Experimental and modeling approaches to Am(III) and Np(V) adsorption on Maoming kaolinite

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The goal of this paper is to study the adsorption of Am(III) and Np(V) on the Maoming kaolinite and to provide corresponding adsorption models. The surface properties of Maoming kaolinite were evaluated by continuous potentiometric titration. Comparisons with the titration curves of KGa-1, KGa-2, Zettlitz, St Austell and Twiggs County kaolinite samples indicate that the Maoming kaolinite may have different surface acid-base chemistry than these kaolinite samples. These titration curves of the Maoming kaolinite can be interpreted with both the diffuse double layer model (DDLM) and a non-electrostatic model (NEM). The adsorptions of Am(III) and Np(V) were studied as functions of contact time, solid-to-liquid ratios, pH, ionic strength, and concentration using a batch method. The adsorptions of Am(III) and Np(V) on kaolinite were rapid and equilibrated within 20 and 10 hours, respectively. The adsorption of 6 × 10−10 mol/L Am(III) was shown to be sensitive to both pH and ionic strength, whereas the adsorption of 1 × 10−6 mol/L Np(V) was sensitive only to pH. The DDLM overestimates the effect of ionic strength on Np(V) adsorption. Finally, the adsorption of Am(III) and Np(V) was quantitatively interpreted using the NEM with the addition of one cation exchange reaction for Am(III) (TABLE 1). These adsorption data and corresponding models are useful for the safety assessment of an HLW repository, which may be built in the clayey formation in China.

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TABLE I. Surface reactions and corresponding equilibrium constants of Np(V) and Am(III) adsorption on kaolinite based on double-layer model and non-electrostatic model.

Reactions	DLM model		NEM model	
Np(V)	K^{int}	log K^{int}	K	log K
$\equiv \mathrm{S^WOH} + \mathrm{NpO_2^+} \rightleftharpoons \equiv \mathrm{S^WONpO_2} + \mathrm{H^+}$	$K_4^{\mathrm{int}} = \frac{[\equiv \mathrm{S^WONpO_2}][\mathrm{H^+}] \cdot \gamma_{\mathrm{H^+}}}{[\equiv \mathrm{S^WOH}][\mathrm{NpO_2^+}] \cdot \gamma_{\mathrm{NpO_2^+}}}$	-1.45	$K_4 = \frac{[\equiv \mathrm{S^WONpO_2}][\mathrm{H^+}] \cdot \gamma_{\mathrm{H^+}}}{[\equiv \mathrm{S^WOH}][\mathrm{NpO_2^+}] \cdot \gamma_{\mathrm{NpO_2^+}}}$	-1.13
$\equiv \mathrm{S^WOH} + \mathrm{NpO_2^+} + \mathrm{H_2O} \rightleftharpoons \equiv \mathrm{S^WONpO_2(OH)^-} + 2\mathrm{H^+}$	$K_5^{\mathrm{int}} = \frac{[\equiv \mathrm{S^WONpO_2(OH)^-}][\mathrm{H^+}]^2 \cdot \gamma_{\mathrm{H^+}}^2}{[\equiv \mathrm{S^WOH}][\mathrm{NpO_2^+}] \cdot \gamma_{\mathrm{NpO_2^+}}} \exp(-F\psi/RT)$	-8.29	$K_5 = \frac{[\equiv \mathrm{S^WONpO_2(OH)^-}][\mathrm{H^+}]^2 \cdot \gamma_{\mathrm{H^+}}^2}{[\equiv \mathrm{S^WOH}][\mathrm{NpO_2^+}] \cdot \gamma_{\mathrm{NpO_2^+}}}$	-8.82
$\equiv \mathrm{S^SOH} + \mathrm{NpO_2^+} \rightleftharpoons \equiv \mathrm{S^SONpO_2} + \mathrm{H^+}$	$K_6^{\mathrm{int}} = \frac{[\equiv \mathrm{S^SONpO_2}][\mathrm{H^+}] \cdot \gamma_{\mathrm{H^+}}}{[\equiv \mathrm{S^SOH}][\mathrm{NpO_2^+}] \cdot \gamma_{\mathrm{NpO_2^+}}}$	1.80	$K_6 = \frac{[\equiv \mathrm{S^SONpO_2}][\mathrm{H^+}] \cdot \gamma_{\mathrm{H^+}}}{[\equiv \mathrm{S^SOH}][\mathrm{NpO_2^+}] \cdot \gamma_{\mathrm{NpO_2^+}}}$	2.52
Am(III)	DLM model			
	K	log K		
$3 \equiv \mathrm{XNa} + \mathrm{Am^{3+}} \rightleftharpoons (\equiv \mathrm{X})_3\mathrm{Am} + 3\mathrm{Na^+}$	$K_7 = \frac{[(\equiv \mathrm{X})_3\mathrm{Am}][\mathrm{Na^+}]^3 \cdot \gamma_{\mathrm{Na^+}}^3}{[\equiv \mathrm{XNa}]^3[\mathrm{Am^{3+}}] \cdot \gamma_{\mathrm{Am^{3+}}}}$	0.85		
$\equiv \mathrm{S^SOH} + \mathrm{Am^{3+}} \rightleftharpoons \equiv \mathrm{S^SOAm^{2+}} + \mathrm{H^+}$	$K_8 = \frac{[\equiv \mathrm{S^SOAm^{2+}}][\mathrm{H^+}] \cdot \gamma_{\mathrm{H^+}}}{[\equiv \mathrm{S^SOH}][\mathrm{Am^{3+}}] \cdot \gamma_{\mathrm{Am^{3+}}}}$	3.68		
$\equiv \mathrm{S^SOH} + \mathrm{Am^{3+}} + \mathrm{H_2O} \rightleftharpoons \equiv \mathrm{S^SOAm(OH)^+} + 2\mathrm{H^+}$	$K_9 = \frac{[\equiv \mathrm{S^SOAm(OH)^+}][\mathrm{H^+}]^2 \cdot \gamma_{\mathrm{H^+}}^2}{[\equiv \mathrm{S^SOH}][\mathrm{Am^{3+}}] \cdot \gamma_{\mathrm{Am^{3+}}}}$	-1.50		
$\equiv \mathrm{S^SOH} + \mathrm{Am^{3+}} + 2\mathrm{H_2O} \rightleftharpoons \equiv \mathrm{S^SOAm(OH)_2} + 3\mathrm{H^+}$	$K_{10} = \frac{[\equiv \mathrm{S^SOAm(OH)_2}][\mathrm{H^+}]^3 \cdot \gamma_{\mathrm{H^+}}^3}{[\equiv \mathrm{S^SOH}][\mathrm{Am^{3+}}] \cdot \gamma_{\mathrm{Am^{3+}}}}$	-9.36		