets and Sabinas coals and attributed that to the cracking of bitumen generated during pyrolysis into solid bitumen and an abundance of light compounds (methane in particular). Given (1984), Derbyshire et al. (1989), and Erdmann and Horsfield (2006) showed that the oil generated by coal tends to be adsorbed in the micropores of vitrinite; eventually this trapped oil is converted to gas as the coal matures under increasing thermal stress. Thus, it is generally accepted that coal can generate an important amount of methane and store a large volume of generated gas, if the entrapment conditions are favourable. Ritter and Grover (2005) related the absorption capacity of generated petroleum (C15+ hydrocarbons) in the coal matrix, the later secondary cracking during subsequent maturation and the generation of light hydrocarbon compounds when cracking becomes more dominant. They revealed that expulsion begins relatively late and that the expelled hydrocarbons consist of C_1 to C_5 hydrocarbons and heavy aromatics.

Prediction of the volume of hydrocarbons (HCs) stored in reservoirs is difficult due to the impossibility of directly quantification of the HCs amounts generated during coalification. Thus, two laboratory methods were proposed. The best known method is heating experiments which allow us to simulate the natural coalification process by applying increased heating temperatures and shortened time (Tissot et al., 1971; Huc and Durand, 1973; Stach et al., 1982). Open, closed, hydrous and confined systems were established to meet this objective. Monthioux (1988) showed that the natural reaction system should be considered as 'quasi confined' in the case of coals because of their physical and chemical properties. In this case, non-hydrous closed pyrolysis system using glass tubes (Horsfield et al., 1989) or gold cells (Monthioux et al., 1985) can be used to simulate natural coalification and estimate HCs generation during basin evolution. Béhar et al. (2003) compared artificial maturation of lignite (Type III) in open non-hydrous, closed non-hydrous and closed hydrous pyrolysis and observed in generally a slight increase in the total HC gas for both non-hydrous pyrolysis with an increase in maturity. Kotarba and Lewan (2004) showed that amounts of thermogenic gases produced by non-hydrous pyrolysis from lignites of Poland are 1.29 times greater than by hydrous pyrolysis. Su et al. (2006) compared artificial maturation of Miocene coal from China (starting 0.35%R_r) with confined hydrous and confined non-hydrous pyrolysis and found 1.6 times more gas generated for non-hydrous than for hydrous pyrolysis. We think that each of these pyrolysis systems must be viewed as additional, not as replacement. An open system pyrolysis experiment enables us to simulate the primary cracking reactions of organic matter, while closed and hydrous pyrolysis systems take into account both primary and secondary cracking. As hydrous pyrolysis is limited by the maximum pyrolysis temperature due to excessive water and its critical point, the complete reaction of gas generation cannot be reached at the end of pyrolysis. Because of the strong adsorption capability, the early generated products (heavy hydrocarbon) will be prevented from expulsion from the coal sample and undergo secondary cracking into gas. Therefore, a dry, confined system pyrolysis was employed in this study to simulate gas generation from coal at three different temperatures (330, 360 and 400 °C).

The second method is through the use of numerical models to estimate the amount of generated HCs by using the changes in major elemental composition (C–H–O) with coal rank. Depending on the elemental data employed, starting rank, and assumptions made about the products, it has been estimated that about 100 to 300 cm^3 CH_4/g can be generated from coals (Jüntgen and Karweil, 1966; Jüntgen and Klein, 1975; Meissner, 1984; Ermakov and Skorobogatov, 1984; Welte et al., 1984; Levine, 1987; Hunt, 1996). Rice (1993) and Clayton (1998) estimated the methane yield by using numerical models method to be in the range of 150–200 cm^3/g, with yield depending strongly on the elemental and maceral composition and also the maturity level of kerogen.

The purpose of this paper is not to compare different pyrolysis methods nor to propose a kinetic study, but to compare the ability of two different coals (Carboniferous and Cretaceous) from two basins (Donets and Sabinas) in order to determine the CBM potential in these basins. This paper is a continuation of the paper (Alsaab et al., 2007b) that concerned the production of oil and pyrobitumen by the same samples under the same artificial maturation by confined-pyrolysis system.