Supplementary material

Surrogate model for multi-component diffusion of uranium through Opalinus Clay on the host-rock scale

Theresa Hennig[a,b,*] and Michael Kühn[a,b]

[a] GFZ German Research Centre for Geosciences, Fluid Systems Modelling, Telegrafenberg, 14473 Potsdam, Germany

[b] University of Potsdam, Institute of Geosciences, Karl-Liebknecht-Str. 24-25, 14476 Potsdam-Golm, Germany

[*] Corresponding author: theresa.hennig@gfz-potsdam.de, Phone: +49 331 288-28723

Supplementary material S-1

Table S1: Summary of the used self-diffusion coefficients D_w (m^2/s).

Species	**D_w (m^2/s)**	**Reference**	**Comments**
H^+	$9.31 \cdot 10^{-9}$	a	
H^2	$5.13 \cdot 10^{-9}$	a	
OH^-	$5.27 \cdot 10^{-9}$	a	
O_2	$2.35 \cdot 10^{-9}$	a	
CO_3^{2-}	$8.12 \cdot 10^{-10}$	b	
$HCO3^-$	$1.18 \cdot 10^{-9}$	a	
CO_2	$1.92 \cdot 10^{-9}$	a	
CH_4	$1.85 \cdot 10^{-9}$	a	
Ca^{2+}	$7.93 \cdot 10^{-10}$	a	
$CaHCO_3^+$	$5.06 \cdot 10^{-10}$	a	
$CaCO_3$	$4.46 \cdot 10^{-10}$	a	
$CaSO_4$	$4.71 \cdot 10^{-10}$	a	
Cl^-	$2.03 \cdot 10^{-9}$	a	
Fe^{2+}	$7.19 \cdot 10^{-10}$	a	
K^+	$1.96 \cdot 10^{-9}$	a	
KSO_4^-	$1.50 \cdot 10^{-9}$	a	
Mg^{2+}	$7.05 \cdot 10^{-10}$	a	
$MgHCO_3^+$	$4.78 \cdot 10^{-10}$	a	
$MgCO_3$	$4.21 \cdot 10^{-10}$	a	
$MgSO_4$	$4.45 \cdot 10^{-10}$	a	

Na^{+}	$1.33 \cdot 10^{-9}$	a	
$NaHCO_3$	$6.73 \cdot 10^{-10}$	a	
$NaCO_3^{-}$	$1.20 \cdot 10^{-9}$	a	
$NaSO_4^{-}$	$1.33 \cdot 10^{-9}$	a	
PO_4^{3-}	$6.12 \cdot 10^{-10}$	a	
HPO_4^{2-}	$6.90 \cdot 10^{-10}$	a	
$H_2PO_4^{-}$	$8.46 \cdot 10^{-10}$	a	
SO_4^{2-}	$1.07 \cdot 10^{-9}$	a	
HSO_4^{-}	$1.33 \cdot 10^{-9}$	a	
S^{2-}	$7.31 \cdot 10^{-9}$	a	
HS^{-}	$1.73 \cdot 10^{-9}$	a	
H_2S	$2.10 \cdot 10^{-9}$	a	
Sr^{2+}	$7.94 \cdot 10^{-10}$	a	
UO_2^{2+}	$7.66 \cdot 10^{-10}$	b	
UO_2OH^{+}	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$UO_2(OH)_2$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$UO_2(OH)_3^{-}$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$UO_2(OH)_4^{2-}$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$(UO_2)_2OH^{3+}$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$(UO_2)_2(OH)_2^{2+}$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$(UO_2)_3(OH)_4^{2+}$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$(UO_2)_3(OH)_5^{+}$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$(UO_2)_3(OH)_7^{-}$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b

$(UO_2)_4(OH)_7^{+}$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
UO_2Cl^{+}	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
UO_2Cl_2	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
UO_2SO_4	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$UO_2(SO_4)_2^{2-}$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
UO_2HPO_4	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$UO_2H_2PO_4^{+}$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$UO_2H_3PO_4^{2+}$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
$UO_2(H_2PO_4)_2$	$7.66 \cdot 10^{-10}$	c	analogous to UO_2^{2+} from b
UO_2CO_3	$6.67 \cdot 10^{-10}$	b	
$UO_2(CO_3)_2^{2-}$	$5.52 \cdot 10^{-10}$	b	
$UO_2(CO_3)_3^{4-}$	$5.57 \cdot 10^{-10}$	b	
$(UO_2)_3(CO_3)_6^{6-}$	$5.57 \cdot 10^{-10}$	b	analogous to $UO_2(CO_3)_2^{2-}$
$(UO_2)_2CO_3(OH)_3^{-}$	$5.57 \cdot 10^{-10}$	b	analogous to $UO_2(CO_3)_2^{2-}$
$UO_2H_2PO_4H_3PO_4^{+}$	$5.57 \cdot 10^{-10}$	b	analogous to $UO_2(CO_3)_2^{2-}$
$(UO_2)_3O(OH)_2HCO_3^{+}$	$5.57 \cdot 10^{-10}$	b	analogous to $UO_2(CO_3)_2^{2-}$
$UO_2(SO_4)_3^{4-}$	$5.57 \cdot 10^{-10}$	b	analogous to $UO_2(CO_3)_2^{2-}$
$MgUO_2(CO_3)_3^{2-}$	$5.06 \cdot 10^{-10}$	b	
$CaUO_2(CO_3)_3^{2-}$	$5.06 \cdot 10^{-10}$	b	
$Ca_2UO_2(CO_3)_3$	$4.6 \cdot 10^{-10}$	b	
$SrUO_2(CO_3)_3^{2-}$	$4.83 \cdot 10^{-10}$	b	
U^{4+}	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}
UOH^{3+}	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}

$U(OH)_2^{2+}$	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}
$U(OH)_3^{+}$	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}
$U(OH)_4$	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}
$UCO_3(OH)_3^{-}$	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}
$U(CO_3)_4^{4-}$	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}
$U(CO_3)_5^{6-}$	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}
USO_4^{2+}	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}
$U(SO_4)_2$	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}
UO_2^{+}	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}
$UO_2(CO_3)_3^{5-}$	$7.66 \cdot 10^{-10}$	b	analogous to UO_2^{2+}

a	taken from database phreeqc.dat distributed with PHREEQC (Parkhurst and Appelo, 2013)
b	Kerisit and Liu (2010)
c	Liu et al. (2011)

References

Kerisit, S.; Liu, C. Molecular simulation of the diffusion of uranyl carbonate species in aqueous solution. Geochimica et Cosmochimica Acta 2010, 74, 4937–4952. doi:10.1016/j.gca.2010.06.007

Liu, C.; Shang, J.; Zachara, J.M. Multispecies diffusion models: A study of uranyl species diffusion. Water Resources Research 2011, 47, 1–16. doi:10.1029/2011WR010575

Parkhurst, D.L.; Appelo, C.A.J. Description of input and examples for PHREEQC Version 3 — A computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations. In U.S. Geological Survey Techniques and Methods; 2013; Vol. book 6, chapter A43, p. 497