

Table S11 Relative total electronic energies E_{rel} (in kcal/mol) of PYH^+/ZO^- adsorption complexes of different Si/Al ratios and Al distributions determined at the PBE and PBE-D3 levels in cluster and periodic models

Cluster model (32T)			Periodic model (96T)		
PYH ⁺ /ZO ⁻ Adsorption complexes	E_{rel}		PYH ⁺ /ZO ⁻ Adsorption complexes	E_{rel}	
	PBE/ 6-31+G(d)	PBE-D3/ 6-31+G(d)		PBE	PBE-D3
(31Si,1Al)			(95Si,1Al)		
Al1 _{ads}	1.6	2.2	Al1 _{ads}	2.2	2.7
Al2 _{ads}	4.5	5.0	Al2 _{ads}	0.0	0.0
Al3 _{ads}	0.0	0.0	Al3 _{ads}	1.5	2.2
Al4 _{ads}	21.6	15.9	Al4 _{ads}	17.0	12.6
(30Si,2Al)			(94Si,2Al)		
Al1 _{ads} Al2	4.4	4.8	Al1 _{ads} Al2	2.9	1.7
Al1 _{ads} Al3	9.0	9.8	Al1 _{ads} Al3	4.5	4.2
Al1 _{ads} Al4	7.6	8.0	Al1 _{ads} Al4	6.6	5.9
Al2 _{ads} Al1	6.7	8.3	Al2 _{ads} Al1	5.5	5.1
Al2 _{ads} Al3	9.1	9.3	Al2 _{ads} Al3	0.0	0.0
Al2 _{ads} Al4	7.6	7.9	Al2 _{ads} Al4	1.8	0.5
Al3 _{ads} Al1	0.0	0.0	Al3 _{ads} Al1	4.9	3.5
Al3 _{ads} Al2	4.5	4.4	Al3 _{ads} Al2	3.9	3.5
Al3 _{ads} Al4	6.2	5.8	Al3 _{ads} Al4	4.5	5.0
Al4 _{ads} Al1	21.8	16.6	Al4 _{ads} Al1	20.3	15.1
Al4 _{ads} Al2	30.3	24.5	Al4 _{ads} Al2	20.7	16.3
Al4 _{ads} Al3	22.8	17.2	Al4 _{ads} Al3	19.8	15.2
(29Si,3Al)			(93Si,3Al)		
Al1 _{ads} Al2Al3	6.3	7.1	Al1 _{ads} Al2Al3	0.8	1.0
Al1 _{ads} Al2Al4	4.5	4.3	Al1 _{ads} Al2Al4	3.3	1.9
Al1 _{ads} Al3Al4	8.6	8.9	Al1 _{ads} Al3Al4	6.6	6.6
Al2 _{ads} Al1Al3	5.1	7.0	Al2 _{ads} Al1Al3	0.8	1.9
Al2 _{ads} Al1Al4	5.6	6.1	Al2 _{ads} Al1Al4	4.4	4.5
Al2 _{ads} Al3Al4	7.3	7.3	Al2 _{ads} Al3Al4	0.0	0.0
Al3 _{ads} Al1Al2	0.0	0.0	Al3 _{ads} Al1Al2	3.1	3.8
Al3 _{ads} Al1Al4	1.3	0.7	Al3 _{ads} Al1Al4	4.3	5.7
Al3 _{ads} Al2Al4	5.6	4.8	Al3 _{ads} Al2Al4	3.9	5.3
Al4 _{ads} Al1Al2	24.7	19.2 ¹	Al4 _{ads} Al1Al2	21.0	16.0
Al4 _{ads} Al1Al3	17.9	12.6	Al4 _{ads} Al1Al3	19.3	15.2
Al4 _{ads} Al2Al3	26.4	20.4	Al4 _{ads} Al2Al3	18.6	15.2
(28Si,4Al)			(92Si,4Al)		
Al1 _{ads} Al2Al3Al4	5.6	6.1	Al1 _{ads} Al2Al3Al4	1.4	0.0
Al2 _{ads} Al1Al3Al4	1.0	2.0	Al2 _{ads} Al1Al3Al4	0.0	0.6
Al3 _{ads} Al1Al2Al4	0.0	0.0	Al3 _{ads} Al1Al2Al4	5.7	4.2
Al4 _{ads} Al1Al2Al3	20.0	14.8	Al4 _{ads} Al1Al2Al3	18.2	12.7