



initial and final radionuclide concentrations in the liquid, an alternative explanation becomes apparent. Figure B illustrates that the change in concentrations in the liquid ( $\Delta$ ) approaches zero as the initial radionuclide concentration in the liquid becomes much greater than the final radionuclide concentration in the solid. Thus, at high radionuclide concentrations in the liquid, calculated  $R_s$  is zero even though the maximum amount of radionuclide is sorbed onto the solid. Disregarding radioactive decay, sorption on the vessel walls, etc., the sorption ratio,  $R_s$ , can be calculated from

$$R_s = (ml/g)((counts_i - counts_f)/counts_f)_1$$

where ml is the milliliters of liquid, g is the grams of solid, counts is the number of decays in the liquid per unit time, i is initial, and f is final. From this equation, it is apparent that at constant  $R_s$  the ratio of phases can be changed so that  $(counts_i - counts_f) \neq 0$ . Thus, by decreasing the water:rock ratio, the region in which sorption ratios can be measured (calculated) is increased.

I suggest that Table 1 (page 3) could have been improved by including a column listing the final Tc concentrations in the liquid. By my calculations, these can be significantly different from the initial concentrations.

Why is it that you consider the microcrystalline components of the mesostasis as the most chemically reactive components? Would not the glass be more reactive (less stable)?

The neptunium study involving various particle sizes is extremely interesting. By varying the duration of these experiments it might be possible to determine if the  $R_s$  values for different particle sizes are kinetically or

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Your concern of isotopic equilibrium is valid especially at low temperatures (e.g. see  $\ln \alpha$  vs  $1/T$  for  $^{18}\text{O}$ - $^{16}\text{O}$ ,  $^2\text{D}$ - $^1\text{H}$ ,  $^{34}\text{S}$ - $^{32}\text{S}$ , etc.). The low temperatures result in considerable partitioning of the isotopes between species. To your advantage, the high molecular weight of the radionuclides should reduce the degree of partitioning as predicted by quantum mechanics.

The action taken by this letter is considered to be within the scope of the current contract FIN B-0290. No change to cost or delivery of contract products is authorized. Please notify me immediately if you believe this letter would result in changes to costs or delivery of contract products.

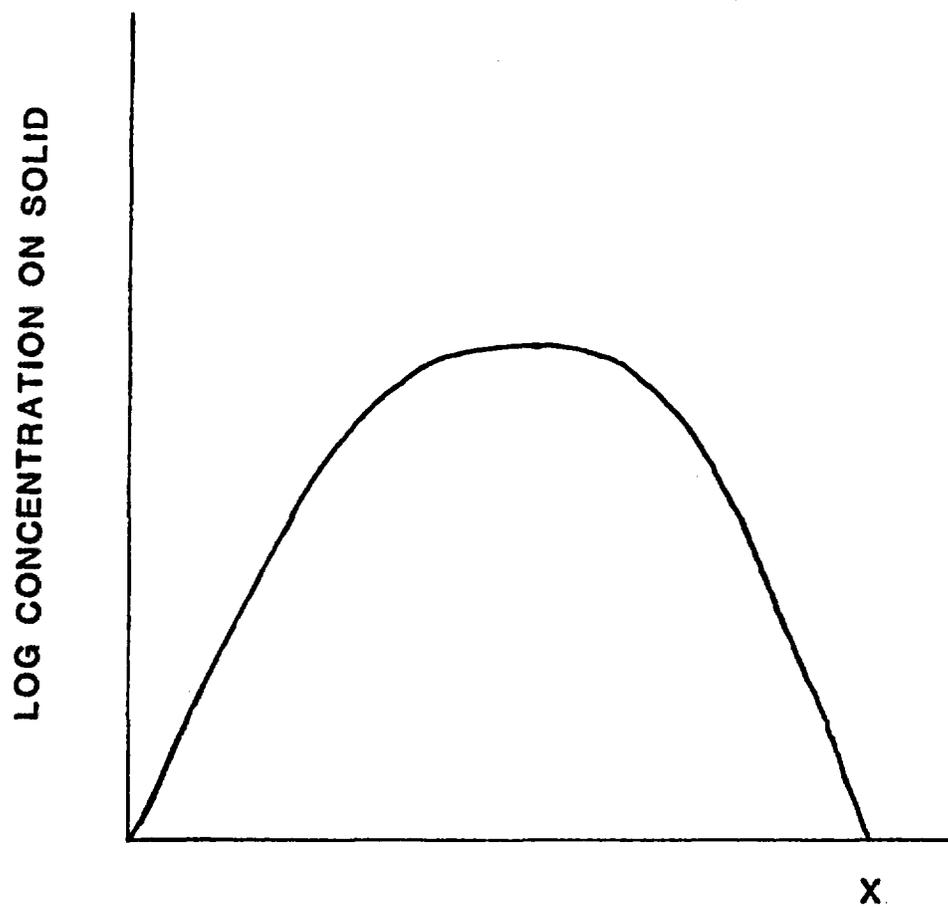
Sincerely,

John W. Bradbury  
Geochemistry Section  
Geotechnical Branch  
Division of Waste Management  
Office of Nuclear Material Safety  
and Safeguards

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OFC	: WMGT <i>JWB</i>	: WMGT <i>RJS</i>	:	:	:	:	:
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**LOG CONCENTRATION IN LIQUID**

Figure A

No measureable sorption at initial concentrations greater than X. Diagram incorrectly illustrates sorption conditions.

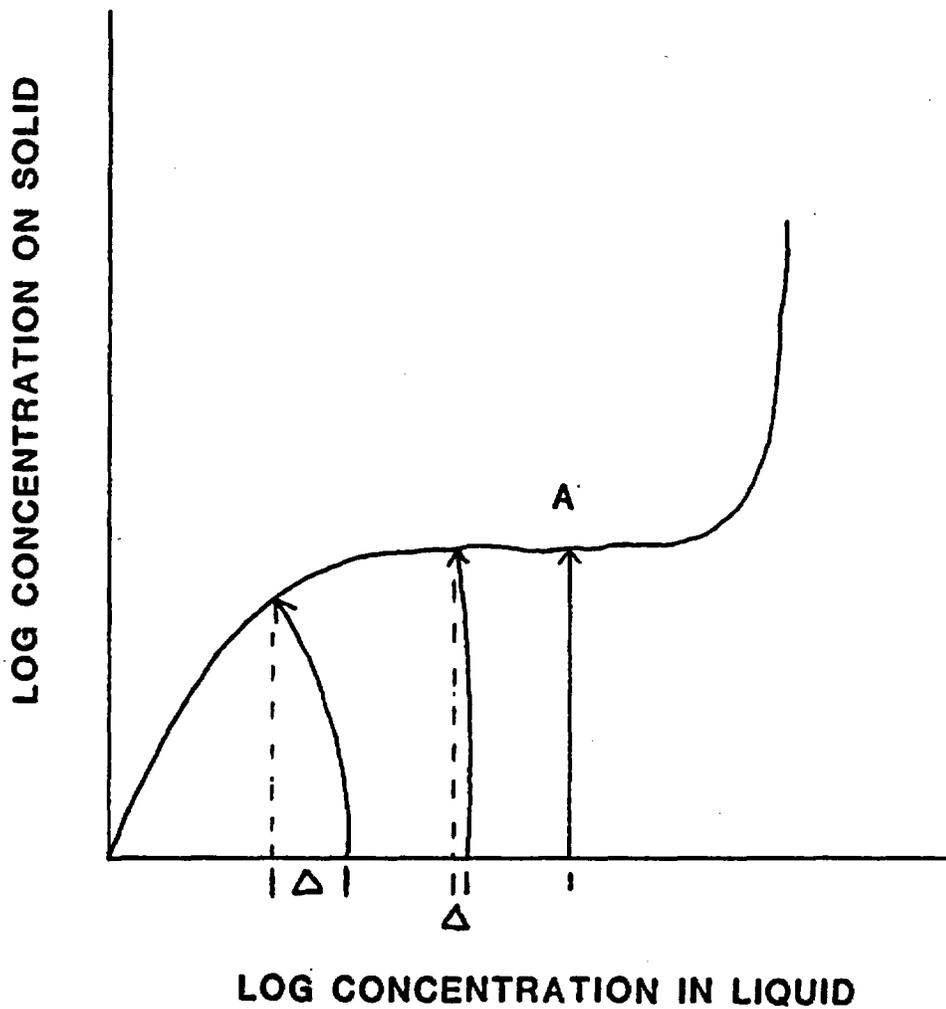


Figure B

Theoretical sorption isotherm. Arrows indicate change of liquid-solid compositions with time. At point A, the amount of radionuclide in the liquid is much greater than the amount on the solid.