1 Introduction

1.1 Motivation

1.1.1 Why CO_2 storage?

Since the end of 19^{th} century, the global surface temperature has increased significantly according to data derived from ice-core and tree-ring samples (IPCC 2007). The average global temperature will increase by 1.1 to 6.4°C by the end of the 21st century as a result of greenhouse gases (GHG) emission (IPCC 2007). Global warming has caused many problems, including climate change and the melting of glaciers, which threatens the food security as well as the living environment of all human beings. The cause of the temperature increase is generally believed to be the increasing emissions of greenhouse gases into the atmosphere.

Greenhouse gases generally contain water vapor, carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), as well as ozone (O_3). In spite of natural existence in the atmosphere, human activities have contributed greatly to the increase of their concentrations. From the beginning of the industrial era (end of 19 century) to 2005 concentrations of these gases have globally increased to 36.148 ppm, corresponding to 18 percent (IPCC 2007).

The greenhouse gases absorb a part of the long-wave heat radiation emitted from the surface of the earth and further cause a heating-up of the atmosphere (Houghton 1997). In order to evaluate the ability of each greenhouse gas to absorb heat in the atmosphere compared to another gases, the IPCC developed the Global Warming Potential (GWP) concept, as listed in Table 1.1. The GWP of the greenhouse gas is defined to be the ratio of the time-integrated radioactive forcing of the instantaneous release of 1 kg trace substance to that of 1 kg reference gas (IPCC 2001).

Table 1.1: Global warming potential (IPCC 2001)

Gas	GWP		
	20 years	100 years	500 years
CO_2	1	1	1
CH_4	62	23	7
N_2O	275	296	156

Figure 1.1 gives the information on the composition of greenhouse gases emissions in a typical year in the 21^{st} century. From this figure, it is shown that CO_2, despite relatively small GWP compared with the other greenhouse gases from Table 1.1, makes the largest contribution to the global warming due to the total volume of CO_2 into the atmosphere from fossil fuels use. Globally, about 35 billion metric tons of CO_2 per year is released into the atmosphere as a result of burning oil, gas, and coal. Estimates predict that CO_2 emissions from oil, gas, and coal will amount to more than 41 billion metric tons by 2020 (World Bank Report 2012).

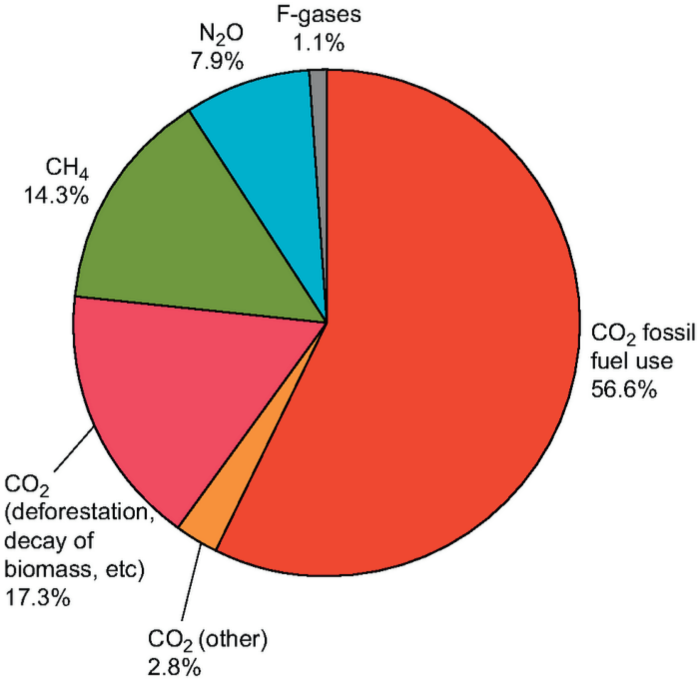

Figure 1.1: Global greenhouse gas emissions from anthropogenic activities in 2004 (IPCC 2007)

The rapid reduction of the CO_2 emission into the atmosphere is the preoccupation of preventing the global warming. Basically, the measures used to reduce the CO_2 emission can be classified into four categories:

1) Using alternative renewable energy instead of fossil fuels, which emit less to no CO_2, such as wind energy, biomass energy, solar energy, and geothermal energy.

2) Developing new technologies which could be used in such fields as motor vehicles to reduce CO_2 emissions.

3) Creating efficient policies to encourage companies and individuals to curb activities which can lead up to CO_2 emissions, including emission tax, which is carbon cap on the total annual emissions which is combined with tradable allowance system, and a cap-and-trade program which has features to decrease the cost to reduce emissions (Congress of the United States 2008).

4) CO_2 capture and storage.

CO_2 capture and storage, generally known as CCS, refers to CO_2 captured from large point sources, like coal-fired plants, then compressed for storage in geological formations. The CCS technology consists of the infrastructure establishment, CO_2 capture technology, transportation as well as storage in locations where CO_2 can be successfully isolated (IPCC 2005).

To reduce CO_2 emissions, the best method is to change fossil fuels as main energy source to low carbon renewable sources as well as reducing energy consumption by using energy effective methods. Dramatic reduction or elimination of anthropogenic CO_2 emissions from the burning of fossil fuels is possible in the far future; however, in the short term it is not a realistic solution because of the enormous cost needed to switch to alternative sources of energy. A man-made CO_2 storage site can reduce CO_2 emission dramatically at a much lower cost, compared with other methods, which can help to gain the time which is needed to develop new technologies to cut the cost of renewable energy. The CCS technology is suggested as one of the effective methods to reduce global CO_2 emissions by the IPCC, and recent presentations of IPCC reports indicate that CCS has in fact higher potential than addressed by the most optimistic IEA (International Energy Agency) scenarios. Therefore, CO_2 capture and storage is considered as an efficient solution to reduce the greenhouse emission in the atmosphere for the next few decades.

1.1.2 Why saline aquifer?

Generally speaking, there are three main types of geological media which are potentially suitable for CO_2 storage: depleted oil and gas reservoirs, unmineable coal seams, and saline aquifers.

1) Depleted oil/gas reservoirs

Depleted oil and gas reservoirs are important potential targets for carbon storage by direct CO_2 injection into geological formations. It is generally believed the sealing capacity of the oil and gas reservoir caprock is sufficient to keep CO_2 safely in the reservoir because the accumulation and entrapment of a light gas, such as methane, testifies to the integrity of reservoirs for containing gas for a long period (Oldenburg *et al*. 2001). In addition, injecting CO_2 into oil and gas reservoirs can be used to increase the amount of oil and natural gas production which is known as Enhanced Oil Recovery (EOR) and Enhanced Gas Recovery (EGR), respectively. The engineering CO_2 injection into geological formations was first carried out in Texas, USA in the 1970s as EOR projects (IPCC 2005). However, pure storage of anthropogenic CO_2 as the mitigation of greenhouse gases was not performed until the end of 1990s, when many demonstrations and commercial EOR and EGR projects were completed, such as the Weyburn EOR Project in Canada in May of 2000, and the In Salah EOR Project in Algeria in 2004 (IPCC 2005).

2) Unmineable coal seams

Unmineable coal seams are another promising value-added target for carbon storage. Coal bed seams are besides source of methane also carbon-dioxide sink. The injection of CO_2 into coal beds works for CO_2 storage and can also replace methane in Enhanced Coal Bed Methane production (ECBM) (Zoback *et al*. 2004). So far, CO_2 has been successfully injected at the Allison Project, which commenced in April 1995 in the northern New Mexico in the USA; another successful example is the Alberta Basin Project in Canada (IPCC 2005).

3) Saline aquifer

Saline formations generally refer to deep sedimentary rocks saturated with formation water or brine which contains high concentrations of dissolved salts. Besides high accumulations of thick sediments and permeable rock formations which can be saturated with water, qualified saline formation for the CO_2 storage should also have characteristics like extensive cover of low permeable rock over the saline formation as well as simple geological structure. Figure 1.2 shows a schematic of CO_2 storage in a saline aquifer.

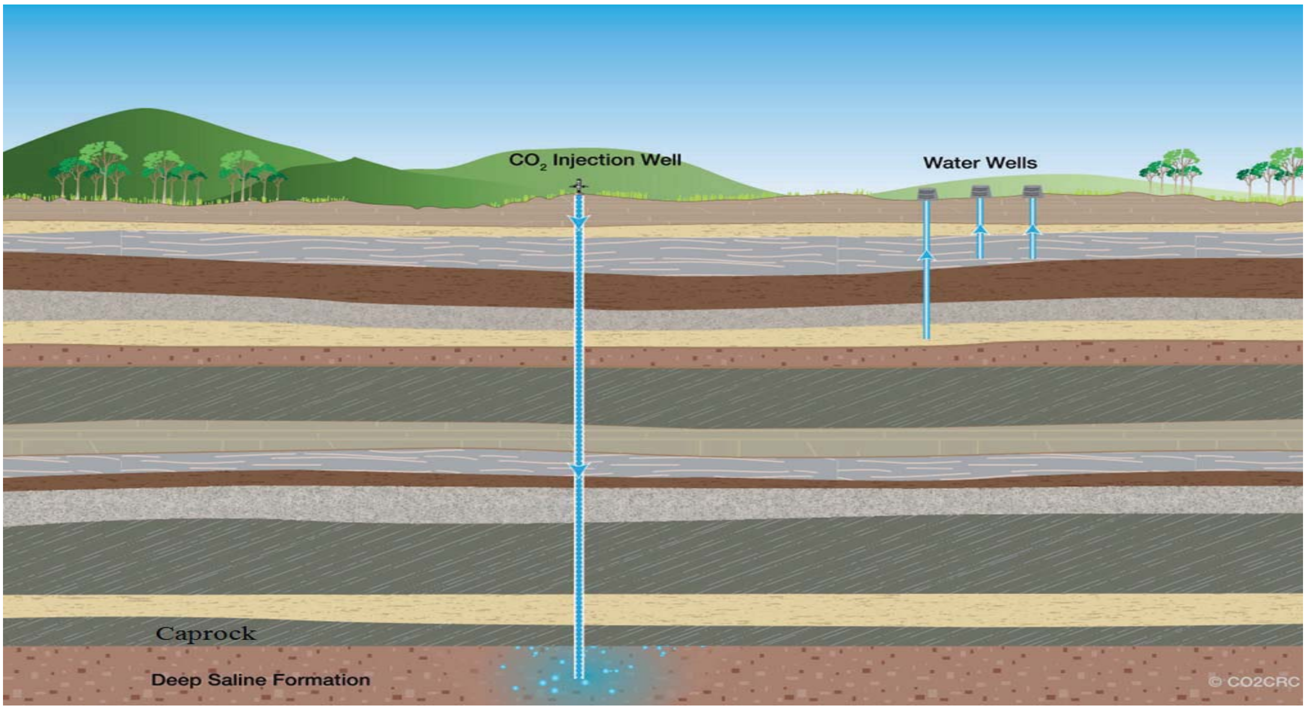

Figure 1.2: Schematic of CO_2 saline formation storage (adapted from IPCC 2005)

Why is the saline aquifer chosen for this work? Compared with the CO_2 storage in depleted oil/gas reservoirs and coal beds, the CO_2 storage in deep saline formations does not produce value-added byproducts. However, deep brine aquifers with a confining layer that serves as a cap-rock exist in many places all over the world. The storage capacities of saline aquifer are significantly higher than those of oil and gas reservoirs and they are more likely to be found close to large CO_2 point sources (Bachu 2003; Bielinski 2006). CO_2 storage capacities (Shukla *et al*. 2010) are listed in Table 1.2.

Table 1.2: Storage capacities and risks (Shukla *et al.* 2010)

Storage option	Capacity (Gt-CO_2)	Storage integrity	Environmental risk
Depleted oil and gas fields	25-30	High	Low
Active oil well (EOR)	Low	High	Low
Enhanced coal bed methane	5-10	Medium	Medium
Deep aquifers	1-150	Medium	Medium
Ocean (global)	1000-10,000	Medium	High
Carbonate storage (no transport)	Very high	Highest	High

Koide *et al*. (1993) stated that global sedimentary basins are capable of holding around 320 Gigatons of carbon dioxide. The CO_2 storage in saline aquifers is chosen as an option to mitigate the CO_2 emission in the atmosphere due to its significant capacity.

1.1.3 Why geological barrier integrity?

The storage safety is the most concerned issue with regards to CO_2 storage in saline aquifer, while the geological barrier integrity is the focus of the storage safety. To make clear the CO_2 trapping mechanisms in saline aquifer is the prerequisite of the geological barrier integrity study. Since the concept of CO_2 underground storage was introduced, the trapping mechanisms have been studied by a number of researchers. Generally speaking, CO_2 underground storage is realized through four main mechanisms:

1) Hydrodynamic trapping (Nghiem *et al*. 2004), CO_2 as a separated phase is trapped beneath an impermeable caprock. The amount of trapping is influenced by the CO_2-brine relative permeability, which depends on the rock composition and may vary between different locations.

2) Solubility trapping (Pruess *et al.* 2003), refers to CO_2 dissolution in saline water. The CO_2 storage in solution is controlled by the phase behavior of water-CO_2 mixture, which is dependent on the salinity of water, temperature and pressure.

3) Structural trapping (Shukla *et al.* 2010), the CO_2 rises to the top of saline aquifer which lies below an impermeable caprock and is stored there.

4) Mineral trapping (Pruess *et al.* 2003), the acidity of the CO_2-water increases as CO_2 dissolves into the brine, which consequently leads to the dissolution from rock minerals and resultant precipitation.

The interaction between CO_2 plume and reservoir as well as caprock can have strong influence on the geo-mechanical and geo-chemical properties of the reservoir and caprock, leading to the alteration of porosity, permeability as well as fracturing and fault reactivation, resulting in the integrity issues, which could possibly lead to the leakage and jeopardize the storage safety.

1.2 State of the art

1.2.1 Ongoing CO_2 sequestration projects

Ever since the first CO_2 saline aquifer storage project was started in Statoil's North Sea Sleipner facility in 1996 (Johnson *et al*. 2002), CO_2 storage in saline aquifers has become an effective solution to reduce the CO_2 emission. Since then, there are numerous CO_2 storage projects in saline aquifers all over the world which are active or under planning. Among those projects are some pilot projects, some of which have been used as commercial CO_2 storage. Table 1.3 provides an overview of the well-known CO_2 storage sites in saline aquifer around the world.

Table 1.3: Major geological sites of CO_2 saline aquifer storage worldwile (modified based on Micheal et al. 2010)

Project name	Location	Scale	Status	Inj.start	Inj.finish	Inj.rate (t/day)	Total storage (kt)
Frio	Liberty County Texas USA	Pilot	Completed	Frio 1 2004 Frio 2 2006	Frio 1 2004 Frio 2 2006	250	1.6
Nagaoka	Nagaoka City, Japan	Pilot	Completed	2003	2005	40	10
Ketzin	Ketzin, Germany	Pilot	Injection	2008	2013	86	60
Alberta Basin (Acid Gas)	Alberta&B.C., Canada	Commercial	Injection	1990		5-190	
Snøhvit	Barents Sea, Norway	Commercial	Injection	2008		2000	23,000
Sleipner	North Sea, Norway	Commercial	Injection	1996		2700	20,000
In salah	Krechba, Algeria	Commercial	Injection	2004		3500	17,000
Gorgon	Barrow Island, WA Austria	Commercial	Approved	2014		12,300	12,900
MGSC Decatur	Decatur, IL,USA	Demonstration	Work underway	2010	2012	1000	1000
MRCSP Cincinnati Arch	Kentucky,USA	Pilot	Monitoring	2009	2009	500	1
MRCSP Michigan Basin	Gaylord, MI,USA	Pilot	Monitoring	2008	2009	300-600	60
SECARB Mississippi	Escatawpta, MS,USA	Pilot	Completed	2008	2008	160	2.75
SECARB Early	Cranfield, MS, USA	Demonstration	Injection	2009	2010	2700	1500
Shenhua Ordos	Ordos China	Demonstration	Injection	2010		274	200,000

1.2.2 Research status

Since the concept of CO_2 storage in saline aquifer was introduced, numerous studies have been performed on the CO_2 storage safety all over the world. Analytical solutions and numerical simulations are two basic methods.

1.2.2.1 Analytical solutions

Analytical solution is the basic method which is used to solve the geological problems. Analytical study of the caprock integrity issues with regard to CO_2 storage has been performed by a few researchers. Streit and Hillis (2004) estimated the stability of fault and sustainable fluid pressure for CO_2 storage in porous media, and the FAST technique was used to estimate the fluid pressures that lead up to failure of the reservoir rock. Hawkes *et al*. (2005) found that if the maximum shear stress acting on the fault zone surpasses the shear strength of the fault, there will be the fault reactivation. Soltanzadeh and Hawkes (2009) developed the ΔCFS (Coulomb Failure Stress) concept to predict the reactivation tendency of fault under normal and thrust stress regimes. Sinha *et al.* (2010) built a 1-D mechanical model using the deformation and strength properties derived from the log data to perform the wellbore stability analysis. Zeidouni *et al.* (2011) introduces an analytical method to evaluate the pressure change in the overlying layers due to leakage from the storage formation.

1.2.2.2 Numerical simulations

Numerical simulations have been used in CO_2 storage ever since the concept was originally proposed. Lindeburg (1997) simulated CO_2 escape in saline aquifers involving CO_2 dissolution, which is one of the earliest numerical simulations of CO_2 storage. Oldenburg *et al*. (2001), and number of other simulations are also performed using this code (Pruess *et al*. 2002; Pruess *et al*. 2003). However, early numerical simulations only focused on the hydraulic process, while the geomechanical effects caused by CO_2 injection are generally ignored.

Meanwhile, few of the existing numerical software programs, which are used for hydro-mechanical simulations, take multiphase flow into account. Guy *et al*. (2010) performed hydro-mechanical coupled simulations of CO_2 storage using CODE_Aster, in which the hydraulic process is treated as single phase flow. The direct Hydro-mechanical coupling is realized in some codes like CODE_BRIGHT and COMSOL, Vilarrasa *et al*. (2010) by conducting simulations of CO_2 injection into saline aquifers with the modified CODE_BRIGHT, which is a finite element code designed for modeling of coupled thermo-hydro-mechanical phenomena in geological media. Bjørnarå *et al* (2010) performed a coupled geomechanical modelling analysis on CO_2 storage in geological media using the modified COMSOL simulator, in which the multiphase flow was taken into account. Nevertheless, these codes are not widely used for HM coupling because of efficiency problems.

More common practice in HM coupling in CO_2 storage is using coupled code like TOUGH2-CODE_Aster or TOUGH2-FLAC3D to realize the multiphase coupling simulation of CO_2 storage in geological media. The coupled codes realize the coupling of the hydro-mechanical process, which provides a more realistic solution. Rohmer and Seyedi *et al*. (2010) used CODE_Aster in combination with TOUGH2 to simulate CO_2 storage in saline aquifers. Meanwhile, more studies were performed by the coupled TOUGH2-FLAC3D simulator. The simulator is successfully used by Rutqvist and Tsang (2002), Rutqvist *et al*. (2007) and Ruqvist *et al*. (2008) in topics with regards to CO_2 storage in geological media. Rutqvist and Tsang (2002) demonstrated the feasibility of the coupled TOUGH2-FLAC3D code in simulating CO_2 injection and storage problems in saline aquifers. Rutqvist *et al*. (2007) estimated the maximum sustainable pressure during the CO_2 geological storage using the shear-slip analysis. Rutqvist *et al*. (2008) found out that the potential of the mechanical failure, the type as well as orientation of the failure, depends mainly on the initial stress field.

1.3 Objectives and structure

1.3.1 Objectives of this dissertation

One of the most important aspects with regards to the long term CO_2 storage in saline aquifers is the integrity of the geological barrier which can trap CO_2 in the saline aquifer. However, the existing integrity studies regarding CO_2 storage into saline aquifers either fail to take the geomechanical effect into account (Oldenburg *et al*. 2001; Pruess *et al*. 2002; Pruess *et al*. 2003) or use a simplified model which is not realistic, like treating CO_2 mixture as single phase flow (Vilarrasa *et al*. 2010). Even both geomechanical effects and multiphase flow are considered, some studies only use the direct hydro-mechanical coupling (Kempka *et al*. 2014), where pore pressure distribution data are transferred to the geomechanical model at each simulation time step to calculate the geomechanical efforts, while ignores the interaction between the pore pressure and the mechanical properties. Therefore, the main objectives of this dissertation are to:

1) Develop an indirect hydro-mechanical coupling concept for CO_2 storage simulation, which can take the hydro-mechanical interaction, especially geomechanical effects, such as permeability, porosity, Biot's coefficient alteration due to CO_2 injection into account, and finally use it to analyze the integrity of geological barrier.

2) Provide a better reference for the ongoing and future CO_2 storage projects in saline aquifer by analyzing the simulation results of CO_2 storage in a generic reservoir.

1.3.2 Structure of this work

Chapter 1 provides a brief overview of the motivation of this dissertation, the state of art, objectives and the structure of the dissertation. Chapter 2 gives definitions and terms which explain hydraulic and mechanical fundamentals as well as the numerical simulator which is to be used in this study. Chapter 3 presents one effective mean stress dependent permeability model based on the experiments results of Albrecht *et al*. (2010) and one effective mean stress dependent Biot's coefficient model developed based on literature data. Implementations of developed permeability and Biot's coefficient models into TOUGH2MP-FLAC3D code are also realized in this chapter. Chapter 4 presents mechanical criteria for the integrity of geological barrier. A real case study of the Ketzin CO_2 storage site in Germany is performed using the modified HM code TOUGHMP-FLAC3D in Chapter 5. The sensitivity analysis is performed in chapter 6 to counteract the result uncertainty due to parameter heterogeneity as well as the scenario uncertainty. A commercial scale CO_2 injection simulation using a generic model is carried out in Chapter 7, to study the integrity geological barrier under commercial scale injection conditions. Discussion and conclusions are presented in chapter 8.

2 Fundamentals of hydro-mechanical coupling

The technology of CO_2 storage in saline aquifer can give rise to many geological processes, such hydro-mechanical(HM), hydro-thermal (HT) and thermal-mechanical (TM) processes, of which TM and HT processes are not considered, while HM process is the focus of this work. The hydraulic process in CO_2 storage is very complicated, and the CO_2 in a deep brine formation will be presented in three forms (Bachu *et al*. 1994): (1) a dense gas phase which is in supercritical state, (2) a dissolved phase in water, (3) an immobilized state from geomechanical reactions with in situ minerals (which is not considered in this dissertation). Hydro-mechanical (HM) coupling refers to the interactions between hydraulic and mechanical processes in geological media. HM coupling contains two fold meanings (Rutqvist and Stephansson 2003): (1) Direct HM coupling, which occurs through deformation and pore pressure changes; (2) Indirect HM coupling, occurring through changes in hydraulic and mechanical properties. HM coupling process in CO_2 underground storage is complex. The CO_2 injection into saline aquifer can result in increase of pore pressure in the formation, especially in the vicinity of the injection source; and the pressure increase will gives rise to changes in the effective stress, which further induces mechanical property changes, such as alteration in porosity and permeability. Figure 2.1 gives a schematic overview of the indirect hydro-mechanical coupling process.

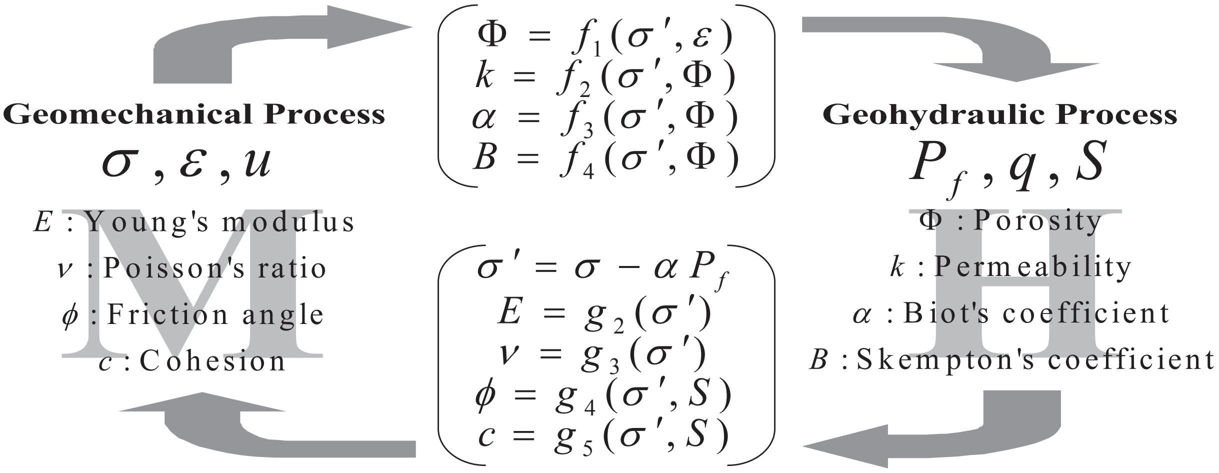

Figure 2.1: Schematic diagram of HM coupling (Hou 2010)

The HM coupling process involves knowledge of many disciplines such as rock mechanics, fluid mechanics and multiphase flow. Basics of multiphase flow and the properties of CO_2 are presented in this chapter.

2.1 Fundamentals of rock mechanics

A sound understanding of the fundamentals of rock mechanics is of great importance for the geological barrier integrity analysis with regards to CO_2 underground storage.

2.1.1 Linear elasticity and Hooke's Law

If changes in the forces are sufficiently small, the response of the formation is linear. This phenomenon is called linear elasticity which can be presented by Hooke's law (Fjaer *et al.* 2008) under uniaxial load:

$$\sigma = E\varepsilon \tag{2.1}$$

Where:

E	Young's modulus, MPa
σ	Stress, MPa
ε	Strain, -

Under the three axial loads, the Hooke's law can be generalized into the following form:

$$\begin{aligned}
\varepsilon_x &= \frac{1}{E}[\sigma_x - \nu(\sigma_z + \sigma_y)],\ \gamma_{xy} = \frac{\tau_{xy}}{G} \\
\varepsilon_y &= \frac{1}{E}[\sigma_y - \nu(\sigma_x + \sigma_z)],\ \gamma_{yz} = \frac{\tau_{yz}}{G} \\
\varepsilon_z &= \frac{1}{E}[\sigma_z - \nu(\sigma_x + \sigma_y)],\ \gamma_{zx} = \frac{\tau_{zx}}{G} \\
\varepsilon_{vol} &= \frac{3(1-2\nu)}{E}\sigma_m = \frac{\sigma_m}{K}
\end{aligned} \tag{2.2}$$

Where:

σ_x, ε_x	Stress and strain in x direction,
σ_y, ε_y	Stress and strain in y direction
σ_z, ε_z	Stress and strain in z direction
τ_{xy}, γ_{xy}	Shear stress and shear strain in xy plane
τ_{yz}, γ_{yz}	Shear stress and shear strain in yz plane
τ_{zx}, γ_{zx}	Shear stress and shear strain in xz plane
σ_m	Mean stress, MPa
ν	Poisson's ratio, -
G	Shear modulus, GPa
K	Bulk modulus, GPa