Electronic Supplementary Material (ESI) for Journal of Materials Chemistry
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ELECTRONIC SUPPORTING INFORMATION

The Intercalation of Flavouring Compounds into Layered Double Hydroxides for Taste Modification

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CONTENTS

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Table S1. Comparison of guest molecule lengths to the observed interlayer separations.

Guest	Guest Length (Å)	Metals	d-spacing (Å)	Interlayer Separation (Å)	(Gallery Height)/ (Guest Length)	Probable Arrangement
Acesulfame	7.5	Li/Al	14.4	9.6	1.3	Interdigitated bilayer
		Mg/Al	14.3	9.5	1.3	Interdigitated bilayer
Cyclamate	10.2	Ca/Al	15.5	10.7	1.0	Monolayer
		Li/Al	37.2 / 20.3	32.4 / 15.5	3.2 / 1.5	Hydrated structure / IB
		Mg/Al	21.8	17.0	1.7	Interdigitated bilayer
Saccharin	8.6	Ca/Al	14.7	9.9	1.2	Interdigitated bilayer
		Li/Al	15.4	10.6	1.2	Interdigitated bilayer
		Mg/Al	15.2	10.4	1.2	Interdigitated bilayer
Ferulate	11.9	Ca/Al	11.1	6.3	0.5	Tilted Monolayer
		Li/Al	17.2	12.4	1.0	Monolayer
		Mg/Al	17.3	12.5	1.1	Monolayer
Theobromine	10.0	Ca/Al	13.6	8.8	0.9	Tilted Monolayer
		Li/Al	17.7	12.9	1.3	Interdigitated bilayer
		Mg/Al	13.0	8.2	0.8	Tilted Monolayer

The 'guest length' is equal to the through-space separation between the charge-bearing atom and the most distant atom in the molecule, with the van der Waals radii of each atom added on. The thickness of a single hydroxide layer is 4.8Å.[1] Values shown for the Li/Al-Cyclamate LDH are those of the expanded and dehydrated structures, respectively. IB = Interdigitated bilayer.

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Table S2. Elemental Analysis Data and Chemical Formulae of the Intercalates

Compound Name	Chemical Formula	Predicted C, H, N (%)	Observed C, H, N (%)
Ca/Al-Cyclamate	$[Ca_2Al(OH)_6](C_6H_{12}NO_3S)_{0.76}(CO_3)_{0.12}\cdot 2.4H_2O$	14.23, 5.08, 2.69	14.20, 5.07, 2.62
Ca/Al-Saccharin	$[Ca_2Al(OH)_6](C_7H_4NO_3S)_{0.76}(NO_3)_{0.24}\cdot 2H_2O$	16.03, 3.30, 3.51	15.99, 3.21, 3.41
Ca/Al-Ferulate	$[Ca_2Al(OH)_6](C_{10}H_9O_4)\cdot 1.2H_2O$	28.33, 4.14, -	28.44, 4.05, -
Ca/Al-Theobromine	$[Ca_2Al(OH)_6](C_7H_7N_4O_2)_{0.21}(OH)_{0.79}\cdot H_2O$	6.35, 3.72, 4.23	6.28, 3.91, 4.22
Li/Al-Acesulfame	$[LiAl_2(OH)_6](C_4H_4NO_4S)_{0.73}Cl_{0.27}\cdot 1.5H_2O$	11.03, 3.78, 3.22	10.98, 3.77, 3.12
Li/Al-Cyclamate	$[LiAl_2(OH)_6](C_6H_{12}NO_3S)\cdot 2.2H_2O$	18.91, 5.93, 3.68	18.93, 5.90, 3.71
Li/Al-Saccharin	$[LiAl_2(OH)_6](C_7H_4NO_3S)\cdot 1.2H_2O$	22.92, 3.41, 3.82	23.01, 3.41, 3.76
Li/Al-Ferulate	$[LiAl_2(OH)_6](C_{10}H_9O_4)_{0.65}Cl_{0.35}\cdot 0.6H_2O$	25.04, 4.22, -	25.01, 4.26, -
Li/Al-Theobromine	$[LiAl_2(OH)_6](C_7H_7N_4O_2)_{0.61}(OH)_{0.39}\cdot H_2O$	17.26, 4.30, 11.51	17.20, 4.25, 11.48
Mg/Al-Acesulfame	$[Mg_2Al(OH)_6](C_4H_4NO_4S)_{0.99}(NO_3)_{0.01}\cdot H_2O$	13.33, 3.38, 3.93	13.35, 3.45, 3.92
Mg/Al-Cyclamate	$[Mg_2Al(OH)_6](C_6H_{12}NO_3S)_{0.7}(NO_3)_{0.3}\cdot 1.3H_2O$	14.65, 4.98, 4.07	14.67, 4.93, 3.97
Mg/Al-Saccharin	$[Mg_2Al(OH)_6](C_7H_4NO_3S)_{0.78}(CO_3)_{0.11}\cdot 0.9H_2O$	19.53, 3.21, 3.19	19.58, 3.04, 3.14
Mg/Al-Ferulate	$[Mg_2Al(OH)_6](C_{10}H_9O_4)_{0.85}(OH)_{0.15}\cdot H_2O$	28.21, 4.40, -	28.20, 4.09, -
Mg/Al-Theobromine	$[Mg_2Al(OH)_6](C_7H_7N_4O_2)_{0.69}(OH)_{0.31}\cdot H_2O$	17.90, 4.09, 11.94	17.91, 3.83, 11.96

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Table S3. (t_{50} and Total Release Values for all LDH intercalates)

Metals	Guest	Release Medium	t_{50} (Minutes)	Release after 180 Minutes (%)
Ca/Al	Cyclamate	H_2O	17	35.2
		Saliva	<10	91.9
	Saccharin	H_2O	<10	59.3
		Saliva	<10	97.2
	Ferulate	H_2O	<10	29.7
		Saliva	<10	99.4
	Theobromine	H_2O	<10	63.7
		Saliva	30	90.5
Li/Al	Acesulfame	H_2O	21	26.5
		Saliva	61	35.8
	Cyclamate	H_2O	<10	38.8
		Saliva	<10	87.9
	Saccharin	H_2O	115	11.2
		Saliva	<10	49.3
	Ferulate	H_2O	47	31.3
		Saliva	<10	81.7
	Theobromine	H_2O	<10	81.8
		Saliva	<10	83.2
Mg/Al	Acesulfame	H_2O	25	21.9
		Saliva	<10	90.8
	Cyclamate	H_2O	<10	50.1
		Saliva	<10	80.9
	Saccharin	H_2O	<10	66.9
		Saliva	<10	88.8
	Ferulate	H_2O	58	6.5
		Saliva	31	92.1
	Theobromine	H_2O	<10	93.1
		Saliva	<10	99.3

The t_{50} value is the time (in minutes) required for release of 50% of the amount of guest present in solution after 180 minutes.

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Table S4. Rate Equations of the Kinetic Models used[2-8]

Kinetic Model	Rate Equation
Avrami-Erofe'ev	$\ln\left[-\ln\left(1-\frac{C_t}{C_0}\right)\right] = n\ln(t) + n\ln(k_r)$
Elovich	$1-\frac{C_t}{C_0} = n\ln(t)+m$
First-Order	$\ln\frac{C_t}{C_0} = -k_r t$
Freundlich	$C_0 - C_t = k_r C_0 t^n$
Parabolic	$\left(1-\frac{C_t}{C_0}\right)\frac{1}{t} = k_r\frac{1}{\sqrt{t}} + n$

t = Reaction time elapsed

C_t = Concentration released at time t

C_0 = Concentration present in the solid at $t = 0$

m = Reaction coefficient

n = Reaction coefficient

k_r = Rate constant

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Table S5. Parameters extracted from the fitting of the kinetic models

Metals	Guest	Deionized H_2O (Avrami-Erofe'ev Kinetics)		Saliva (First Order Kinetics)
		Rate Constant (k_r) (x 10^{-3})	Reaction Exponent (n)	Rate Constant (k_r) (x 10^{-3})
Ca/Al	Cyclamate	560.1	0.17	3.1
	Saccharin	3.50	0.27	26.6
	Ferulate	0.00079	0.04	31.4
	Theobromine	6.78	0.11	14.8
Li/Al	Acesulfame	0.081	0.30	2.7
	Cyclamate	0.0011	0.08	15.3
	Saccharin	0.087	0.63	5.8
	Ferulate	2.16	0.80	12.7
	Theobromine	293.9	0.14	13.2
Mg/Al	Acesulfame	0.025	0.28	16.3
	Cyclamate	0.110	0.11	12.3
	Saccharin	43.7	0.30	9.9
	Ferulate	14.2	0.66	13.5
	Theobromine	557.6	0.21	26.3

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