



Review

Enhanced coalbed methane recovery

Marco Mazzotti^{a,*}, Ronny Pini^a, Giuseppe Storti^{b,c}^a ETH Zurich, Institute of Process Engineering, Sonneggstrasse 3, CH-8092 Zurich, Switzerland^b ETH Zurich, Institute for Chemical and Bioengineering, Wolfgang-Pauli-Strasse 10, CH-8093 Zurich, Switzerland^c Dipartimento di Chimica, Materiali e Ing. Chimica "Giulio Natta", Sede Mancinelli, Via Mancinelli 7, I-20131 Milano, Italy

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ABSTRACT

The recovery of coalbed methane can be enhanced by injecting CO₂ in the coal seam at supercritical conditions. Through an in situ adsorption/desorption process the displaced methane is produced and the adsorbed CO₂ is permanently stored. This is called enhanced coalbed methane recovery (ECBM) and it is a technique under investigation as a possible approach to the geological storage of CO₂ in a carbon dioxide capture and storage system. This work reviews the state of the art on fundamental and practical aspects of the technology and summarizes the results of ECBM field tests. These prove the feasibility of ECBM recovery and highlight substantial opportunities for interdisciplinary research at the interface between earth sciences and chemical engineering.

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Contents

1. Introduction	619
2. Competitive adsorption	620
3. Swelling and permeability	621
4. Mass transfer and injection/displacement simulations	623
5. Field tests and ECBM modeling	623
6. Concluding remarks	625
Acknowledgements	625
References	625

1. Introduction

Carbon dioxide capture and storage (CCS) is a set of technologies for the capture of CO₂ from its anthropogenic point sources, its transport to a storage location, and its isolation from the atmosphere [1]. This is only one, though very important, option in a portfolio of actions to fight the increase of atmospheric CO₂ concentration and to mitigate climate change, while at the same time allowing for the continued use of fossil fuels [2]. Deployment of CCS technologies is expected to be limited in the next 5–10 years, but to contribute significantly to the reduction of CO₂ emissions 20 years from now. Capture of CO₂ using existing separation techniques can be applied to large point sources, i.e. power plants or industrial plants; CO₂ can be easily transported over large distances using pipelines and ships; finally CO₂ can be permanently stored in suit-

able deep geological formations, namely deep saline aquifers, oil or gas reservoirs, and unmineable coal seams, or it can be fixed in carbonates [1].

Unmineable coal seams are estimated to have a smaller potential for CO₂ storage than other geological formations but still significant, i.e. from 3 to 200 GtCO₂ capacity to be compared to current anthropogenic CO₂ emissions of almost 30 GtCO₂ per year [1]. Once injected in the coal seam, CO₂ is adsorbed and retained permanently, if a sealing caprock is present. At the same time, thanks to its higher affinity for coal [3], CO₂ displaces coalbed methane, which is formed during coal formation, and enhances its primary recovery, which is performed by pumping out water and degassing the reservoir and is typically limited to only 20–60%. Such enhanced coalbed methane recovery (ECBM, see Fig. 1) allows in principle offsetting the costs associated to CCS operation, as it is the case for enhanced oil recovery [1].

Once injected underground, CO₂ is trapped as a dense gas in the coal cleats, adsorbed on and absorbed in the coal (see Sections 2 and 3), and solubilized in the formation water. Optimal storage

* Corresponding author. Tel.: +41 44 6322456; fax: +41 44 6321141.

E-mail address: marco.mazzotti@ipe.mavt.ethz.ch (M. Mazzotti).

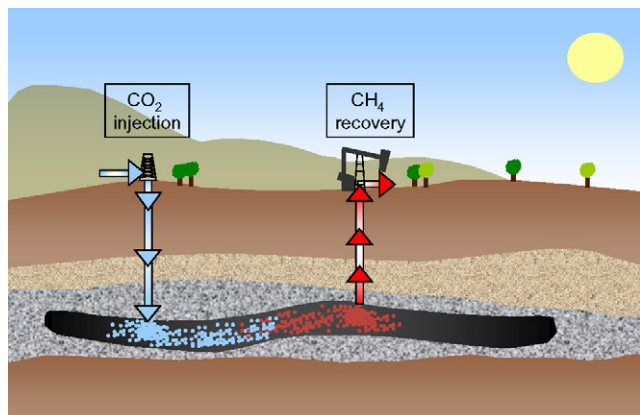


Fig. 1. Schematic of an ECBM operation, where captured CO₂ from a power plant is injected into the coal seam and CH₄ is produced. Injection and production wells can in general be more than one.

conditions are attained at high density, i.e. at a depth of more than 750 m, where pressure is more than 75 bar and temperature is about 40 °C or more, and therefore CO₂ is supercritical. From an engineering point of view ECBM recovery is thus an adsorption/desorption process at supercritical conditions in a natural underground coal formation, which is accomplished by injecting CO₂ in one or more injection wells and by collecting CH₄ from one or more production wells.

ECBM recovery is not yet a mature technology, in spite of the growing number of pilot and field tests worldwide that have shown its potential and highlighted its difficulties [4–8]. Research is active both in the laboratory and in the field in order to address these issues, and to assess storage capacity (see Section 2), to understand adsorption/desorption dynamics during injection (Section 2), to characterize coal swelling and permeability (Section 3), to develop predictive tools for ECBM operations (Section 4). A thorough understanding of the different mechanisms acting during ECBM achieved through the aforementioned experimental and theoretical studies will allow to critically assess the success or the failure of the field tests (Section 5) as well as the feasibility of future demonstration projects, thus paving the way for ECBM's commercial deployment.

This review puts more emphasis on the fundamental mechanisms involved in ECBM recovery, which are quite typical of chemical engineering phenomena. Geophysical and geochemical aspects are not in the focus as they have been recently reviewed in detail elsewhere [9].

2. Competitive adsorption

Being ECBM controlled by adsorption of CO₂ and by desorption of CH₄, competitive adsorption equilibria involving these two gases and nitrogen are of key importance. Nitrogen is of interest as it has been used for ECBM already [4] and might be considered for co-injection with CO₂ in the future (see Section 4). Moreover, pure CO₂ and pure CH₄ adsorption isotherms provide an estimate of the coalbed capacity for CO₂ storage and of the maximum theoretical amount of coalbed methane, i.e. the so-called maximum Gas In Place, GIP_{max}, respectively.

The gravimetric [10] and volumetric [26,31] methods are commonly used to measure single component adsorption isotherms, mostly using a powdered coal sample, in order to minimize the equilibration time. This must be complemented in the case of multicomponent adsorption measurements by gas chromatography in order to measure the exact gas phase composition at equilibrium [34,35]. To be useful, these measurements have to cover a broad range of pressure conditions, from subcritical to supercritical, and should involve both dry and wet samples since coal seams are saturated with water. As we will see, unfortunately techniques for the measurement of adsorption isotherms on wet samples are not as well established as those on dry samples.

In the case of coal, the uptake of CO₂, CH₄ and N₂ is a combination of adsorption on its surface and penetration (sorption) into its solid matrix, the latter resulting in coal swelling [36]. In the case of non-swelling commercial adsorbents, e.g. zeolites, the only truly measurable quantity at high pressure is the excess adsorbed amount, i.e. the amount adsorbed minus the theoretical quantity held up in the adsorbed phase volume at the bulk density [37]. In the case of coal therefore, the only truly measurable quantity accounts for the effect of both adsorption and sorption, whose contributions cannot be separated [10,18,38–40]. This is not always acknowledged in the literature that reports adsorption data on dry and wet samples from different coal mines around the world, a summary of which is given in Table 1.

As an example of general validity, Fig. 2 shows the single component adsorption isotherms of the three gases on the same dry Australian coal at 55 °C, measured at CSIRO (Newcastle, Australia) as reported earlier [13] and in our lab using the equipment and procedure described elsewhere [19]. The experimental data are in excellent agreement and exhibit the usual behavior of excess adsorption isotherms. They show that CO₂ adsorbs more than CH₄, and CH₄ more than N₂, in accordance with several other studies [10,18,19,26]. This property is of key importance for ECBM application and is confirmed by several multicomponent adsorption

Table 1
Studies reporting high-pressure gas adsorption measurements on coal at typical coalbed temperature

Coal origin	Single component adsorption					Competitive adsorption				
	Dry/wet	Gas	P_{\max}	Method	Reference	Dry/wet	Mixtures	P_{\max}	Method	Reference
Bowen Basin, Australia	Dry	CO ₂ /CH ₄	200	G	[10]	–	–	–	–	
Sydney Basin, Australia	Wet	CO ₂	60	G	[11]	Dry	CO ₂ /CH ₄ /N ₂	52	V–C	[12]
Australia	Dry/wet	CO ₂ /CH ₄ /N ₂	200	G	[13–15]	–	–	–	–	
Qinshui Basin, China	Wet	CO ₂ /CH ₄	100	V	[16]	–	–	–	–	
Warndt Colliery (Saar), Germany	Dry/wet	CO ₂	200	V	[17]	–	–	–	–	
Nottinghamshire Coal Field, UK	Dry/wet	CO ₂	200	V	[17]	–	–	–	–	
Sulcis Coal Province, Italy	Dry	CO ₂ /CH ₄ /N ₂	200	G	[18–20]	Dry	CO ₂ /CH ₄ /N ₂	150	G–C	[18,20]
Ishikari Coal Field, Japan	Dry	CO ₂ /CH ₄ /N ₂	60	V	[21]	Dry	CO ₂ /CH ₄ /N ₂	60	V–C	[21]
Silesian Basin, Poland	Dry/wet	CO ₂ /CH ₄	200	V	[17,22]	Dry/wet	CO ₂ /CH ₄ /N ₂	230	V–C	[23,24]
Achterhoek Area, The Netherlands	Dry/wet	CO ₂ /CH ₄	200	V	[25]	Dry/wet	CO ₂ /CH ₄	230	V–C	[24]
San Juan Basin, USA	Wet	CO ₂ /CH ₄ /N ₂	150	V	[26–28]	Wet	CO ₂ /CH ₄ /N ₂	150	V–C	[26,27,29,30]
Argonne Premium, USA	Dry/wet	CO ₂	150	V/G	[17,31,32]	Dry/wet	CO ₂ /CH ₄	180	V–C	[24,32,33]
Black Warrior Basin, USA	–	–	–	–	–	Wet	CO ₂ /CH ₄ /N ₂	100	V–C	[28]

Method: V, volumetric; G, gravimetric; C, chromatographic. Maximum pressure, P_{\max} is given in bar.

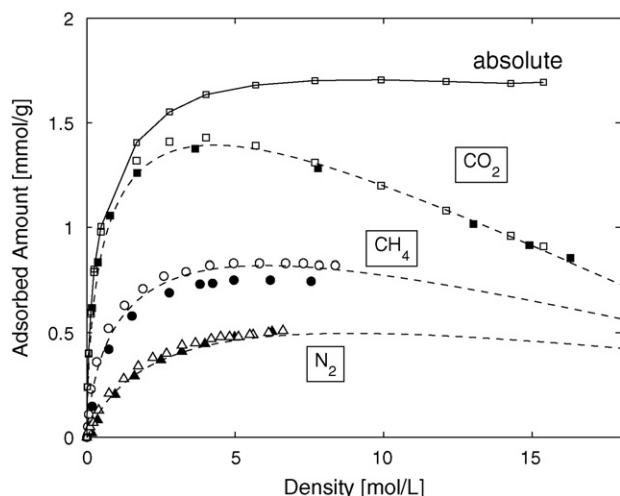


Fig. 2. Excess adsorption isotherm of CO_2 , CH_4 and N_2 on a dry Australian coal (type A) at 55°C as a function of the bulk density ρ^b measured at CSIRO (Newcastle, Australia) (open symbols) [13] and in our lab (closed symbols) using a Rubotherm magnetic suspension balance as described elsewhere [19]. Dashed lines are calculated with a modified Dubinin–Radushkevich equation [13]. The total CO_2 loading capacity (absolute sorption, solid line) is obtained by correcting the excess adsorbed amount with the slope of the linear descending part of the excess adsorption isotherm as explained in detail earlier [19].

studies [12,18,20,21,28,30]. Only in two cases, preferential adsorption of CH_4 over CO_2 on low rank coals has been observed [24,33]. It is worth noting that some early studies hinted at an irregular behavior in the adsorption of CO_2 , particularly close to its critical density [25,41]. It is believed that these were artifacts due to the experimental set-up and procedure, as demonstrated by the fact that when repeating one of the measurements in another setting the irregularities disappeared [41].

In order to assess the CO_2 storage capacity of coal, the absolute adsorption isotherm can be estimated from the excess. This can be done in principle by correcting the adsorption data with the swelling measurements, e.g. by accounting for the buoyancy due to the equilibrium coal volume in the case of the gravimetric method, and by assuming a value for the adsorbed phase density [10,42,43]. Alternatively, one can add to the excess adsorption isotherm a term proportional to the bulk density and to the slope of the linear part of the excess isotherm, which corresponds to the sum of the volume of the adsorbed phase and of the swollen part of the coal volume, as discussed in detail elsewhere [19]. The outcome of this procedure is also shown in Fig. 2, thus indicating a maximum capacity of dry coal of 0.08 g/g.

All adsorption measurements show that wet coal uptakes CO_2 always less than dry coal, because of competitive water adsorption. However, the quantitative effect of moisture on CO_2 uptake is less certain because of the intrinsic difficulty of the measurements. It has been shown for instance that a coal containing about 3% moisture, corresponding to a relative humidity of 50%, adsorbs 30% less CO_2 than the corresponding dry one [14]. Some have measured adsorption on coal samples as received [17,26]; others have prepared wet coal samples by equilibrating them in a sealed chamber with a saturated salt-solution of known water partial pressure [14,25]. In all cases one has to make the assumption that the adsorbed amount of water on coal remains constant as the gas adsorbs, i.e. that the experiment is carried out in such a way to prevent the gas from drying the sample. This is believed to be one of the reasons why an inter-laboratory study using different techniques has shown unsatisfactory reproducibility of CO_2 adsorption data on wet coal, particularly above 80 bar [31,44]. In this context, the

Table 2

Studies reporting swelling measurements on coal

Coal origin	Method	Gas	P_{max}	Reference
Australia	Optical	CO_2	150	[42]
Canada	Strain	$\text{CO}_2/\text{CH}_4/\text{N}_2$	50	[53]
Sulcis Coal Province, Italy	Optical	$\text{CO}_2/\text{CH}_4/\text{N}_2/\text{He}$	140	[18]
South Island, New Zealand	Strain	$\text{CO}_2/\text{CH}_4/\text{N}_2/\text{He}$	40	[36]
Poland	Dilatometric	CO_2	40	[56]
San Juan Basin, USA	Strain	CH_4/He	104	[52]
Kentucky, USA	Dilatometric	CO_2	15	[57]
USA	Dilatometric	$\text{CO}_2/\text{N}_2/\text{He}$	48	[58]

Maximum pressure, P_{max} is given in bar.

impact of several sources of error on the measured high-pressure adsorption isotherms have been recently discussed in detail [45].

A variety of semiempirical isotherm models have been used to describe the experimental adsorption data: for single gas adsorption these include the Langmuir, Toth, Dubinin–Radushkevich (as in Fig. 2) and Dubinin–Astakhov isotherms [10,13,46], whereas for multicomponent gas adsorption their combination with the ideal adsorbed solution (IAS) theory [12,16,27,46] or the extended Langmuir equation [28,29] have been applied. Other methods use an equation of state (EOS), namely a 2D EOS, such as the Eyring and Virial equation of state [27] or the Zhou–Gasem–Robinson two-dimensional equation of state [26]. The relatively simple form of all these equations allow for their direct implementation in reservoir simulators describing ECBM dynamics. An alternative approach uses fluid–fluid and fluid–solid molecular interaction energies and a microscopic description of the pore geometry in the framework of statistical thermodynamics [20,30,47]. When quantitative information about the pore size distribution of the coal are incorporated, insights on the behavior of the adsorbed gas in pores of different sizes, particularly under near- and super-critical conditions, can be obtained [20,48]. Other fundamental approaches, the so-called dual-sorption models, treat the coal as a microporous copolymer containing both elastic polymer-like chains and crystal-like domains and describe the sorption phenomenon as a combination of both penetration of gas molecules into the elastic matrix (absorption) and adsorption of gas into the micropores [38]. The results in the literature show that experimental data can be described well with one or the other of these models, which may have however limited predictive capability when extrapolated.

The research on adsorption isotherms aims on the one hand at providing data to analyze and to predict the behavior of ECBM operations, and on the other to develop understanding and to establish correlations that guide in choosing coal seams for ECBM based on coal properties only, before carrying out lengthy adsorption studies. In this context, there have been useful comparative studies that show important trends, e.g. that CO_2 maximum adsorption capacity is always between 4 and 10 wt.% and depends on coal rank in a non-monotonic way [10,15,19,22,49,50].

3. Swelling and permeability

Uptake and release of many gases and liquids are associated with swelling and shrinking of coal, respectively [51]. Volume changes of unstressed coal samples of a given shape upon exposure to a gas at high pressure for several days can be measured using the dilatometric, optical or strain measurement methods [18,42,52]. A summary of the studies reported in the literature together with the corresponding experimental conditions is given in Table 2. For the sake of illustration and without loss of generality, swelling data measured in our lab on a Sulcis coal are shown in Fig. 3[18]. In the figure, likewise all other studies, the extent of swelling increases monotonically with pressure up to a few percents for adsorbing gases, with

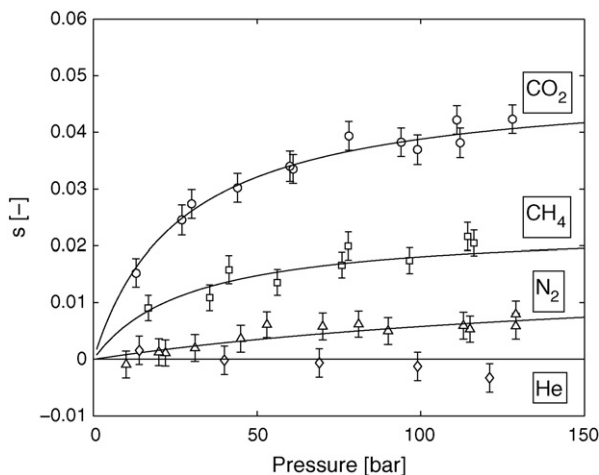


Fig. 3. Swelling of an unconstrained dry disc of the same coal from the Sulcis Coal Province used in previous studies [19,18], as a function of the pressure, P , of CO_2 , CH_4 , N_2 and He at 45°C . Measurements have been carried out in our lab in a high pressure view cell equipped with a sapphire window by monitoring the change in geometrical dimensions of the disc upon pressurization with pure gases. Swelling is assumed to be isotropic. The experimental data (symbols) are well described as a function of pressure by Langmuir-like equations (solid lines), i.e. $s = s^{\max}bP/(1 + bP)$, where s^{\max} and b are temperature dependent, gas and coal specific constants.

CO_2 swells coal more than CH_4 that swells more than N_2 , whereas for helium, a non-adsorbing gas, volume changes are negligible [42,53,36]. Therefore, in view of an ECBM operation, the displacement of CH_4 by CO_2 would lead to a net coal swelling, whereas its displacement by N_2 to a net shrinking. As shown in Fig. 3 swelling isotherms can be effectively described with Langmuir-like equations [53,54]. Coal swelling is reversible, with the sample returning to its original size when the pressure is released [42,53]. In a recent study, repeated CO_2 swelling measurements on coal showed that changes are greater in the direction perpendicular to the bedding plane than in that parallel to it [42]. In another study, the observed differences between the two directions were very limited [54]. The results from the first case suggest that the anisotropic nature of the coal remains unchanged upon repeated exposure to the high-pressure gas, which is different from the behavior observed when organic solvents are used, where after the first exposure the coal behaved isotropically [51,55]. It is clear that further measurements are needed to clarify whether one or the other conclusion can be drawn. However, we believe that the data presented in Fig. 3 are useful and the assumption of isotropic expansion acceptable, being the error associated to the experimental technique similar to the difference due to assuming in the two mentioned works either isotropic or anisotropic behavior.

According to the dual nature of CO_2 uptake, i.e. surface adsorption and absorption into the solid matrix, also swelling can be interpreted in two complementary ways. On the one hand, adsorption induces a change of the coal specific surface energy, which can be compensated by the elastic energy change associated to the volume change [59,60]. On the other hand, as a glassy, strained, cross-linked macromolecular system, coal undergoes structural changes in the presence of high pressure CO_2 that can be explained only by penetration of CO_2 into the coal matrix [51,61]. Through this mechanism, CO_2 uptake may lead to weakening and plasticization phenomena, as well as to changes of coal mechanical properties, e.g. its softening temperature and its Young's elastic modulus, possibly over the long time horizon of CO_2 storage [51,62–64].

Volume changes of coal during ECBM operations are of key importance because they affect coalbed permeability, which in turn controls injection pressure and gas production. In order to clarify

this effect, let us consider the so-called matchsticks model, which describes coal as an ensemble of parallel elongated matrix elements separated by fractures, i.e. cleats, that constitute its transport porosity [65,66]. On the one hand, the external lithostatic pressure tends to press the matrix elements together, and to reduce porosity [53]. On the other hand, gas absorption swells the coal matrix elements, and therefore consumes the space between them, thus reducing porosity as well. Both effects lead to a decrease of permeability, which scales with the cube of the porosity as given by Eq. (1) [53,67]. With reference to Fig. 3, displacement of CH_4 by CO_2 at a coalbed pressure of around 100 bar yields a net coal swelling of about 2%, which for a reference porosity of 5% leads to a fivefold decrease of permeability. Being dependent on the specific stress situation in the coal seam, porosity changes should be calculated by solving a 3D elasticity problem. However, it is common procedure to simplify it by describing the coalbed as an isotropic linear poroelastic medium under uniaxial strain and constant overloading [67,68]. Therefore, porosity, ε , and permeability, k , in the coalbed can then be predicted as [53,69,70]:

$$\frac{k}{k_0} = \left(\frac{\varepsilon}{\varepsilon_0} \right)^3 = \exp[-C_1(P_c - P) - C_2s(P)] \quad (1)$$

where P and P_c are fluid and confining pressure, respectively; s is the pressure dependent swelling (see for instance Fig. 3); C_1 and C_2 are coefficients depending on coal properties. In the above equation, the reference values of porosity and permeability apply to an unstressed coal in contact with a non-swelling gas at atmospheric pressure. It is apparent that the physical description of porosity changes based on the matchsticks model is fully reflected in the functional form of Eq. (1).

The parameters in Eq. (1) have to be determined through experiments. The permeability of a geological structure can be determined in situ through a standard protocol, but experiments with coal are complicated by its interactions with sorbing and swelling gases like CO_2 hence by the possible dynamic variation of permeability in the course of the experiment. Alternatively, 1D dynamic flow experiments can be carried out in the lab using a coal core placed in a hydrostatic or a triaxial cell. In a hydrostatic cell the coal core is surrounded by a rubber sleeve, through which it is exposed to an external confining pressure, P_c , and it is connected through on-off valves to an upstream and a downstream reservoir containing the gas at the desired pressure levels [71–75]. In a triaxial cell additionally vertical and horizontal stresses on the coal core are imposed independently, so as to better reproduce the in situ conditions [64,76]. After saturating the coal sample with a gas at the given pressure and after setting the desired stresses on it, typical experiments in both cells are carried out either by imposing a constant gas pressure difference across the sample and by measuring the corresponding gas flow rates (constant pressure difference method), or by imposing an initial pressure difference between the two reservoirs and by monitoring pressure during equilibration (transient step method) [77].

To estimate the parameters in the permeability Eq. (1), the experimental behavior in terms of pressure or flow rates is compared to the results obtained using a mathematical model of the process [64,75,78,79]. With reference to Fig. 4, the model consists of the following material balances in the fluid and in the adsorbed phase, where we assume a lumped linear mass transfer driving force with controlling resistance in the micropores of the coal matrix (see Section 4 for more details):

$$\frac{\partial(\varepsilon\rho)}{\partial t} + \frac{\partial[(1-\varepsilon)q]}{\partial t} + \frac{\partial(u\rho)}{\partial z} = 0 \quad (2)$$

$$\frac{\partial[(1-\varepsilon)q]}{\partial t} = (1-\varepsilon)k_m(q^* - q) \quad (3)$$

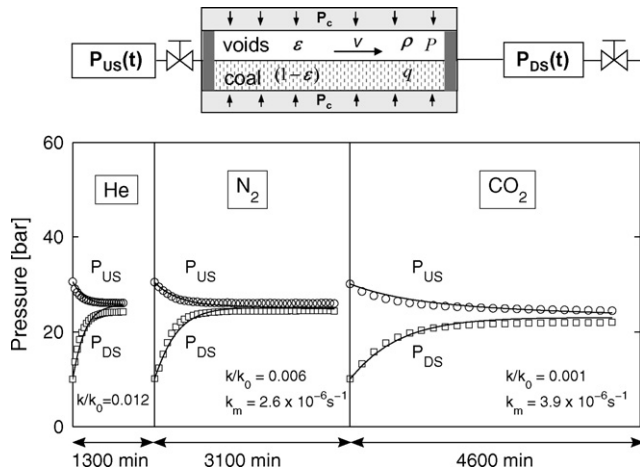


Fig. 4. Scheme of a hydrostatic cell for dry coal core testing, and results of transient step experiments carried out on a Sulcis coal core (diameter 2.54 cm, length 3.60 cm) at 45 °C with He, N₂ and CO₂ under an external confining pressure of 100 bar. Symbols: experimental (○) upstream and (□) downstream reservoir pressures; (—) corresponding simulation results. The estimated values of the mass transfer coefficients k_m and of the relative permeabilities k/k_0 at the end of each transient step are also given, with $k_0 = 0.052$ mD [75].

where ρ and q are the gas density and adsorbed phase concentration, respectively; q^* is its equilibrium concentration in the adsorbed phase; k_m is its mass transfer coefficient; u is the superficial velocity; t and z are time and space coordinates. These equations are completed by the constitutive equations: Darcy's law, expressing velocity as a function of pressure gradient and permeability; the equation of state, e.g. Peng–Robinson EOS, to relate gas density to pressure and temperature; the adsorption and the swelling isotherms, giving q^* and s as a function of pressure and temperature (see Section 2 and the caption of Fig. 3); finally, the Eq. (1) expressing permeability and porosity as a function of pressure and swelling. Upstream and downstream boundary conditions are either constant pressure values in the case of the constant pressure difference method, or simple reservoir material balances in the case of the transient step method.

In Fig. 4 three transient step experiments carried out in our lab are illustrated [75]. When setting the same initial upstream and downstream pressure values, the time to reach pressure equilibration increases going from He, to N₂, to CO₂, i.e. going from the non-swelling gas to the one that swells coal the most. With reference to Eq. (1) and using a value of the reference porosity that has been determined independently through helium pycnometry, namely 5.1%, the reference permeability and the coefficient C_1 are determined by fitting He experiments, whereas C_2 and the mass transfer coefficient k_m for each adsorbing gas, N₂ or CO₂, is estimated from the corresponding experiments. Fig. 4 shows that the model results and the experimental data are in excellent agreement, and that the final permeability with CO₂ is clearly the smallest, as reported also in all other similar studies including those reporting field data [71,80]. Note also that the estimated values of the mass transfer coefficients correspond to time constants of about 3 and 4.5 days for CO₂ and N₂, i.e. of the same order of magnitude of those used in the literature [81,82].

4. Mass transfer and injection/displacement simulations

Laboratory studies of CO₂ injection with concomitant CH₄ displacement can be carried out in the high pressure cells presented above and can be described with models such as the one presented in the previous section when extended to multicomponent gas mix-

tures. The complexity of the pore structure of coalbeds impacts also mass transfer mechanisms and how to describe them in ECBM models. There are up to four types of pores in coal, namely cleats where gas and water are present, macro- and mesopores where there is only free gas, and micropores where adsorption takes place.

Traditionally mass transfer is described through a linear driving force model either by lumping gas diffusion in the different types of pore using a single mass transfer coefficient, or the corresponding time constant, as in Eq. (3)[83,84], or by using a bidisperse pore diffusion model and the corresponding time constants for diffusion in macro-/mesopores and in micropores [78,85]. If matrix swelling and permeability changes are not included in the model, it was shown that an empirical dependence of the rate-limiting micropore diffusivity on the adsorbed phase concentration was required to match the results of a laboratory core flush test [78].

When this relatively simple description of mass transfer is complemented by the description of porosity and permeability changes in the coal during injection and displacement through Eq. (1), one obtains the model presented and used in the previous section, which has proven to be effective both in describing laboratory experiments and in constituting the nucleus of reservoir models used to describe field tests (see Section 5).

Recently, a more detailed model has been proposed, which accounts for the coal pore size distribution and for three mechanisms of coal swelling [64]. Mass transfer includes convective flow in cleats, convective and diffusive flows in meso- and macropores, adsorption and surface diffusion in micropores, whereby diffusion is described using the Maxwell–Stefan equations. A more compact version of such a model has been shown to describe with good agreement a number of displacement experiments carried out on coal cores in different labs [79,86].

A completely different approach is based on neglecting dispersion phenomena, mass transfer resistance, i.e. assuming local equilibrium between fluid phases (gas and water) and coal matrix, and swelling effects [87–89]. In this case a powerful mathematical technique, i.e. the method of characteristics, can be used to calculate the multiphase multicomponent flow in a coalbed in a way that sheds light on the complex injection/displacement dynamics. Such model brings important conceptual insight even though it represents a strong simplification of the real coal seam. In agreement with field observations and more detailed simulations, it has been shown that CO₂ injection allows for a complete displacement of CH₄ through a sharp injection front, the so-called “shock front”, and its recovery as a pure gas thanks to the stronger CO₂ adsorptivity. On the contrary, injection of a N₂ rich gas mixture displaces CH₄ at a higher rate, through a broad injection front, the so-called “simple wave”; as a drawback, the produced gas is a mixture of N₂ and CH₄.

5. Field tests and ECBM modeling

Towards demonstration of its feasibility and as a first step in the direction of its commercial deployment, the ECBM technology has been implemented in a number of field tests, which are reported in Table 3. Key performance parameters of an ECBM operation used to assess the outcome of a field test are the amount of CO₂ stored and its rate of injection, as well as the amount and purity of CH₄ produced.

The first ECBM project, the Coal-Seq project, was the one at the largest scale and the most meaningful; it took place at the San Juan Basin in New Mexico (USA), where pure CO₂ and pure N₂ were injected in the Allison and Tiffany Unit, respectively, while CH₄ was successfully produced in a multi-well configuration over a period of more than 5 years [4]. It was shown that gas injection indeed enhanced methane recovery. CO₂ injection yielded a reduction in permeability and injectivity, whereas N₂ injection lead to a much

Table 3
ECBM field tests

Location	Project	Year	Gas	# of wells	Injection	Reference
Fenn-Big Valley, Canada	Alberta CO ₂ /ECBM	1999	CO ₂	sw	0.19 kt	[5]
			13% CO ₂ /87% N ₂	sw	0.11 kt	
			53% CO ₂ /47% N ₂	sw	0.12 kt	
South Qinshui Basin, China	–	2004	N ₂	sw	≈ 0.1 kt	[7]
Ishikari Coal Field, Japan	JCOP	2004	CO ₂	sw	0.19 kt	
Upper Silesian Basin, Poland	RECOPOL	2004	CO ₂	2w	0.15 kt	[8]
			CO ₂	2w	0.76 kt	
San Juan Basin, USA	Coal-Seq	1995	N ₂	mw	–	[4]
			CO ₂	mw	370 kt	

Well configuration: sw, single-well; 2w, two-well; mw, multi-well.

more rapid breakthrough thus reducing product purity. The former effect was attributed to the porosity reduction associated with coal swelling upon CO₂ injection, particularly evident near the well, where the CO₂ pressure is high. In the case of N₂ coal undergoes a net shrinking, as gathered from the data shown in Fig. 3, but the N₂ injection front and the CH₄ desorption front overlap so as the injected gas pollutes methane much more than in the case of CO₂. These observations are in perfect agreement with the theoretical considerations made in the previous sections.

The other field tests in Table 3 were on a much smaller scale, exploiting a single well [5,7] or a two-well configuration [6,8]. The goal of these projects was that of testing the ECBM technology in reservoirs with different geological characteristics. In all cases CH₄ production was enhanced in response to gas injection; CO₂ injection lead to a reduction of injectivity that could be compensated through shut-in periods in the Alberta CO₂ ECBM project [5], or through a frac job in the RECOPOL project, at least partially and temporarily [6]. On the contrary, during the injection of flue gas in the Canadian project a steady increase of well injectivity was observed [5].

The knowledge of the CH₄ amount within the coalbed before starting its primary and enhanced recovery, i.e. the so-called Gas In

Place (GIP), is important for several reasons. A reliable estimation of it can be achieved through the so-called pressure coring method, which allows obtaining a coal sample at reservoir pressure [90]. The GIP can then be evaluated by letting the coal sample desorb CH₄ and by measuring its desorption rate and the overall amount released. First, the GIP is used for the design of the ECBM operation because, when compared to the CH₄ adsorption isotherm, it defines the reservoir pressure level where gas starts to be released during primary recovery [9]. Secondly, it provides an indication of the amount of CH₄ that has left the coalbed, and therefore an indirect information about the sealing efficiency of the caprock. Coalbeds with a CH₄ content close to the maximum theoretical value given by the adsorption isotherm are therefore promising for a future ECBM project. Finally, the GIP is used as input data for reservoir models to predict the evolution of the ECBM operation, particularly gas injection, CO₂ storage and methane production.

Reservoir models for ECBM recovery simulations incorporate an extension of the 1D single component description of a dry coal core presented in Sections 3 and 4 to a 3D multi-component multi-phase (coal, gas, and water) model. Such model has to be solved in a 3D domain that comprises the coalbed and accounts for its geological structure and possibly heterogeneous physical features

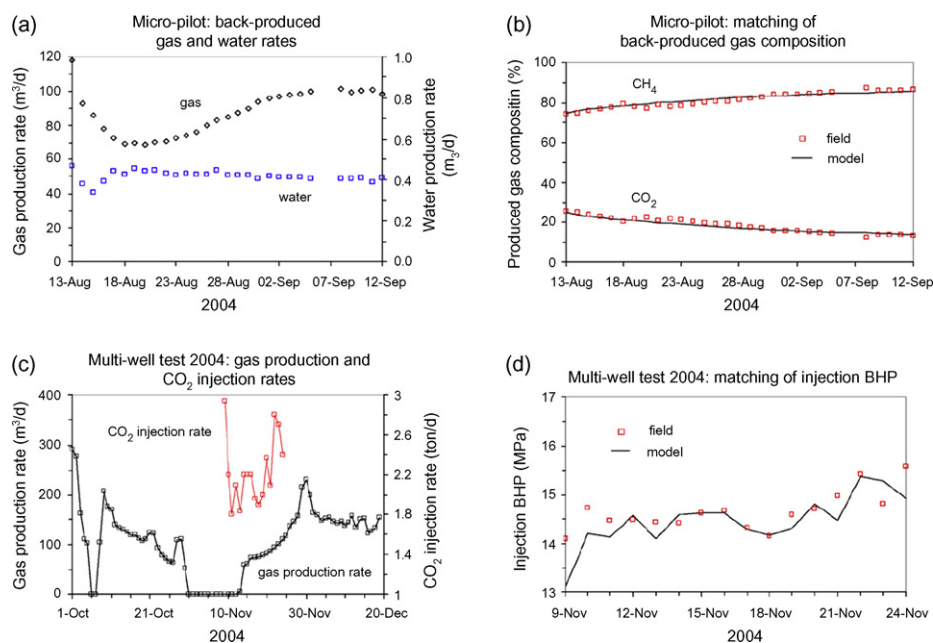


Fig. 5. Results from the field test at the Ishikari coalfield CO₂ storage pilot project in Japan [8]. Single well test: (a) field-back produced gas and water rates; (b) field gas composition and model match; multi-well test: (c) CO₂ injection and gas production rates; (d) field injection well bottomhole pressure and model match (with permission from Shi et al. [81]).

as well as for the configuration of the injection and production wells.

One of such reservoir simulators, namely PSU-COALCOMP developed at The Pennsylvania State University and based on a 2D description of the coalbed that assumes vertical homogeneity, has been used to study the effect of the well configuration and design on the stored amount of CO₂ as compared to its theoretical amount, that is given in principle by the sorption isotherm [83,84]. It was shown that the stored amount of CO₂ predicted by the model could be as small as 50–70% of the theoretical one. Depending on the parameters of the adsorption isotherm, the mass transfer kinetics and the well configuration, at the end of the ECBM project simulations indicate that there might be a significant fraction of the swept region, which is still far from equilibrium, where CO₂ storage is far below its thermodynamic limit. Useful design criteria can be derived from this type of study.

The model parameters used by the reservoir simulators can be tuned to history match field test data and then used for design or optimization purposes. This very important approach has been applied to the data of the South Qinshui Basin single well micro-pilot test, where after successful history matching a larger scale multi-well field test has been designed and planned using the simulator [7]. Similarly, a reservoir simulator has been used in support of the operations of the Coal-Seq project [4].

Durucan and Shi have consistently and successfully used the Imperial College in-house ECBM simulator METSIM2 to history match field data from the Coal-Seq project (Allison unit) [91], from the Alberta project (flue gas injection) [92], and from the JCOP project for both single and multi-well tests [81]. Some of the results referring to the JCOP project are illustrated in Fig. 5 in terms of time resolved gas and water production rate and of produced gas composition for the single well test, and in terms of injection rates and field injection well bottomhole pressure for a multi-well test. The METSIM2 simulator has also been used to analyze the effect of variabilities in the reservoir data and of uncertainties in the data and in the model on the quality of the results of ECBM simulations [82], and to analyze and optimize ECBM performance [80]. In the latter work, different ECBM schemes, comparing pure CO₂ injection with mixed N₂/CO₂ injection were investigated for a three spot pattern of horizontal wells using the Alberta Fenn-Big Valley reservoir data. For a 5 years time period, mixtures rich in N₂ improved significantly CH₄ production compared to primary recovery, whereas pure CO₂ injection lead to no enhancement of CH₄ production. This effect was attributed to the higher permeability following N₂ injection in the coalbed compared to CO₂. For the same reason, injection of a 25% N₂/75% CO₂ mixture led to a larger amount of CO₂ stored compared to that stored if pure CO₂ were injected. Due to the early breakthrough of N₂ at the production well, however, there is a trade-off between the enhanced CH₄ recovery and the purity of the produced gas.

6. Concluding remarks

Field tests supported by fundamental studies and reservoir simulations have shown that storage of CO₂ into deep, unmineable coal seams with simultaneous recovery of CH₄ is technically possible and environmentally attractive. There are therefore excellent research opportunities at the interface between earth sciences and chemical engineering, particularly supercritical fluid science and technology, to fill the gaps in knowledge and to improve ECBM understanding. Some issues that require further studies are the long term stability and fate of CO₂ stored, the effect of impurities such as SO_x, NO_x and O₂ on the process, the heterogeneity of the coal seam that makes the extrapolation of the lab results to the field challenging, and the effect of water on the storage capacity

and displacement dynamics. Besides, in developing a new field test innovative methods have to be applied to characterize the 3D coal formation and to monitor properly the ECBM operation.

An important aspect that is subject of intensive investigations is the design of gas injection and coalbed methane displacement so as to optimize the economics and the effectiveness of the ECBM project as a whole, in terms of injectivity, amount of CO₂ stored and amount of CH₄ produced. Based on the observations summarized in Sections 4 and 5, it would be extremely attractive to inject flue gas directly without the expensive CO₂ capture step and as a way to keep permeability sufficiently high. Answering whether this is indeed possible, in spite of the increased gas compression costs due to the additional nitrogen, requires more research work and field tests under different conditions and in different geological settings. Efforts in this direction are justified by the ultimate goal of finding ways for reducing greenhouse gases emissions.

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