

Shale Gas Formations and Their Potential for Carbon Storage: Opportunities and Outlook

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Abstract Shale gas resources are proving to be globally abundant and the development of these resources can support the geologic storage of CO₂ (carbon dioxide) to mitigate the climate impacts of global carbon emissions from power and industrial sectors. This paper reviews global shale gas resources and considers both the opportunities and challenges for their development. It then provides a review of the literature on opportunities to store CO₂ in shale, thus possibly helping to mitigate the impact of CO₂ emissions from the power and industrial sectors. The studies reviewed indicate that the opportunity for geologic storage of CO₂ in shales is significant, but knowledge of the characteristics of the different types of shale gas found globally is required. The potential for CO₂ sorption as part of geologic storage in depleted shale gas reservoirs must be assessed with respect to the individual geology of each formation. Likewise, the introduction of CO₂ into shale for enhanced gas recovery (EGR) operations may significantly improve both reservoir performance and economics. Based on this review, we conclude that there is a very good opportunity globally regarding the future of geologic storage of CO₂ in depleted shale gas formations and as part of EGR operations.

Keywords Shale gas · Shale formations · EGR · CO₂ storage in geological formations · Natural gas production

1 Introduction

Global and national energy outlooks to 2030 and beyond indicate growing global energy demand, particularly in non-OECD countries, and a continued dominant role for fossil fuels in the world's energy mix, even as utilization of renewable energy sources grows faster than utilization of fossil fuels (British Petroleum 2013; Exxon Mobil 2013; Shell 2013). The future energy landscape will see natural gas play an increasing role for energy supply, particularly in light of the substantial quantities of unconventional gas reserves now being exploited in the

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United States and plans to exploit similar unconventional resources in many other countries. Even though natural gas is a cleaner burning fossil energy source than coal or oil (Carbon emissions from electricity generation are approximately 400 gCO₂/kWh for natural gas, 785 to 1,005 gCO₂/kWh for coal, depending on type, and 635 to 715 gCO₂/kWh for oil products, depending on type) (International Energy Agency 2013), it still produces greenhouse gas (GHG) emissions and will only complement rather than fully replace coal and oil for energy in the stationary and transportation sectors. Therefore, addressing the joint challenge of growing energy demand and the potential of climate change consequences from the resulting GHG emissions requires coordinated action at the global scale. Immediate and effective actions are needed to maintain the atmospheric concentration of GHGs to 450 ppm or less, which is the level consistent with maintaining global temperature change below 2 °C relative to pre-industrial levels and the associated social, economic and environmental consequences (IPCC 2014). The International Energy Agency (IEA) estimates that key actions include large-scale investments in carbon capture and storage (CCS) to mitigate the impact of CO₂ (carbon dioxide) emissions from the power and industrial sectors. In fact, IEA explicitly states that unless CCS technology is widely deployed by 2050, approximately two-thirds of proven fossil fuel reserves need to remain un-extracted if the world is to achieve the 2 °C goal (International Energy Agency 2012).

Carbon sequestration options include storage in depleted oil and gas reservoirs, saline formations, un-mineable coal seams, basalts, and shales. Perhaps the most commonly considered target repository for CO₂ is deep saline aquifers. However, documented concerns for storage in saline formations include cost, long-term storage, impact on seismicity, and the logistics of sequestration (Tao and Clarens 2013). Enhanced recovery of tight oil, coal bed methane and other unconventional fossil fuels, known as enhanced gas recovery (EGR) or enhanced oil recovery (EOR), using CO₂ is receiving interest as an opportunity for carbon sequestration because of the economic benefits it affords and the existing infrastructure that can be leveraged (Tao and Clarens 2013).

The potential for the geologic storage of CO₂ in shale formations that have undergone hydraulic fracturing for extraction is being explored for several reasons (Rodosta et al. 2013) such as: shales are broadly distributed; availability of infrastructure of wells, pipeline and so on; decrease in pore pressure caused by gas production in the shale formation before CO₂ injection.

Development of shale resources may benefit CO₂ storage because the innovations developed are directly transferable, particularly those that relate to well completion, like new methods for well cementing, advanced horizontal drilling techniques, and enhancement of field treatment skills for saline water (Nicot and Duncan 2012). Thus, understanding the behavior of CO₂ in shale is an important part of advancing the opportunity for the economic geologic storage of CO₂, particularly because of the fact that the geological characteristics of a particular storage site often have important influences on the design of related CO₂ capture and transportation infrastructure (International Energy Agency 2013).

This paper examines the global unconventional hydrocarbon landscape with a focus on the assessment of recoverable shale gas resources. The opportunity for geologic storage of CO₂ in shale formations (as well as other natural gas reservoirs that include shales) is then considered through a review of the types of shales globally, and the CO₂ storage capacity and EGR opportunities in shale formations.

2 Global Shale Gas Resources

According to the World Energy Outlook (International Energy Agency 2012), the remaining technically recoverable natural gas resources by the end of 2011 globally reached 790 Tcm

Table 1 Technically recoverable natural gas resources by end 2011 (International Energy Agency 2012)

Region	Conventional (Tcm)	Unconventional (Tcm)				Total (Tcm)
		Tight gas	Shale gas	Coalbed methane	Sub-total	
E. Europe/Eurasia	144	11	12	20	44	187
Middle East	125	9	4	–	12	137
Asia-Pacific	43	21	57	19	94	137
OECD America	47	11	47	9	67	114
Africa	49	10	30	0	40	88
Latin America	32	15	33	–	48	80
OECD Europe	24	4	16	2	22	46
World	462	81	200	47	328	790

(trillion cubic meters). This includes both conventional and unconventional resources and is slightly greater than the estimate of 719 Tcm provided by McGlade et al. (2013) in their recent review of regional and global unconventional gas estimates. A breakdown of the amount of each type of resource globally (International Energy Agency 2012) is shown in Table 1.

The U.S. Energy Information Administration (EIA) estimates that roughly 32 % of total estimated global natural gas resources are in shale formations (Kuuskraa et al. 2013). The top 10 countries for technically recoverable shale gas resources for which EIA has made a shale resource assessment are shown in Table 2 (Kuuskraa et al. 2013).

Although the data shown in Tables 1 and 2 suggest a significant CO₂ storage opportunity via exploitable shale resources, the data represent technically recoverable resources which differ from economically recoverable resources. According to the EIA's definitions (Kuuskraa et al. 2013): technically recoverable natural gas is that which can be produced with current technologies, regardless of natural gas prices and production costs; economically recoverable resources are resources that can be produced profitably under current market conditions. The costs of well drilling and completion, the amount of natural gas produced from an average well over its lifetime, and the prices received for gas production define the economic recoverability of gas resources.

As specifically stated by the EIA, the economic recoverability is influenced by factors such as ownership of mineral rights that provide economic incentive for development, availability of

Table 2 Top 10 countries with technically recoverable shale gas resources (Kuuskraa et al. 2013)

	Rank	Country	Shale gas (Tcm)
	1	China	31
	2	Argentina	22
	3	Algeria	20
	4	United States	18.60 (EIA), 32.50 (ARI)
	5	Canada	16
	6	Mexico	15.20
	7	Australia	12.25
	8	South Africa	11
	9	Russia	8
	10	Brazil	6.80

Estimates for the United States include those from the Energy Information Administration (EIA) and Advanced Resources International (ARI)

experienced independent operators and contractors, existing infrastructure, and the availability of water resources for use in hydraulic fracturing (Kuuskraa et al. 2013). It should also be noted that estimates of both technically and economically recoverable shale gas resources are uncertain and continually undergoing revision based on updated information (Tables 3 and 4).

Given the substantial variation across the world's shale formations in both geological and non-geological factors, the extent to which technically recoverable shale resources will prove to be economically recoverable is very uncertain. China, for instance, holds the most technically recoverable reserves of shale gas globally (Table 2) and was once considered well positioned to replicate the United States' shale gas success. However, problems comprising poor infrastructure, political disputes, and the fact that the shale gas formations are much deeper than expected, have made economic shale gas recovery extremely challenging in China (Lux Research 2013). Although not assessed in EIA's coverage, large amounts of potentially productive shales exist in other countries around the world, including most of the countries in the Middle East and North Africa (MENA) (Ibrahim 2013; Martin 2012). According to Baker Hughes, Saudi Arabia (Baxter 2013) has more than 16.8 Tcm of shale gas reserves. However, several of the obstacles already noted make MENA countries hamper the development of tight and shale gas. The obstacles stated by Martin (2012) include: high cost of MENA field-development operations (i.e., the cost of drilling and completing in the MENA countries is said to be 2.5 to 5 times higher than similar operations in the United States); lack of fiscal incentives; lack of infrastructure; competition from alternative fossil sources; discomfort with wellbore-scale experimentations; lack of wellbore-specific information; and difficulty in doing business generally. The conclusion, therefore, is that shale gas resources are abundant but many factors must converge in order to make shale gas extraction profitable. These factors include technology innovation, government policy, private sector entrepreneurship, land and mineral rights ownership, market structure, geology, water availability, and natural gas pipeline infrastructure, to name a few. Hence, the policy and market environment are key factors in determining the potential for profitable shale gas production in a particular country. Therefore, the opportunity for geological storage of CO₂ in shale resources is dependent on success in overcoming a large number of development challenges that would make the shale resources

Table 3 Shale Gas Formations in the United States and Canada with their Characteristics (Brathwaite 2009; Chalmers et al. 2012; Fowler et al. 2003; Montgomery et al. 2005; Wust et al. 2013)

Formation	Period	Location	TOC wt%	Vitrinite reflectance	Color
Antrim Shale	Late Devonian	Michigan Basin, US	0.5–24	0.4–0.6	Black
Barnett Shale	Mississippian	Fort Worth and Permian Basin, US	3.2	2.25	Dark Gray
Marcellus Shale	Devonian	New York, Ohio, Pennsylvania, West Virginia, US	3.8	1.56	Black
Haynesville Shale	Late Jurassic	Louisiana, East Texas, US	4.2	2.37	Black
Woodford Shale	Devonian-Mississippian	Oklahoma, Texas, US	2	1.51	Gray
Duvernay Shale	Devonian	Alberta, Canada	4–11	0.4–1.41	Dark Brown

Characterization of shales is provided by Total Organic Carbon (TOC) and Vitrinite Reflectance (VR). VR indicates maturity of the organic matter and the onset of oil generation in hydrocarbon source rocks is generally correlated with a VR of 0.5–0.6 % and the termination of oil generation with VR of 0.85–1.1 %. The onset of gas generation is typically associated with VR values of 1.0–1.3 % and terminates around 3.0 %

Table 4 CH₄ sorption amount on shale

Author	Year	Gas	Sample	Method	CH ₄ sorption capacity
Ross and Marc Bustin (2007)	2007	CH ₄	Shale	V	0.002 mmol/g
Ross and Marc Bustin (2009)	2008	CH ₄	Shale	V	0.4 to 4 cc/g at $T=30\text{ }^{\circ}\text{C}$
Weniger et al. (2010)	2010	CH ₄	Shale	M	$= 0.009 \text{ TOC}(\%) + 0.026 \text{ mmol/g}$
Chareonsuppanimit et al. (2012)	2011	CH ₄	Shale	V	$= 0.035 \text{ mmol/g at } T=55\text{ }^{\circ}\text{C}$
Gasparik et al. (2012)	2012	CH ₄	Shale	M	0.05 to 0.3 mmol/g at $T=65\text{ }^{\circ}\text{C}$
Gasparik et al. (2013a)	2013	CH ₄	Shale	M	For carboniferous sample, 0.03 to 0.09 at $T=65\text{ }^{\circ}\text{C}$ and different TOC
Rexer et al. (2013)	2013	CH ₄	Shale	M	0.042–0.176 mmol/g at $T=27$ to $T=200\text{ }^{\circ}\text{C}$
Khosrokhavar et al. (2014b)	2014	CH ₄	Shale	M	0.02–0.04 mmol/g at $T=30.40$ and $63\text{ }^{\circ}\text{C}$

The effects are shown with three abbreviations: *TOC* Total Organic Content, *V* Volumetric and *M* Manometric

available for storage. If these challenges can be overcome, then not only will abundant hydrocarbon resources become available, but potentially very good repositories for geologic storage of CO₂ will also result.

3 Current Status of Shale Gas Development

World production of unconventional gas, of which shale gas is a subset, was approximately 472 Bcm in 2010, with approximately 76 % of production coming from the United States and 13 % of production coming from Canada (Peduzzi and Harding Rohr Reis 2013). The opportunity to produce substantial amounts of gas from shale formations in both the United States and Canada is based on the development and application of new technologies for economic shale gas extraction (Soeder 2012; Wang et al. 2014). Unlike China and the MENA region previously discussed, the United States and Canada currently have regulatory and political climates generally conducive for shale gas development. Both countries are predicted to drill thousands of wells in the next decade as shale gas development becomes increasingly economical (Speight 2013).

During recent years, because of development of shale gas plays, the production of natural gas has been significantly increased in the United States (Fig. 1). The U.S. is now the largest producer of natural gas in the world. The most growth in economic extraction of shale gas has been made by Canada and the United States which are producing 25 % of global natural gas (British Petroleum 2012). Indeed, shale gas is expected to account for 49 % of total gas production in the United States by 2035, up from 23 % in 2010 (Fig. 1). Advances in shale gas extraction technologies have increased shale gas production in the United States and this production is believed to be able to support the United States' energy demand for about 60 years at the current level of use (NPC 2011). The main shale resources in the United States are provided in Fig. 2 (GAO 2012; Kuuskraa et al. 2013). Among these formations, the Marcellus Shale in the Appalachian Basin is the largest one (Kuuskraa et al. 2013).

In Canada, a large potential value for gas-in-place is estimated in shale formations (~28Tcm) (NEB 2009). Gas production from Canadian shales began modestly in 2005 (Fig. 3) and has increased rapidly afterwards. Natural gas is mainly being produced from Devonian shales in the Horn River Basin and from the Triassic Montney shales and siltstones, both located in northeastern British Columbia and in the Devonian Duvernay Formation in Alberta (Rivard

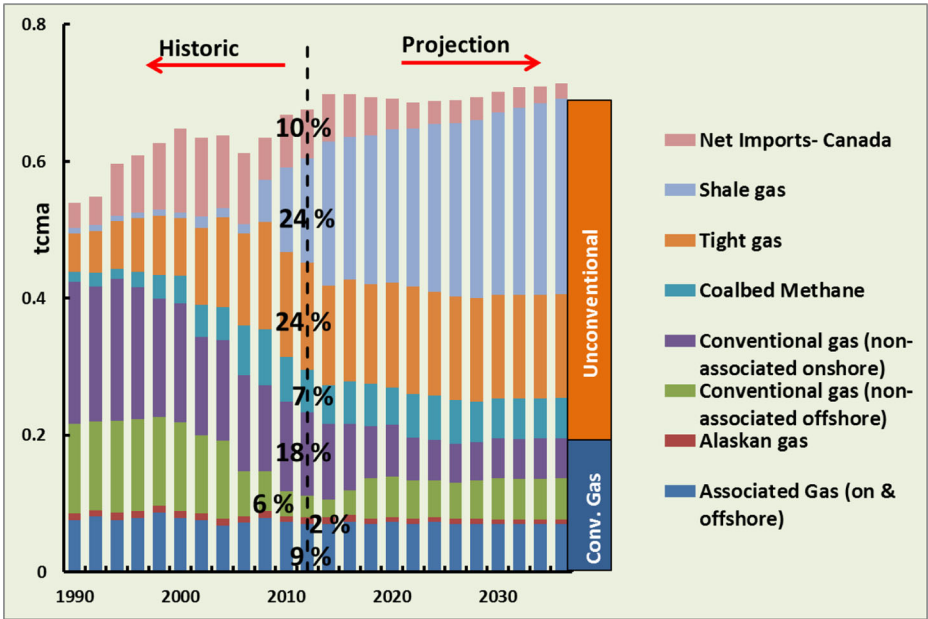


Fig. 1 US historic gas production record (in Trillion cubic meter per year – Tcma) and forecast until 2035 (Weijermars 2014)

et al. 2013). The important gas-bearing reserves in Canada are shown in Fig. 4. Gas production from the Utica Shale in Quebec, Canada was placed under a moratorium until the possible risks for groundwater contamination could be evaluated (Lavoie et al. 2013).

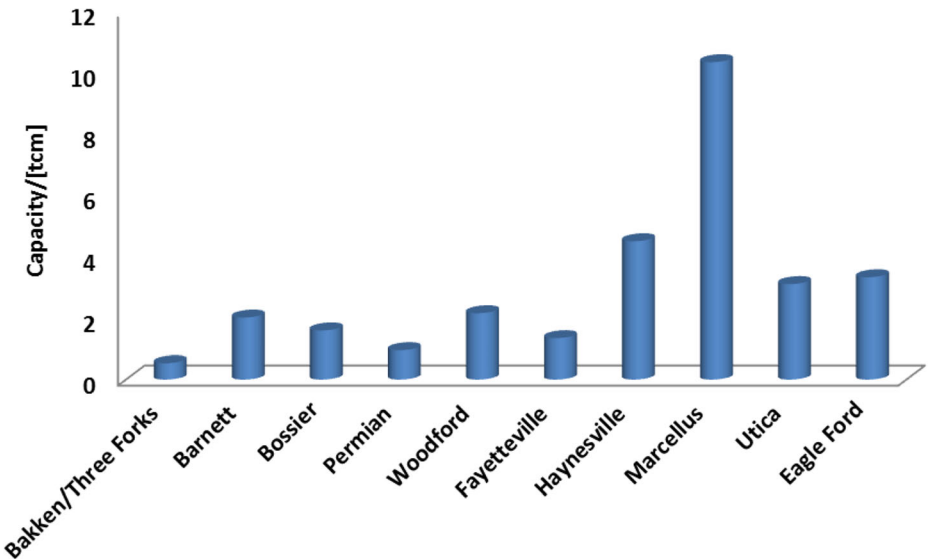


Fig. 2 Remaining shale gas reserves and undeveloped resources in the United States (Kuuskraa et al. 2013)

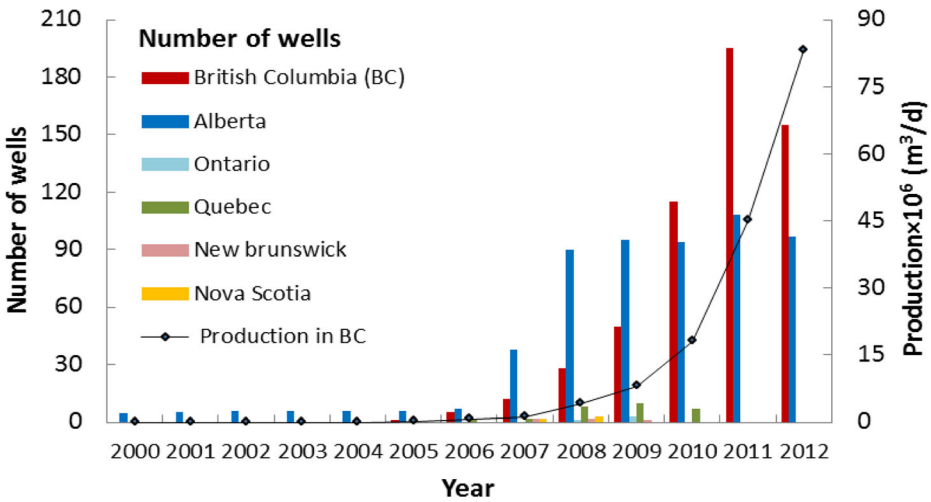


Fig. 3 Total number of wells drilled yearly for unconventional hydrocarbons per year in Canada and yearly production of shale gas for British Columbia (Rivard et al. 2013)

China is at the early stages of evaluating its shale gas resources (Wang et al. 2014). China is planning to increase its shale gas output from nearly zero in 2012 to 6.5 Bcm per year by 2015 and to 80 to 100 Bcm per year by 2020 (Wang et al. 2014). In China, the Sichuan Basin, located in south central China, accounts for 40 % of the country’s shale resource (Speight 2013).

Argentina’s Neuquén Basin, on Argentina’s border with Chile, is the country’s largest source of hydrocarbons, accounting for 35 % of the country’s oil reserves and 47 % of the gas reserves. Argentina’s biggest energy company, YPF (Yacimientos Petrolíferos Fiscales), was successful in discovering shale gas in the Mendoza province (Speight 2013).

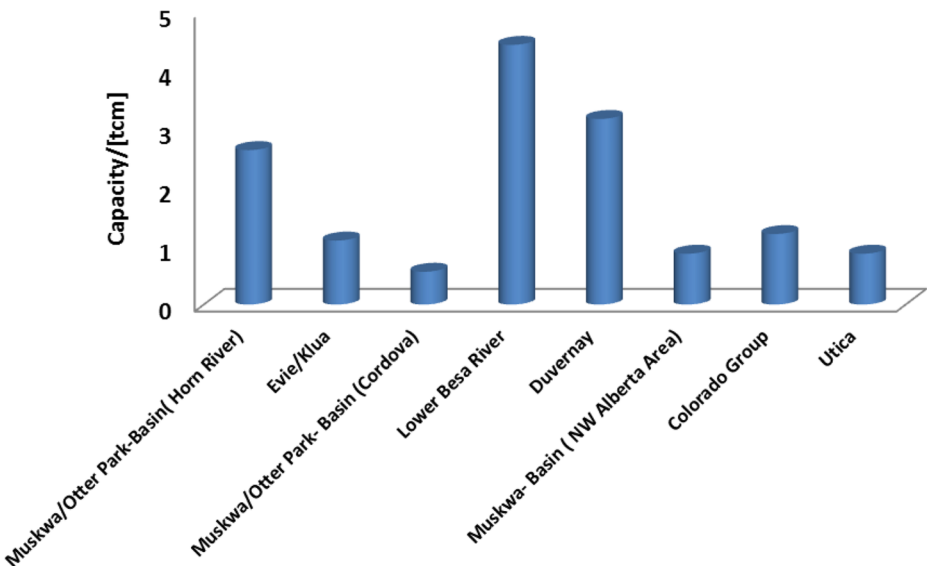


Fig. 4 Technically recoverable shale gas reserves in Canada (Kuuskraa et al. 2013)

Australia's natural gas resources are shown in Fig. 5. They are dominated by unconventional gas, particularly shale gas and coal seam gas. This indicates the significance of shale gas resources in long-term gas supply for Australia (Leather et al. 2013).

Eastern Europe is estimated to have ~12 Tcm of shale gas resources. This potential provides a great opportunity for Europe to decrease its dependency on natural gas imports. There have been estimates that Poland, for example, would not need to import natural gas for 300 years at the country's current level of use if shale gas reserves were exploited to meet demand (Speight 2013). Recently, intense shale gas exploration has occurred in Poland (Karcz et al. 2013) with over 42 wells drilled. Kiersnowski and Dyrka (2013) present a brief history and a comparison with other reports of shale gas resources in Poland. According to McGlade et al. (2013), Poland and France are expected to have the largest shale gas resources among European countries. The development of shale gas has been largely unsuccessful so far in European countries (Geny 2010) for several reasons (Soeder et al. 2014), including uncertainty in estimating technically recoverable shale gas resources, a lack of field data, perceived environmental risks, lack of infrastructure, operational costs, water scarcity, and land access, similar to issues in the MENA region discussed earlier. The status of shale gas development as of 2013 is shown in Table 5.

4 Types of Shale Gas

Unconventional hydrocarbons in the context of this paper are those hydrocarbons obtained from reservoirs difficult to produce due to physically limiting factors such as low permeability. We focus our attention on shale and tight hydrocarbon formations, with particular consideration given to natural gas. However, it is important to acknowledge that shale oil and liquids play an equally important role in the broader realm of unconventional hydrocarbons. Although

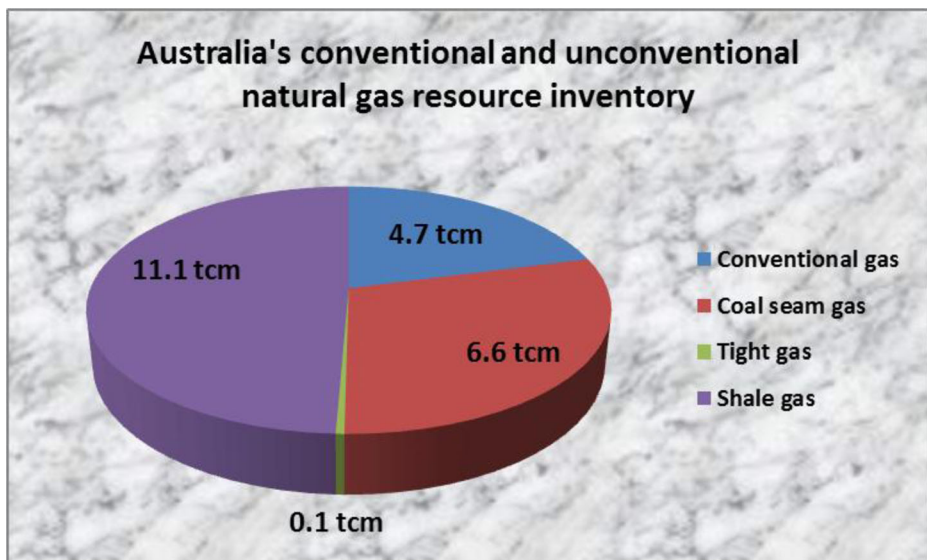


Fig. 5 Australia's natural gas resources (Leather et al. 2013). The differences depend on the methodology (hypotheses) used for the estimation

Table 5 Shale gas development status in European countries (as of February 2013) in allowing or banning shale gas development (Economist 2013)

Allowed	Banned/moratorium	Allowed and permits issued	
Norway	Netherlands	Britain	Ukraine
Latvia	Luxemburg	Spain	Romania
Belarus	France	Portugal	Slovenia
Moldova	Czech Rep.	Denmark	Hungary
Belgium	Bulgaria	Sweden	Serbia
Croatia		Germany	Greece
Bosnia		Poland	Turkey
		Estonia	Austria
		Lithuania	Slovakia

the terms “shale” and “tight” are often used interchangeably, we adopt the U.S. Energy Information Administration (EIA) definitions (Kuuskraa et al. 2013) that specifically states that shale formations are a subset of all low permeability tight formations, which includes not just shales but also sandstones and carbonates. Tight gas reservoirs are geologically similar to conventional reservoirs except that they have much lower permeability of less than 0.1 millidarcy. Shale gas reservoirs have permeability three orders of magnitude (or more) lower than tight gas reservoirs (Mokhatab et al. 2006) and also differ geologically since shale is a geological rock formation containing clay minerals and organic particles where the shale source rock is also the reservoir. Unconventional gas typically cannot be produced economically unless the well from which it is extracted is stimulated by a hydraulic fracture treatment, a horizontal wellbore, or some other technique to expose more of the reservoir to the wellbore (McGlade et al. 2013; Wang et al. 2014).

Shales are divided into dark and light forms based on organic content (Hosterman and Whitlow 1981). The dark form is organic rich whereas the light form is organic lean. A good illustration is in the United States where the Barnett shale, Marcellus shale, Haynesville shale, Fayetteville shale are dark (gray dark) shale (Blatt et al. 1996). Shale formation defining characteristics are based on age and physical characteristics, including era, period, depth and thickness. A shale gas reservoir is a fine grained shale with organic content (Bustin et al. 2008). Criteria that are important in characterizing shale gas hydrocarbon potential include: permeability, Total Organic Carbon (TOC) content, thermal maturity of the organic matter and the type of hydrocarbons available in the reservoir (Brathwaite 2009; Chalmers et al. 2012; Fowler et al. 2003; Montgomery et al. 2005; Wust et al. 2013). Characteristics of Shale gas formations in the United States and Canada are summarized in Table 3. This suggests that each shale reservoir is unique with regard to the optimal approach to well drilling, completion and production. Shale gas is a dry gas in many formations, albeit some reservoirs produce wet gas. To clarify the terms dry and wet in quantitative measures, dry natural gas is primarily a CH₄ vapor with less than 0.014 m³ of gasoline vapor per 28.32 m³. Wet natural gas contains natural gas liquids, such as ethane and butane, in the gas at concentrations of more than 0.014 m³ per 28.32 m³ (Speight 2013). The gas from shale is generated in two ways: thermogenic (generated from cracking of organic matter or the secondary cracking of oil), and biogenic (generated from microbes such as in the Antrim shale gas field in Michigan) (Martini et al. 2003). Mature organic matter typically generates the most gas in the formation. Gas in the formation is in sorbed and free phases. Thermal maturity, the thickness of organic shale and TOC are effective parameters for the economic viability of a shale gas play. The required thickness to economically develop a shale gas target may decrease by improving and

developing drilling and completion techniques (Speight 2013). For a more detailed discussion of shale gas geology, see Bruner and Smosna (2011).

5 CH₄ Capacity and CO₂ Storage in Shales

Currently, natural gas is the third largest energy source in the world and its consumption is predicted to raise substantially in coming years (Leather et al. 2013). From an environmental perspective, CH₄ is a hydrocarbon with a carbon footprint that is generally smaller than that of other hydrocarbons used in energy production, although various studies suggest that shale gas production and utilization may result in significant GHG emissions, depending on the total fugitive CH₄ emissions during extraction and utilization (Peduzzi and Harding Rohr Reis 2013). Additionally, extraction of shale gas may have negative impacts on water resources, public health, biodiversity, food supply and soil, if not adequate technical precautions are taken and there is lack of regulatory oversight.

The sorption capacity of both CO₂ and CH₄ on shales is important to understanding the storage capacity of natural gas in different type of shales, as well as the extent to which CO₂ can displace CH₄ in EGR. Shale gas is a combination of sorbed and free natural gas, which is mainly composed of methane (CH₄) and to a much lesser extent ethane, propane and other gases (Soeder 1988; Wang et al. 2013). The sorption capacity of shales is affected by its TOC content, clay minerals and micropore structure. There is a large body of literature investigating the sorption capacity (Amann-Hildenbrand et al. 2013; Ambrose et al. 2012; Bowker 2007; Chalmers and Bustin 2007; Curtis 2002; Diaz-Campos et al. 2010; Gasparik et al. 2012, 2013a, b; Gensterblum et al. 2014; Harris et al. 1970; Hartwig and Schulz 2010; Jarvie et al. 2007; Ji et al. 2012; Khosrokhavar et al. 2014b; Loucks et al. 2012; Lu et al. 1995; Rexer et al. 2014; Ross and Marc Bustin 2009; Shabro et al. 2011; Slatt and O'Brien 2011; Zhang et al. 2012). Beaton et al. (2010) measured CH₄ sorption isotherms on several shale samples from Alberta. They showed that the data could be fitted by a Langmuir isotherm. The samples originated from depths between 2,000 and 3,000 m in western Canada at in situ temperatures between 60 and 80° C. They illustrated that maximum sorption capacities of CH₄ vary between 0.012 and 0.029 mmol/g. Weniger et al. (2010) studied the gas sorption behavior of CH₄ and CO₂ in coals and shales in the Parana Basin, Brazil. They demonstrated a correlation between the sorption capacity and the TOC. They found for sorbed CH₄ that $[CH_4]_{ads} = 0.009 \text{ TOC}(\%) + 0.026 \text{ (mol/kg)}$, but that $[CO_2]_{ads} = 0.008 \text{ TOC}(\%) + 0.183 \text{ (mol/kg)}$. Therefore, the sorption capacity on mineral matter for CO₂ is much stronger but not very dependent on the TOC. The TOC, however, is the dominating factor for CH₄ sorption. Wang et al. (2013) investigated the CH₄ sorption capacity of Paleozoic shales from the Sichuan Basin, China. They showed that the TOC content is the controlling factor for CH₄ sorption capacity in these shales. CH₄ sorption experiments were carried out by Ji et al. (2012) on clay-rich rocks at 35, 50 and 65 ° C, and at CH₄ pressure up to 15 MPa under dry conditions. The experimental results illustrate that the clay mineral type greatly affects CH₄ sorption capacity under experimental conditions. In terms of relative CH₄ sorption capacity: montmorillonite » illite/smectite mixed layer > kaolinite > chlorite > illite. Physisorption is the dominant process for CH₄ adsorption on clay minerals and, as a result, in the clay-mineral dominated rocks exists a linear correlation between CH₄ sorption capacity and surface area. Ross and Marc Bustin (2009) showed the effect of shale composition, pore structure and CH₄ sorption for potential shale gas reservoirs in the Western Canadian Sedimentary Basin (WCSB). They demonstrated that CH₄ sorption on dried and moisture equilibrated shales increases with total organic carbon (TOC) content. The results of recent work are summarized in Table 4.

In various studies, the total CO₂ storage capacity of unmineable coalbeds is estimated to range between 100 and 300 Gt CO₂ (Wilcox 2012) and the total storage capacity of deep saline aquifers is estimated to range between 1,000 and 10,000 Gt CO₂ (Wilcox 2012). Gas-bearing shales are also a strong candidate for CO₂ storage. To illustrate the importance of shale formations for CO₂ storage, we can refer to work done concerning the Netherlands. The Dutch resource of shale gas is estimated between 48,000 and 230,000 Bcm. If one assumes that technology can be developed to store 1 m³ of CO₂ for 1 m³ of CH₄ produced, and that only one percent of the resource can be recovered, a substantial 480–2,300 Bcm of CO₂, corresponding to 0.9 to 4.35 Gton CO₂, could be stored (Khosrokhavar et al. 2012). This compares very favorably with the annual Dutch emission of 0.17 Gton/year and would allow for several decades of the Dutch yearly CO₂ emissions to be stored (Khosrokhavar et al. 2012). This indicates the potential importance of CO₂ storage in geological formations, and many studies have been undertaken on this topic (Bachu 2002; Bachu et al. 1994; Busch et al. 2008, 2009; Class et al. 2009; Elder 1968; Elenius and Johannsen 2012; Ennis-King et al. 2005; Foster 1965; Gasda 2010; Iglaier 2011; Khosrokhavar et al. 2014a; Lahann et al. 2013; MacMinn et al. 2012; Godec et al. 2013a, b; Myint and Firoozabadi 2013; Neufeld et al. 2010; Nuttall et al. 2005; Pau et al. 2010; Riaz et al. 2006; Van Duijn et al. 2004; Walker and Homsy 1978). Methods have been proposed to decrease CO₂ emission through storage in geological formations and criteria for the site selection for CO₂ sequestration in geological formations have been given by (Bachu 2002). Nuttall et al. (2005) estimated the CO₂ sequestration capacity in the organic-rich Devonian black shales in the Big Sandy gas field of Eastern Kentucky to be about 6.8 Gt. Gas sorption behaviour of Muderong shales in Australia were investigated by (Busch et al. 2008, 2009). Their experiments indicate significant CO₂ storage capacity in the shale formations. For Belgium black shale, Khosrokhavar et al. (2014b) investigated the sorption capacity for CO₂ is about 7.9 kg/t of the formation, while Busch et al. (2008) showed the sorption capacity for CO₂ is 5.5 kg/t of the formation for dry samples. These results are consistent with those by Weniger et al. (2010), discussed previously regarding the gas sorption behavior of CH₄ and CO₂ in coals and shales in the Parana Basin, Brazil.

Godec et al. (2013a, b), based on numerical investigation, concluded that the maximum CO₂ storage capacity for eastern U.S. shale gas is 1.12 million metric tons per square kilometer, and the sorbed CO₂ storage capacity is estimated to be 0.72 million metric tons per square kilometer. The total CO₂ storage capacity of organic-rich shales at supercritical conditions as a function of pore pressure was measured by Kang et al. (2011) by considering the pore compressibility and sorption effects. The results state that kerogen, the organic part of the shale, acts as a molecular sieve and accounts for the gas sorption on shales.

6 Enhanced Gas Recovery in Shales

Most work on sequestration of CO₂ in depleted gas reservoirs is based on storage. Recently some studies have illustrated that CO₂ injection can be considered as a method of EGR (Blok et al. 1997; Câmara et al. 2013; Oldenburg et al. 2001; Schepers et al. 2009). The injection of CO₂ may increase the rate and the volume of natural gas recovered from a reservoir (Regan 2007; Schepers et al. 2009) as CO₂ can be trapped in shale gas through sorption onto organic materials and clay minerals as a sorbed gas with the parallel displacement of natural gas. Schepers et al. (2009) showed that continuous CO₂ injection (300 t) over a period of 45 days can lead to significant improvement in recovery. As previously discussed, shale gas formations include organic materials. Large amounts of sorbed natural gas could be stored by organic

materials from 20 to 80 % of original-gas-in-place. Recent work demonstrates that CO₂ could displace CH₄ with up to a 5:1 ratio on a molecule-by-molecule basis (Liu et al. 2013) or 14:1 on a mass basis. 1 kg of CH₄ produces approximately 55 MJ (Perry et al. 1984) of energy. To store 1 kg of CO₂, 1 MJ is consumed (Iijima et al. 2011) on compression costs. This suggests that for storage, 55 MJ of energy (Perry et al. 1984) is obtained while spending only about 14 MJ of energy for compression, indicating the potential benefit of EGR from an energy balance perspective. If we take into account the consumption of energy in separation of CO₂ in CCS projects, there is still a net energy gain, even considering storage of CO₂ (Iijima et al. 2011). This means that by storing CO₂ in a shale gas formation, chances for gas recovery increase without significant energy penalty, and this benefit is in addition to those previously discussed regarding storage of CO₂ in shales as opposed to saline aquifers. Godec et al. (2013a, b) showed that by injecting CO₂ in the Marcellus Shale in the Eastern United States, a 7 % enhancement in gas production can be realized. Al-Hasami et al. (2005) investigated the main criteria for CO₂ EGR and found that the density of CO₂ is 2 to 6 times greater than CH₄ at reservoir conditions and this caused the effect of gravity to accelerate the displacement of CH₄. The dissolution of CO₂ in water is higher than the solubility of CH₄ in water and this helps to delay CO₂ breakthrough. The mobility ratio and viscosity of CO₂ are other important parameters for CO₂ EGR projects. Recently, Ishida et al. (2012) investigated the application of CO₂ in hydraulic fracturing and found that use of CO₂ may decrease the environmental risk (i.e., contaminated water) and increase the production rate. As a final and critically important consideration, CO₂ EGR projects are only economically feasible when an available supply of CO₂ is close to the shale gas formation as this decreases the cost of infrastructure, transportation, and operation (Regan 2007).

7 Conclusions

Carbon capture and storage to mitigate the impact of CO₂ emissions from the power and industrial sectors receives increased interest in importance, and sequestration of CO₂ in shale gas formations has significant potential. Shales are widely distributed. Fracked shales have infrastructure in place that can be used for CO₂ storage. The CO₂ storage in shale formations has reduced potential for induced seismicity relative to CO₂ storage in saline aquifers. Furthermore, EGR using CO₂ presents an interesting opportunity to produce a relatively clean burning fuel while simultaneously promoting environmental sustainability through CO₂ storage. We recognize, however, that various studies suggest that shale gas production and utilization may result in significant GHG emissions depending on the total fugitive CH₄ emissions during extraction and utilization. Additionally, extraction of shale gas may have negative impacts on water resources, public health, biodiversity, food supply and soil without adequate technical precautions and regulatory oversight. Hence, EGR and storage of CO₂ in shale gas formations must be further explored before the environmental benefits can be decisively concluded.

As discussed, technically recoverable shale gas is abundant globally, although only the United States and Canada have made substantial progress in the development and production of shale gas reservoirs. Several countries with substantial technically recoverable shale gas, such as China and countries in the MENA region, are struggling not only with technical challenges for shale gas development, but also financial and regulatory challenges. Nonetheless, there is a great global opportunity for developing large, economically recoverable shale gas resources, and so it is necessary to investigate now how such reservoirs may perform for both CO₂ storage and EGR. Our review of the literature on this topic suggests that CO₂ storage

and EGR are viable, yet dependent on shale physical characteristics. Hence, knowledge of shale properties is critical to assessing the economic viability of CO₂ storage in shales.

The literature review in this paper indicates that shales are in fact a good medium for CO₂ storage with capacities of 5 to 10 kg/t of formation or approximately 1 million metric tons per square kilometer. This level of storage capacity suggests a substantial opportunity for CO₂ storage in shales. The opportunity for CO₂ storage in shales via EGR becomes even more attractive as significant energy can be extracted with relatively modest energy input. As discussed in this paper, EGR can be used to exploit the high affinity of shales for CO₂ relative to CH₄ (14:1 on a mass basis).

Although widespread exploitation of shale gas resources and the long-term economic viability of carbon capture and storage are not certain, the state of knowledge regarding geologic storage of CO₂ and EGR in shale formations needs to be improved in order to make these processes viable as the extraction of gas from shale resources gains traction globally. Based on the studies reviewed and information presented in this paper, we conclude that there is a very good opportunity globally regarding the future of geologic storage of CO₂ in depleted shale gas formations and as part of EGR operations.

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