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Reactive transport modeling: An essential tool and a new research approach for the Earth sciences

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Abstract

Reactive transport modeling is an essential tool for the analysis of coupled physical, chemical, and biological processes in Earth systems, and has additional potential to better integrate the results from focused fundamental research on Earth materials. Appropriately designed models can describe the interactions of competing processes at a range of spatial and time scales, and hence are critical for connecting the advancing capabilities for materials characterization at the atomic scale with the macroscopic behavior of complex Earth systems. Reactive transport modeling has had a significant impact on the treatment of contaminant retardation in the subsurface, the description of elemental and nutrient fluxes between major Earth reservoirs, and in the treatment of deep Earth processes such as metamorphism and magma transport. Active topics of research include the development of pore scale and hybrid, or multiple continua, models to capture the scale dependence of coupled reactive transport processes. Frontier research questions, that are only now being addressed, include the effects of chemical microenvironments, coupled thermal–mechanical–chemical processes, controls on mineral–fluid reaction rates in natural media, and scaling of reactive transport processes from the microscopic to pore to field scale.

Keywords: reactive transport; water–rock interaction; contaminant transport; magma transport; coupled processes

1. Introduction

In recent years, the Earth sciences have benefited from the strong interest in fundamental research that focuses on individual features and mechanisms considered to play a critical role in the overall system behavior (e.g., mineral or bacterial surfaces, the morphology of nanoparticles, or the rate of a process as a function of a single environmental variable). While such an approach is absolutely crucial in advancing our fundamental scientific understanding of natural processes, there is also a need to take a broader view of integrated system behavior. Often thought of as an engineering concept, system integration also has a role in scientific investigations of complex natural systems where individual time and space-dependent processes are linked and where the relative importance of individual sub-processes cannot be fully assessed without considering them in the context of the other dynamic processes at work. It can be argued that the complex interplay of

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material flow, transport, and reactions at multiple spatial and time scales which characterize most Earth systems requires an integrated approach.

A key task in developing an integrative scientific approach is to build capabilities for computer simulation of complex and often large-scale natural systems and then to link these to laboratory and field studies. In the Earth sciences, the complex natural systems that are accessible to direct study include soils, groundwater reservoirs, volcanoes, the ocean floor, faults, mountain ranges, oceans, glaciers, the atmosphere, and ecosystems. Deeper in the Earth are the mantle and core, and within them dynamical features like subduction zones and mantle plumes. These systems are characterized by mass transport processes such as diffusion and flow of gases, fluids, and solids, and within the material flow and transport system there are chemical, mechanical, and/or biological interactions. In many cases, the transport, chemical, mechanical, and biological processes in these Earth systems are coupled—for example, the metabolic activity of a biofilm at the pore scale may depend on some combination of advective and diffusive transport combined with local biogeochemical reactions providing electron donors and acceptors. These processes may then result in changes in the physical properties of the medium through biological growth and/or mineral precipitation or dissolution, providing a feedback between flow, transport, and reaction (Fig. 1).

What is referred to as "reactive transport modeling" is then an important set of interpretive tools for unraveling complex interactions between coupled processes and the effects of multiple space and time scales in the Earth. But reactive transport modeling can also be viewed as a research approach, a way of organizing

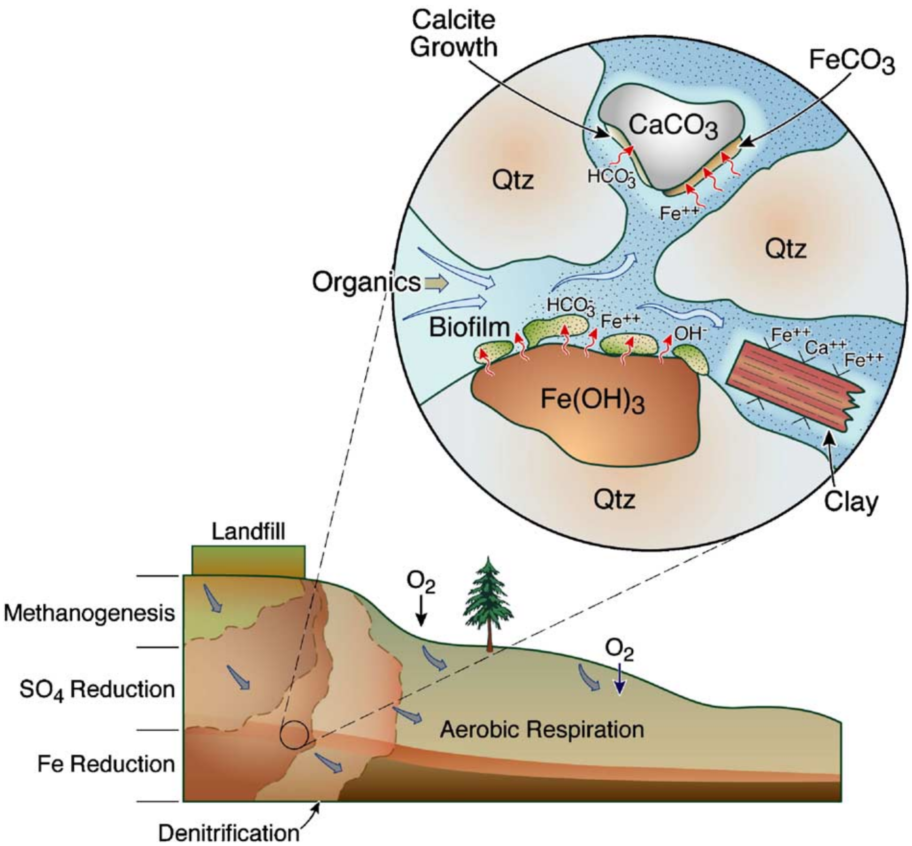

Fig. 1. Schematic representation of the oxidation–reduction zones that may develop in an aquifer downstream from an organic-rich landfill. Closest to the landfill may be a zone of methanogenesis, which is progressively followed downstream by zones of sulfate reduction, dissimilatory iron reduction, denitrification, and aerobic respiration that develop as the plume becomes progressively oxidized through the influx of oxygenated water. Within the dissimilatory iron reduction zone, a pore scale image is shown in which the influx of dissolved organics provides electrons for dissimilatory iron reduction mediated by a biofilm. The dissolution of this phase leads to the release of Fe^{2+}, HCO_3^-, and OH^- into the pore fluid, potentially driving siderite or calcite precipitation downstream, and thus reducing the porosity and permeability of the material. Sorption of Fe^{2+} may also occur on clays, displacing other cations originally present on the mineral surface. Where reactions are fast relative to local transport, gradients in concentration, and thus in reaction rates, may develop at the pore scale.