



Diffusive leakage of brine from aquifers during CO₂ geological storage

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ABSTRACT

The area of investigation in this study is designed around an improved understanding of fundamentals of the diffusive leakage of brine from a storage aquifer into overlying and underlying low permeability layers during geosequestration of carbon dioxide (CO₂) through development of a theoretical model. Here, we consider a two-dimensional domain in cylindrical coordinates, comprised of an aquifer and an overburden, where the interaction between the two media is handled by imposing the continuities of pressures and fluid fluxes at the aquifer-overburden interface. This coupled problem is solved by successive implementation of the Laplace and finite Hankel transforms. The developed solutions can be used to analyze diffusive leakage of brine from the aquifer into overburden and generate type curves for average pressures in the aquifer and overburden during injection and post injection periods. The results show that the leakage rate at early times is scaled with $t^{1/2}$ while it remains constant at late times. It is also shown that the average pressure in the aquifer is scaled with t for short and long times. Moreover, the average pressure in the overburden is scaled with t at late times while it is scaled with $t^{3/2}$ at early times. In addition, the results reveal that factors affecting diffusive leakage rate through intact overburden during CO₂ storage are, in decreasing order of significance, thickness of overburden, thickness of aquifer, aquifer to overburden permeability ratio, and aquifer to overburden porosity ratio. However, thickness of aquifer has minimal effect on diffusive leakage of brine within post injection period. To evaluate the theoretical model, case studies for two potential sites in United Kingdom, one in Lincolnshire and the other one in the Firth of Forth, are conducted. The field studies show that the diffusive leakage from the aquifer into the overburden diminishes ~40 years after the injection has ceased for Lincolnshire while it stops after ~12 years for Firth of Forth. The average amount of the brine leaked from the aquifers per standard cubic meter (Sm³) of the injected CO₂ through diffusive leakage was found to be 6.28×10^{-4} m³ of brine (or 0.330 kg of brine/kg of CO₂) over ~70 years for Lincolnshire and 4.59×10^{-4} m³ of brine (or 0.242 kg of brine/kg of CO₂) over ~42 years for Firth of Forth.

1. Introduction

Leakage of carbon dioxide (CO₂) and brine from deep storage reservoirs is a major risk factor associated with geosequestration (Metz et al., 2005; Celia and Nordbotten, 2009; Shaffer, 2010). Under typical subsurface storage conditions, injected CO₂ is in supercritical state and it is less dense and less viscous than the resident aquifer brines. Consequently, CO₂ migrates upward and spreads laterally under an overburden, which increases the risk of CO₂ and brine leakage into overlying regions through faults, fractures, or leaky wells (Metz et al., 2005; Damen et al., 2005; Celia and Nordbotten, 2009; Neufeld et al., 2011; Sun et al., 2013; Zeidouni, 2014; Huang et al., 2015; Woods et al., 2015; Shakiba and Hosseini, 2016). The leakage of CO₂ and brine may gradually occur. However, such a leakage can remain undetected for a long

time while it has the greatest potential to cause environmental issues in a broad scale (Metz et al., 2005; Damen et al., 2005). Thus, it is very important to understand the fundamentals of fluid flow and transport phenomena involved in the CO₂ and brine leakage associated with geological storage.

Various physical testing techniques can be used for detecting leakage of CO₂ and brine (Benson, 2006). Since the pressure propagation is fast, well testing (which monitors pressure) has been studied as a common potential tool for detection and characterization of CO₂ and brine leakage from the storage aquifer into overlying layers (Sun and Nicot, 2012; Jung et al., 2013; Sun et al., 2013; Zeidouni, 2014; Huang et al., 2015). Shaffer (2010) suggested that leakage of less than 1% of the injected CO₂ over 1000 years from a storage aquifer allows concentrations of carbon dioxide in atmosphere close to those projected for

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Nomenclature

B	formation volume factor
c	compressibility (LT^2M^{-1})
$\mathbf{e}$	unit vector (L)
h	thickness (L)
H	dimensionless thickness
j	diffusive leakage rate of brine (L^3T^{-1})
J	dimensionless diffusive leakage rate of brine
k	permeability (L^2)
K	ratio of aquifer to overburden permeabilities
n	variable of finite Hankel transform
p	pressure ($\text{ML}^{-1}\text{T}^{-2}$)
P	dimensionless pressure
q	injection rate of CO_2 (L^3T^{-1})
r	radial coordinate (L)
R	dimensionless radius
s	complex argument of Laplace transform
S	storativity ($\text{L}^2\text{T}^2\text{M}^{-1}$)
t	time (T)
t_i	time for injecting CO_2 (T)
T	temperature (K)
T	dimensionless time
T_i	dimensionless time for injecting CO_2
$\mathbf{v}$	Darcy velocity vector (LT^{-1})
v	velocity component (LT^{-1})
z	compressibility factor
z	vertical coordinate (L)
Z	dimensionless height

Greek letters

ρ	density (ML^{-3})
ϕ	porosity (fraction)
Φ	ratio of aquifer to overburden porosities
μ	viscosity ($\text{ML}^{-1}\text{T}^{-1}$)
η	hydraulic diffusivity (L^2T^{-1})

ξ	ratio of aquifer to overburden hydraulic diffusivities
ℓ	sign for Laplace transform
$\hbar$	sign for finite Hankel transform
Λ	ratio of aquifer to overburden thicknesses
ω	ratio of aquifer to overburden storativities
Δt	time period for shutting down CO_2 injection well (T)
ΔT	dimensionless time period for shutting down CO_2 injection well

Subscripts

0	early-time asymptotic behaviour
1	aquifer
2	overburden
∞	late-time asymptotic behaviour
e	external
f	fluid
F	Firth of Forth
i	initial
L	Lincolnshire
p	pore
$post$	post injection
r	radial direction
R	finite Hankel transform with respect to R
t	total
T	Laplace transform with respect to T
w	well
z	vertical direction
$\alpha = 1, 2$	aquifer and overburden

Superscripts

—	Laplace transform
~	finite Hankel transform
•	average well pressure
*	average aquifer or overburden pressure

decreasing emissions. By ignoring geomechanical impacts (as a result of deformation) under controlled field circumstances, the pressure differences in the overlying media will be mainly due to the leakage of CO_2 and brine from the storage aquifer (Hsieh, 1996; Kim and Hosseini, 2013). Therefore, pressure measurements for the overlying and underlying layers are critical in leakage detection (Hovorka et al., 2013).

From the start of efforts for reducing CO_2 emissions, several numerical, semi-analytical, and analytical tools have been developed to study the leakage of CO_2 and brine from the storage aquifer into the overlying media (through faults, fractures, or well linings) and the corresponding pressure changes in these two regions. The International Energy Agency (IEA) published a comprehensive report regarding overburden systems for CO_2 geological storage (IEAGHG 2011). All CO_2 leakage rates discussed in the IEA report are in terms of leakage via faults, fractures, and microfractures within the overburden rather than the overburden itself. Instead, Cihan et al. (2011) presented a set of analytical solutions for coupled diffuse and focused leakage of groundwater in a multi-layered system consisting of any number of aquifers, alternating aquitards, pumping/injection wells, and leaky wells. Their governing equations are one-dimensional radial flow in aquifer (including the rate of diffuse leakage through the aquifer-aquitard interface from aquifer into the overlying or underlying aquitard) and one-dimensional vertical flow through aquitard in terms of hydraulic head buildup. They also used the continuity of hydraulic head buildup as the boundary condition at the aquifer-aquitard interface (Cihan et al., 2011). Later, Hou et al. (2012) used the STOMP

(Subsurface Transport Over Multiple Phases) simulator to investigate the sensitivity of CO_2 leakage (from a storage aquifer into an intact overburden) to different parameters of concern by running an extensive numerical simulation. From visual inspection as well as statistical analyses of the fraction of CO_2 leakage after 200 years, Hou et al. (2012) found that critical factors determining CO_2 leakage rate through overburden are, in decreasing order of significance, the overburden thickness, overburden permeability, reservoir permeability, overburden porosity, and reservoir porosity. Subsequently, Chen et al. (2014) investigated the CO_2 leakage through overburden by a combination of experimental studies and numerical simulation. They found that the thickness and permeability of the overburden affect the CO_2 leakage significantly. Chen et al. (2014) also concluded that the leakage rate has a power relationship with the overburden thickness for certain overburden permeability. Recently, Mosaheb and Zeidouni (2017) used the analytical solutions developed by Zeidouni (2012) for leaky fault in a two-layer system and Cheng and Morohunfolu (1993) and Cihan et al. (2011) for fluid leakage through a low-permeability barrier to distinguish between fault and caprock leakage using above-zone pressure response.

However, a limited number of semi-analytical and analytical works in literature addressed mathematical modeling of the diffusive leakage of brine from the aquifer into the overburden and underburden where the three media interact with each other at the aquifer-overburden and aquifer-underburden interfaces. Under these conditions, the pressure does not only vary in the aquifer (from which brine leakage happens)

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