
Hanford 300 A IFC

Multi-scale Mass Transfer as the Key Science Issue at the Hanford IFC

Office of Science
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Environmental Remediation Sciences

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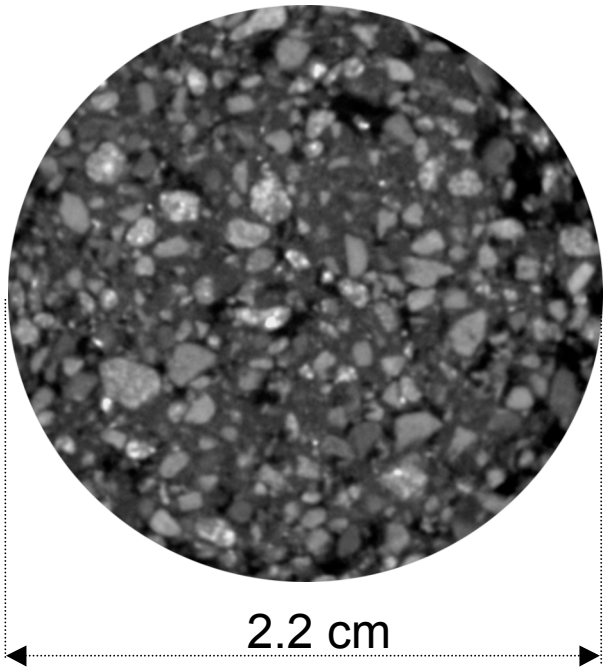
U.S. Department of Energy

Mass Transfer in Hanford 300A Sediments

Column experiments were performed to investigate U mass transfer in unsieved sediment (300A NPP1-14) and its < 2 mm fraction.

Small Column (< 2 mm grains)



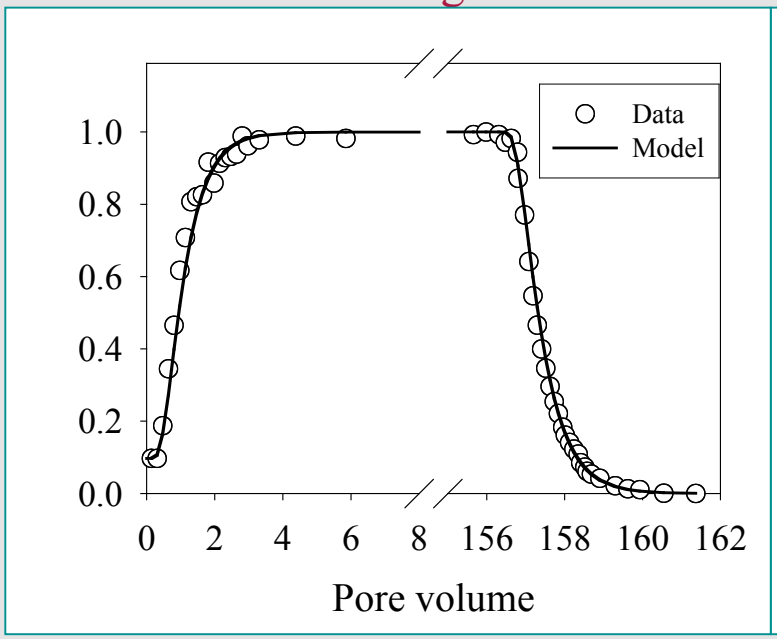
Large Column (80 kg unsieved materials)

Sediment Size and U(VI) Distribution

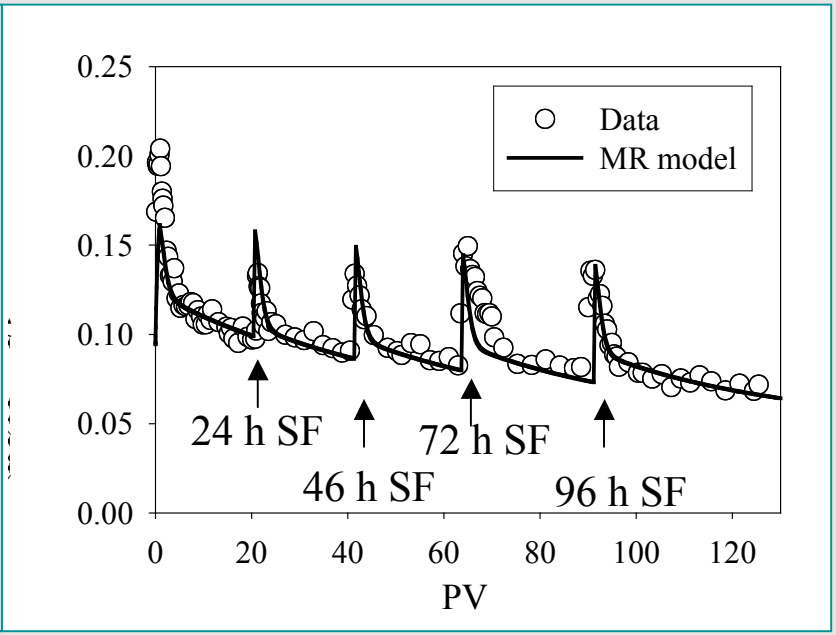
	Size Range (mm)	Mass Fraction (%)	Total U(VI) (nmol/g)
Cobbles			
	>12.5	74.5	<22
	2.0 – 12.5	17.2	<19
Sand			
	1.0 – 2.0	2.64	26
	0.5 – 1.0	2.34	<18
	0.25 – 0.5	0.78	<21
	0.149 – 0.25	0.33	37
	0.106 – 0.149	0.19	<23
	0.053 – 0.149	0.20	<23
Silt+Clay			
	<0.053	1.78	125
Sand+Silt+Clay	<2.0	8.30	47.81

Mass Transfer in Small Columns (<2mm)

Br breakthrough



U(VI) Desorption

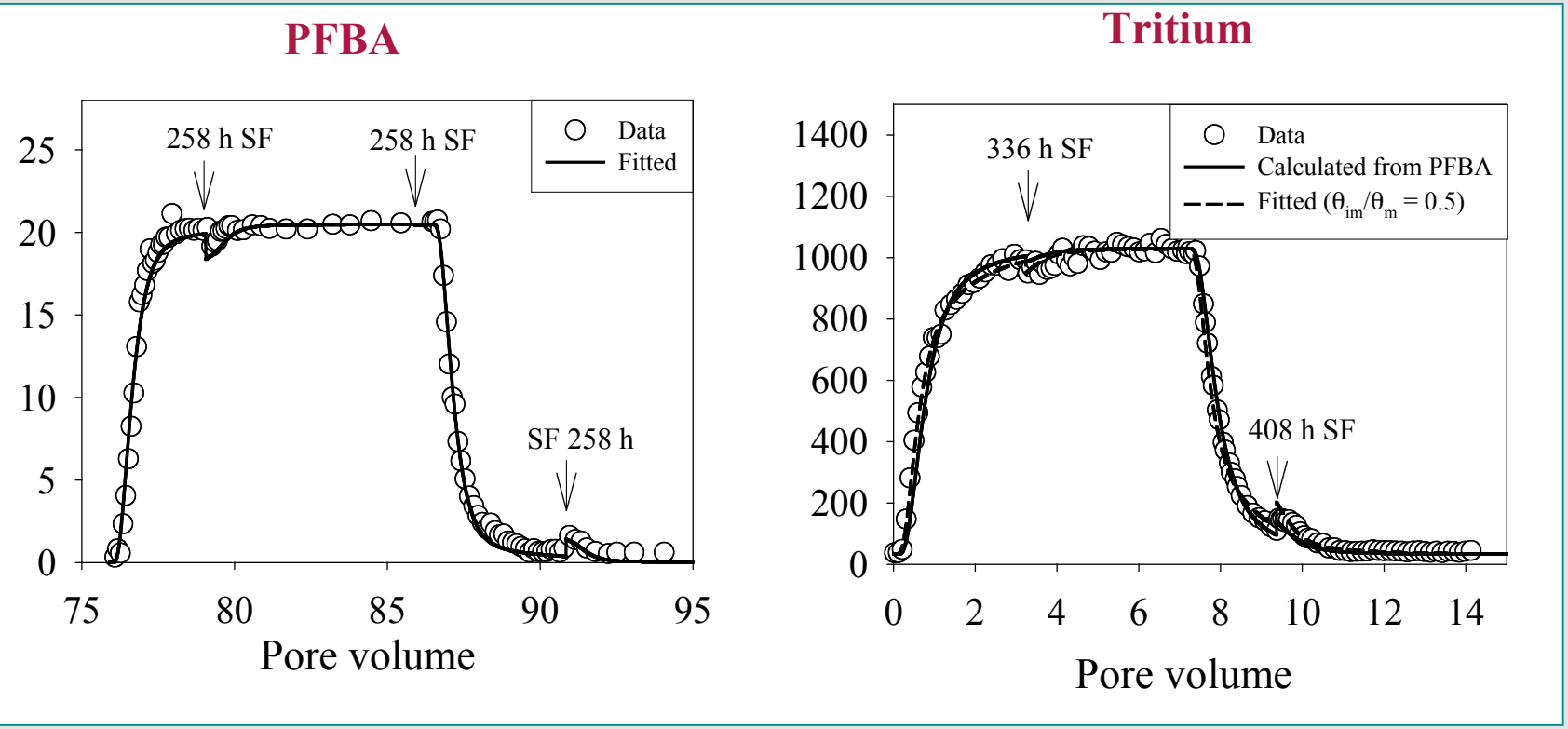


Multi-rate Model: $\frac{\partial q_i}{\partial t} = \int_0^\infty f(\alpha)\alpha(Q_i - q_i)d\alpha$

q_i: sorbed component i, Q_i: equilibrium sorbed component i, α: rate constant, $f(\alpha)$: rate probability distribution.

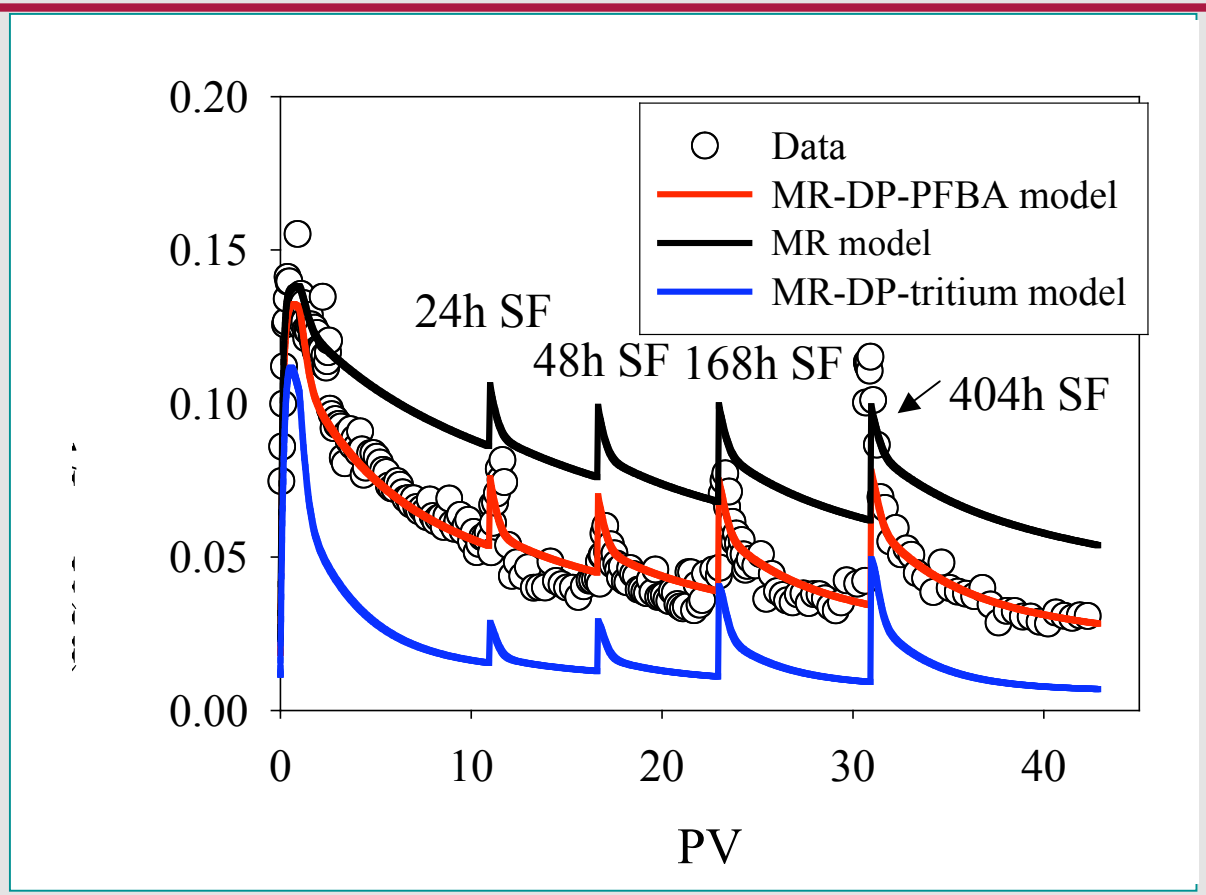
Surface Complexation Reactions	log K
$>SOH + UO_2^{2+} + H_2O = >SOUO_2OH + 2H^+$	-4.72
$>SOH + UO_2^{2+} + CO_3^{2-} = >SOUO_2HCO_3$	16.79

Dual-Domain Mass Transfer in Large Column (Unsieved)



- Tracer breakthrough curves revealed dual domain mass transfer properties;
- Mass transfer is species (tracer)-dependent.

U(VI) Mass Transfer in Large Column (Unsieved)



- Multi-scale mass transfer processes superimposed with increasing scale.
- Importance of selection of tracers in exploring physical mass transfer properties.

Upscaling of Mass Transfer Kinetics

Transport Equation:

$$\theta_m \frac{\partial C_i^m}{\partial t} + (1-\theta_m)\rho_s \frac{\partial q_i^m}{\partial t} = \theta_m AD(C_i^m) - \theta_{im}\beta(C_i^m - C_i^{im}) \qquad i = 1, 2, \ldots, N$$

Mass Exchange Between Mobile and Immobile Domains:

$$\theta_{im} \frac{\partial C_i^{im}}{\partial t} + (1-\theta_{im})\rho_s \frac{\partial q_i^{im}}{\partial t} = \theta_{im}\beta(C_i^m - C_i^{im})$$

Mobile Domain Multi-Rate Equation:

$$\frac{\partial q_i^m}{\partial t} = \int_0^\infty f(\alpha)(Q_i^m - q_i^m)d\alpha \qquad f(\alpha): \text{lognormal distribution} \qquad Q_i^m: \text{From surface complexation}$$

Immobile Domain Multi-Rate Equation:

$$\frac{\partial q_i^{im}}{\partial t} = \int_0^\infty f(\alpha)(Q_i^{im} - q_i^{im})d\alpha \qquad f(\alpha): \text{lognormal distribution} \qquad Q_i^{im}: \text{From surface complexation}$$

- Total site density was calculated based on the mass weight of the reactive size fraction in the large column;
- Sorbed U(VI) was splitted in mobile and immobile domains based on their porosity values.

Parameters in Modeling U(VI) Desorption in Column Experiments

Parameters	Symbol	Unit	SC-1	SC2	LC
Column Length	L	cm	10.5	10.5	80
Pore velocit y	v	cm/h	8.60	7.75	3.52
Dispersion coefficient	D	cm2/h	15.25	3.64	46.52
Porosity	θ	/	0.41	0.46	0.32
Soil bulk density	ρ_b	kg/L	1.56	1.42	1.88
Immobile porosity	θ_{im}	/	0.00	0.00	0.064
Logarithm mean rate	μ	$\log(h^{-1})$	-9.99	-9.99	-9.99
Standard deviation	σ	$\log(h^{-1})$	2.66	2.66	2.66
Two domain mass exchange rate constant (h^{-1})			PFBA 1.45×10^{-2}	Tritium 4.28×10^{-2}	Br 3.87×10^{-2}	U(VI) 1.45×10^{-2}