Table S4. Optimized Intermolecular Geometrical Parameters of PYH^+/ZO^- Adsorption Complexes with Two Al Atoms Calculated at the PBE and PBE-D3 (in brackets) Levels in Cluster and Periodic Models.

PYH^+/ZO^- Adsorption Complexes	Cluster Model (32T)				Periodic Model (96T)			
	O…H[a]	N–H[a]	O…H–N[b]	α[bc]	O…H[a]	N–H[a]	O…H–N[b]	α[bc]
$Al1_{ads}Al2$	1.614(1.615)	1.079(1.077)	175.4(176.3)	35.7(39.9)	1.706(1.716)	1.064(1.063)	177.5(172.3)	27.8(24.8)
$Al1_{ads}Al3$	1.565(1.576)	1.085(1.082)	169.1(166.2)	64.6(66.2)	1.714(1.698)	1.064(1.064)	174.8(171.9)	34.8(38.4)
$Al1_{ads}Al4$	1.614(1.635)	1.076(1.072)	173.9(170.0)	37.5(45.7)	1.732(1.715)	1.060(1.062)	174.1(176.4)	36.3(33.3)
$Al2_{ads}Al1$	1.620(1.618)	1.073(1.074)	167.0(168.2)	51.7(52.1)	1.606(1.607)	1.076(1.074)	171.0(171.1)	73.7(72.5)
$Al2_{ads}Al3$	1.583(1.581)	1.081(1.082)	171.8(172.1)	66.2(60.7)	1.612(1.610)	1.074(1.075)	173.5(173.6)	68.2(67.9)
$Al2_{ads}Al4$	1.606(1.605)	1.074(1.074)	171.5(170.7)	74.6(71.0)	1.610(1.637)	1.070(1.069)	174.2(173.9)	68.7(69.7)
$Al3_{ads}Al1$	1.670(1.670)	1.069(1.068)	173.3(170.6)	64.1(72.1)	1.586(1.566)	1.074(1.075)	174.4(174.8)	73.4(73.8)
$Al3_{ads}Al2$	1.696(1.688)	1.067(1.066)	172.7(172.9)	49.2(47.7)	1.666(1.621)	1.065(1.070)	168.0(168.6)	76.6(78.3)
$Al3_{ads}Al4$	1.655(1.650)	1.070(1.069)	178.7(176.4)	75.5(75.8)	1.703(1.618)	1.061(1.068)	164.5(171.1)	77.0(76.5)
$Al4_{ads}Al1$	1.673(1.668)	1.069(1.069)	158.4(159.5)	62.8(61.1)	1.764(1.770)	1.051(1.051)	148.4(147.9)	69.7(69.0)
$Al4_{ads}Al2$	1.630(1.631)	1.076(1.077)	157.9(160.4)	63.4(60.7)	1.716(1.719)	1.059(1.057)	150.4(149.8)	68.5(67.8)
$Al4_{ads}Al3$	1.668(1.668)	1.070(1.070)	159.3(160.8)	63.0(60.4)	1.742(1.734)	1.054(1.055)	149.7(149.0)	71.3(70.7)

[a]In Å; [b]In deg; [c]Angle between the straight channel axis and the vector normal to the PY plane.