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A STUDY OF POTENTIAL OCCURRENCE OF BIOGENIC METHANE IN COAL SEAMS

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ABSTRACT: A significant proportion of the total gas emitted from coal mining, particularly for shallow seams (<300 m depth), is believed to have been generated from microbial activities within the coal seams and water filling the pores and fractures in coal. To investigate the potential and extent of gas generation in coal due to microbial activities, we developed a method to culture and monitor the production of biogenic methane in coal. We then applied the method to study the process of biogenic methane generation in coals from a mining region in New South Wales. Fresh coal core samples were collected from an exploration borehole drilled into a sequence of coal seams at a greenfield site where five coal seams were located between the depths of 50 to 250m. The formation water was collected from an adjacent borehole drilled into the same sequence of coals. The coal samples were crushed and mixed with formation water and other solutions in glass vials, and then placed in pre-designed incubator at in-situ temperature to allow the production of methane over the life of the project. The results of measurements show that biogenic activities take place and that methane is generated. Methane continued to be produced throughout the life of the project for the studied coals.

INTRODUCTION

Primary origin of gas in coal is the result of coalification, which occurs under high pressure and temperature conditions generating thermogenic gas. This gas consists predominantly of methane (CH_4), carbon dioxide (CO_2), potentially some higher hydrocarbons such as ethane (C_2H_5), and nitrogen (N_2). However, a major source of gas, particularly at shallow depths, is believed to be through microbial activities and is labelled biogenic gas.

The occurrence of gas (mostly CH_4) of potentially biogenic origin has commonly been reported in Australian coalfields (see for example Smith *et al*, 1982; Smith and Pallasser, 1996; Boreham *et al*., 1998; Faiz *et al*, 1999; Faiz and Hendry, 2006; Faiz *et al*, 2007; Li and al., 2008; Flores *et al.*, 2008; Formolo *et al.*, 2008; Hamilton *et al*, 2014). Numerous overseas researchers have also reported on biogenic processes for the generation of coal seam gas (see for example Rice, 1993; Whiticar, 1994, 1996; Clayton, 1998; Rice *et al*, 2008; Green *et al*., 2008; Moore, 2012; Ritter *et al*, 2015; Park and Liang, 2016).

Overall it is believed that CH_4 is generated as a result of the microbial degradation of coal. Macromolecules in coal are broken down during acetate fermentation, providing nutrients essential for microbial metabolic functions. The extent of gas produced by microbial activities depends on the type of methanogenic microorganisms present in the coal seams and whether the in-situ conditions are aerobic or anaerobic, which in turn depends on the coal seam depth. Other factors believed to influence the biogenic gas production are redox potential and pH levels, temperature and coal properties such as porosity, composition and rank.

A new laboratory method for monitoring the generation of CH_4 in coal through microbial activities is developed to simulate the *in situ* process of biogenic generation of gas in coal seams. Using this method the production of biogenic CH_4 is accelerated so that the process of methane generation can be assessed in reasonable time periods (months). Details of the new method is described in the

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following sections, followed by the descriptions of coal core samples used, generated data analyses and interpretation of the results.

METHODOLOGY

Field procedure

The field procedure consisted of collecting fresh coal cores and formation water from a sequence of coal seams traversed by the designated surface drilling for this study. A suitable greenfield site was selected for the project where a sequence of coal seams could be intercepted by the surface exploration boreholes so that the effect of coal properties variation could also be investigated. Core coal samples were placed in CSIRO-designed, purposely built stainless steel gas tight canisters to allow rapid and secure sealing of the sample and inertisation of the headspace by flushing helium gas (He) into and out of the canister. The formation water was also collected from the same borehole, and from other boreholes in the vicinity of the sampled holes, to be used with coal samples in a pre-designed microbial culture procedure. Formation water was placed in glass laboratory fluid containers, which were modified to suit the requirements of water collection at the drilling site. Formation water provides nutrients and supports metabolic activities of microbial populations living in coal reservoirs, leading to the generation of various gases. Specific procedures were designed for collecting and testing the required coal and water samples. Both coal and water sampling containers were fully sterilised before use in the field to eliminate the risk of introducing microbial populations other than those from the reservoir. For the anaerobic microbial populations to survive, oxygen (O_2) was removed from the water by adding deoxygenating solutions to the collected formation water as well as by bubbling a stream of ultra-pure He through the water for a sufficient amount of time.

Laboratory procedure

Once the coal samples were brought to the laboratory, they were crushed and pulverised in a helium atmosphere using a purposely built crusher, on which the stainless steel canister containing coal could be mounted and coal crushed without opening the canister. For some samples, the molecular and isotopic composition of gas desorbed during crushing was analysed. Crushed coals were then removed from the canisters, inside a sterilised, anaerobic chamber filled with helium gas, and partitioned into multiple small subsamples. The coal subsamples were mixed with other solutions and placed in gas tight, pre-sterilised 50 mL glass vials. The vials were then incubated in the dark and at *in situ* coal seam temperature (~30 °C) for extended period of time.

Coal subsamples produced in this way were then used for two types of microbial gas generation experiments:

- Type 1 or treatment experiments: in these experiments coal subsamples were mixed with a solution of formation water, a reducing agent (to create anaerobic conditions by removing any O_2 in the solution) and a nutrient. The gas evolving from this mixture was then measured after a period of time to quantify the total volume of gas which would be produced by microbial activities but also produced from desorption of any residual gas remaining in the crushed coal.
- Type 2 or control experiments: in these experiments coal was mixed with a biocide solution (usually a solution of 70% ethanol and 30% distilled water) to stop any microbial activities. These experiments were conducted to quantify any gas produced from desorption of residual gas remaining in the crushed coal.

Concurrent treatment and control experiments allows for investigating the effects of microbial activities on the nature of gas evolving from the coal. Molecular and isotopic composition analyses were conducted to characterise gas evolved from these experiments.

COAL SAMPLES

The surface exploration borehole drilled for this study intercepted five coal seams in a coal sequence between depths of 68 and 206 m. Proximate and petrology analyses of all samples were also undertaken to characterise coals for their type and maturity. Petrology results show that vitrinite reflectance (VR) for these samples range between 0.64 to 0.70% and hence these coals can be considered high volatile bituminous rank. The vitrinite content of the samples varied from 50 to 70% on a mineral-free basis. Table 1 reports some of the petrology data and proximate analysis data of coal core samples collected for this study.

Table 1: Petrology and proximate analyses of coal samples used for experiments

		Proximate analysis (w/w %)						
Coal seam	Depth (m)	Moisture	Volatile matter	Fixed carbon	Ash yield	Volatile daf	Mineral free vitrinite (%)	Vitrinite reflectance (%)
Seam1	68	2.0	35.0	43.6	19.4	44.5	66.9	0.65
Seam2	115	2.1	32.7	44.4	20.8	42.4	59.0	0.64
Seam3	124	2.4	28.2	46.0	23.4	38.0	59.1	0.69
Seam4	160	2.8	27.3	57.4	12.5	32.2	70.3	0.68
Seam5	206	2.6	27.4	58.5	11.5	31.9	50.0	0.70

EXPERIMENTS

As explained in the methodology section, treatment and control subsamples were prepared for coals from five seams. For each coal sample, a total of 12 treatment and 12 control sub-samples were prepared resulting in a total of 120 sub-samples for the five seams studied. It was planned to have three harvests (sampling periods for analysis) for the life of the project spaced about four to five months apart.

For each coal, at the completion of each successive harvest, four treatment and four control subsamples were analysed. These replicates were set up to allow for statistical analysis of the results. The remaining subsamples were kept in the incubator for the following harvests. All samples were prepared in a He atmosphere (with no O_2 present in the system). Treatment and control samples were placed in gas tight glass vials and incubated in a pre-designed incubator at *in situ* conditions.

At each harvest the gas evolved in each experimental vial was measured for the produced volume as well as for the molecular and isotopic composition of the gas. Isotopic composition data are not reported in this paper.

EVALUATION OF GAS GENERATED IN COAL DUE TO MICROBIAL ACTIVITIES

To verify whether microbial activities had taken place and biogenic gas had been produced, the concentration and volume of CH_4 in the headspace of all treatment and control vials were measured. Note that treatment experiments gave an indication of the total gas evolved from a mixture of coal and solutions in the vials. To estimate the volume of CH_4 produced by microbial activities alone, the volume of gas in treatment vials was reduced by the volume of CH_4 in control vials.

The CH_4 production data, in terms of net biogenic gas generated from harvests, are presented in Table 2. Note that the amounts of gas produced during harvests are presented in terms of volume of gas per unit mass of coal. As the amount of gas in coal is commonly expressed in cubic meters per tonne(m^3/t). This unit was used to quantify the volume of biogenic gas production in these experiments. Note that the volume of gas is estimated by measuring the concentration of gas in each experimental vial and the void volume of that vial.

Table 2: CH_4 produced from microbial activities from coals used in this study

Coal seam	Net biogenic CH_4 produced (m^3/t)		
	Harvest 1	Harvest 2	Harvest 3
Seam1	1.32	1.56	5.11
Seam2	0.03	0.04	0.12
Seam3	0.06	0.05	0.22
Seam4	0.11	0.10	0.11
Seam5	0.55	0.75	10.52

The results show that gas was produced for all coals from all five seams and in all three harvests. The data also show that, with the exception of Seam 4, more gas was produced for the longer harvest periods. The most microbial gas was generally produced in the third or longest harvest (about a year). An interesting observation is that the volume of gas produced does not depend on the seam depth. In fact the coal samples from the shallowest and deepest coals (Seam1 and Seam5) produced the most gas. A maximum of 10.5 m^3/t was produced by coals from Seam 5 and about half of this amount (5.1 m^3/t) was produced by coals from Seam1. The other three seams produced much smaller volumes of gas generally about or below 0.1 m^3/t. Figure 1 shows a plot of the amount of CH_4 produced for each of the five seams for the three harvests.

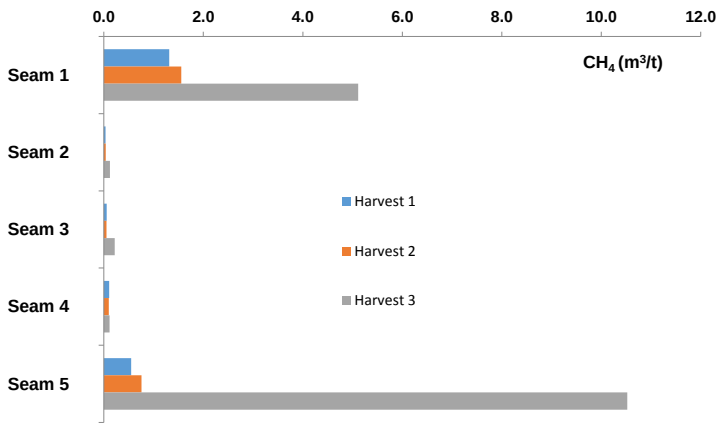

Figure 1: Net volume of CH_4 produced from microbial activities for the coals studied

Using available petrology and coal quality data for these coals (Table 1), relationships were established between the amount of microbially produced CH_4 and these parameters. However, the petrology data (such as vitrinite content and reflectance) and coal quality data (such as ash yield, moisture content and volatile matter content) do not show large variations across these coals. Therefore, no trends could be established between these properties and average CH_4 production from microbial activities.

CONCLUSION

The analysis of CH_4 gas produced from microbial culture experiments on coals from the sequence of coal seams at a mining zone in New South Wales suggests that CH_4 has been produced for all the seams although at very different rates. The shallowest and deepest coals produced the most gas with about 5.1 and 10.5 m^3/t for the longest harvests (~one year), respectively. Other coals from the three remaining seams in the sequence produced small amounts of gas (~0.1 m^3/t or less). Overall the results show that microbial activities produce CH_4 in these shallow coals, however, large variations exist between the rates of gas generation for the coals studied.

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