

Coupled fluid flow with calc-silicate reactions around plutons: Results of numerical simulations

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Mineral assemblages in contact aureoles are the products of the interplay between heat and fluid flow that are induced by intrusion of magma into the upper crust. Metamorphism of calcareous shales causes metamorphic reactions that produce CO₂, which then becomes part of the hydrodynamic system. Flow of reactive H₂O-CO₂ fluids and progress of metamorphic reactions in an aureole of a granite pluton were simulated using a modified version of the US Geological Survey SUTRA code that allows calculation of metamorphic reactions in a transient P - T - XCO_2^{fluid} field. Several permeability structures of the aureole were considered, including: a) homogeneous, b) layered and c) reaction-enhanced. Reactions were assumed to occur only in horizontal calcareous shale beds. Magmatic, metamorphic, and sedimentary fluid sources were considered. The rates of reactions were assumed to be related to their ΔG 's at the prevailing P - T - XCO_2^{fluid} conditions at each node, an Arrhenian rate constant, and grain size.

Results show that after magma intrusion, overall fluid flow is away from the intrusion as the hydrostatic pressures in the inner aureole rise with temperature increase. Fluid composition in the inner aureole evolves rapidly toward high XCO_2^{fluid} as metamorphic reactions initiate before water begins to exsolve out of the pluton. Only after tremolite and diopside-forming reactions come to completion and local fluid pressures drop, infiltration of H₂O from the pluton becomes significant and can drive production of minerals such as wollastonite and vesuvianite. Further away from the intrusion, fluid composition initially reflects the CO₂ production in the inner aureole, and only later it is influenced by local reactions.

In the case of a homogeneous permeability structure of the aureole, fluid flux and composition are dispersed, while in a layered permeability structure, the highest fluid fluxes are confined to high permeability layers and fluid compositions remain discrete. When permeability increase is related to reaction progress, the highest fluid flux is confined to the inner aureole. The reaction-enhanced permeability model is in agreement with field and geochemical observations that suggest confinement of reactive fluid flow largely to inner calc-silicate contact aureoles.

Fate of ¹⁴C organic compounds released from sub-surface low-level radioactive waste repository

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¹⁴C is a major radionuclide of concern in the sub-surface low-level radioactive waste disposal due to its long half-life (5730 years) and relatively high mobility in an aquifer. It is mainly released from the disposal site in the form of low-molecular-weight organic compounds such as acetate, acetaldehyde and ethanol that are generated abiogenically by Fischer-Tropsch reactions.

These low-molecular-weight organic compounds are degraded by anaerobic microorganisms in many subsurface environments and are therefore considered to be easily metabolized by indigenous microorganisms that alter the migration behavior of carbon-14 in an aquifer.

In the laboratory experiments, however, acetate was not anaerobically degraded by microorganisms indigenous to a pumice-tuff rock, even after incubation for at least 3 months; however, it was aerobically degraded to bicarbonate within a short period, i.e., 7 days. 16S rDNA analysis of the rock revealed clones related to the sulfate reducer, *Desulfovibrio* sp. and methanogens, *Methanosarcina* sp. and *Methanogenium frigidum*, indicating the possibilities of anaerobic microbial degradation of acetate in the subsurface environments. However, when growth media specific for sulfate reducers and methanogens were used, there was no evidence of anaerobic microbial degradation of acetate.

Because of the differences in the experimental conditions between the laboratory and aquifer environments, more definitive tracer test using ¹³C-labeled acetate is now used to analyze its bioavailability at a site in Rokkasyo, Japan.