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Effects of N_2 and H_2S binary impurities on CO_2 geological storage in stratified formation – A sensitivity study

Didi Li[a], Hongcheng Zhang[a], Yang Li[a], Wenbin Xu[a,*], Xi Jiang[b,*]

[a] Guangzhou Key Laboratory of Environmental Catalysis and Pollution Control, School of Environmental Science and Engineering, Institute of Environmental Health and Pollution Control, Guangdong University of Technology, Guangzhou 510006, China
[b] School of Engineering and Materials Science, Queen Mary University of London, Mile End Road, London E1 4NS, UK

HIGHLIGHTS

- Effects of the binary impurities N_2-H_2S on carbon storage in stratified formations were investigated.
- The plume spread and injection-induced pressure build-up in the geologic reservoirs were predicted.
- The binary impurities had more significant effects on geological storage after injection completion.
- Increasing ratio of N_2/H_2S resulted in larger plume spread enhancing the dissolution trapping.
- Permeability distribution and multiple injection on the geological carbon storage were investigated.

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ABSTRACT

Impurities are unavoidable during CO_2 geological storage, and they would potentially affect the plume spread as well as storage capacity and/or efficiency of CO_2. The current study numerically investigated the effects of binary impurities comprising typical components N_2 and H_2S on CO_2 geological storage in stratified formations. For a fixed total content of the binary impurities, increasing ratio of N_2/H_2S resulted in larger plume spread which meant a higher dissolution trapping efficiency. Because of the backflow of formation brine during the post-injection period, the residual trapping efficiency decreased while the dissolution trapping efficiency increased. This tendency was reinforced with increasing ratio of N_2/H_2S. Besides, this work examined the effects of the ratio of vertical permeability (k_v) to horizontal permeability (k_h) and the addition of an injection point in the stratified formation. It was found that lower k_v/k_h shrunk the plume spread and intensified the maximum pressure build-up. However, in the case of two injection points, the plume in the vertical direction was elongated and the maximum pressure build-up was lessened. The results should be taken into consideration to determine the types and concentrations of impurities allowed in the injected CO_2 stream as well as the site selection and injection design for impure CO_2 geological storage in stratified formation.

1. Introduction

CO_2 capture and storage (CCS) is considered as the most promising technology to reduce anthropogenic greenhouse gas emissions into the atmosphere [1,2]. Geological storage of CO_2 in deep saline aquifers represents the best option in the short-to-medium term [3]. Because of deposition and erosion in the processes of strata forming, most potential geological reservoirs consist of alternating high-permeability (high-k) layers such as sandstones and low-permeability (low-k) layers such as shales. The characteristics of the stratified layers are supposed to affect the behaviours of the injected CO_2 stream, including the plume footprint, the amount of CO_2 dissolved and residually trapped, storage capacity and/or efficiency, etc. [4].

The footprint of the injected CO_2 plume plays an important role in the security and permanence of CO_2 geological storage. Because of both the injection-induced pressure and buoyancy force resulted from the density difference between CO_2 plume and the formation brine in situ, CO_2 plume will migrate upwards after injection. If running into faults or abandoned wells in the ascending process, CO_2 may leak into the atmosphere. In stratified formations, the vertical propagation of the CO_2 plume depends on both the layer thickness and the permeability contrast between the low-k layers and the high-k host formation [5]. Apart

* Corresponding authors.
E-mail addresses: xuwenbin@gdut.edu.cn (W. Xu), xi.jiang@qmul.ac.uk (X. Jiang).

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Nomenclature

K	permeability
S	phase saturation
X, Z	Cartesian coordinates
y	component mole fraction in gas phase

Abbreviations

CCS	CO_2 capture and storage
DTI	dissolution trapping index
EOS	equation of state
IP	injection point
IP2	additional injection point 39 m above IP
PR	Peng-Robinson
PT	Patel–Teja
RTI	residual trapping index
SL	shale layer
SRK	Redlich–Kwong–Soave
TTI	total trapping index

Subscripts

g	gas phase (supercritical fluid)
h	in the horizontal direction
v	in the vertical direction

from layer thickness and permeability, sensitivity analyses performed by Kano and Ishido [6] implied that geothermal gradient, capillary pressure and relative permeability were also influential factors on the long-term fate of the CO_2 plume in the multi-layered aquifer. Higher capillary force would slow down CO_2 plume spreading in both the vertical and horizontal directions [7]. Viscosity difference between CO_2 and formation brine as well as the buoyancy force also played a role in the vertical migration of the CO_2 plume in the stratified formations [8]. Core-flooding tests conducted by Oh et al. [9] indicated that stratified heterogeneity including anisotropy and configuration of the embedding low-k layers dominated the upward movement of the CO_2 plume in multi-layered cores. Furthermore, the migration of CO_2 plume was demonstrated to be significantly affected by the sloping of stratified formations [10]. If injection rates and temperature were taken into account, Liu et al. [11] suggested that they were the two most influential factors determining the plume spread in multi-layered formations.

Storage capacity and/or efficiency is one of the most essential issues associated with CO_2 geological storage [12]. In stratified formations, the number of layers was indicated to increase CO_2 dissolution since the low-k layers hindered the upward movement of the CO_2 plume and increased the contact area with the formation brine [13,14]. Oh et al. [9] suggested that storage capacity in the low-k layers was more sensitive to the increase of injection rate than in the high-k layers. Small tank experiments by Agartan et al. [15] indicated that the amount of dissolved CO_2 increased in the low-k layers because of the slow diffusion rates in them. Simulation investigations based on the experiments confirmed that the embedding low-k layers in the multi-layered formations might contribute to the long-term storage of dissolved CO_2 [16]. It was indicated that placing both injection and production well in the lowest permeable layer would result in the maximum CO_2 storage [17]. Although water alternate gas injection was considered to be the least attractive economically, it resulted in the highest overall dissolution and residual trapping in layered reservoirs [18].

The above investigations have contributed to the understanding of CO_2 storage in stratified formations. However, they only considered pure CO_2. In practical CCS projects, captured CO_2 streams from power plants and other large industrial stations usually contain a certain amount of non-CO_2 species, such as N_2, O_2, Ar, H_2S and SO_2 [19,20]. Since the separation of CO_2 from the captured streams was estimated to comprise the major cost of CO_2 capture [21], impure CO_2 injection is often practiced to reduce the total cost of CCS. On the other hand, the inclusion of non-CO_2 species would affect CO_2 plume migration as well as storage capacity and/or efficiency in the geological formations. For instance, the inclusion of low-viscosity impurities such as N_2 and/or CH_4 would result in higher mobility and thus faster migration of the injected CO_2 mixture [22]. Specifically, upward movement of CO_2 plume tended to be enhanced because of the low-density impurity such as N_2 [23].

Generally speaking, the presence of non-condensable impurities and inert impurities such as N_2, O_2, CH_4, Ar as well as the mixture of them would decrease the structural trapping capacity of CO_2 [20]. On the contrary, the inclusion of condensable impurities such as SO_2 would increase the structural storage capacity. Apart from the structural storage capacity, storage capacity in deep saline aquifers also involves solubility trapping mechanism [24]. According to the model of Ziabakhsh-Ganji and Henk Kooi [25], most impurity gases, including N_2, O_2, Ar, CH_4, H_2S, decreased CO_2 storage capacity for solubility trapping mechanism, while SO_2 would enhance it. Furthermore, it was demonstrated that N_2 impurity would impede the convective mixing process in the solubility trapping mechanism, leading to reduced dissolved CO_2 inventory while the effects of SO_2 impurity were opposite [26].

For CO_2 storage in the multi-layered formations, however, there were very few investigations devoted to study the effects of co-injected impurities. Our previous study [23] investigated the effects of one specific impurity (N_2 or H_2S) on CO_2 geological storage in stratified formations. The footprint of injected CO_2 plume extended with N_2 impurity, resulting in larger dissolved CO_2 mass while the effects of H_2S were contrary to N_2 and less significant. In practical CO_2 storage projects, usually more than one kind of impurities would be co-injected. The primary goal of present study was to investigate the effects of N_2 and H_2S binary impurities on CO_2 geological storage in stratified formations, which is the first of its kind. Firstly, the effects of concentration combinations of the binary impurities were evaluated. Secondly, the impact of the ratio of vertical permeability (k_v) to horizontal permeability (k_h) in the stratified formations was estimated. Last but not least, the number of injection points was tested to find its influence on the plume spread and injection-induced pressure build-up. Results from this study are supposed to provide insights on impure CO_2 geological storage in stratified formations, which may be beneficial to the practical deployment of CCS technology.

2. Simulation methodology

2.1. Conceptual model

The dynamics of injected CO_2 stream in the geological formations may involve many processes, including multiphase flow, geochemical reactions, heat transfer as well as mechanical deformation. The present study was mainly focused on important processes of the multiphase flow and the only reactive chemistry considered was the dissolution of gas species in the aqueous phase while other reactive processes were not taken into consideration explicitly. A simplified model in the Utsira Sand Formation of the Sleipner project was adopted [23,27], as shown in Fig. 1. Embedded in the high-k sand, there were four layers of 3 m-thick low-k shale, naming as SL1-4 from top to bottom. The injection point (IP) was located 30 m beneath SL4. Fluid properties and model parameters were listed in Table 1.

To simplify the problem, reservoir temperature was supposed to vary linearly with depth. Specifically, a temperature of 37 °C was used