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# COAL RESERVOIR PARAMETERS REGULATING GAS EMISSIONS INTO AND FROM COAL MINES

Abouna Saghafi<sup>1</sup>

**ABSTRACT:** A few number of gas reservoir parameters regulate the intensity and the extent of gas emissions during and following the mining of coal. Gas content is one of the most important of these parameters. Depending on the purpose of its quantification its accurate determination could be vital to the mining activities. For instance, if this parameter is used to evaluate the outburst and its value falls near the threshold limit it needs to be accurately measured. Similarly when seam gas emissions from coal mines, is to be calculated, an accurate measurement of this parameter in a carbon constraint economy has a very important economic impact. The other challenge associated with gas content is the lower limit of measurability of the standard systems. For instance for low to very low gas content ( $<0.1 \text{ m}^3/\text{t}$ ) encountered in 'non-gassy' underground and open cuts, the standard method is unable to deliver accurate values. A different methodology is then required to evaluate the gas content in these conditions.

Another parameter, important in evaluation of the intensity of the emissions and its time dependency nature is the gas diffusivity parameter. While the saturation indicates the onset of gas desorption, the diffusivity parameter controls the rapidity of gas movement from the micro storage sites into the larger fractures and voids. Diffusivity is, therefore, the primary rate limiting factor in the intensity of gas emissions. The diffusivity is not often measured directly and a diffusion time constant, called Tau, is often used to indicate the speed of diffusion flow in coal. This parameter is also used in numerical simulators which use a simplified model of gas diffusion, namely pseudo state diffusion models.

This paper discusses the current and new methodologies to determine the main parameters of coal reservoirs including gas content and gas diffusivity and potential errors associated with current measurement methods.

## INTRODUCTION

Coal is a porous rock and can contain large volumes of gas, hence it is considered a gas reservoir. The major components of coal seam gas are methane ( $\text{CH}_4$ ) and carbon dioxide ( $\text{CO}_2$ ). Subsidiary volumes of ethane ( $\text{C}_2\text{H}_6$ ) and higher hydrocarbons ( $\text{C}_{2+}$ ), and nitrogen ( $\text{N}_2$ ) can also be present in some coal seams ( $\text{C}_{2+}$  at high depths).

Gas currently present in a coal seam can be of primary or secondary origin. Primary gas has been generated as a by product of the coalification process during which large volumes of  $\text{CH}_4$  and  $\text{CO}_2$  are produced (thermogenic gas). Some volumes of gas generated would be adsorbed by coal but most would escape the site.

Methane gas can be also generated within the coal seam, as a result of microbial activities. In this case coal seams have to act as a permeable aquifer, allowing the movement and storage of the methanogenous micro organism and nutrients. Thus, most of the coal seam methane at fairly shallow depths is of biogenic origin. Igneous activities over geological time have also resulted in the injection of  $\text{CO}_2$  into coal seams replacing methane in some places.

Coal seam gas is stored in pore volume and surface spaces in free and adsorbed phases. The greatest portion of the stored gas, at shallow to medium depths, is held on the pore surfaces in adsorbed phase. The largest part of the pore surfaces are located in the micro pores system (size  $<2 \text{ nm}$ ) which are only a few times larger than the coal seam gas molecular sizes. The adsorbed phase storage in the micro pore system follows a pore filling mechanism and therefore reaches its maximum value (adsorption capacity) when the pore system is fully filled. The stored gas is then retained due to a combination of adsorption and capillary forces.

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Emissions from coal are intensified during mining due to generation of multitude of fractures and fissures within the coal seam and in the strata above and below the coal mined seams. Total volume of gas liberated depends basically on gas content of coal. The rate of gas liberation, however, depends on both gas content and gas diffusivity of coal.

The methodologies used and the accuracy of measurement of these two parameters, i.e., gas content and diffusivity, would influence the results and the estimation of emissions. In particular in case of coals of low gas content (<0.1 m<sup>3</sup>/t), the relative error of the measurement can be very high because the lower limit of measuring system is attained. While measurement of low gas content may not be of any importance to safety issues in underground mining it is of quite importance for greenhouse gas emissions inventory which would be required in carbon retrained economy of the near future.

This paper describes the general mechanism of diffusion in coal and methodologies and accuracy issues inherent to the current methods of measuring the desorption and gas content of coal and the effect of gas diffusivity.

### GAS DIFFUSION IN COAL

Gas is diffused in coal under the forces of gas concentration gradient. The desorption of gas is limited by the diffusivity property of coal. A higher diffusivity allows faster desorption of gas from coal. The diffusivity affects the evaluation of gas content particularly the estimated value of the lost gas during drilling and at the surface. Diffusivity is also an important input to gas reservoir models which is directly used or indirectly in terms of a diffusion time constant  $\tau$  (Kolesar et Ertekin, 1986; King et al, 1986; King, 1993).

Gas diffusion in coal can be mathematically expressed by Fick's law and by assuming that the change in gas concentration per unit of time and in a unit volume of the medium (coal) is equal to the difference between the volumes of gas diffused into and out of the elemental volume of the medium. In its general form it is written as,

$$\frac{\partial c}{\partial t} = \text{div}(D \cdot \nabla c) \quad (1)$$

where  $c$  is the gas concentration (gas content) and  $D$  is the diffusion coefficient (or diffusivity of gas in coal). Solution of Eq(1) is not straightforward and often numerical methods should be employed in particular for cases where  $D$  changes in time and space or the medium is of complex layout (for exact solutions see Crank, 1975).

Barrer and Brook (1953) investigated the molecular diffusion and adsorption of gases in powdered zeolites. They found that irrespective of shape of the powder (cube, parallelepiped, sphere or cylinder) the diffusion in early stages of gas flow can be explained by,

$$\frac{Q}{Q_m} = \frac{2A}{V} \sqrt{\frac{Dt}{\pi}} \quad (2)$$

where  $Q$  is the volume of gas desorbed since the start of diffusion,  $Q_m$  is the total gas initially contained in coal and  $t$  is the time elapsed.  $A$  and  $V$  are the surface area and volume of powders.

Some researchers have used this equation for gas desorption from coal at early stage of desorption. Gunther (1965) used this equation for gas desorption up to release of 20% of the total gas in coal. Others extend the use of the equation for up to 50% of total gas initially in coal (Smith and Williams, 1984).

For spherical grains, this equation can be simplified to,

$$\frac{Q}{Q_m} = \frac{6}{\sqrt{\pi}} \sqrt{\frac{Dt}{a^2}} \quad (3)$$

where  $a$  is the radius of spheres approximating coal grains (half of the average cleat spacing). The diffusion coefficient is expressed in unit of square m per second (m<sup>2</sup>/s) and radius is in meter (m).

A full and exact solution for the unsteady diffusion of gas in coal can be obtained provided the assumption of uniformity of the diffusion coefficient in space and time and that coal can be seen as an

assembly of spherical grains. These assumptions are justified by the fact that coal is highly and uniformly fractured (cleats) so that the matrix can be thought of spheres delineated by cleats. The solutions have been given various authors (Carslaw and Jaeger, 1959; Crank, 1975) and have been used by coal workers (Walker and Mahajan, 1978; Smith and Williams, 1984), The cumulative volume of gas desorbed from coal, using these solutions, is,

$$\frac{Q}{Q_m} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp(-n^2 \pi^2 t / \tau) \quad (4)$$

where  $\tau$  is the diffusion time constant, defined as,

$$\tau = \frac{a^2}{D} \quad (5)$$

The parameter  $\tau$  (Tau) is an important property of gas and coal and often is used to replace the diffusion coefficient when the direct measurement of  $D$  is not possible. Intuitively Tau can be thought as the time required for the diffusion flow to advance a distance 'a' in the porous medium. This parameter is used in many of gas flow simulators where a pseudo steady state diffusion flows approximates the true unsteady diffusion flow considered. In a full unsteady diffusion flow model the diffusivity  $D$  can be directly used to estimate the free gas volumes released from coal matrix into fractures.

### MEASUREMENT OF GAS CONTENT OF COAL

Gas content is measured using either a slow desorption or a fast desorption method. Both methods have been used in various forms over the years (Bertard et al, 1970; Kissell et al, 1973; Williams et al., 1992; Diamond et Schatzel, 1998; Saghafi et al, 1998; Australian Standard, 1999). Though both methods consist of similar steps to determine the gas content coal, the length of the procedure is significantly longer in slow desorption method. In fast desorption method the time of testing is significantly reduced by accelerating the desorption rate (diffusivity). Coal is crushed and all gas is released in space of an hour or two long before if it were naturally to desorb its gas.

The slow desorption and current Australian fast desorption methods are both based on measurement or estimation of volume of gas desorbed from coal in several stages. For fast desorption there are three stages which delivers the three components of the 'measured' gas content. In slow desorption the last stage may not exist depending on whether a residual gas content testing is required or not.

The three components of gas content in the slow desorption method correspond to three regimes of gas desorption, i.e., 1- loss gas (initial desorption), 2- desorbed gas and 3- residual gas. In the fast desorption method, however, these stages are basically three steps in gas content testing and are not related to gas desorption kinetics of coal. The three components of gas content measured are commonly represented by  $Q_1$ ,  $Q_2$  and  $Q_3$  terms. The 'measured gas content',  $Q_m$ , is the sum of the 3 components (Australian Standard, 1999),

$$Q_m = Q_1 + Q_2 + Q_3 \quad (6)$$

The  $Q_1$  or the lost gas is the volume of gas desorbed from coal during the drilling and prior to its seal in gas tight canisters. This stage is identical for the two methods. The  $Q_2$  is the gas desorbed during transport and in the lab. It is called desorbed gas and is the main component of the gas content in slow desorption method. For this method this stage is allowed to continue until no further measurable gas desorption is observed. In fast desorption method  $Q_2$  step is generally short as coal may be crushed any time depending on the availability of measuring system and proper conditions. The last component of gas content is  $Q_3$  which is the gas desorbed from crushed coal.  $Q_3$  measurement is the most important stage of gas content testing for the fast desorption method. Coal gives away most of its gas in this stage. For slow desorption this stage is often of no importance as  $Q_3$  is expected to be low (very low residual gas content).

The measurement of the volume of gas released in the three stages is usually done by using a measuring cylinder. The released gas is admitted into a water filled inverse 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 including gas partial pressure effect and dissolution of gas in water. These have been addressed over the years and improvements have been suggested and applied (Saghafi and Williams, 1998; Saghafi et al, 1998; Danell et al, 2003).

The volume of gas released from a coal sample can be low either because the low gas content or because of the small sample size. The latter may happen when the cost of coring is high (deep coal) and/or a large part of the core section is used for other more urgent testings including quality and geotechnical measurements. In these cases the method of measurement of volume by using water displacement could not be applied.

To illustrate the limit of measurability using the standard method it should be noted that the smallest volume that can be confidently measured by using the water displacement in measuring cylinder, is about 2-5 cm<sup>3</sup>. Therefore for a sample of 100 g size the method can measure low gas content of about 0.02 - 0.05 m<sup>3</sup>/t. These are the very real limits of measurement and though they have no impact on gas content testing for safety purposes they have important impact on coal mining economics whence the mitigation of greenhouse and carbon tax are introduced into the mining economy.

The other issue is the estimation of  $Q_1$  which is based on initial desorption measurement in the field. Theoretically the measurement should start as soon as the core is retrieved from the exploration gas hole and visually logged by field geologist. However, this is not always the case and the kinetics of desorption may change if the delay is large. In addition there are debates on the effect of measurement temperature and whether the in-situ temperature should be used.

In the next section these questions are discussed and methodologies for overcoming the issues are suggested.

### Measurement of low gas content

For very low gas content coals ( $Q_m < 0.1$  m<sup>3</sup>/t) or when the coal mass available for crushing is very small the water displacement method of measuring the volume is not adequate. In such cases normally there would be no measureable  $Q_1$  and often no measurable. Coal is crushed as soon as it reaches the gas lab and residual gas ( $Q_3$ ) is determined. This is the case for most of non-gassy underground mines and surface mining.

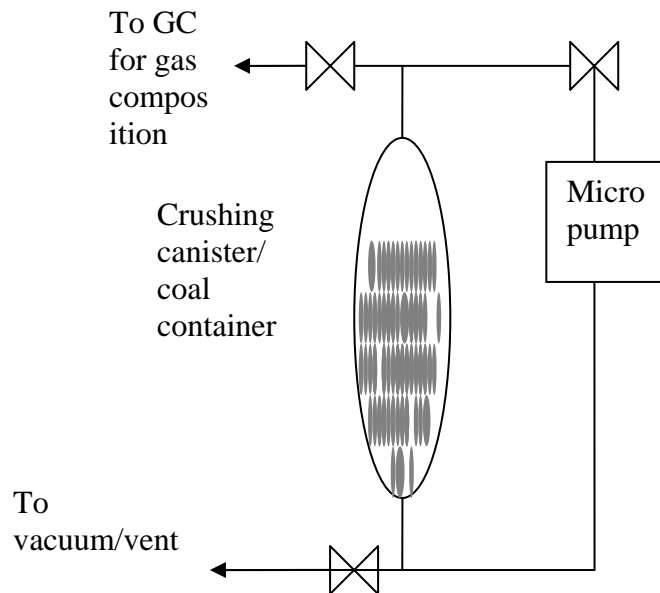
For measurement of low gas content (non-gassy coals) the best practice is to seal the fresh sample in purposely gas tight canister in the field, and then dispatch it to the lab for crushing. Ideally coal should be sealed in a canister which can be directly mounted on the crusher so that there would be no need to open the coal canister before crushing. The total desorbed gas can then be indirectly evaluated using a gas composition testing method.

The indirect method of determining gas volume by measuring gas composition had been used over the years in some old coal mining countries in Europe. The method was used to obtain the 'total gas content' which is theoretically larger than the 'measured gas content' and includes post  $Q_3$  component of gas content. The method consists of keeping the crushed coal in the crusher container for sometime after the completion of crushing. Then gas composition in crusher canister is measured. The volume of desorbed gas is determined from the knowledge of void volume in the crusher canister and gas composition values.

This method was used by CSIRO in the course of a number of ACARP projects to deliver the total gas content. This new component of gas content is called  $Q_3'$  ( $Q_3$  prime)

For routine measurement of low gas content of coal we suggest a similar approach. The set up is conceptually illustrated in Figure 1. The crushing canister is initially flushed with nitrogen and then coal is placed in the crusher and crushed. After the completion of the crushing and allowing time for temperature equilibrium, the canister is opened to a closed circuit with an in-line pump. A gas sample is collected from the system after a sufficient period of time and gas composition is measured using gas chromatography. Knowing the volume of the total void space in the system the gas content is determined.

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 volume of void is about 500 cm<sup>3</sup> (typical void in the CSIRO quick crush canister for a 100g coal sample), and a GC which can measure accurately a concentration of 100 ppm of methane (many GC's can measure concentration values below 10 ppm of methane) is used, then the gas content of about 0.0005 m<sup>3</sup>/t can be determined. This method, therefore, can measure gas content values of at least 100 times smaller than the standard method.



**Figure 1 - Schematic diagram of measurement of residual gas content for very low gas content coal**

#### Estimation of lost gas, $Q_1$

The  $Q_1$  component of gas content is determined by extrapolating back the gas desorption curve to the time when the drill bit hit the coal. Measurement of initial desorption is undertaken in the field as soon as the core is available after its retrieval from the borehole.

Based on the discussion in previous sections in the early stages of gas desorption the cumulated volume of gas follows a linear equation of square root of time. As discussed this linearity is analytically demonstrated from the solution of the equation of gas diffusion for broken spherical pieces of coal, i. e.,

$$\frac{Q}{Q_m} = \frac{6}{\sqrt{\pi}} \sqrt{\frac{t}{\tau}} \quad (7)$$

where  $\tau$  is the diffusion time constant or Tau. If the cumulative volume of desorbed gas is plotted against the square root of time, generally the desorption curve has the following mathematical expression,

$$q(t) = k\sqrt{t} - Q_1 \quad (8)$$

In this equation  $q(t)$  is the volume of gas desorbed since the start of measurements but  $t$  is the time since the start of desorption in the borehole (in practice time zero is the mid time between the time the drilling hit the coal and the end of the coring run). From Eq (8) at time  $t_0$ ,  $q(t_0) = 0$ . Therefore the lost gas  $Q_1$  which obtained from the intercept of the regression line;  $Q_1 = -q(0)$ . The lost gas can also be obtained from the slope of the regression line,

$$Q_1 = k\sqrt{t_0} \quad (9)$$

$t_0$  is the lost time or the time elapsed since the drill bit had hit the sample in the borehole until the sample is sealed in the canister at the surface for desorption measurement. The released gas during the field measurement is added to the  $Q_2$  component of gas content.

In Figure 2 a typical field measurement of the initial gas desorption rate for estimation of lost gas is shown. In this case some 20-25 minutes ( $t_0$ ) had passed before the sample could be sealed and measured.

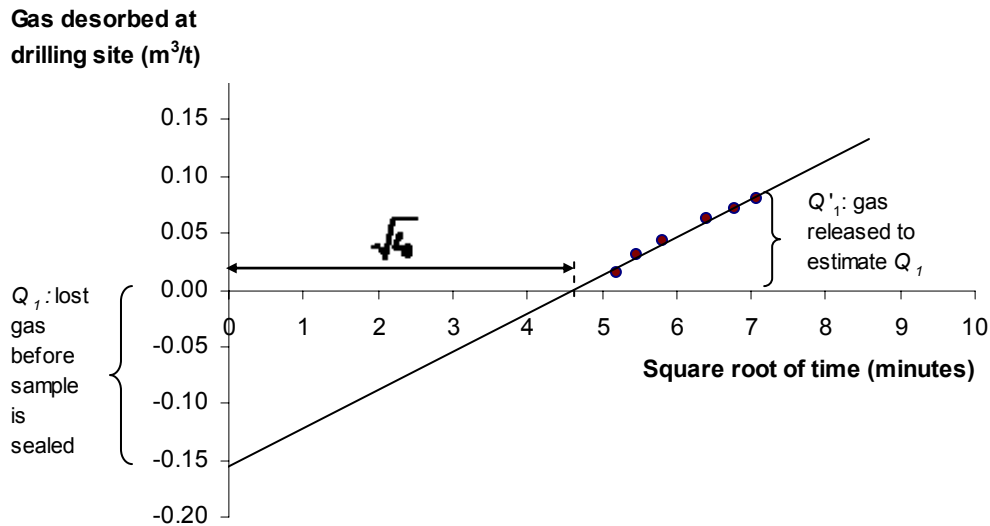


Figure 2 - Measurement of initial desorption rate for estimation of lost gas ( $Q_1$ )

Note that initial desorption rate can be used to estimate the diffusion time constant  $\tau$ . This is possible after all three component of gas content are evaluated,

$$\tau = \frac{36}{\pi} \left( \frac{Q_m}{k} \right)^2 \quad (10)$$

In the above equation  $Q_m$  is the 'measured gas content ( $Q_m = Q_1 + Q_2 + Q_3$ )

#### Accuracy of estimating $Q_1$ from the initial desorption rate data

Two of the concerns that have been raised in relation to the accuracy of estimating the lost gas are - the maximum length of the time that can be tolerated before starting the measurement ( $t_0$  time) and - at what temperature the field measurement of desorption rate should be carried out.

#### Effect of the length of the lost time, $t_0$

The estimation of  $Q_1$  is based on the assumption that desorbed gas volume is a linear function of the square root of time and therefore the loss gas can be estimated by extrapolating back the regression line. However, Eqs. (7) and (8) which are used for the extrapolation are only valid for short values of  $t_0$  or more accurately for small values of  $t_0/\tau$ . Therefore, the length of the lost time ( $t_0$ ) would directly affect the magnitude of the error. The error of estimation is greater for larger  $t_0$ . The acceptable values of  $t_0$  depend primarily on the diffusion time constant ( $\tau$ ). This constant in turn depends on diffusion coefficient and fracture/cleat spacing. Imposing a condition of validity for use of these equations such as  $Q/Q_m < r_0$  then the maximum value of  $t_0$  for each individual case can be assessed.

#### Effect of the temperature of measurement

The temperature of desorption measurement at the drilling site is believed to affect the results. Some authors (Mavor and Pratt, 1996) recommend measuring gas desorption at in-situ coal temperature. This, however, can be a source of error by itself. This is because gas would not desorb fully until coal is pulled out from water. Secondly as soon as gas start desorbing the temperature rapidly fall the core is being pulled out from the borehole. Hence, the measurement of desorption rate at in-situ

temperature can falsely increase the value of the lost gas content. The effect of temperature on desorption rate is due principally to effect of temperature on the diffusivity of gas in coal. It can be shown that the relative error in  $Q_1$  estimate is equal to half of the relative variation in diffusivity, i.e.

$$\frac{\delta Q_1}{Q_1} = 0.5 \frac{\delta D}{D} \quad (11)$$

The potential error of determination of  $Q_1$  due to using a different temperature can be estimated by studying the effect of temperature on gas diffusivity.

### DIRECT MEASUREMENT OF GAS DIFFUSIVITY IN SOLID COAL AND CALCULATION OF DIFFUSION TIME CONSTANT (TAU)

Gas diffusion coefficient can be indirectly estimated either from desorption curves generated in slow desorption method of gas content testing or from desorption isotherm data. In both cases numerous assumptions are required. For pulverized coal used in sorption tests the indirect results of diffusivity may differ considerably from gas diffusivity for solid coal. The diffusivity, however, can be directly measured. Recently new methods are presented (Saghafi et al, 2007) allowing direct measurement of diffusivity of coal. In these methods gas is flown through solid coal by maintaining a gas concentration gradient across a coal disk of small thickness (<5mm). The small thickness allows the diffusion test to take place in reasonable time (about a week). The diffusivity obtained from this method can be used in evaluating the diffusion time constant tau. For full unsteady state models the diffusion coefficient can be used in the models.

#### Estimation of Tau from gas content data

Tau ( $\tau$ ) is physical parameter related to diffusion rate and in the absence of a direct measurement of diffusion coefficient can be used in simplified diffusion flow models to simulate the flow of gas from microspores into fractures. It also gives a feel for the speed of diffusion of gases in coal. Tau can be derived from initial desorption curve which is established in previous section Eq (10). The value of Tau ( $\tau$ ) obtained in this way can be used both in unsteady and steady models. This is also an economic way of measuring  $Q_1$ . However if the gas desorption can not be measured at site because of logistics or very low gas content of coal the method can not be applied. A direct diffusivity test should then be undertaken in the laboratory.

The numerical models originated from petroleum and conventional gas industries often use a pseudo steady state diffusion mechanism presented initially by Warren and Root (1963). This is to simulate gas desorption from primary porosity (coal matrix) into fractures.

According to the pseudo-steady state diffusion model, the desorption/diffusion rate out of coal is proportional to the difference between the average gas concentration in the coal matrix and the gas concentration in the fractures, i.e.,

$$\phi = \frac{-D}{a^2}(c - c_f) \quad (12)$$

where  $\phi$  is the gas desorption rate from a unit volume of matrix,  $c$  is the matrix average gas concentration and  $c_f$  is the gas concentration in fractures.  $D$  is the diffusion coefficient and  $a$  is the radius of coal grain (half of cleat spacing). Gas desorption reduces the gas content of matrix and the change in unit time should be equal to the flow rate of desorbed gas out of matrix, i.e.,

$$\frac{dc}{dt} = -\frac{c - c_f}{\tau} \quad (13)$$

Note that  $a^2/D$  in Eq (12) is replaced by its other representation, namely parameter  $\tau$ . The solution of equation (13) yields the variation of gas concentration in the coal matrix as a function of time,



$$\frac{C - C_f}{C_m - C_f} = e^{-t/\tau} \quad (14)$$

$C_m$  is the initial gas content of coal which should all fall to gas concentration in fractures if sufficient time is allowed.

In terms of the volume of gas released to total volume of gas initially in coal the above relation can be rewritten as,

$$r = \frac{Q}{Q_m} = 1 - e^{-t/\tau} \quad (15)$$

Based on Eq(15) when desorption time equals  $\tau$ , the value of  $(\tau)r$  would be  $\sim 0.63$  which means that coal would release more than 63% of its initial gas after a time  $\tau$  passed start of diffusion.

If gas desorption from coal follows a pseudo steady state mechanism then it is legitimate to evaluate the diffusion time constant  $\tau$  from gas content testing data. In this case the time required for coal to release 63% of its total gas would be obtained from various gas content desorption curves data and an average value of Tau ( $\tau$ ) is determined. This method is, however, costly because the slow desorption measurement should be carried out on its totality which may take weeks or sometime months.

Some gas workers had presented empirical relations similar to the Eq (15). For example Airey (1968) had suggested an equation for emission from broken coals as follows,

$$r = \frac{Q}{Q_m} = 1 - e^{(-t/t_0)^n} \quad (16)$$

Based on the measurement of gas desorption for different rank coals these workers suggest  $n$  values varying from  $1/3$  to  $1/2$ . Note that  $t_0$  in Eq (16) is similar to  $\tau$  in Eq (15).

## CONCLUSIONS

Gas content is the most important parameter to be evaluated for any study of coal seam gas irrespective of the end use. For low gas content conditions the current methodologies are not accurate. While from a mine safety viewpoint the accuracy of measurement of low gas content is not an issue, it is of vital for calculation of the greenhouse gas emissions. Accurate measurement of gas content is required to calculate the emissions from 'non-gassy' or class B mines (underground) and open cut mines. Because of the large volume of coal mined any small error at measurement point can be magnified significantly in the final results of emission calculation. In the context of a carbon constraint mining economy, a new methodology of gas content measurement is required. A method of measurement for low gas content coals is presented. The method is based on the measurement of gas concentration rather than gas volume in the standard methods. The analysis of the method indicates that gas content of a hundred times smaller can be measured using the new method.

The importance of diffusion flow in gas content measurement and in particular in measurement of loss gas ( $Q_1$ ) was discussed. The desorption rate is a function of diffusivity which is sensitive to temperature. However, measuring the initial gas desorption at in-situ temperature can be a source of larger errors and therefore overestimating the gas content.

The desorption time constant (Tau  $\tau$ ) was analysed and its relation to the diffusion coefficient (diffusivity) was discussed. It is possible to estimate this parameter from the initial rate of desorption which is required for estimation of  $Q_1$ . A less accurate, and more costly, is to measure gas content using the slow desorption method and obtain a desorption curve. In this method Tau ( $\tau$ ) is the time

that 63% of total gas is desorbed. It was shown that Tau can also be determined from direct measurement of diffusivity of gas in solid coal.

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