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1 Introduction

1.1 Motivations and Objectives

With the strong increase in economics and a rising population, the world's energy consumption is increasing. According to the analysis by EIA (2016), the total world's energy consumption is supposed to increase by 48% between 2012 and 2040, in which the non-OECD nations (most are developing countries) will contribute a huge part. Currently fossil fuels and renewable energy are both developing. Although renewable energy is increasing very fast, fossil fuels are still the main source of energy and will account for 78% of the total energy use in 2040 (EIA 2016). It could be predicted that the most important sources of energy will be found underground for a long period, and geo-energy must play a major role in today's energy production (Hou et al. 2015a, Kolditz et al. 2015). Geo-energy includes not only the fossil fuels including gas, oil and coal, but also renewable energy such as geothermal energy.

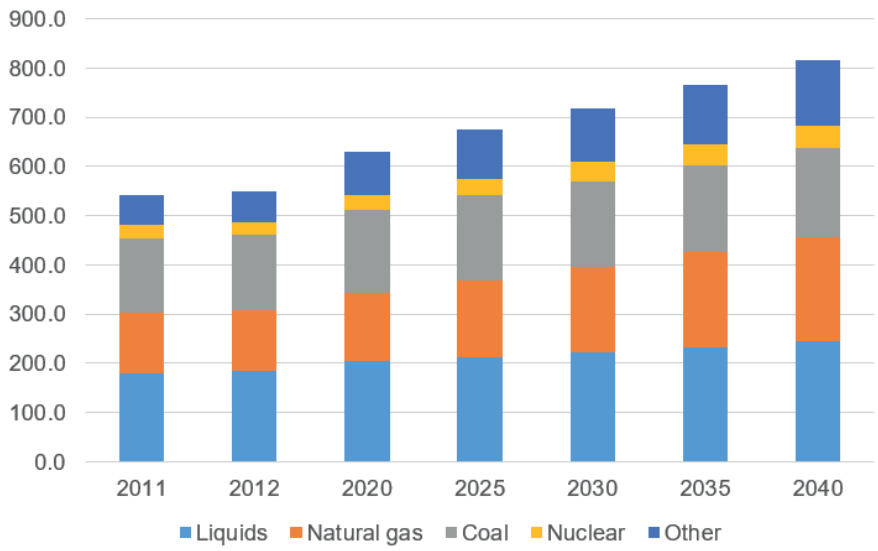

Figure 1.1 World energy consumption in quadrillion Btu (EIA 2016)

Geo-energy is being produced and delivered directly to the consumers or stored underground in interim storage facilities like depleted reservoirs, rock caverns, to cover the seasonal fluctuation in demand (Rutqvist et al. 2012, Kabuth et al. 2017). In order to reduce emissions,

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greenhouse gases like CO_2, which are generated by the combustion of fossil fuels, will be injected and stored in the subsurface (Kolditz et al. 2012a, Liu et al. 2014). All such activities require an intrusion of the underground area. For this purpose, an understanding of the geosystem and related processes is necessary. Four geo-processes are determinant, namely the thermal (T), hydraulic (H), mechanic (M) as well as chemical (C) processes.

For the past hundred years, these processes have been studied separately. However, researchers have found that they are coupled with each other, and sometimes the coupling effects are so strong that they cannot be neglected. An example of this is deep geothermal energy utilization. In order to utilize the geothermal energy underground, cold water was injected into the subsurface, warmed underground and then produced from another well. For such a system, not only the heat exchange but also the fluid flow, and the interactions between them are important. On one hand the heat stored in the rocks will be extracted by the fluid. The increase in temperature leads to a change in the fluid properties or and also its phase state, which will alter the flow behavior of the fluids. On the other hand, the heat transfer is strongly dependent on the flow behavior and the geomechanical processes have also influenced the geothermal system. During the construction and operation phase, there is a stress change and deformation of the rock formations, which is closely connected to the hydraulic pathways of the rocks. The fluid injection may affect the stability of the system or induce microseismicity, which can be evaluated using geomechanics. The geochemical processes are one of the influence factors, which will affect the heat exchange and flow process. The rock-fluid interactions, e.g. dissolution and precipitation, should also be considered. The production of underground energy and resources highlights out the requirement to study the coupled THM/C processes.

Modern experimental apparatus and the fast development of computer techniques as well as simulation methods make the study of coupled THM/C processes possible. On one hand, modern techniques such as large-scale three axial geomechanical test system, acoustic emissions test devices, computer tomography (CT) scanners, nuclear magnetic resonance (NMR) device are utilized in the experiments (Xie et al. 2011, Zhang et al. 2015), while large-scale experiments were carried out in the underground layers to study the coupled THM/C processes in situ (Jing et al. 2016). On the other hand, different numerical codes have been developed, which are able to consider all the process together, such as OpenGeoSys (Kolditz

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et al. 2012b), DuMu^x (Flemisch et al. 2011), COMSOL Multiphysics (COMSOL 2012) etc. More attempts have been made through the coupling of two individual simulators which are well established in their own research fields, e.g. TOUGH-FLAC (Rutqvist et al. 2002), ECLIPSE-VISAGE (Khan et al. 2010), MoReS-PHREEQC (Tambach et al. 2013), OpenGeoSys-GEM (Kosakowski & Wanatabe 2014), etc. Based on the experimental results and analysis a bank of benchmarks of coupled THM/C processes have been set up (Kolditz et al. 2016). However, due to the complexity of the coupled processes as well as the new challenges from "Energiewende", much work is still to be done in the future.

The objective of this thesis therefore is the study of the coupled thermo-hydro-mechanical-chemical processes related to the geo-energy production using numerical simulations.

1.2 Thesis outline

In this thesis the numerical study of the coupled thermo-hydro-mechanical-chemical processes was based on TOUGH2MP and FLAC3D. The following contents have been integrated in this thesis.

Chapter 2 gives the fundamentals of the geosystem and geo-processes. The theoretical background to the coupled THM/C processes is introduced in this chapter. First the underground multiphase system is explained from a thermodynamic point of view. The important terms are the thermodynamic system, thermodynamic equilibrium, phase determination and phase change, properties of pure fluids as well as mixtures. Next the mass and heat transport processes are introduced, including the various mechanisms. Then the mechanics of intact rocks and fractured rock mass are described, including the stress and strain analysis, constitutive models, etc. After that the coupled process is explained in pairs for some basic models.

Three different areas related to geo-energy production were studied using a coupled THM/C simulation. Since different study areas had different focuses, the simulation concept and numerical tools had to be adjusted.

Chapter 3 covers the study of CO_2 enhanced gas recovery in the Altmark gas field. This study area focused on the gas displacement processes, pore pressure increase in the reservoir as well

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as the induced stress change, deformation and caprock integrity. It began with the development of numerical tools. In this chapter an improved equation of state for CO_2-CH_4-H_2O-NaCl system was first developed. The new EOS considered the salt precipitation, the effect of salt on gas solubility as well as other fluid properties, which had been implemented in TOUGH2MP. Verifications were made to both the experimental data and two simple examples. Next TOUGH2MP was linked with FLAC3D as the coupled simulator TOUGH2MP-FLAC3D. It was also verified with two simple examples. After that the in-situ stress and rock mechanical parameters were determined through both laboratory measurement and correlation study. A 3D model was generated in FLAC3D based on geological information and existing models, and coupled numerical simulations were carried out to investigate the various injection/production scenarios.

Chapter 4 discusses hydraulic fracturing in tight reservoirs. This study area focused mainly on the fracture propagation in tight formations, the fracture geometry (length, height and width), the induced stress change and rotation, distribution of the fracturing fluid in the formation etc. The study began by adjusting the linked simulator TOUGH2MP-FLAC3D. FLAC3D played the role of simulating the hydraulic fracturing propagation, while TOUGH2MP was modified to simulate the multiphase multicomponent leak-off processes. Then a generic model was generated for the study. Numerical simulations were carried out for different types of geo-reservoirs under comparable reservoir conditions and the same treatment schedule.

Chapter 5 covers the induced seismicity during the geothermal energy utilization in Unterhaching, the Greater Munich area. This study area focuses mainly on the temperature change in the underground, the induced stress change, especially the stress change of fault zones, fault stability, microseismicity etc. The study began with the development of a thermo-hydro-mechanical model based on crack tensor which could be used to simulate the behavior of faults. The model was verified with some simple examples and implemented in TOUGH2MP-FLAC3D. After that the in-situ stress and rock parameters were determined by both laboratory measurement and literature. A 3D model was generated based on the measured geological structure and stratigraphic information. Coupled numerical simulations were carried out to investigate both scenarios and measured production data.

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2 Fundamentals of a geosystem and geo-processes

2.1 Overview of a geosystem and geo-processes

The geosystem in this study mainly concerns the subsurface system, the outer part of the lithosphere, in which the solid rocks form the main supporting structure (Fig. 2.1). Rocks are primarily porous media, made of various mineral grains and cementations. Rocks without fractures are defined as intact rocks, while those with fractures are called a fractured rock mass. The fractures haven been formed due to tectonics, volcanics, weathering etc. Other structures include joints, cavities etc. Such voids and pores in the rocks are the main space for many other abiotic and biological substances in the subsurface. Abiotic substances include gases (steam, N_2, CO_2, CH_4 etc.) and liquids (brine), while biological substances encompass heavy hydrocarbons (oils) and animals, plants, and microorganisms which build an ecosystem. The subsurface contains both resources and energy, including precious metals, oil, gas, coal as well as heat from inside the earth. It continues to be the main energy source for human beings, especially oil and gas.

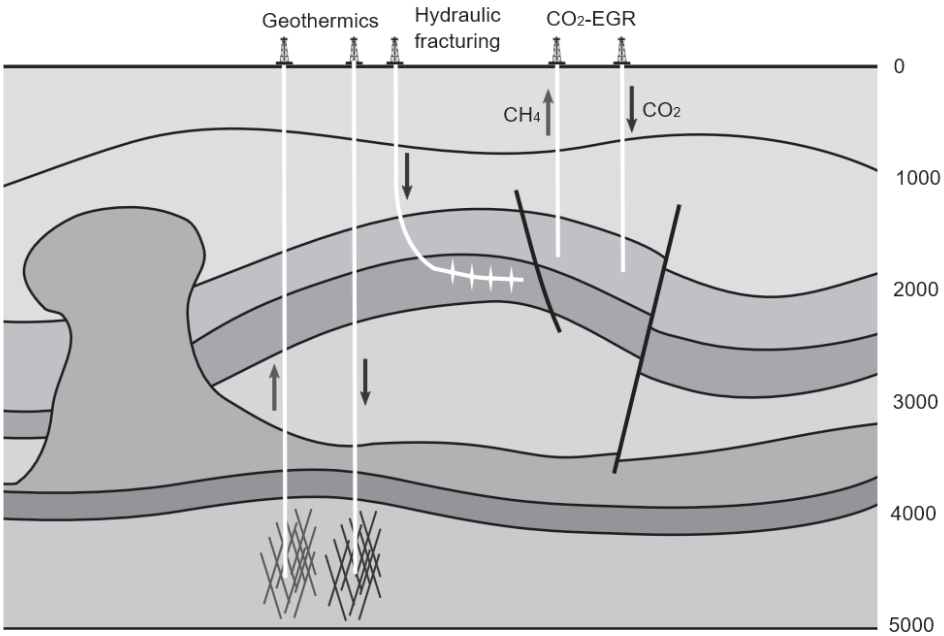

Figure 2.1 Geosystem for the energy production

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There are many processes occurring underground, including both natural and anthropogenic procedures, which can be divided into different categories, e.g. thermal (T), hydraulic (H), mechanical (M), chemical (C) and biological (B). Although the geosystem is normally treated as being stationary, it is not actually in equilibrium but moves at a very low velocity. This is a natural mechanical process (deformation). Underground gas and liquid will migrate under a pressure gradient, a natural hydraulic process. The earth's internal heat will be transferred through the lithosphere to the earth's surface by conduction and convection, which are natural thermal processes. While human activities lead to additional processes occurring underground. E.g. the production of hydrocarbons will deplete the reservoir, which causes fluid flow and deformation processes. The operation of a geothermal system will cause a fluid flow by injection and withdrawal, and temperature reduction due to heat exchange between solid rocks and circulated fluids. These processes are coupled with each other. A schematic of such coupling effects in the subsurface is shown in Fig. 2.2 and will be addressed in this chapter.

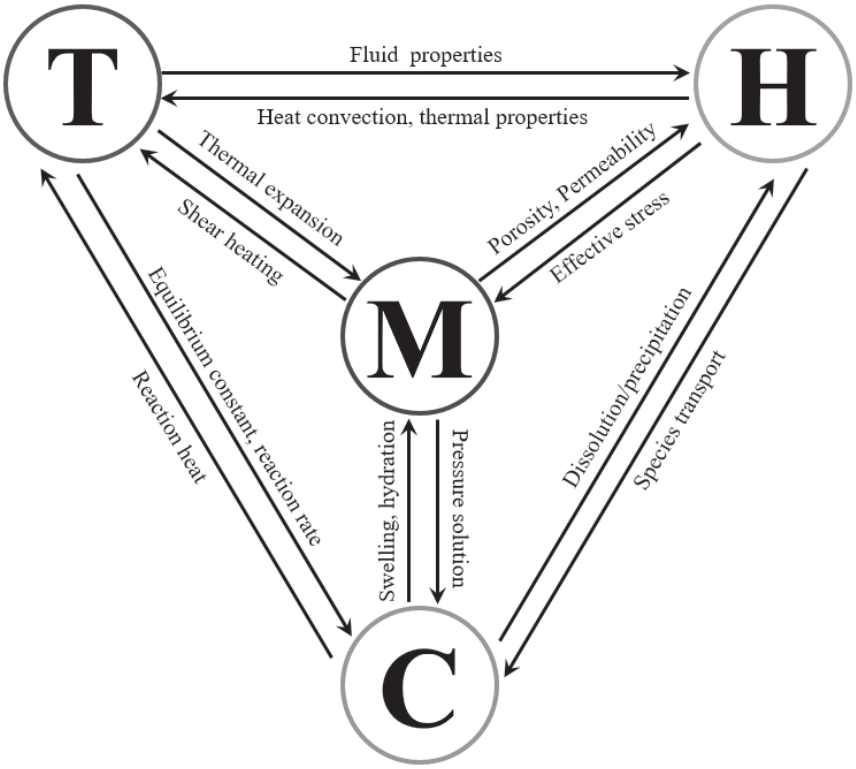

Figure 2.2 Overview of the coupled THM/C Processes in the subsurface

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2.2 Multiphase underground system

2.2.1 Basic concepts of thermodynamics

A system has different boundaries, through which it can exchange mass and energy with the surrounding area. Underground systems can be divided into the following four groups according to their interaction with the environments (Baehr & Kabelac 2016):

- Isolated systems: The boundary of this system does not allow any exchange of mass or energy. This type of system is meaningless to technology.
- Closed systems: The boundaries of such system are impermeable. There is no mass exchange. However, energy transfer can occur both through heat flow across the system boundaries and also through mechanical energy (work) (e.g. closed gas tank).
- Open systems: The boundaries of the system are permeable for matter and heat. Mass and heat exchange are enable (e.g. atmosphere).

Technically the subsurface is an open system, e.g. for geothermal energy utilization, since both natural and anthropogenic mass and energy exchange take place (Fig. 2.1). If the injection or production stops, the system can be treated as a closed system. During the investigation of the geo-processes the underground area was often divided into different volume elements. Such an open subsystem is called a control volume, and is considered to include not only the interior volume but also its mass and heat exchange with the environs.

A thermodynamic system can contain a single phase or multiple phases. It can also contain single component or multiple components. Two important terms that should be explained are:

- Phase (α): A Phase is defined as a quantity of matter that is homogeneous (Borgnakke & Sonntag 2009, Baehr & Kabelac 2016). The chemical and physical properties of a phase are locally constant and have only one value, regardless of how its state is reached. There are boundaries between two or more phases, through which their properties can suddenly change. A Phase can be composed of one pure substance or a mixture of various substances.
- Component (i): The number of components defines the minimum number of independent substances, which are necessary to describe the structure of a multiphase system (Helmig 1997).

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A single-phase system has only one phase but can contain two or more components, while a single-component system contains only one component, but can has two or even three phases. The underground is a multiphase system, including solid (rock), gas (air, vapor, gaseous hydrocarbons) as well as liquid phase (water, brine, oil). The gases and liquids are fluids and stored in the void volumes of the solid rocks, like pores, fractures and cavities. The porosity ϕ is defined as the pore volume V_{pore} divides the considered total volume of the rocks V_{tot},

$$\phi = \frac{V_{pore}}{V_{tot}} \tag{2.1}$$

The fluids inside the pores include gas, liquid or even oil. The saturation S^{α} describes the occupied volume in the pore by the phase V^{α} divides the total pore volume V_{pore}, namely:

$$S^{\alpha} = \frac{V^{\alpha}}{V_{pore}} \tag{2.2}$$

In each of the phases there may be various substances like nitrogen, water, etc. Their quantities are often described with their mass fractions x_i^{α} [-] (Borgnakke & Sonntag 2009),

$$x_i^{\alpha} = \frac{m_i^{\alpha}}{\sum_j m_j^{\alpha}} \tag{2.3}$$

where m_i^{α} is the mass of component i in phase α in [kg], or mole fractions y_i^{α} [-],

$$y_i^{\alpha} = \frac{n_i^{\alpha}}{\sum_j n_j^{\alpha}} \tag{2.4}$$

where n_i^{α} is the amount of substance for component i in phase α in [mol].

A thermodynamic system can be quantified by its state properties. There are two types of state variables (Baehr & Kabelac 2016):

- Extensive state variables

State variables are considered to be extensive, if the value of the whole system is equal to the sum of the state variables of the subsystems. Examples include volume and mass.

- Intensive state variables

State variables are considered to be intensive, if the values of the whole system cannot be calculated through adding the values of the subsystems. Temperature is an example.

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There are also special state variables, namely specific and molar state variables. The specific state variables are defined with regard to their mass, e.g. the specific volume, specific enthalpy etc. while molar state variables are defined with regard to the number of substances, e.g. molar volume, molar mass etc. These variables are all intensive state variables.

The state variable of a system stays constant, if the system is in equilibrium. In order to describe the geo-processes a system is normally considered to be in equilibrium. During the investigation, the subsystems (control volumes) of the underground are normally assumed to be in local equilibrium. Equilibrium will be discussed in the next section.

2.2.2 Thermodynamic equilibrium

In a multiphase system, thermodynamic equilibrium implies not only thermal equilibrium but also mechanical and chemical equilibrium (Helmig 1997, Baehr & Kabelac 2016).

Thermal equilibrium

A system in thermal equilibrium has the same temperature throughout: $T^{\alpha} = T^{\beta} = T$.

Mechanical equilibrium

Mechanical equilibrium means that the pressures on both sides of the phase boundaries are equal: $p^{\alpha} = p^{\beta} = p$.

However, a rock formation is a porous media. There are small void spaces connected to narrow throats and the capillary effect is significant. The capillary effect is induced by the molecular forces between two fluids and a solid phase, through which an angle is formed between the fluid interface and the solid surface (Helmig 1997). A phase which has contact angle < 90° is called a wetting phase (more adhesive to solid) and the other phase is called a non-wetting phase. The capillary pressure is then defined as the pressure difference between them, namely

$$p_{cap} = p^{w} - p^{nw} \tag{2.5}$$

The capillary pressure is dependent on the surface tension, the contact angle as well as the geometry of the capillary tube or pores in a microscopic scale. It is often related to the phase saturation with capillary function in the application. There are various types of capillary

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pressure functions, two types of which are very often used under two-phase conditions. Both use the effective saturation S_e^α as a variable

$$S_e^\alpha = \frac{S^\alpha - S_r^\alpha}{1 - \sum_{i=1}^{\varphi} S_r^i} \tag{2.6}$$

The first function is the van Genuchten capillary pressure function

$$p_{cap}(S^w) = \frac{1}{\alpha}\left(S_e^{-\frac{1}{m}} - 1\right)^{\frac{1}{n}} \tag{2.7}$$

where the parameters m ([-]), n ([-]), α ([Pa^{-1}]) are constants. The other is the Brooks-Corey capillary pressure function

$$p_{cap}(S^w) = p_d S_e^{-\frac{1}{\lambda}} \tag{2.8}$$

where the parameter λ ([-]) is constant and p_d ([Pa]) is the air entry pressure. Some example curves are shown in Fig. 2.3. In three phase cases the capillary effects are more complicated. Capillary pressure occurs between gas and oil, oil and water as well as gas and water phases. As an example of Parker's capillary pressure function is shown in Appendix A.

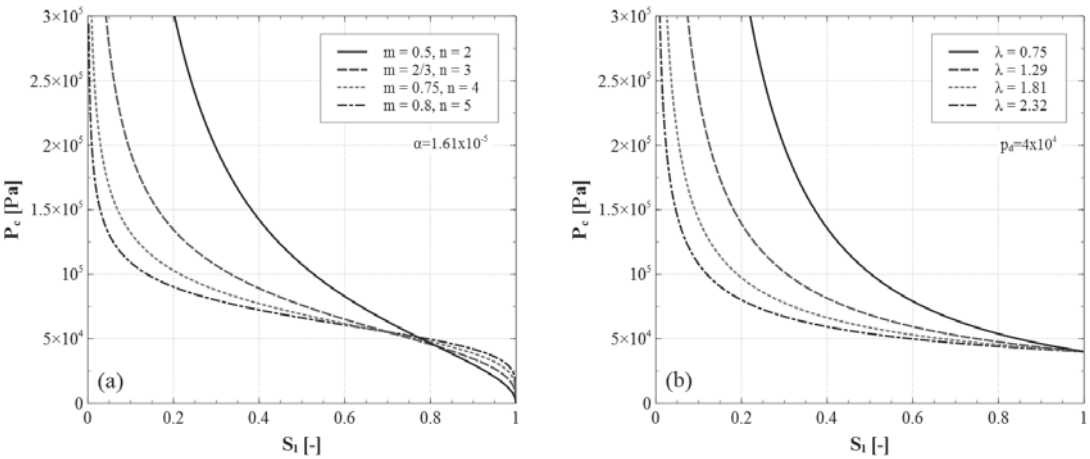

Figure 2.3 Capillary pressure function after (a) van Genuchten; (b) Brooks-Corey

<u>Chemical equilibrium</u>

Chemical equilibrium occurs when the chemical potential of each of the components in all the phases are equal: $\mu_i^\alpha = \mu_i^\beta$ (i = 1, 2, ..., N).

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