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and W.W-L. Lee,

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Mass Transfer and Transport in Geologic Repositories:

Analytical Studies and Applications

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Assessing the long-term performance of geologic repositories for radioactive waste requires reliable quantitative predictions of rates of release of radionuclides from the waste into the rock, transport through the geologic media, cumulative release to the accessible environment, and maximum concentrations in ground water and surface water. Here we review theoretical approaches to making these predictions and issues that require resolution.

Far-Field Transport

Predicting the hydrogeologic transport of dissolved radionuclides to the environment is a problem of "far-field" transport. Most radionuclides migrate as single decaying species, but the transport of the toxic radionuclides Ra-226 and Pb-210 is controlled by the simultaneous transport of their precursors U-234 and Th-230. Exact analytical solutions have been developed for the transport of radioactive decay chains, with and without dispersion, in a steady one-dimensional flow field in a porous medium of infinite or finite extent^{1,2}. Some of these equations were used to predict the benchmark results for the Intracoil-1 calculations³ and to predict environmental releases for model repositories in basalt, salt, and tuff for the Waste Isolation System Study⁴. The analyses have been extended to predict dispersion-free transport in a steady two-dimensional flow field and transport in a one-dimensional flow field with a spatially dependent dispersion coefficient^{5,6,7}. Analytical solutions have been developed to predict transport in fractured media with diffusion into and out of the rock matrix, including the effect of parallel multiple fractures^{8,9}. Many of these equations for porous media have now been programmed into computer codes by repository projects. The fracture-flow analyses will be useful in making more detailed calculations of radionuclide

migration to the environment.

Near-Field Transport

"Near-field" transport is concerned with the analysis of radionuclide mass-transfer from waste solids into ground water and into surrounding rock. This provides the source term for far-field calculations. Mass-transfer analysis in the U.S. programs was stimulated when the National Research Council's Waste Isolation System Study⁴ questioned the assumptions that long-term dissolution rates in a geologic repository could be predicted from laboratory data on leach rates of waste samples. The actual processes of dissolution are reaction of the waste solid with ground water at the waste surface and diffusion and convection of the dissolved species from the surface into ground water in surrounding rock. Recognizing that the waste-solid matrix and most of its solid constituents are of low solubility, we first sought an upper limit on the dissolution rate by assuming that the solution at the waste surface is saturated with individual waste constituents.

Exact analytical solutions^{10,11} for the time-dependent mass transfer rate from a waste solid in contact with saturated porous rock show that the limiting mass-transfer rates for the buried waste are so slow that the actual dissolution rate is expected to be limited by diffusive-convective transport in pores in the rock and not by chemical reaction at the waste surface. For the low ground-water flows expected in U.S. repository programs the near-field mass transfer from waste in contact with rock is expected to be controlled by molecular diffusion in the surrounding media and little affected by convection. A more detailed analysis¹² used experimental chemical reaction rate data as a boundary condition for the diffusion mass-transfer analysis and showed the conditions and time span during which chemical reaction rate could control the dissolution rate. This time span is so short as to be unimportant for borosilicate glass, and this seems likely to be true also for spent fuel. These predictions were confirmed by a more recent general analysis¹³ of steady-state diffusive-convective mass transfer from a waste sphere that covers the entire velocity range of ground water flow. The near-field analytical solutions have been extended also to include the effect of a backfill layer between waste solid and rock^{14,15,16} and the mass-transfer of the highly soluble cesium and iodine "gap" activity in spent fuel¹⁷. These basic mass-transfer equations and extensions have been incorporated into PNL's AREST code¹⁸ for predicting waste-form performance.

Further extensions include analytical solutions for the effect of repository heating on mass transfer of both low-solubility and soluble species¹⁹, the time-dependent diffusive release of radionuclide chains through backfill into rock²⁰, the time-dependent diffusion of low-solubility species through

backfill into a rock fracture²¹, and the time-dependent diffusive release of low-solubility species into porous rock matrix and rock fractures²². The near-field analyses are applicable to all repository projects, including a repository in unsaturated tuff wherein sediments or moist rock contacts waste packages. Studies underway include the effect of water flow in backfill and the effects of nonuniform chemical environment that can cause precipitation of dissolved species in the porous medium away from the waste surface.

Our studies of radionuclide releases from waste solids in a salt repository led to the expectation that, except for a short time after waste emplacement, releases will be controlled by diffusive-convective mass transfer into brine in grain boundaries of the consolidated salt surrounding a waste package²³. Our analyses of the time-dependent migration of grain-boundary brine after consolidation²⁴ can be used to predict the transient convective effects in salt, which may be so small as to be negligible compared to mass transfer by molecular diffusion. Examples of diffusion calculations of release rates into salt have been given²⁵. The solution describing the transient release to porous and fractured rock can be applied to non-halite interbeds in a salt repository.

Intermediate Field-Transport

"Intermediate-field" transport concerns radionuclide migration from arrays of discrete waste packages. Our analytical solutions^{6,26,27}, of the multidimensional advective transport from waste-package arrays show a near region in which the concentrations vary greatly in the direction transverse to ground-water flow, an intermediate region in which the array can be treated as an infinite plane source of dissolving species, and a far-field region in which the array can be treated as a plane source of finite extent. The array equations have been developed for both porous and fractured media. These intermediate-field analyses will be useful to repository projects in making detailed predictions of releases to the environment.

Summary

An extensive body of analytical theory exists to predict the isolation performance of geologic repositories. Special features of analytic solutions for this application are:

1. They identify the functional forms and importance of the design variables and parameters.
2. Their theoretical framework can be examined and tested, essential to developing valid predictions of long-term performance.

3. Their generality provides flexibility for multi-variable system optimization.

4. They can be used to benchmark nonanalytic numerical calculations.

Only a limited number of these analytical tools have yet been utilized by repository projects.

Further analyses will address details such as fracture networks, release of gaseous species, nonuniformities in chemical environment, layered media, unsaturated media, precipitation away from waste surfaces, and transition from near-field to far-field transport.

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