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Introduction

The motion of aqueous and magmatic liquids has a large influence on the evolution of the cooling and chemically differentiating Earth. In many geological environments, these fluids percolate through the pores and fractures of a solid matrix, which is composed of (un)consolidated rocks or solidified magma. The major driving forces within the liquid arise from the pressure, temperature and compositional variations present within the liquid, or from the density difference between liquid and solid states. Buoyancy driven flow (also called convection) in these porous or fractured media is capable to efficiently transport heat and chemical elements within and between geological environments. As a consequence, convection in porous or fractured media plays a significant role in, or even dominates, a wide variety of geological processes [Jaupart and Tait, 1995; Ingebritsen and Sanford, 1998].

In the oceanic crust, for instance, hydrothermal circulation accounts for about one third of the heat flux through the ocean floor or 25 percent of the global heat loss [Lowell *et al.*, 1995]. To dissipate this heat, estimated at 10^{13} W, the mass of the oceans is circulated through the crust approximately every one million years [Elderfield and Schultz, 1997]. These sub-seafloor hydrothermal systems are driven by heat loss from magma lenses beneath spreading centers and from already solidified basaltic crust. Advective fluxes are important for all the major ocean basins to seafloor ages of 65 ± 10 Myr [Stein and Stein, 1994]. Ninety percent of the oceanic heat flux occurs on the sedimented flanks of the spreading centers [Morton and Sleep, 1985]. It was also at these flanks that convective circulation was first invoked as an explanation for the missing heat in the measured heat flow values, as compared with those predicted by theoretical models of conductively cooling oceanic crust (see Figure 1.1, process A).

On the mid-ocean ridge axes, the direct observations of high temperature sulfide-rich fluid vents, so-called 'black smokers' plumes, have readily demonstrated the presence of active hydrothermal convection (process B in Figure 1.1). The temperature and geochemistry of the vent fluids indicate water-rock reactions at high temperature and pressure suggesting seawater penetration even within the gabbroic rocks, several kilometers below the seafloor [Seyfried, 1987]. During the frequent volcanic events below the ridges, the circulating seawater may pass into a supercritical mixture of a chemically diluted vapor and a dense brine [Bischoff and Rosenbauer, 1989]. Analysis of fluid inclusions found in obducted ophiolites [Nehlig, 1991] and measurements of the chemistry of vent fluids [Butterfield and Massoth, 1994; Von Damm *et al.*, 1997] support this hypothesis. While the vapor vents quickly through the seafloor, the salty brine may accumulate into a continuous layer at the base of the system. The evolution of

a convecting brine layer has implications for the thermal and chemical evolution of the ridge-crest system. Moreover, venting of the brine phase significantly reduces the productivity of most biological communities at the seafloor.

In the continental crust, cooling of plutonic regions occurs mainly by hydrothermal convection and this results in the formation of large mineral deposits (process C in Figure 1.1) [Norton and Knight, 1979; Furlong *et al.*, 1991; Barnes, 1997]. Oxygen isotope systematics and analysis of exhumed metamorphic terrains, geothermal fields, and of mesozonal batholithic rocks have indicated that fluids circulate down to depths of 5 to 15 kilometer from the surface [Valley *et al.*, 1986; Nesbitt and Muehlenbachs, 1991]. Major consequences of the flow are the deformation and chemical alteration of the rocks [Ferry and Gerdes, 1998]. Large-scale groundwater flow in and perhaps also beneath sedimentary basins influences a variety of geological processes, including anomalous heat flow and basin tectonics (see Figure 1.1, processes C and D) [Person *et al.*, 1996]. Moreover, the flow may lead to the dissipation of excess pore pressures built up within the basin, to hydrothermal ore genesis, sediment diagenesis, and to petroleum generation and migration.

The pressures in continental hydrothermal systems are generally lower, as compared to seafloor hydrothermal systems. Subcritical boiling is therefore a common feature in the continental areas and this leads to the generation of hypersaline brines at the base of these systems. In the Salton Sea Geothermal System, California, for example, a sharp salinity interface has been observed at a depth of about 1.5 kilometers [Williams, 1997]. This interface follows the isotherm of 260° C, rather than any structural or stratigraphic feature, and separates a relatively homogeneous liquid with up to 28 wt% total dissolved solids from the overlying fresh fluids. These observations speak very well for a layered convective system [Oldenburg and Pruess, 1998].

The solidification and melting of magmatic environments are also geological phenomena, which are influenced by convective flow in porous media. Since magmas are complex mixtures of several chemical components, the solidified outcrops of magmas inevitably have crossed that regime in which the magma was partially solidified. The layered structures commonly observed in these relicts may well have developed during the final stages of solidification, in which the system resembles a porous medium. While thermochemically driven convection of purely viscous fluids is proposed to be a common prominent layer forming mechanism in the oceans (for a review see [Schmitt, 1994]) and, later, in magma chambers [McBirney and Noyes, 1979; Spera *et al.*, 1986; Hansen and Yuen, 1995], it is by no means clear whether layers can develop in porous environments. Finally, flow through partially solidified boundaries of magmatic intrusions may also determine the dynamics of the totally melted region. An example is the convection driven by solidification of heavy components at the base of a liquid environment, such as the outer core or magmatic intrusions [Jaupart and Tait, 1995; Campbell, 1996]. The understanding of layer formation and other magmatic processes necessitates an improved knowledge of the stability and dynamics of thermochemical convection in porous media.

Information on the presence, dynamics and geological consequences of aqueous liquids within the crust is obtained first by drill-hole measurements in active systems, which extend up to almost 13 km depth in continental crust [Manning and Ingebritsen, 1999] and about one km in the oceanic basement [Fisher, 1998]. Permeability, which may be qualitatively described as the ease with which fluid can move through porous material, is one of the most critical parameters governing flow in natural porous environments. However, permeability is also one of the most difficult parameters to characterize due to problems of heterogeneity and scale. Moreover, preferred fracture or pore orientations within the rocks generally lead to anisotropy of the permeability field. Nonetheless, these in-situ measurements together

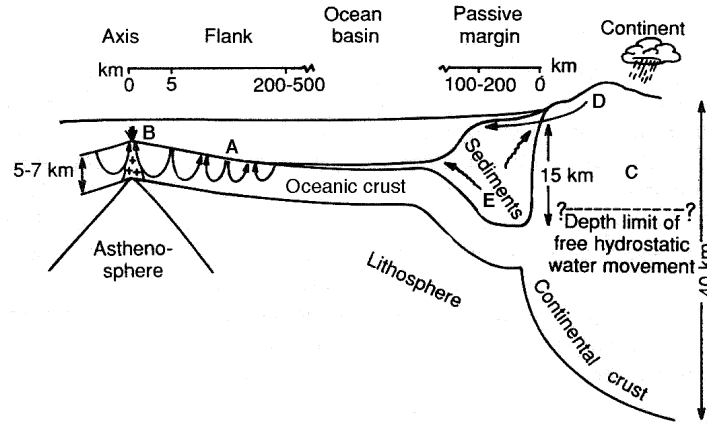


Figure 1.1: (a) Various processes of aqueous liquids within the earth's crust (modified from *Cathles* [1990]). A. Convection on the sedimented flanks of the oceanic ridge, B. convection at the mid-ocean axis spreading center, C. free convection within the continental crust, D. topographically driven flow, and E. compactive expulsion in passive margins.

with oxygen isotope systematics and analysis of exhumed metamorphic terrains, geothermal fields, mesozonal batholithic rocks and obducted ophiolites have indicated that the rocks are permeable up to large depths, and aqueous fluids may circulate even within the deepest areas. Finally, seismic and electrical conductivity measurements provide information on boundaries between hydrologically distinct areas and the averaged fluid/rock properties up to larger depths, respectively. Employing these techniques can only give a rough picture of the involved processes.

A complimentary approach to obtain fundamental insight into the nature of thermochemical convection in geological porous media is the mathematical analysis of the conservation equations of the system [*Phillips*, 1991; *Nield and Bejan*, 1992]. A major issue in these analyses is that solute, unlike heat, cannot diffuse through solid matrix. As a result, temperature perturbations advect more slowly than salinity fluctuations by the factor ϕ , but diffuse more rapidly by the factor κ/D and so are smoothed out more efficiently. Here, ϕ is the void space or porosity of the medium, while κ and D denote the thermal and chemical molecular diffusivity, respectively. Double-advective, double-diffusive instabilities play a significant role in the transport of heat and solute in geological environments, where porosities are typically very low.

A third approach is performing fluid dynamical experiments, either by laboratory experiments or by numerical simulation. The laboratory experiments can be divided into two categories. First, two closely spaced glass plates are considered, in which the small gap between the plates is filled with liquid. In these so-called Hele-Shaw cells, the viscous drag of the plates on the the fluid can be described with Darcy's law. As a result, experiments of this kind give insight in the stability criteria and flow processes involved in porous media [*Cooper et al*, 1997; *Wooding et al*, 1997ab]. Major differences with flow in natural porous media are, first, the flow is irrevocably two-dimensional, secondly, heat and solute advect with the same velocity and, finally, dispersive effects are lacking.

In a second category, a three-dimensional box is filled with glass beads and saturated with liquid. The porosity of the medium depends on the size of the beads and is typically between

10 and 70 volume percents. In the context of thermochemical convection, these experiments have primarily shed light on the onset of convection and the three-dimensional flow patterns [Murray and Chen, 1989]. Moreover, it was shown that a thin density interface between two convective layers of different composition can be maintained against diffusive thickening [Griffiths, 1981], which is essential for the persistence of these layers.

In this thesis, a numerical approach has been chosen to study thermochemically driven convective flow in porous media. The investigations described here are devoted to the study of (1) the formation and evolution of layered structures from an initially non-layered state, and (2) the stability and dynamics of thermochemical convection in geological porous media. Focus was both on the fundamental characteristics of this type of flow and on some hydrogeological applications.

Modeling convective fluid dynamics in porous media involves solving time-dependent, strongly coupled, nonlinear partial differential equations of the flow behavior through heterogeneous and anisotropic media. Although being limited by a macroscopic mathematical description of flow through a porous medium and by the available computer power, numerical experiments help us to distinguish between different geological processes or to unravel them. Numerical simulations unlike laboratory experiments enable the researcher to study quantitatively the flow through anisotropic and heterogeneous permeability fields. Moreover, fundamental aspects of flow through natural porous media, such as double-advective instabilities and mechanical dispersion, can be studied numerically on scales which may never be reached in the laboratory. Finally, numerical experiments allow for accurate monitoring of the physical properties of the fluid, like its temperature and chemical concentration, and the transport properties.

1.1 Outline of this thesis

This thesis is organized as follows. **Chapter 2** presents the basic equations for flow in porous media, along with a discussion of the assumptions made in this dissertation. Moreover, the system of partial differential equations describing thermochemically driven flow in porous media is given. In order to solve this system of equations, a numerical method has been developed which is based on the control volume theory. The characteristics and accuracy of this method are discussed in **Chapter 3**. The methodology has been applied to investigate two research topics in the subsequent chapters. First, flow in porous media has been investigated with respect to its potential to being a layer-forming mechanism in Hele-Shaw cells (**Chapter 4**) and in a medium of low porosity (**Chapter 5**). The results have been used in an applied study of the evolution of a brine-saturated fluid layer at the base of mid-ocean ridge hydrothermal systems (**Chapter 6**). The second topic, which has been addressed in this thesis, is that of the stability and dynamics of thermochemical convection in geological media of low porosity. It is shown that double-advective instabilities give rise to an almost intrinsically unsteady behavior of the flow (**Chapter 7**). In **Chapter 8**, finally, the criteria for the onset of thermochemical convection in heterogeneous and anisotropic media are investigated. Furthermore, the thermal, chemical and subsidence effects of free thermochemical convection in and beneath intracratonic sedimentary basins are discussed from simulations of a large-scale hydrogeological study of the Michigan basin.