

Summary

This thesis describes the study of buoyancy driven flow of fluid in rigid porous media, in which the density differences arise from thermal and chemical variations in the fluid. The fundamental characteristics of this type of flow, which is also called thermochemical convection, and some hydrogeological applications were studied by means of numerical simulation. In **Chapter 1**, the geophysical applications of this study are introduced, along with a discussion of the current scientific methods.

The experiments described in this thesis were performed with a two-dimensional second order accurate numerical method, which is based on the control volume theory. In this method, the conservation equations of mass, momentum, energy, and species of the fluid in a porous medium are discretized in a conservative way. A multigrid iterative method was employed to solve the discretized equations line-by-line. As a result, the computation time needed for one iteration scales linearly with the number of cells. The basic equations and the discretization methodology are described in **Chapters 2** and **3**, respectively. The methodology has been applied to investigate two research topics: (1) the formation and evolution of layered structures within the liquid, and (2) the stability and dynamics of free thermochemical convection in geological low-porosity media.

The formation and evolution of layered structures in porous media

Layering is perhaps the most common feature among all geological structures, on almost every scale. These structures vary from relatively small scales, like in magmatic intrusions, to the global scale structures of planetary interiors. Beyond geological settings, the formation and evolution of chemically distinct layers plays a significant role in the oceans and in the evolution of hydrothermal systems. At present, it is unclear if and in which manner layers can be generated in porous or partially solidified systems.

Therefore, first the basic processes through which layers can form and evolve from an initially non-layered state were investigated (**Chapter 4**). A numerical model of a Hele-Shaw cell was used, in which a compositionally stably stratified liquid was heated from below. In this Hele-Shaw approximation, both heat and solute advect with the same velocity. The most important processes in the layer formation are (a) the growth of a convective layer at the bottom of the domain through convective entrainment of fluid from above (b) the formation of a horizontal density interface on top of this layer and (c) the destabilization of the next layer above the interface. These processes appeared to be intimately linked. The growth of the first convective layer stops once the thermal equilibrium is reached. Since heat (solute) transfer across the thin interfaces becomes purely diffusive (dispersive), the separately convecting layers can persist on a compositionally dispersive time scale. While the number of layers that develop is determined by the magnitude of the thermal Rayleigh number, the final height of

a newly formed layer is inversely proportional to the ratio between the chemical and thermal components of the buoyancy. Besides the ultimate disappearance of any interface through diffusional homogenization, the experiments have shown that (1) the vertical migration and (2) the breakdown of an interface can also lead to sudden changes of the layer structure. On a time scale short as compared to the dispersive one, interface migration and breakdown are the key mechanisms determining the layered structure, rather than the initial evolution toward the thermal equilibrium.

Since solute unlike heat cannot diffuse through solid matrix, temperature perturbations advect more slowly than salinity fluctuations but diffuse more rapidly. Especially in geologically relevant media of low porosity, this may lead to double-advective, double-diffusive instabilities. In **Chapter 5**, therefore, the layer-forming mechanism was studied in a medium with a porosity of ten volume percents. The major features of the layer formation appeared to be similar to those observed in Hele-Shaw cells. By monitoring the heat (solute) fluxes, however, it was observed that the transport of heat (solute) across the interface has become purely diffusive (dispersive) before the moment that the lower layer arrives at the thermal equilibrium. It was argued that the layer growth stops when the density interface on top of the layer has grown sufficiently strong to keep the ascending plumes in the lower layer from convectively entraining fluid from above. A simple balance between the most important forces, exerted on a fluid parcel in the lower convective layer, was proposed as to determine this transition. The proposed force balance serves as indicator whether a density interface keeps intact, migrates upwards or breaks down during the further evolution of the layered sequence. The porosity of the medium and the buoyancy ratio are the most important parameters in determining the stability of the interfaces. Finally, the sensitivity of the layer-forming mechanism to a mixing process called mechanical dispersion was investigated. Mechanical dispersion is due to obstructions and the fact that all pores may not be accessible to a fluid element after it has entered a particular flow path. An increase of the dispersion lengths of the medium leads to an increase of the solute transport across the density interface. As a result, the initial layer thickness increases.

The evolution of layered structures may be particularly important in the formation of new the formation of new basaltic crust at oceanic spreading centers. Seawater penetrates the crust to a depth of several kilometers, transporting heat and chemicals from the magmatic intrusion and the newly formed basalts to the seafloor. Direct chlorinity measurements of fluids ventilating through the seafloor are indicative of mixing of hydrothermal seawater with fluids that have undergone supercritical phase separation. While a chemically diluted vapor phase vents in condensed form through the seafloor shortly after a magmatic event, a brine phase may generate a continuous layer within the lowermost part of system. The evolution of this brine-saturated layer plays a significant role in the thermal evolution of the hydrothermal system. Moreover, brines alter the chemistry of the basaltic rocks and lead to variations in the vent chemistry. As a first approach to study this complex process, numerical simulations were performed to study the evolution of the brine layer once it has returned into the subcritical regime (**Chapter 6**).

For realistic geological parameters, the interface between the brine and the overlying liquid is not very stable, but vanishes by the dynamical mechanisms mentioned before: convective breakdown or vertical migration. This contradicts the conventional picture of a steadily layered convective system, in which the brine is depleted purely by diffusion/dispersion across the interface. The type of mechanism involved with the brine depletion depends critically on the dynamical behavior of the brine layer. Convection of the brine layer results either in the convective breakdown (when the brine salinity is low) or the upward migration of the interface (for higher brine salinities). The upward migration process leads to a catastrophic

heat and mass release through the seafloor. However, the vent chemistry is not in agreement with the chemical signature of so-called megaplumes, huge warm-water bodies observed up to one kilometer above the seafloor just after a magmatic event.

In case the brine layer is static, on the other hand, high-chlorinity liquid is entrained slowly by the convection currents in the overlying fluid. This results in a downward migration of the interface. The thermal and chemical signatures of the vent fluids is consistent with those observed in chronic high-salinity seafloor vents. Since these vents may also be the result of present-day phase separation, however, this agreement should be verified with a two-phase numerical method.

The stability and dynamics of thermochemical convection in low-porosity media

Convective flow of aqueous liquids in the upper crust is driven by both thermal and chemical buoyancy; the former in response to the geothermal gradient and the latter due to differences in salinity that appear to be ubiquitous. **Chapter 7** considers the stability and dynamics of the double-advective double-diffusive instabilities, which may develop in geological media with a porosity of ten volume percents or less. In the experiments, an initially cold and chemically depleted porous medium was heated from below, while solute provided a stabilizing influence. For typical geological parameters, the thermochemically driven flow appears intrinsically unsteady. The chaotic dynamical behavior of the flow is due to a dominance of advective and dispersive chemical transfer over the more moderate convective heat transfer that actually drives the flow. Fast upward advective transport and lateral mixing of solute leads to the spontaneous formation of horizontal chemical barriers at depth. These gravitationally stable interfaces divide the domain in several layers of distinct composition and lead to significantly reduced heat flow for thousands of years. The intrinsic unsteadiness of thermochemical convection in low-porosity media have implications for heat transport in both oceanic and continental hydrothermal systems, for ore genesis, for metasomatism and metamorphic petrology, and the diagenetic history of sediments in subsiding basins.

Heat transport plays an important role in the thermomechanical evolution of intracratonic sedimentary basins. Observations of an episodic subsidence history and elevated levels of organic maturity of/in the Michigan intracratonic basin cannot be explained by simple conductive models of a cooling crust. In **Chapter 8**, it was investigated whether free thermochemical convection within a fractured igneous rift body beneath the sediments provides an explanation for these deviations.

First, it was investigated under which circumstances convection initiates from the geologically relevant situation in which both temperature and salinity of the liquid increase linearly with depth. From linear stability analysis is known that, under these conditions, the fluid is almost unconditionally stable against infinitesimal perturbations in homogeneous, isotropic media. However, crustal rocks are typically characterized by a heterogeneous and anisotropic permeability field. By means of simulations with simplified models of the upper crustal rocks, with a stochastic representation of the permeability field and by introducing anisotropy on the subgrid scale, it was shown that the stringent stability conditions become slightly reduced in such media. The most critical parameters, which determine the onset of convection are the permeability of the (fractured) rocks underlying the sediments, and the initial salinity distribution in the fluid.

The results of these simple simulations were used in a model study of the hydrogeological evolution of the Michigan basin. From the results of these large-scale simulations, it was observed that the temperatures in the sediments increase substantially during periods of ther-

mochemical convection within the fractured igneous body underneath. Therefore, it explains the high organic maturity observed in the sediments. Furthermore, the convective heat transport leads to an excessive subsidence of approximately 100 meters per five million years, as compared to subsidence due to purely conductive cooling. As a result, the flow explains up to a quarter of the fast subsidence rates, as obtained by backstripping well data of the Michigan basin. Finally, it was postulated that hydrological interaction between the igneous rift body and sediments may have altered the chemistry of the sedimentary pore fluids. This provides an explanation for the formation of Ca-rich, MgSO₄-poor evaporites after closure of the basin from the open sea.