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Recalibration of CO_2 storage in shale: prospective and contingent storage resources, and capacity

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ABSTRACT

Storing CO_2 in shale formations is an essential complement to the resources of geological CO_2 sequestration and the achievement of carbon neutrality. However, previous estimations have reported significant discrepancies in estimating CO_2 storage resources. To rationalize and address the broad range of these resource estimates, we introduce a categorization framework inspired by the SRMS (CO_2 Storage Resources Management System). Thus, we reclassify estimates according to the categories of *prospective* storage resources, *contingent* storage resources, and *capacity* estimates for CO_2 storage in shales, in rank order considering the decreasing challenge associated with their attainment (such as energy/pressure requirements and time-consumed in injection). Classical volumetric and production-based methods are employed for assessing *prospective* and *contingent* storage resources, respectively. The *capacity* is estimated by analyzing the historical records of hydraulic fracturing, focusing on the dominant role of fractures in the storage process. A significant disparity (two to three orders of magnitude) is revealed between *capacity* and *contingent* or *prospective* storage resources, which aligns with known challenges encountered during field injections. This disparity highlights the importance of further efforts and advanced techniques to secure CO_2 injection in fields and recalibrate the geological CO_2 inventories to achieve carbon neutrality.

1. Introduction

Geological storage of CO_2 is an essential strategy for mitigating climate change and meeting the Paris climate goals [1–3]. The International Energy Agency (IEA) estimates that approximately 1 GT (Gigaton) of CO_2/year must be sequestrated by 2030 and 6 GT by 2050 [4]. Storing CO_2 in subsurface formations has been recognized as the most effective engineered approach to sequestration, compared to other methods with a limited resource of CO_2 sink, such as using CO_2 as fertilizer or as a feedstock for chemical production [5,6]. Among the array of geological hosts, oil and gas reservoirs are optimal due to their well-characterized geological and hydrodynamic features, availability of pipeline and injection infrastructure and pre-depletion to reduce the likelihood of injection-triggered earthquakes (both subsurface and ground) [7–9]. This infrastructure potentially saves time and reduces characterization costs compared to other formations, such as saline aquifers [10]. Surprisingly, the development of hydrogen (playing an essential role in the new energy transition) may aggravate the CO_2 storage load because most of the hydrogen will be produced from fossil fuel (coal or natural gas) in the foreseeable future [11]. In the United States, for example, approximately 99 % of hydrogen is currently produced from natural gas, accommodating factors such as land use, infrastructure, and the cost of electrolysis-based hydrogen [12]. In such conditions, approximately 10 kg of CO_2 is co-produced for every kg of hydrogen through SMR (steam methane reforming) or ATR (auto thermal reforming) processes, which may result in an additional 2 GT CO_2/year burden on the CO_2 storage goal by 2050 [7].

Although storing CO_2 in conventional reservoirs accompanied by petroleum engineering operations (CO_2-enhanced oil recovery, CO_2-huff-*n*-puff, and CO_2 fracturing) is technically mature, the cumulative storage resource by 2030 is estimated as ~0.244 GT CO_2/year, well short of the 1 GT goal [13]. Therefore, unconventional shale formations,

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with their unique storage features, become an indispensable supplement to augment the inventory capacity [14,15]. This evolves from the shale gas revolution that emerged only a couple of decades ago when advanced well-logging and surface survey techniques (such as the 3-D seismic) were applied to characterize the geological circumstances more precisely than conventional reservoirs with longer histories [16]. Furthermore, higher standards of well-completion, including high integrity wellbore and cement, were deployed for shale wells to secure the large-scale and high-pressure hydraulic fracturing [17], which provides both a high-quality underground infrastructure and extensive hydrodynamic experience for injecting and sealing CO_2. Second to this, the extremely low permeability of the shale matrix naturally prevents CO_2 leakage [18]. New conceptual strategies, integrating CO_2 capture and storage with hydrogen production and storage in shale, are also important to improve scale, efficiency and economy for carbon neutrality [19]. Moreover, most of the CO_2 (~80 %) is found to be fixed in adsorbed, dissolved and mineral phases and in a shale reservoir may be stable over thousands of years [14]. In contrast, ~95 % of the CO_2 may remain in a free phase in an aquifer formation even after millions of years [19].

Theoretically, the enormous reserves and worldwide distribution of shale reservoirs provide a substantial inventory for CO_2 storage. Previous estimates have reported CO_2 storage resources at the Gt-level even in single regional shale formations (28–171 Gt of CO_2 in Ohio, New Albany, and Marcellus Shale) [20–22], and a resource of ~740 Gt of CO_2 in 69 shale formations worldwide [23]. These prospects consider fractures as the flow path and the shale matrix as the terminal storage site, assuming that *in situ* oil and gas will be replaced by CO_2 in a free or absorbed form. However, field injectivity tests of CO_2 in the Bakken have reported low injection rates (~25 tons/day) even at high pressures (bottom-hole pressures of ~65.5 MPa – but still below the breakdown pressure) [24]. Recent efforts argue that the fracture system in shale provides the major space for CO_2 storage on a timescale of decades [19,25]. This fracture-dominated mechanism, distinct from the connected-pore-system in conventional reservoirs, is supported by field trails of CO_2 fracturing in the Yanjian and Qingshankou shales (China), where new artificial fractures were continuously fractured to achieve a relatively high CO_2 injection rate (~4.8 m^3/min at ~60 MPa of wellhead pressure) [26,27]. This highlights that a consensus is yet to be reached regarding the resource of CO_2 storage in shale, which affects the confidence and prospects of underground CO_2 sequestration.

The following clarifies the prospective storage resources, contingent storage resources, and capacity available for CO_2 storage in shales and considers the challenges related to parasitic losses in injection and time rates of injection. This classification framework refers to the SRMS (CO_2 Storage Resources Management System) published by the Society of Petroleum Engineers [28]. The prospective and contingent CO_2 storage resources are differentiated based on different evaluation methods (the volumetric versus production-based methods). For the estimation of CO_2 storage capacity, we propose a new data-driven method that evaluates the artificial fractures created during hydraulic fracturing. We assume that the volume of the artificial fracture system represents the initial CO_2 storage reservoir in a produced shale formation. A case study is performed to demonstrate estimates based on different methods and assumptions, which highlights the disparities between theoretical analyses and field trials. Potential approaches are also discussed to bridge this gap and to increase CO_2 storage capacity in shales – to guide future research and development efforts.

2. Methodology

Referring to the classification framework used in the SRMS, the CO_2 storage resource in shales can be categorized into prospective and contingent resources, and capacity based on the difficulty and the commerciality associated with attaining each level, as illustrated in Fig. 1. The volumetric and production-based methods are employed in evaluating the prospective and contingent resources [23,29], respectively. In contrast, the CO_2 storage capacity is assessed by analyzing historical data from hydraulic fracturing considering fracture-dominated mechanisms. The recalibration of CO_2 storage

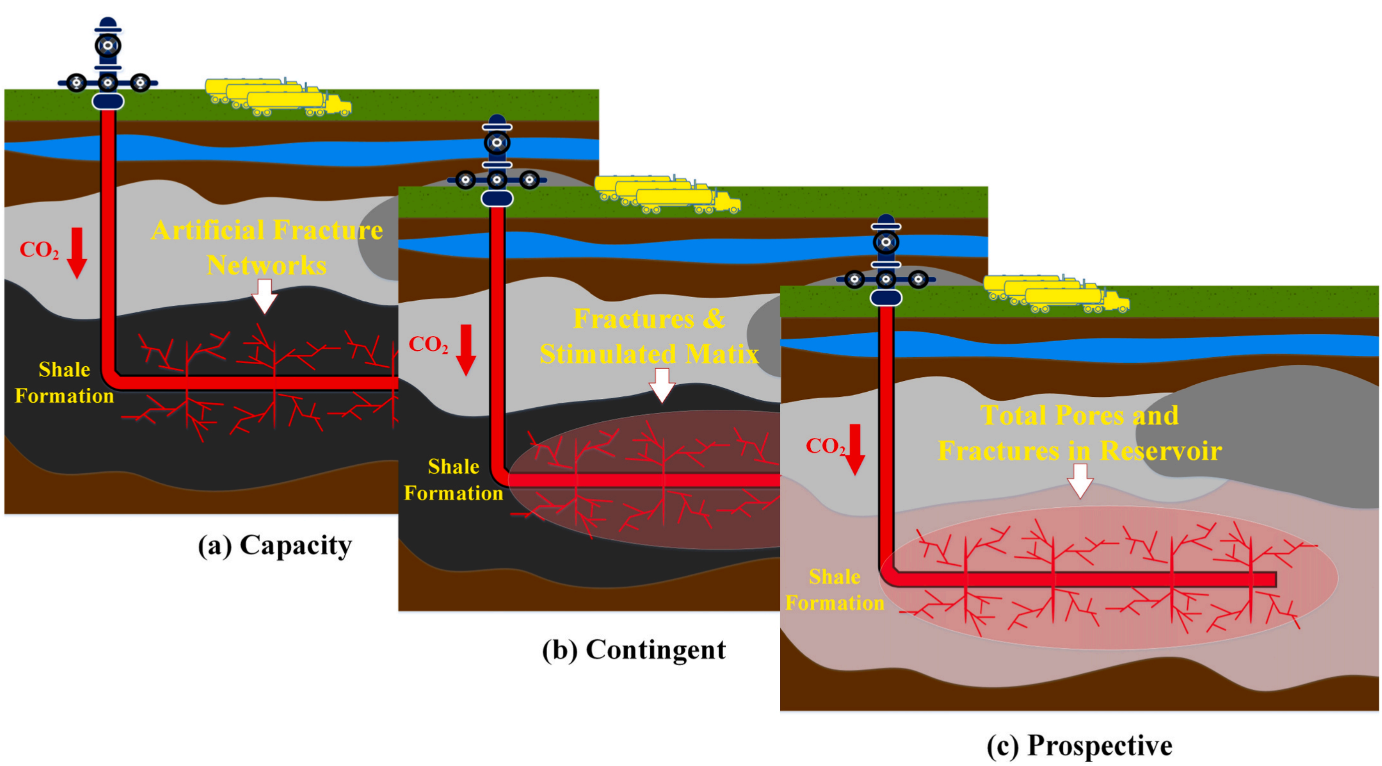

Fig. 1. Schematic of CO_2 storage resources in the shale formation. (a) capacity of CO_2 storage in artificial fracture networks; (b) contingent storage resources in fractures and their stimulated matrix; (c) prospective storage resources in total porosity and fracture systems of the reservoir.

resources aims to facilitate a more accurate assessment of the feasibility and potential of CO_2 storage in shales and provide a valuable reference for field trials, thereby potentially enhancing the development of CO_2 sequestration in shales.

2.1. Prospective CO_2 storage resource of shale

We propose a redefinition of the prospective storage resource in shale as the maximum theoretical resource of CO_2 storage, assuming that all *in situ* oil or gas present in the porosity and fracture systems is completely replaced by CO_2 in either free or absorbed phases (Fig. 1 c). This definition focuses on the shale formations with the prospective hydrocarbon resources. The hydrocarbon resource and the CO_2 storage resource should be evaluated consistently. Consequently, the volumetric method is applied to estimate the prospective CO_2 storage resources, which can be evaluated as [30].

$$G_{CO_2} = A_t E_A h_g E_h [\rho_{CO_2}\varphi + \rho_{sCO_2}(1-\varphi)] \tag{1}$$

where G_{co2} is the mass of the CO_2 storage resource of the shale; A_t is the geographical area of the shale formation; E_A is the fraction of the total shale area available for CO_2 storage; h_g is the gross thickness of the formation; E_h is the fraction of total thickness of the formation available for CO_2 storage; ρ_{co2} is the density of free CO_2 under formation conditions; φ is the percentage of the bulk volume that is a void volume for storing CO_2; ρ_{sco2} is the mass of CO_2 absorbed per unit volume of shale matrix. US-DOE-NETL (United States, Department of Energy, National Energy Technology Laboratory) modified the volumetric equation by incorporating coefficients that quantify the effective volumes of pores (E_φ) and matrix (E_s) accessible for CO_2 storage, as [23],

$$G_{CO_2} = A_t E_A h_g E_h [\rho_{CO_2}\varphi E_\varphi + \rho_{sCO_2}(1-\varphi)E_S] \tag{2}$$

This improved equation excludes disconnected pores and isolated matrix regions with nano-Darcy permeability within shales. However, it is important to note that the effective volumes are dynamic and subject to changes resulting from oil and gas exploitation activities that involve drilling new wells, creating new fractures and stimulating previously inaccessible formations. In theory, the introduced coefficients can approach unity if sufficient exploitation activities are carried out to connect all the pores and natural fractures within the shale formation. We, therefore, suggest Eq. (1) for estimating the prospective resources of CO_2 storage in shales.

2.2. Contingent CO_2 storage resource of shale

We define contingent storage resources in shale as the potential resource for CO_2 storage based on current or developing technologies. This definition sets limitations compared to the prospective resources as presented in Fig. 1 (b) and (c). Correspondingly, the production-based method is recommended for estimating the contingent CO_2 storage resources in shale. This method focuses on gas-producing wells, which serve as indicators of the technological advancements in shale formation exploitation and, subsequently, the potential for CO_2 storage [29]. It assumes that the injection of CO_2 follows the reverse pathway of the produced CH_4, involving advection through fracture networks, diffusion through shale matrix and then adsorption in the kerogen and clays. The core components of this method are the historical CH_4 production data, CH_4/CO_2 sorption equilibria and kinetics models, assuming a ratio of 1:1 between the adsorbed and free phases of CO_2. The sorption models are expressed as

$$\begin{cases} [CH_4](cm/g) = 3.04 + 0.35 * (TOC(\%)) \\ [CO_2](cm/g) = 0.08 + 1.72 * (TOC(\%)) \end{cases} \tag{3}$$

The kinetics model is given by

$$\frac{V_t}{V_\infty} = 1 - \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left(\frac{-n^2 D\pi^2 t}{r_p{}^2}\right) \tag{4}$$

where V_t is the accumulated gas desorption (or adsorption) at time t; V_∞ is the total gas desorption (or adsorption) capacity of the shale matrix; r_p is the diffusion path length; and D is the diffusivity coefficient.

The production of CH_4 from shale formations not only indicates the presence of *in situ* gas but also provides insights into the connectivity of pore volumes within the shale. The production-based method, therefore, enables the estimation of a technically feasible CO_2 storage resource in shale, referred to as contingent resources. For instance, considering a gas recovery rate of 10 %–25 % observed in the Marcellus Shale [20], the estimated contingent CO_2 storage resources are one order of magnitude smaller than the prospective resources, but still remain at the gigatonne level, representing more than 50 % of total U.S. CO_2 emissions over the same gas-producing period [31].

2.3. Capacity of CO_2 storage in shale

2.3.1. Definition of CO_2 storage capacity

We define the capacity of CO_2 storage in shale as the potential that is both technically and commercially feasible. This definition takes into consideration the nano-Darcy permeability of the shale matrix, which presents significant challenges in terms of the time and energy required for CO_2 to permeate and diffuse. While kinetic analyses suggest that CO_2 injection rates could surpass the rate of CH_4 production, the injection period still spans several years to fill the void spaces left following gas production [29]. However, continuous artificial injection of CO_2 into shale matrix over such a long duration is neither technically feasible nor economically efficient and it could result in substantial additional carbon emissions to sustain the operation. For instance, a CO_2 injectivity test conducted on the intact Bakken shale matrix reported a high bottom-hole pressure of 65.5 MPa under an injection rate of 25 tons/day [24]. In contrast, the CO_2 injection rate in a sandstone formation at the Sleipner storage site (located off the western coast of Norway) approaches 3000 tons/day with a wellhead pressure below 8 MPa [32]. This, however, is improved in the case of a shale formation that has been previously fractured. The injection rate is significantly increased, approaching 132.5 tons/day [33]. Recent studies suggest that the fracture system within shale formations not only provides a flow pathway but also serves as the primary volume for CO_2 storage over a decade-long timescale, given the low permeability of the shale matrix [19]. Therefore, the evaluation of the fracture system dominates the capacity of CO_2 storage within the shale formation, as illustrated in Fig. 1 (a).

2.3.2. Estimation of CO_2 storage capacity in shales

The fracture system within shale formations comprises both natural fractures and induced fractures resulting from hydraulic fracturing. Natural fractures in proximity to the wellbore may be reactivated or developed by hydraulic injection, whereas those located at a distance may not be affected. In this study, we disregard the contribution of natural fractures to CO_2 storage due to the current challenges in their detection and quantification. This simplification leads to a conservative estimation of the capacity, focusing primarily on artificial fractures. Nevertheless, characterizing the artificial fractures presents its own difficulties, as they typically have widths on the millimeter scale, based on core drilling [34]. Precise characterization is challenging through techniques such as micro-seismic monitoring or bottom-hole fiber monitoring, as well as numerical simulations [35,36]. To address this issue, we propose a data-driven method for estimating the volume of artificial fractures by analyzing historical data related to proppant injection and transport within underground fractures. The evaluation process is outlined in Fig. 2.

This new method evaluates the artificial fractures at field-practical scales by first training a deep learning algorithm based on a database

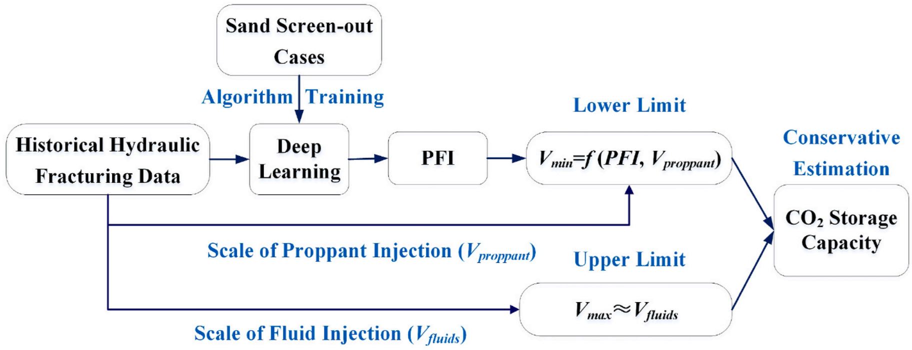

Fig. 2. The workflow for data processing to evaluate the CO_2 storage capacity in shales. The lower limit of the capacity (V_{min}) is assessed based on PFI (the proppant filling index) and injected volume of proppant ($V_{proppant}$). The upper limit (V_{max}) is estimated by the injected volume of fracturing fluids (V_{fluids}).

of multiple sand screen-out cases (assuming that the volume of injected proppant approximately equals the volume of artificial fractures in these cases). A newly defined index, the proppant filling index (PFI), is applied to predict the volume ratio between injected proppant and fractures that are accessible for proppant [37], as illustrated in Fig. 3. During shale gas fracturing, high pump rates (~20 m^3/min), coupled with low viscosity fluids (mainly slickwater) generate fractures that are typically partially filled by proppant [38]. A proppant filling zone and a flowing zone are formed, as shown in Fig. 3, enabling high-pump-rate injections under controllable pressures [39]. We assume that the injected CO_2 will access the initially cracked spaces within the artificial fractures. Then, the fracture volume can be calculated using the predicted PFI and the total amount of injected proppant, as depicted in Figs. 2 and 3. To make this method sufficiently robust for regional then nationwide estimations, we use a finite number of cases to obtain an averaged value of PFI. Subsequently, the fracture volume is calculated by integrating the volume of the injected proppant with the averaged PFI.

We define the PFI-estimated fracture volume as the lower limit of CO_2 storage capacity (V_{min}, Fig. 2), as it is based on the filling degree within fractures. However, there are also many induced fractures with widths too small to accommodate proppant, as well as shear-reactivated fractures that are inaccessible to proppant but are accessible for CO_2. Therefore, we evaluate the upper limit of CO_2 storage capacity (V_{max}, Fig. 2) by assuming 100 % fluid efficiency – all injected fluid remains within artificial fractures without leak-off. By assuming no leak-off or flowback of the injected fluid, the volume of induced fractures is approximately equal to the volume of injected fluid. This assumption is supported by the field observations, in which a pre-fractured shale reservoir can be successfully injected with an amount of CO_2 equivalent to the prior fracturing fluid [33].

3. Distinctions among different categories

The distinctions among prospective resources, contingent resources, and capacity of CO_2 storage in shale are schematically presented in Fig. 1 and summarized in Table 1. The prospective resource assessment focuses on evaluating the shale formation (both exploited or unexploited) at macro- or regional-scale. The volumetric method is employed to estimate the potential storage resource within the pore/fracture volume,

Table 1
Distinctions between prospective resources, contingent resources, and capacity.

	Prospective	Contingent	Capacity
Geological Assessing Range	Exploited and unexploited formations (with hydrocarbon resources)	Exploited formation with production histories	Exploited formation near wellbores
Assumption	All *in situ* oil and gas are replaced by CO_2	Produced oil and gas are replaced by CO_2	Artificial fractures are filled by CO_2
Method	Volumetric	Production-based	Fracture-volume based
Calculations	Eq (1)	Eq (4)	Fig. 1 (Data-driven)
Magnitude	~Gigatonne	~Gigatonne	~Megatonne
Notes			A conservative estimation ignoring natural fractures

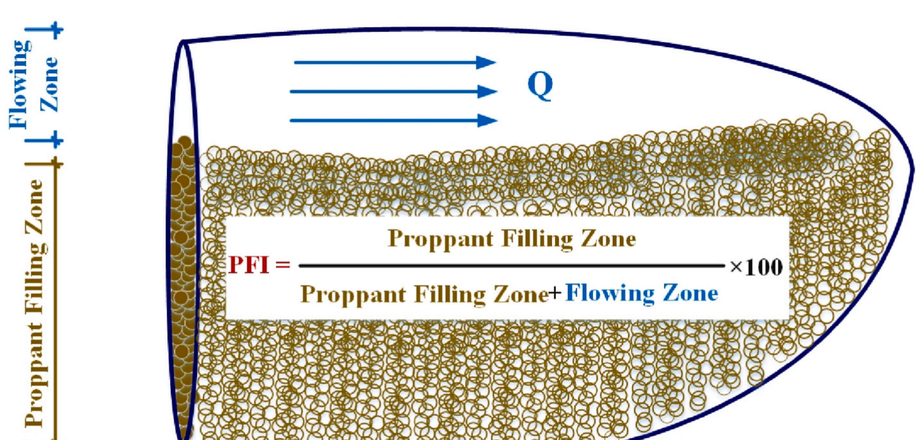

Fig. 3. Schematic of proppant filling state in the artificial fracture.

assuming a complete replacement of *in situ* oil and gas with CO_2. Subsurface characterization data are usually applied to generate the prospective resources of CO_2 storage, which can be refined through further exploration and subsurface characterization during subsequent exploitation activities. The technical and economic feasibilities are generally neglected in this evaluation process, resulting in the largest estimates, often in the gigatonne range among the three categories presented in Table 1.

The contingent resources and the capacity take into account the technical and economic feasibility of CO_2 storage in shale, to varying degrees. Both categories focus on the exploited shale formation, in contrast to the prospective resources. The contingent storage resource assumes that the injected CO_2 will flow through the reverse pathway of the produced shale gas, thus probing and evaluating the limited extent of the formation surrounding the wellbore, as illustrated in Fig. 2 (b). Based on an average shale gas recovery rate of approximately 20 %–30 % [20], the portion of the shale formation accessible for the contingent resource estimation corresponds roughly to the same percentage. Consequently, the estimated contingent resources typically have a similar magnitude but are smaller compared to the prospective resource and are dynamic, with results based on evolving cumulative production data.

The capacity of CO_2 storage in shale formations is subject to stricter criteria for technical and economic feasibility. This evaluation takes into account the time and energy required for CO_2 to penetrate deeply into the shale matrix. Even within the production zone where flowing pathways exist (as assumed for contingent storage resources), shale gas recovery is driven by *in situ* stresses and overpressures. In contrast, CO_2 injection is constrained by the capacity of surface equipment and infrastructure, which may primarily confine CO_2 injection into fractures. As a result, the estimated capacity of CO_2 storage in shale represents the lowest potential among the different categories, particularly due to the omission of natural fractures in the inventory estimation process – defining a conservative storage estimate. However, such estimates are crucial for field implementation, such as in the pre-design stage of injection design for the scaling of pump capacity and wellhead capability, among other parameters.

4. Recalibration for CO_2 storage in Marcellus Shale

4.1. Estimation of artificial fracture volumes based on injected proppant

A pre-trained deep learning model is employed to predict the proppant filling index (PFI) for hydraulic fracturing in shale formations and to thereby establish the correlation between PFI and injected proppant volume. This model is trained using 47 stages of fracturing records, comprising approximately ~10,000 data groups per second during each fracturing stage, obtained from shale gas fracturing wells in the Longmaxi Formation in the Sichuan basin, China. Our previous studies have presented the tuning, training and verification of the deep learning model [37,40]. This study uses the pre-trained model directly to predict the PFI for 10 fracturing stages from 5 different wells located in the same region as the original training data. The results are shown in Fig. 4, along with the corresponding volume of injected proppant for each operation. The PFI is defined as an index ranging from 0 to 100, as illustrated in Fig. 3. A higher PFI value indicates a greater degree of proppant infilling within fractures. An inverted U-shaped correlation between PFI and the volume of injected proppant is observed, as in Fig. 4. Both the low- and high-volumes of proppant injection tend to exhibit relatively low PFIs. Low-volume injections represent the case where the shale formation is difficult to be fractured, due to geological conditions that restrain the development of artificial fractures, limit fracture width and thereby restrict proppant access to the fractures – resulting in a relative filling and low PFI. Conversely, the fractures are well-developed during high-volume injections, with a constantly increasing fracture volume in response to proppant injection. In such instances, the rate of fracture

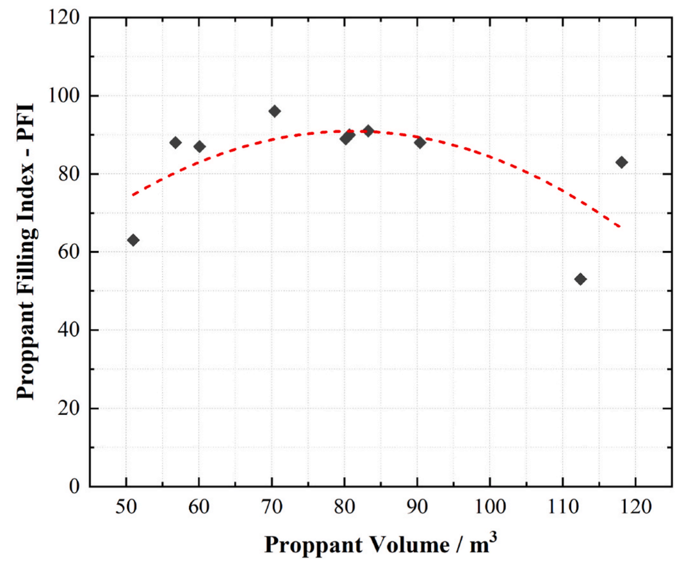

Fig. 4. Correlation between PFI and volume of injected proppant per stage.

propagation exceeds the ability of proppant injection to keep up, leading to a relatively low PFI despite a substantial volume of injected proppant, as shown in Fig. 4.

Using the deep learning model to predict the PFI for each stage of shale gas fracturing at a field-practical scale is a time-consuming and data-intensive process – since typical shale gas fields often include hundreds of thousands of fracturing stages. To ensure the robustness of the PFI-based method in evaluating the capacity of CO_2 storage at basin scale, we adopt the average PFI value (82.8) from Fig. 4 as a representative indicator of the proppant filing status (Fig. 3). Then, the volume of artificial fractures can be estimated by combining the volume of injected proppant, which serves as the lower limit of CO_2 storage capacity in shale, as evaluated from Eq. (5). Simultaneously, the total volume of injected fluids for hydraulic fracturing is utilized to represent the upper limit of the capacity, as specified in Eq. (5). This approach ensures a comprehensive estimation of the CO_2 storage capacity within the shale formation as

$$\begin{cases} V_{min} = V_{fracture} = \dfrac{V_{proppant}}{PFI} * 100 \\ V_{\max} = V_{fluid} \end{cases} \tag{5}$$

4.2. Prospective and contingent resources, and capacity for CO_2 storage in Marcellus Shale

The CO_2 storage potential in Marcellus Shale is compared among the prospective, contingent, and capacity categories in Fig. 5. Previous studies have assessed the prospective storage resource based on the volumetric method, resulting in a resource of 171 Gt for CO_2 storage in Marcellus Shale, assuming complete replacement of in-place CH_4 with injected CO_2 [21]. Considering the average recovery rate of CH_4, the technically achievable storage resource is estimated to be approximately 55 Gt [20], as shown in Fig. 5. Furthermore, the contingent resource of CO_2 storage in Marcellus Shale was evaluated using the production-based method, which projected a resource ranging between 10.4 Gt and 18.4 Gt based on predicted CH_4 production until 2030 [29]. The contingent resource (~18.5 Gt) is roughly one order-of-magnitude smaller than the prospective resource (~171 Gt), but continuously increases in proportion to the cumulative CH_4 production. Similarly, the capacity of CO_2 storage in Marcellus Shale, estimated using Eq (5), exhibits a gradual annual growth in accordance with the cumulative fluid injection and proppant volume, as shown in Fig. 5. The lower limit of the capacity, limited by data availability, is represented to 2014 and

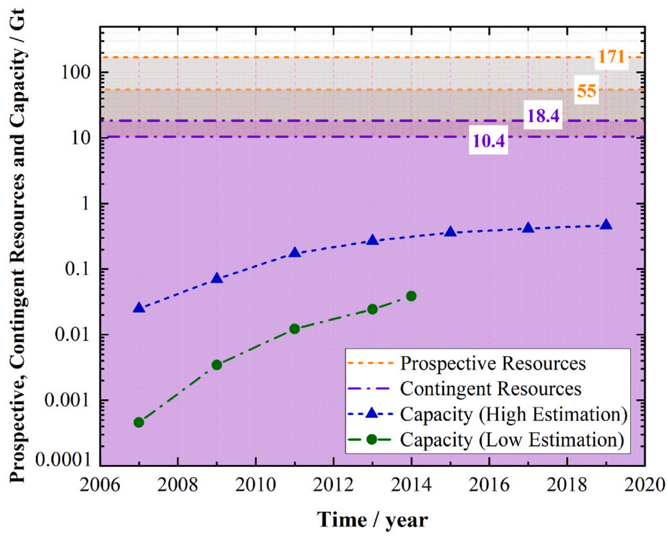

Fig. 5. Prospective [20] and contingent [29] resources, and capacity for CO_2 storage in Marcellus Shale.

amounts to approximately 38.5 Mt (Megaton) [41]. By 2019, the upper limit of the capacity will reach 466 Mt, based on well-drilling and logged volumes of fluid injection [42]. Regardless, this is two to three orders of magnitude smaller than contingent and prospective resources, respectively. A significant disparity between the capacity and the contingent or prospective resources highlights the need for additional efforts and advanced techniques to enhance the efficiency and economic viability of CO_2 storage in shale formations. Closing this gap necessitates further research and innovation in the field.

5. Discussions

5.1. Limitations and implications

The classification framework developed in this study is specifically designed for highly fractured and exploited shale reservoirs. The methodology confronts limitations when applied to undeveloped or emergent shale formations, where the paucity of historical production data restricts the robust assessment of both contingent resources (depending on extant production records) and capacity (based on measurements of hydraulic fracturing) of CO_2 storage. Considering the substantial expenditure for horizontal drilling and fracturing operations, we assume that shale formations lacking demonstrable economic viability are presently unsuitable for CO_2 storage due to the tightness of shale matrix. In such instances, only the category of prospective resources is recommended. Conversely, for economically promising shale formations, initial assessments of contingent resources and capacity can be derived from projected production schedules and anticipated fracturing activities.

In addition, this study utilizes only a single but representative average value of PFI to assess the capacity of CO_2 storage due to the data limitation. This, however, introduces uncertainties due to variations in geological conditions and fracturing operations across different shale gas fields. Predictions of PFI are derived from field records of hydraulic fracturing. Thus, the estimate is more accurate when training a model based on data from a specific region, such as the Marcellus Shale, and then using that model to predict PFI for individual wells within the same region. Applying such data from one region to another requires the adoption of transfer learning if deep learning methods are to be applied. Establishing a correlation between PFI and proppant injection volume, as demonstrated in Fig. 4, can further enhance the accuracy of PFI prediction for each well. The primary objective of this study is to illustrate how the utilization of a definable parameter, PFI, may enable prediction and provide a preliminary estimate of capacity, as well as provide inferred comparisons among prospective and contingent resources, and capacity of CO_2 storage in shale formations. However, it is important to note that the precise prediction of PFI is constrained by the availability of data. While variations among different shale fields introduce some degree of error to the capacity, these are generally minor when contrasted with the substantial differences, spanning two to three orders of magnitude, between the estimated capacity and the contingent or prospective storage resources (Fig. 5).

5.2. Uncertainties in estimating the CO_2 storage capacity

Estimating the potential CO_2 storage capacity in shale formations is subject to many uncertainties – primarily due to ignoring the impact of natural fractures and the innate simplification in using a single average PFI for the full reservoir. Natural fractures play a crucial role not only in shale gas production but also in CO_2 storage in shale formations. These natural pathways enhance the permeability of the mass and provide proximal access to the shale matrix, connect isolated pores to gas and increase the fracture surface area, which in turn facilitates CH_4/CO_2 competitive desorption and exchange/replacement [43,44]. Additionally, natural fractures can be reactivated in shear and connected with the artificial fractures created by hydraulic fracturing by injection. This is an essential mechanism for maximizing the stimulated reservoir volume and thus gas production then CO_2 storage. However, characterizing natural fractures in shale formations remains challenging. Analyzing drill core with optical microscopy or X-ray CT imaging, or utilizing borehole image logs, can directly identify and quantify the presence of fractures within and therefore adjacent to the wellbore. These methods, however, have a limited range (beyond the wellbore) for fracture characterization. Micro-seismic monitoring can reach deep into the shale formation and distant from wells, but only indirectly represents fractures through the proxy of energy release of microearthquakes [45]. Moreover, these techniques currently face challenges in distinguishing between pre-existing natural fractures and induced fractures created by drilling or fracturing activities. Therefore, estimates of CO_2 storage capacity are conservatively underestimated and can be improved by gaining a more comprehensive understanding of the natural fracture networks within shale formations.

5.3. Improving the capacity of CO_2 storage in shale

The capacity of CO_2 storage in shale mainly relies on the volume of fluid-driven artificial fractures – *viz.* hydraulic fractures. This volume increases as injected fluid and proppant volumes increase with time for shale gas development. Moreover, the capacity can be further enhanced by directly using CO_2 as a fluid for fracturing the initial shale reservoir or refracturing the depleted reservoir. Field trials involving CO_2 fracturing in shale formation indicate that CO_2, with a lower viscosity than water-based fluids, can create more complex fracture networks and yield a higher stimulated reservoir volume [46]. Additionally, the flowback of injected CO_2 is significantly reduced compared to the post-fracturing CO_2 injection (for example, as in CO_2 enhanced oil recovery), indicating a higher retention efficiency of CO_2 storage in the shale matrix [27,47]. Moreover, various uses of CO_2 in shale reservoirs, such as CO_2 huff-*n*-puff, hybrid CO_2 fracturing and energy storage in shale associated with CO_2 storage, are also crucial for improving the capacity of CO_2 storage.

5.4. Commerciality of CO_2 storage in shales

The commercial mode (both technical and economic paradigms) of CO_2 storage in shale remains nascent. Field trials have demonstrated the technical viability of CO_2 huff-and-puff and CO_2 fracturing for the purpose of enhancing oil/gas production [24,48]. However, the recovery of injected CO_2 along with oil/gas production presents a significant challenge yet to be fully addressed – a major issue of permanent storage efficiency [25]. Technically, more advanced CO_2 injection techniques in shale are essential, especially the rarely reported security measures (including maintenance of long-term well integrity and monitoring of CO_2 migration) [33,49]. Economically, CO_2 storage in shale is presently incentivized by tax benefits and bolstered hydrocarbon recovery [50, 51]. Therefore, the capacity for CO_2 storage in shale formations is classified by referring to the commerciality criteria of the SRMS. The commerciality may be optimized by the development of carbon trading markets. The elaborations of these technical and economic elements will further subclassify each category on the basis of maturity, yet is currently beyond the scope of this study.

6. Conclusions

As a promising geological sink, the potential for CO_2 storage in shale formations has been extensively evaluated using different perspectives and methods and leading to significant variations in estimated inventory capacities. To provide further characterization and clarification for CO_2 storage capacities in shale, we propose subdividing the storage estimates into three categories. Firstly, the maximum theoretical resource is defined as the *prospective resource* of CO_2 storage in shale formations. The volumetric method is employed to assess the prospective resource, assuming a complete replacement of *in situ* oil or gas in the porosity and fracture systems by CO_2 – and is the most optimistic estimate of performance, possibly unattainable. Secondly, the technically achievable resource is defined as the *contingent resource* of CO_2 storage in shale formations. A production-based method is recommended for estimating the contingent resource, considering that the injected CO_2 follows the reverse pathway of the produced CH_4 and refills the pore/fracture-space left as a result of gas production. Thirdly and finally, the combined constraints of technical and economical storage potential define the *capacity* for CO_2 storage in shale formations. We propose a data-driven method that analyzes the historic record of hydraulic fracturing to evaluate the capacity. This method considers the injected volumes of both proppant and fluids and assumes that the injected CO_2 remains confined within the fracture system of the shale formation over a decadal timescale. By inserting these three categories of evaluation into the lexicon, we provide a comprehensive understanding of CO_2 storage resources in shale formations, considering different aspects of technical feasibility, economic viability and the relevant theoretical limits.

Estimation of the *capacity* for CO_2 storage in shale formations is subject to uncertainties primarily arising from the characterization of natural fractures and the quality of field data (historic records of hydraulic fracturing). Improving the characterization of the *in situ* natural fracture system and utilizing high-quality field data will enhance the accuracy of *capacity* estimates. Moreover, *capacity* for CO_2 storage is two to three orders-of-magnitude smaller than the *contingent* and *prospective* resources, indicating a significant gap between the theoretical resource and the technologically and economically limited capacities. To enhance the efficiency and economic viability of CO_2 storage in shale formations, various forms of CO_2 application in shale reservoirs are crucial, including CO_2 fracturing/refracturing, huff-*n*-puff, and hybrid CO_2 fracturing.

CRediT authorship contribution statement

Lei Hou: Conceptualization, Funding acquisition, Investigation, Methodology, Writing – original draft. **Derek Elsworth:** Conceptualization, Supervision, Writing – review & editing. **Lei Zhang:** Investigation, Methodology, Writing – review & editing. **Peibin Gong:** Resources, Validation, Writing – review & editing. **Honglei Liu:** Methodology, Resources, Validation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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