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June 11, 1984

Everett A. Wick
High Level Waste
Licensing Management Branch
Division of Waste Management
Mail Stop SS 965
U. S. Nuclear Regulatory Commission
Washington, DC 20555

Dear Mr. Wick:

Enclosed please find copies of the abstract and "review summary" of the paper we would like to submit for presentation at the MRS meeting in Boston this fall.

The theme of the paper is that the sorption capacity of the packing materials is a controlling factor in the transport of radionuclides out of the waste package and the engineered barriers. Two points will be made: (a) the presently-used constant K_d approach (e.g., the WAPPA code) assumes that the packing materials never exhaust their ability to sorb radionuclides. Therefore, release estimates based on this approach are non-conservative; (b) in the future, if the constant K_d approach is abandoned in favor of a concentration-dependent K_d, i.e., if the whole isotherm will be used, sorption experiments should be done to take sorption capacity into account. At present, the DOE does not expressly scope for the sorption capacity of the packing materials. Typically, the experiments are stopped before the medium fills to capacity. Therefore, the resulting isotherm is incomplete and unsuitable for modeling.

We have already done some preliminary analysis of points (a) and (b) above. We would need approximately one extra week of Terry Sullivan's and my own time to complete these analyses. In addition, I foresee a limited amount of computer usage, about 500 dollars.

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The issue under consideration and its future developments will be incorporated in the EA review which is presently being funded under FIN-3164.

I would appreciate your reviewing the enclosed documents promptly, as the deadline for "review summary" submission is June 15.

Thank you for your attention.

Regards,

Claudio Pescatore

Claudio Pescatore, PhD.

CP:ep
enc.
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SORPTION-CAPACITY LIMITED RETARDATION OF RADIONUCLIDES TRANSPORT IN WATER-SATURATED PACKING MATERIALS. C. Pescatore and T. M. Sullivan, Brookhaven National Laboratory, Building 703, Upton, NY 11973

Radionuclides breakthrough times and fluxes as they are presently calculated through constant retardation factors are non-conservative. The constant retardation approach regards the packing materials as having infinite sorption capacity at all sections. On the contrary, as the packing materials become locally saturated, such as in the proximity of the waste form-packing materials interface, they will exhibit no retardation properties, and transport will take place as if the radionuclides were locally non-reactive. The magnitude of the effect of finite sorption capacity for the packing materials on radionuclide transport is discussed with reference to high-level waste package performance. Examples are given based on literature sorption data and isotherms.

Sorption-Capacity Limited Retardation of Radionuclides Transport in Water-Saturated Packing Materials

by

C. Pescatore and T. M. Sullivan

Brookhaven National Laboratory

Typical partition functions for the uptake of solute by a water saturated porous solid are of the form displayed in Figure 1. Namely, two regions can be clearly identified: one region where the amount on the solid varies linearly with concentration in solution, and a second region where the curve bends over as the amount on the solid nears its limiting value S_{max}. The constant K_d approach is justified only in the first region; in the second region the full expression for the retardation factor, R, should be used, that is:

$$R = 1 + \frac{\rho_b}{n} \frac{dS}{dC}. \qquad (1)$$

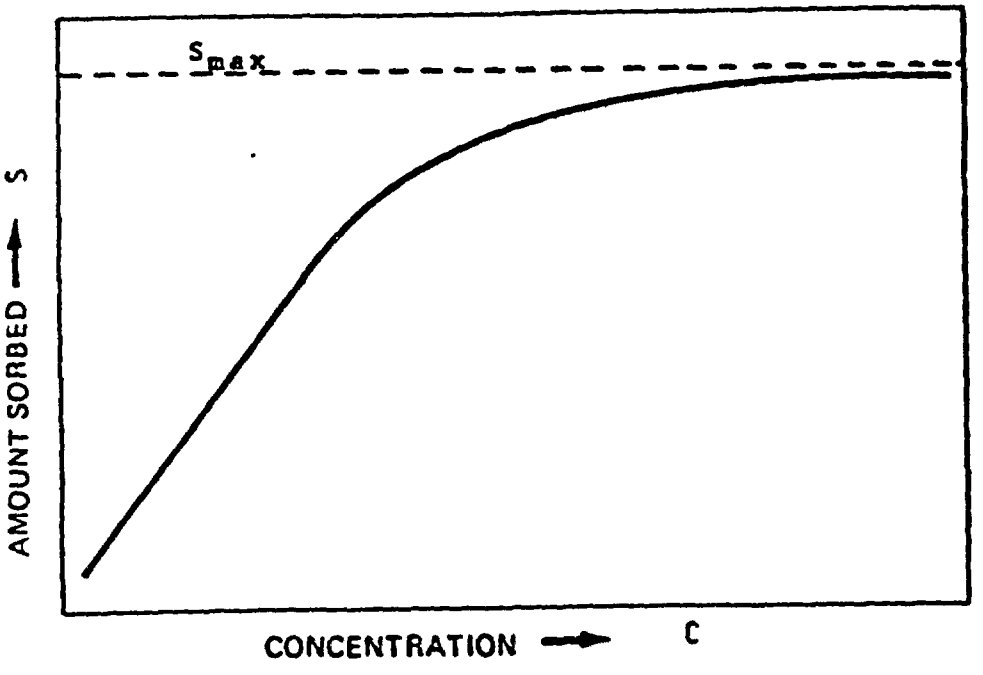

Figure 1. Typical partition of a solute between a porous solid and water (adapted from Ref.[1]).

Indeed, as Equation (1) shows, in the limit as $\frac{dS}{dC}$ tends to zero the retardation factor becomes smaller and smaller, i.e., when the porous medium is close to saturation, little or no uptake of solute is accomplished and transport occurs as if the solute were locally non-reactive.

It should be appreciated that if the partitioning of the solute between the solid and the liquid is an equilibrium process, the value S_{max} will be associated with solution concentrations below solubility. Thus, as long as the equilibrium condition is warranted, if one assumes a solubility concentration at the waste form-packing materials interface, one must also assume that the packing materials are saturated locally and that transport is locally unretarded.

The above considerations have a direct bearing on waste package performance. By taking into account the limited sorption capacity of the solid, we show that breakthrough times and radionuclide fluxes are respectively smaller and larger than the same quantities calculated with a constant retardation model. The magnitude of this effect will be discussed for both diffusion-dominated and advection-dominated systems. Examples will be given based on reported literature data and partition functions (adsorption isotherms). Comments shall also be presented on data applicability and analysis.

A general conclusion regarding waste package performance is the following: if conservatism in the prediction of radionuclide transport is desired, retardation should either be disregarded or non-linear, concentration-dependent retardation factors should be used.

References

1. Smith, M. J., et al., 1980. Engineered Barrier Development for a Nuclear Waste Repository in Basalt: An Integration of Current Knowledge, RHO-BWI-ST-7, Rockwell Hanford Operations, Richland, Washington 99352.