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## GAS CONTENT AND EMISSIONS FROM COAL MINING

## Abouna Saghafi

ABSTRACT: Gas content can be considered the most important parameter for assessing emissions from coal seams during and post mining. Traditionally, the purpose of gas content determination was to assess gas outburst potentials and to quantify the magnitude of gas emissions into underground headings and at the coal face. Therefore, it's a traditional definition; measurement and determination are based on these objectives. However, the calculation of emissions from mining for the purpose of establishing a greenhouse gas inventory would require new definitions for gas content and new measurement methods. Moreover, errors inherent in measuring gas content need to be quantified so that the uncertainty of emissions inventory can be evaluated. Therefore, various definitions of gas content coals is discussed. Moreover, various parameters influencing the value of measured gas content are discussed and errors of estimation using the direct method are evaluated.

#### INTRODUCTION

Mining leads to large disturbances of coal seam reservoirs as fractures develop both in coal and rock strata. Gas trapped in the coal seam and enclosing strata escapes to the atmosphere via fractures and exposed coal surfaces. The intensity of emissions depends on the flow properties and the volume of gas present in the coal seam and strata at the time of mining. The method of mining affects the extent and density of induced fractures which could increase the permeability by several orders of magnitude. This increase in permeability can in turn accelerate the discharge of ground water from mining which leads to a further increase in the relative permeability to gas. The rate of gas liberation during mining, therefore, depends primarily on the virgin gas content of coal and the extent of induced fracture permeability. However, the magnitude of post mining emissions depend mainly on residual gas content, matrix permeability and gas diffusivity through coal and non-coal strata.

It can be shown that if coal mining proceeds at a relatively constant rate during the life of mining, annual emissions from mining can be evaluated by using virgin gas content and production, besides the lithology of strata and geometry of mining (Saghafi, 2010). Gas content is therefore the most important parameter for evaluation of fugitive emissions from mining. Moreover, in view of large outputs of coal in Australia, even small errors in measuring the gas content could lead to large errors in calculated estimates of emissions. In addition to the accuracy of gas content determination, the limit of measurability is also an issue for low shallow coals such as in open cut and 'non gassy' underground coal mines. Traditionally the requirement for gas content testing had been limited to the 'gassy' mines where the safety is the major driving force aimed at evaluation of outburst potentials and high gas emissions. Therefore, low gas content determination has not been a focus of research. At this time the lowest measurability level is about 0.1 to  $0.5 \text{ m}^3/t$ .

Thus the development of more suitable and accurate methods of gas content testing for low gas content coals an important task ahead for coal researchers and the coal mining industry. Another beneficiary of a more accurate method for low gas content coals is the CBM industry which is also active in producing gas from low rank coal regions where coals of low gas contents are present.

#### GAS CONTENT OF COAL

Gas content is generically defined as the volume of gas contained in a unit mass of coal and is generally expressed in cubic metres, at standard pressure and temperature conditions, per tonne of coal (m<sup>3</sup>/t, STP). In Australia the standard conditions are a temperature of 20°C and an absolute pressure of 101.325 kPa (Standards Australia, 1999).

Gas in coal is stored mainly in the adsorbed phase but also in the free phase. Though the contribution of the latter to the total gas volume is small, particularly at shallow depths (<500 m), at greater depths the

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volume of gas in the free phase can be large due to higher density at such depths. From a viewpoint of gas storage in coal, gas content should include both the free and adsorbed volumes, however, the current method of determination does not allow for the measurement of free gas in coal. The current method mainly targets the 'desorbable gas content' and to some extent the 'residual gas content'.

#### Desorbable and residual gas content

As soon as coal is brought to the surface it desorbs its gas. Desorption of gas from coal could continue for days or weeks until there is no 'measurable' gas. The total volume of gas released from coal, when the gas pressure outside the coal is at atmospheric pressure, is called the desorbable gas content ( $Q_d$ ). Gas remaining in coal at this stage is the residual gas content ( $Q_r$ ). Total gas content ( $Q_t$ ) is the sum of the desorbable and residual gas contents (Figure 1).



# Figure 1 - Desorbable and residual gas content defined based on natural release of gas from coal at atmospheric pressure

The rate of gas release from coal depends on the gas concentration gradient (between the inside and outside of coal sample) and the diffusivity of coal. The process of gas release from coal can be considered a combination of instantaneous desorption of gas from the internal surface of pores and diffusion to the fractures. The release of gas from coal can be expressed mathematically assuming a diffusion mechanism as shown in Equation 1,

$$\frac{\partial Q_r(t)}{\partial t} = D \frac{\partial^2 Q_r(t)}{\partial x^2} \tag{1}$$

Where  $Q_r(t)$  is the gas remaining in coal (temporal gas content) at any time *t* (s) after the start of the desorption process and *D* is the gas diffusivity in coal (diffusion coefficient, m<sup>2</sup>/s). Assuming a 'pseudo steady' diffusion mechanism the solution of Equation 1 yields equation 2.

$$Q_r(t) = Q_r + Q_d e^{-t/\tau}$$
<sup>(2)</sup>

 $Q_r$  and  $Q_d$  are the residual and desorbable gas contents (Figure 1). Parameter *r* (tau) is a characteristic time parameter (s) related to the diffusivity of gas in coal, it is sometime called diffusion time constant or desorption time. It is expressed in terms of the diffusivity parameter (*D*) and a diffusion characteristic length (*a*) shown in Equation 3

$$\tau = \frac{a^2}{D} \tag{3}$$

#### Evaluation of parameter *t*

Tau ( $\tau$ ) is a physical parameter related to the diffusivity and the characteristic length for a given gas and coal. If the diffusion coefficient and characteristic length values are not available,  $\tau$  can be evaluated from

the gas desorption testing curve. Based on Equation 2, the volume of gas desorbed from coal since the start of the desorption process is given in Equation 4.

$$Q_d(t) = Q_d(1 - e^{-t/\tau})$$
(4)

Where  $Q_d(t)$  is the volume of gas released since the start of desorption (m<sup>3</sup>/t). In Eq (4), if the time t is replaced by  $\tau$ , the volume of gas released from coal after a period of  $\tau$  would be:  $Q_d(\tau) = 0.63Q_d$ . In other words  $\tau$  is the time required for coal to release 63% of its desorbable gas. Hence,  $\tau$  can be estimated from the gas content testing desorption curve (Figure 2). This method, however, is costly because the slow desorption measurement could take weeks to complete.



Figure 2 - Determination of diffusion characteristic time from the desorption curve

#### MEASUREMENT OF GAS CONTENT OF COAL

Gas content of coal is determined by direct measurement of the volume of gas desorbed from coal. In Australia two methods are routinely used, namely the 'slow desorption' and 'fast desorption' methods (Williams, *et al.*, 1992; Saghafi, *et al.*, 1998). Variants of both methods have been used in other coal mining countries. In France a variant of the fast desorption was used since early 1960's (Bertard, *et al.*, 1970). In the USA variants of the slow desorption method have been used over the years (Kissell, *et al.*, 1973; Diamond and Levine, 1981; Diamond and Schatzel, 1998). The Australian slow desorption method was a developed form of the USBM method after some enhancement (Australian Standard, 1991). The Australian fast desorption method otherwise, known as the quick crash method, was developed in the early 1990's (Williams, *et al.*, 1992; Saghafi, *et al.*, 1998) and is currently the method of choice for assessment of gas emissions and outburst risk in underground coal mines (Australian Standard, 1999).

Though both methods consist of similar steps to determine the gas content of coal, the length of the procedure is significantly longer in the slow desorption method. In the fast desorption method coal is crushed after a short period of natural desorption so that all its gas is forced to release rapidly (in the space of minutes to an hour). An advantage of the fast desorption method besides its rapidity is a reduction in the risk of  $CO_2$  dissolution in the measuring water for mixed gas conditions (when both  $CH_4$  and  $CO_2$  are present in seam gas).

The slow desorption and fast desorption methods are both based on measurement or estimation of the volume of gas desorbed from coal in several stages. Both methods start with estimating 'lost' gas during drilling and retrieval of the coal sample to the surface. In the slow desorption method coal is allowed to desorb its gas 'naturally' until no further desorption is detected. In the fast desorption method, however, after a short time allowed for gas to be released naturally during coal transport to the laboratory and in the lab, the coal is crushed. Note that in the slow desorption method the crushing stage may be also included to determine the 'residual' gas content of coal ( $Q_r$ ).

The three components of gas contents from three stages of gas content testing in the fast desorption method are commonly represented by  $Q_1$ ,  $Q_2$  and  $Q_3$  parameters. The 'measured gas content',  $Q_m$ , is the sum of the 3 components (Australian Standard, 1999) shown in Equation 5,

$$Q_m = Q_1 + Q_2 + Q_3$$

(5)

The  $Q_1$  or the lost gas stage is identical for the two methods. The  $Q_2$ , gas desorbed during transport and in the lab is also called desorbed gas in the slow desorption method and is the main component of the gas content in this method. In this method this stage is allowed to continue until no further measurable gas

desorption is observed. In the fast desorption method the  $Q_2$  step is generally short as coal is crushed soon after reaching lab depending on the availability of a measuring system and proper conditions. The last component of gas content  $Q_3$ , which is the gas desorbed from crushed coal in the fast desorption method, is also usually the largest volume of desorbed gas in this method. For the slow desorption method this stage is often of no importance as residual gas is expected to be small.

#### Errors associated with the standard method of gas content testing

The measurement of the volume of gas released (in all three stages) is done by using a measuring cylinder. The released gas is admitted into a water filled inverted cylinder. The displacement of water provides the measure of the volume. This system has worked well over the years and is used routinely in Australia. There are, however, some problems with this way of measuring the volume of gas including coal oxidation, gas partial pressure effects and dissolution of desorbed gas, particularly CO<sub>2</sub>, into the measuring water. Some of these issues have been addressed over the years and some improvements have been suggested and applied (Saghafi and Williams, 1998; Saghafi *et al.*, 1998; Danell *et al.*, 2003). Measurement of mass (or weight) produces relatively small errors, hence, the accuracy (or error) of measured gas content depends mainly on the accuracy of determination volume. The accuracy of the graduation of the measuring cylinder is, therefore, of primary importance. Accuracy of the graduation depends on the quality and resolution of measuring cylinders. For example glass cylinders are less prone to error than plastic cylinders, which are affected by temperature and other environmental conditions. Burettes have higher resolution but are delicate and their use is limited to the laboratory.

The total error of measurement should be reported in gas content results. It depends on individual errors of  $Q_1$ ,  $Q_2$ , and  $Q_3$ . Measured gas content inherits all these errors and the total error of gas content ( $\varepsilon_m$ ) could be calculated as follow,

$$\varepsilon_m = \varepsilon_1 \frac{Q_1}{Q_m} + \varepsilon_2 \frac{Q_2}{Q_m} + \varepsilon_3 \frac{Q_3}{Q_m}$$
(6)

Where  $\varepsilon_1$ ,  $\varepsilon_2$  and  $\varepsilon_3$  are the errors produced in measuring  $Q_1$ ,  $Q_2$  and  $Q_3$  components. For example in one case we have

 $Q_1 = 0.5 \text{ m}^3/\text{t}, \epsilon_1 = 20\%,$   $Q_2 = 3.0 \text{ m}^3/\text{t}, \epsilon_2 = 15\%,$  $Q_3 = 2.5 \text{ m}^3/\text{t}, \epsilon_2 = 5\%.$ 

The measured gas content is then presented as  $Q_m = 6.0 \pm 0.67 \text{ m}^3/\text{t}$ .

#### **GREENHOUSE GAS ESTIMATION AND GAS CONTENT**

The gas content of coals from shallow seams in open cuts and 'non-gassy' underground mines can be very low. For these coals the conventional method of measurement of volume by using water displacement may not deliver anything meaningful and errors of measurement can be larger than the gas content itself. Currently by using the standard method, the lower limit of gas content can be shown to be about 0.1 to  $0.5 \text{ m}^3$ /t depending on the sample size and the measuring cylinder used. Another source of error for low gas content testing is the correct determination of nitrogen (N<sub>2</sub>) in the seam gas. As N<sub>2</sub> is not a greenhouse gas it is important that its volume be corrected in seam gas. The current method of determination of N<sub>2</sub> content of seam gas is to calculate the 'excess N<sub>2</sub>' volume from the gas composition of the samples collected from desorbed gas. In calculating the N<sub>2</sub> content of seam gas it is assumed that the O<sub>2</sub> deficiency in the desorbed gas is due to coal oxidation. Accordingly, a reliable method of gas content testing for low gas content coal is required, as accurate gas composition and gas content measurements for these coals would have an important impact on large coal mining operations. For low gas content coals ( $Q_m < 0.5 \text{ m}^3/t$ ) the water displacement method of measuring the volume is inadequate. Often for these coals there would be no measureable  $Q_1$  and sometime no measurable  $Q_2$ .

#### Suggestions on measuring low gas content coals

For the measurement of gas content of these coals it is suggested that the best practice would be to seal the fresh sample in a gas tight canister in the field, and then dispatch it to the lab for crushing. Ideally coal should be sealed in a canister which can be directly mounted on the crusher so that there would be no

need to open the coal canister before crushing. The total desorbed gas can then be indirectly evaluated using a method based on measuring of the gas composition rather than measuring gas volume. If coal oxidation is an issue the sample should be flashed by helium gas (He) before sealing the coal canister assuming that there is no  $Q_1$ . If  $Q_1$  is to be estimated then He flushing should take place after measurement for the  $Q_1$  estimate.

Once in the lab coal is crushed in the sealed canister (the CSIRO lab is equipped to crush sealed coal without opening the canister on arrival from the field). Crushed coal in the container is then kept for a period of time to allow for equilibrium and desorption of gas. A gas sample is then taken from the headspace to measure gas composition. Knowing the volume of the canister and the gas composition, gas content can be evaluated. Note that the increase in gas pressure would be minimal due to the low gas content of the coal. However, a gas pressure sensor may be connected to the container to measure the gas pressure. This method was used at the CSIRO in a number of ACARP projects on the determination of residual gas content of coal. The method was applied after the completion of the three stages of gas content testing ( $Q_1$ ,  $Q_2$  and  $Q_3$ ). This new component of gas content was called  $Q_3$ ' ( $Q_3$  prime).

For routine measurement of low gas content coals a design similar to the set up conceptually illustrated in Figure 3 is suggested. After completion of  $Q_1$  and  $Q_2$  stage of testing coal is crushed using the lab standard crusher. Note that the crushing canister should be initially flushed with He gas to reduce air in the system and also to reduce the seam gas partial pressure. After completion of the crushing and allowing time for temperature equilibrium, the canister is opened to a closed circuit with an in-line vacuum pump. More helium can be allowed into the system to further reduce the partial pressure of desorbed seam gas. Gas samples are collected from the system after sufficient periods of time for gas composition measurement. Knowing the volume void space and the seam gas composition the gas content can be determined.



#### Figure 3 - Schematic diagram of proposed system for low gas content testing

The lower limit of gas content which can be determined using this method can be evaluated from the knowledge of void volume in the system (crusher and piping) and the lowest or optimal lower limit of gas chromatography in use. For instance, if the void volume is about 500 cm<sup>3</sup> (typical void in the CSIRO quick crush canister for a 100 g coal sample), and a GC which can accurately measure a concentration of 100 ppm of methane is used, then the gas content of about 0.001 m<sup>3</sup>/t can be theoretically determined. This method can, therefore, theoretically measure, gas content values of 100 times smaller than the standard method.

#### Gas content determination and excess nitrogen

Nitrogen is frequently reported in the desorbed gas composition. While nitrogen is also a by-product of the coalification process, its high values in reported measurements is of concern. For the purpose of greenhouse gas emissions inventory it would be required to quantify the N<sub>2</sub> component of the seam gas as accurately as possible. One cause of high N<sub>2</sub> reported for 'non gassy' coals could be O<sub>2</sub> absorption and oxidation. Coal and carbonaceous materials oxidise in the presence of air and slow oxidation occurs at ambient temperatures. Laboratory and field measurements of gas emitted from coal in the presence of air show that the overall low temperature oxidation (<75°C) of coal and carbonaceous materials produce mainly CO<sub>2</sub> and to a lesser extent CO (Carras *et al.*, 1994; Saghafi *et al.*, 1995; Roberts *et al.*, 2003). CO<sub>2</sub> is the main result of the reaction and uses about 90% of the total consumed O<sub>2</sub> (see for example Wang *et al.*, 2003). If seam gas did not contain CO<sub>2</sub> then it was possible to estimate N<sub>2</sub> content of coal seam gas

from the knowledge of the  $O_2$  and  $CO_2$  composition. However, seam gas in Australia is often rich in  $CO_2$  and new methods are required to identify and quantify the true  $N_2$  content of coal seam gas.

#### CONCLUSIONS

The current method of determination of gas content for the low gas content of non gassy coals is prone to error. As the gas content is the most important parameter for estimating mine emissions, new methods are required to increase the accuracy and measurability of the gas in coal. The high nitrogen content of seam gas reported in gas content testing using the standard method, particularly for low gas content coals, is also an important issue which requires attention and development of new methods to quantify the true  $N_2$  content of coal seam gas.

A new method of measurement for low gas content coals is suggested. The new method is based on lowering seam gas partial pressure in the coal canister and the measurement of gas composition. The new method should largely increase the limit of measurability and accuracy of the gas content testing. Analysis of the method indicates that theoretically gas content of two orders of magnitude smaller can be measured by using the new method instead of current methods.

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