Supporting Information for

Effects of Li/Mn arrangement on the electrochemical and transport properties of Na-Li-Mn cathodes

Wenjing Qin[1, 2*], Yinjun Xiong[3, 4]

[1]*School of Electrical Engineering, Jiangxi Polytechnic University, Jiujiang 332000, China*

[2]*Laboratory of Computational Materials Physics, Department of Physics, Institute of Condensed Matter, Jiangxi Normal University, Nanchang 330022, China*

[3]*School of Electrical Engineering, Xinjiang University, Urumqi 830049, China*

[4]*State Grid Jiangxi Electric Power Company Jiujiang Power Supply Company, Jiujiang 332000, China*

Table S1. The energies of all Na-vacancy configurations for the line configuration of $Na_xLi_{0.17}Mn_{0.83}O_2$ (x = 0, 0.17, 0.33, 0.5, 0.67 and 0.83).

Disinsertion concentration	Disinsertion configuration	Energy (eV)
$Na_{0.83}Li_{0.17}Mn_{0.83}O_2$	1	-143.00
	2	-142.90
	3	-143.00
	4	-142.90
	5	-142.98
	6	-142.60
$Na_{0.67}Li_{0.17}Mn_{0.83}O_2$	1 + 2	-138.59
	1 + 3	-138.51
	1 + 4	-138.94
	1 + 5	-138.79
	1 + 6	-138.83
	2 + 3	-138.59
	2 + 4	-138.32
	2 + 5	-139.13
	2 + 6	-138.73
	3 + 4	-138.83
	3 + 5	-138.79
	3 + 6	-138.94
	4 + 5	-138.99
	4 + 6	-138.84

	5 + 6	-138.99
$Na_{0.5}Li_{0.17}Mn_{0.83}O_2$	1 + 2 + 3	-133.28
	1 + 2 + 4	-134.03
	1 + 2 + 5	-134.34
	1 + 2 + 6	-134.03
	1 + 3 + 4	-134.11
	1 + 3 + 5	-134.27
	1 + 3 + 6	-134.11
	1 + 4 + 5	-134.51
	1 + 4 + 6	-134.29
	1 + 5 + 6	-134.16
	2 + 3 + 4	-134.03
	2 + 3 + 5	-134.34
	2 + 3 + 6	-134.03
	2 + 4 + 5	-134.26
	2 + 4 + 6	-133.90
	2 + 5 + 6	-134.26
	3 + 4 + 5	-134.16
	3 + 4 + 6	-134.28
	3 + 5 + 6	-134.51
	4 + 5 + 6	-134.05
$Na_{0.33}Li_{0.17}Mn_{0.83}O_2$	1 + 2 + 3 + 4	-127.65
	1 + 2 + 3 + 5	-128.11
	1 + 2 + 3 + 6	-127.65
	1 + 2 + 4 + 5	-128.56
	1 + 2 + 4 + 6	-128.22
	1 + 2 + 5 + 6	-128.20
	1 + 3 + 4 + 5	-128.43
	1 + 3 + 4 + 6	-128.50
	1 + 3 + 5 + 6	-128.43
	1 + 4 + 5 + 6	-128.25
	2 + 3 + 4 + 5	-128.20
	2 + 3 + 4 + 6	-128.22
	2 + 3 + 5 + 6	-128.56
	2 + 4 + 5 + 6	-128.36
	3 + 4 + 5 + 6	-128.25
$Na_{0.17}Li_{0.17}Mn_{0.83}O_2$	1 + 2 + 3 + 4 + 5	-122.00
	1 + 2 + 3 + 4 + 6	-121.87
	1 + 2 + 3 + 5 + 6	-122.00
	1 + 2 + 4 + 5 + 6	-122.22

	1 + 3 + 4 + 5 + 6	-122.23
	2 + 3 + 4 + 5 + 6	-122.22
$Na_0Li_{0.17}Mn_{0.83}O_2$	1 + 2 + 3 + 4 + 5 + 6	-116.41

Table S2. The energies of all Na-vacancy configurations for the honeycomb configurations of $Na_xLi_{0.33}Mn_{0.67}O_2$ (x = 0, 0.17, 0.33, 0.5, 0.67 and 0.83). The total energies listed in this table were taken from Ref. [43].

Disinsertion concentration	Disinsertion configuration	Energy (eV)
$Na_{0.83}Li_{0.33}Mn_{0.67}O_2$	1	-137.25
	2	-137.25
	3	-137.25
	4	-137.24
	5	-137.24
	6	-137.25
$Na_{0.67}Li_{0.33}Mn_{0.67}O_2$	1 + 2	-131.86
	1 + 3	-131.45
	1 + 4	-131.79
	1 + 5	-131.70
	1 + 6	-131.67
	2 + 3	-131.79
	2 + 4	-131.45
	2 + 5	-131.92
	2 + 6	-131.70
	3 + 4	-131.80
	3 + 5	-131.64
	3 + 6	-131.64
	4 + 5	-131.60
	4 + 6	-131.64
	5 + 6	-131.67
$Na_{0.5}Li_{0.33}Mn_{0.67}O_2$	1 + 2 + 3	-125.80
	1 + 2 + 4	-125.79
	1 + 2 + 5	-126.01
	1 + 2 + 6	-125.87
	1 + 3 + 4	-125.85
	1 + 3 + 5	-125.80
	1 + 3 + 6	-125.20
	1 + 4 + 5	-124.59
	1 + 4 + 6	-124.59
	1 + 5 + 6	-125.87
	2 + 3 + 4	-125.85

	2 + 3 + 5	-124.59
	2 + 3 + 6	-124.59
	2 + 4 + 5	-125.20
	2 + 4 + 6	-125.80
	2 + 5 + 6	-126.01
	3 + 4 + 5	-125.94
	3 + 4 + 6	-125.94
	3 + 5 + 6	-126.01
	4 + 5 + 6	-126.01
$Na_{0.33}Li_{0.33}Mn_{0.67}O_2$	1 + 2 + 3 + 4	-119.83
	1 + 2 + 3 + 5	-119.60
	1 + 2 + 3 + 6	-119.11
	1 + 2 + 4 + 5	-118.02
	1 + 2 + 4 + 6	-118.56
	1 + 2 + 5 + 6	-119.88
	1 + 3 + 4 + 5	-118.37
	1 + 3 + 4 + 6	-118.15
	1 + 3 + 5 + 6	-119.47
	1 + 4 + 5 + 6	-119.77
	2 + 3 + 4 + 5	-119.11
	2 + 3 + 4 + 6	-118.56
	2 + 3 + 5 + 6	-120.00
	2 + 4 + 5 + 6	-119.47
	3 + 4 + 5 + 6	-119.78
$Na_{0.17}Li_{0.33}Mn_{0.67}O_2$	1 + 2 + 3 + 4 + 5	-113.35
	1 + 2 + 3 + 4 + 6	-113.35
	1 + 2 + 3 + 5 + 6	-113.39
	1 + 2 + 4 + 5 + 6	-113.15
	1 + 3 + 4 + 5 + 6	-113.15
	2 + 3 + 4 + 5 + 6	-113.39
$Na_0Li_{0.33}Mn_{0.67}O_2$	1 + 2 + 3 + 4 + 5 + 6	-107.69

Table S3. The energies of all Na-vacancy configurations for the non-honeycomb configurations of $Na_xLi_{0.33}Mn_{0.67}O_2$ (x = 0, 0.17, 0.33, 0.5, 0.67 and 0.83).

Disinsertion concentration	Disinsertion configuration	Energy (eV)
$Na_{0.83}Li_{0.33}Mn_{0.67}O_2$	1	-136.33
	2	-136.31
	3	-136.55
	4	-136.47
	5	-136.55

	6	-136.31
$Na_{0.67}Li_{0.33}Mn_{0.67}O_2$	1 + 2	-131.35
	1 + 3	-131.15
	1 + 4	-131.38
	1 + 5	-131.15
	1 + 6	-131.35
	2 + 3	-131.33
	2 + 4	-131.18
	2 + 5	-131.49
	2 + 6	-131.27
	3 + 4	-131.42
	3 + 5	-131.40
	3 + 6	-131.49
	4 + 5	-131.42
	4 + 6	-131.18
	5 + 6	-131.33
$Na_{0.5}Li_{0.33}Mn_{0.67}O_2$	1 + 2 + 3	-124.11
	1 + 2 + 4	-125.46
	1 + 2 + 5	-125.66
	1 + 2 + 6	-125.26
	1 + 3 + 4	-125.65
	1 + 3 + 5	-125.51
	1 + 3 + 6	-125.66
	1 + 4 + 5	-125.65
	1 + 4 + 6	-125.46
	1 + 5 + 6	-124.11
	2 + 3 + 4	-124.06
	2 + 3 + 5	-125.78
	2 + 3 + 6	-124.67
	2 + 4 + 5	-125.80
	2 + 4 + 6	-125.67
	2 + 5 + 6	-124.67
	3 + 4 + 5	-124.28
	3 + 4 + 6	-125.80
	3 + 5 + 6	-125.78
	4 + 5 + 6	-124.06
$Na_{0.33}Li_{0.33}Mn_{0.67}O_2$	1 + 2 + 3 + 4	-118.06
	1 + 2 + 3 + 5	-119.42
	1 + 2 + 3 + 6	-118.25
	1 + 2 + 4 + 5	-119.79

	1 + 2 + 4 + 6	-119.74
	1 + 2 + 5 + 6	-118.25
	1 + 3 + 4 + 5	-118.42
	1 + 3 + 4 + 6	-119.79
	1 + 3 + 5 + 6	-119.42
	1 + 4 + 5 + 6	-118.06
	2 + 3 + 4 + 5	-118.23
	2 + 3 + 4 + 6	-118.58
	2 + 3 + 5 + 6	-119.87
	2 + 4 + 5 + 6	-118.58
	3 + 4 + 5 + 6	-118.23
$Na_{0.17}Li_{0.33}Mn_{0.67}O_2$	1 + 2 + 3 + 4 + 5	-112.09
	1 + 2 + 3 + 4 + 6	-113.41
	1 + 2 + 3 + 5 + 6	-113.16
	1 + 2 + 4 + 5 + 6	-113.41
	1 + 3 + 4 + 5 + 6	-112.09
	2 + 3 + 4 + 5 + 6	-112.31
$Na_0Li_{0.33}Mn_{0.67}O_2$	1 + 2 + 3 + 4 + 5 + 6	-107.73

Table S4. The desodiation voltages (V) for the line, honeycomb and non-honeycomb configurations of $Na_xLi_yMn_{1-y}O_2$ (x = 0.17, 0.33, 0.5, 0.67, 0.83 and 1.0, y = 0.17 and 0.33).

Desodiation voltage (V)	line	honeycomb	non-honeycomb
$Na_1 \rightarrow Na_{0.83}$	3.05	4.28	3.14
$Na_{0.83} \rightarrow Na_{0.67}$	2.56	4.02	3.75
$Na_{0.67} \rightarrow Na_{0.5}$	3.31	4.60	4.38
$Na_{0.5} \rightarrow Na_{0.33}$	4.64	4.70	4.62
$Na_{0.33} \rightarrow Na_{0.17}$	5.02	5.30	5.15
$Na_{0.17} \rightarrow Na_0$	4.51	4.39	4.37

Table S5. Magnetic moment of each O atom for the line, honeycomb and non-honeycomb configurations of $Na_xLi_yMn_{1-y}O_2$ (x = 0 and 1, y = 0.17 and 0.33).

Magnetic moment (μ_B)	line		honeycomb		non-honeycomb	
	Na1	Na0	Na1	Na0	Na1	Na0
O_1	-0.09	-0.27	-0.10	-0.53	-0.10	-0.51
O_2	-0.11	-0.50	-0.11	-0.54	-0.10	-0.51
O_3	-0.07	-0.27	-0.10	-0.53	-0.10	-0.51
O_4	-0.01	-0.50	-0.11	-0.54	-0.10	-0.51
O_5	-0.10	-0.48	-0.10	-0.53	-0.10	-0.56
O_6	-0.05	-0.27	-0.11	-0.54	-0.10	-0.56
O_7	-0.08	-0.48	-0.10	-0.53	-0.10	-0.56

O_8	0.05	-0.27	-0.11	-0.54	-0.10	-0.56
O_9	-0.08	-0.48	-0.05	-0.85	-0.10	-0.56
O_{10}	-0.06	-0.27	-0.11	-0.32	-0.10	-0.56
O_{11}	-0.10	-0.48	-0.05	-0.85	-0.10	-0.56
O_{12}	-0.06	-0.27	-0.11	-0.32	-0.10	-0.56

Table S6. Total energies of the initial (ini) and final (fin) states for Li interlayer migration in the line, honeycomb and non-honeycomb configurations of $Na_xLi_yMn_{1-y}O_2$ (x = 0.17, 0.33, 0.5, 0.67 and 0.83, y = 0.17 and 0.33).

Energy (eV)	line		honeycomb		non-honeycomb	
	ini	fin	ini	fin	ini	fin
$Na_{0.83}$	-143.00	-141.81	-137.25	-136.38	-136.55	-134.98
$Na_{0.67}$	-139.13	-138.41	-131.91	-131.34	-131.49	-130.79
$Na_{0.5}$	-134.51	-133.11	-126.01	-125.62	-125.80	-125.58
$Na_{0.33}$	-128.56	-128.11	-119.95	-120.00	-119.79	-119.69
$Na_{0.17}$	-122.23	-121.73	-113.39	-113.78	-113.41	-113.17

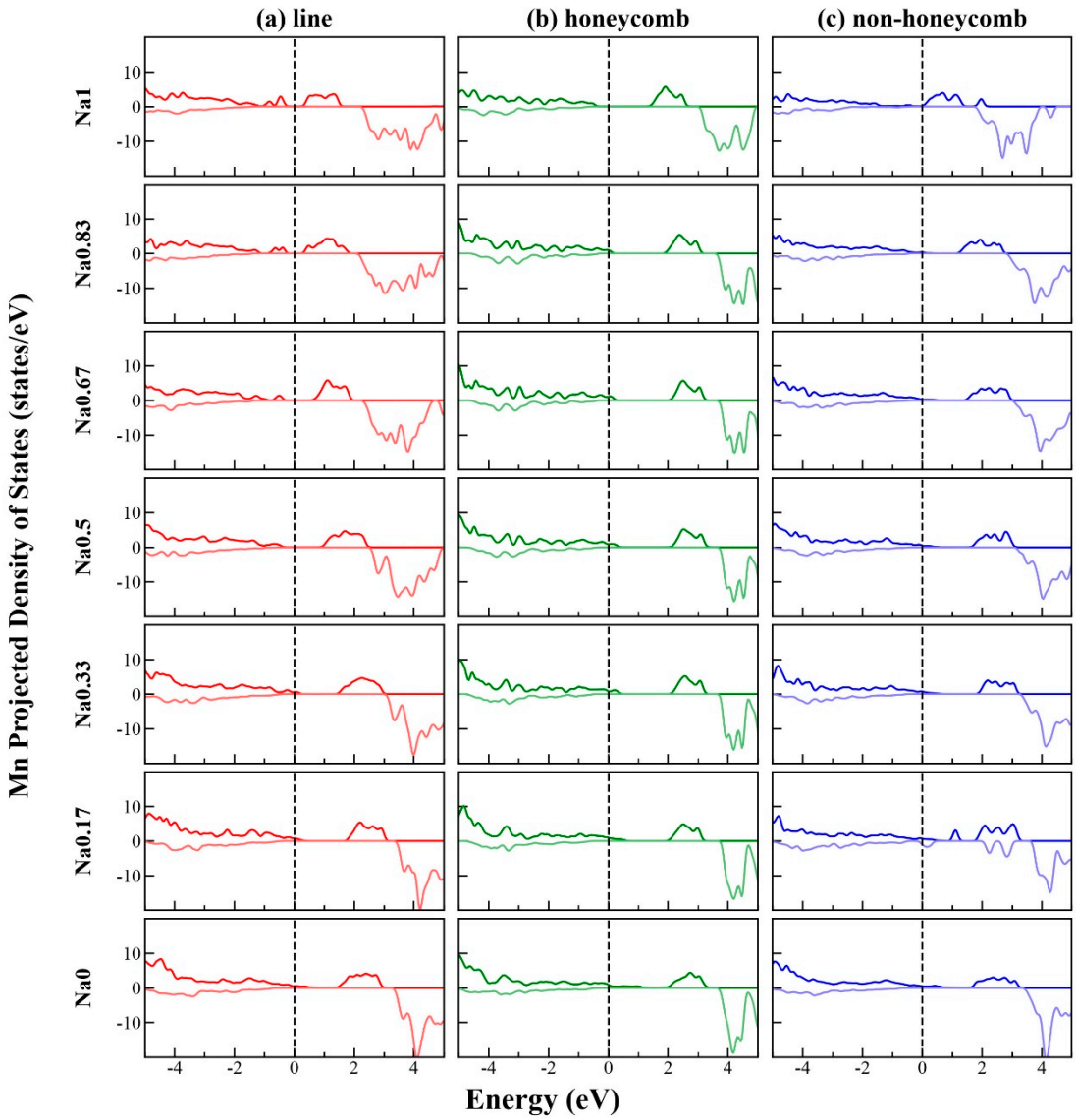


Figure S1. Spin-polarized projected density of states (PDOS) of Mn at different Na contents for the (a) line, (b) honeycomb and (c) non-honeycomb configurations of $Na_xLi_yMn_{1-y}O_2$ (x = 0, 0.17, 0.33, 0.5, 0.67, 0.83 and 1, y = 0.17 and 0.33).

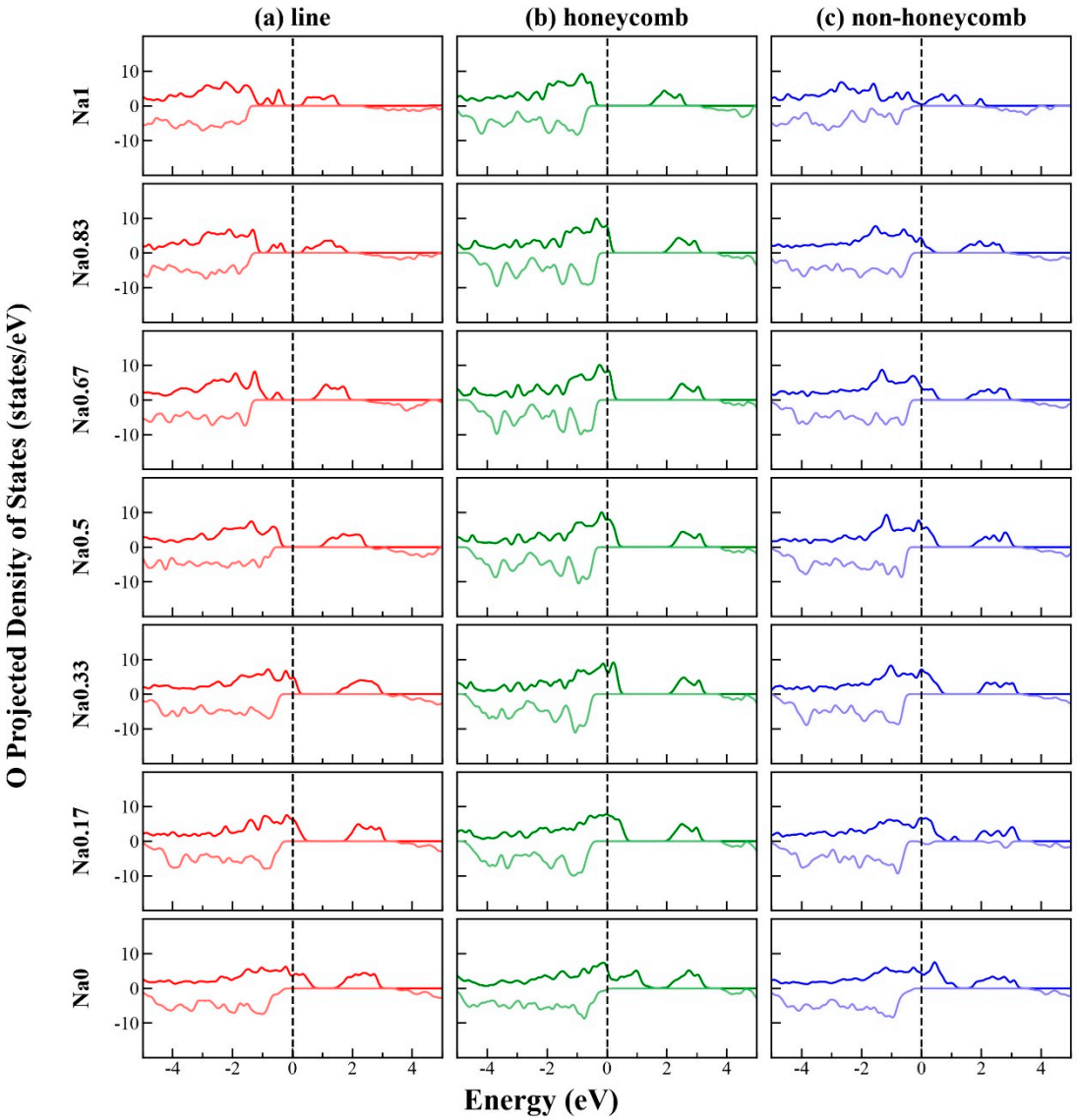


Figure S2. Spin-polarized projected density of states (PDOS) of O at different Na contents for the (a) line, (b) honeycomb and (c) non-honeycomb configurations of $Na_xLi_yMn_{1-y}O_2$ (x = 0, 0.17, 0.33, 0.5, 0.67, 0.83 and 1, y = 0.17 and 0.33).

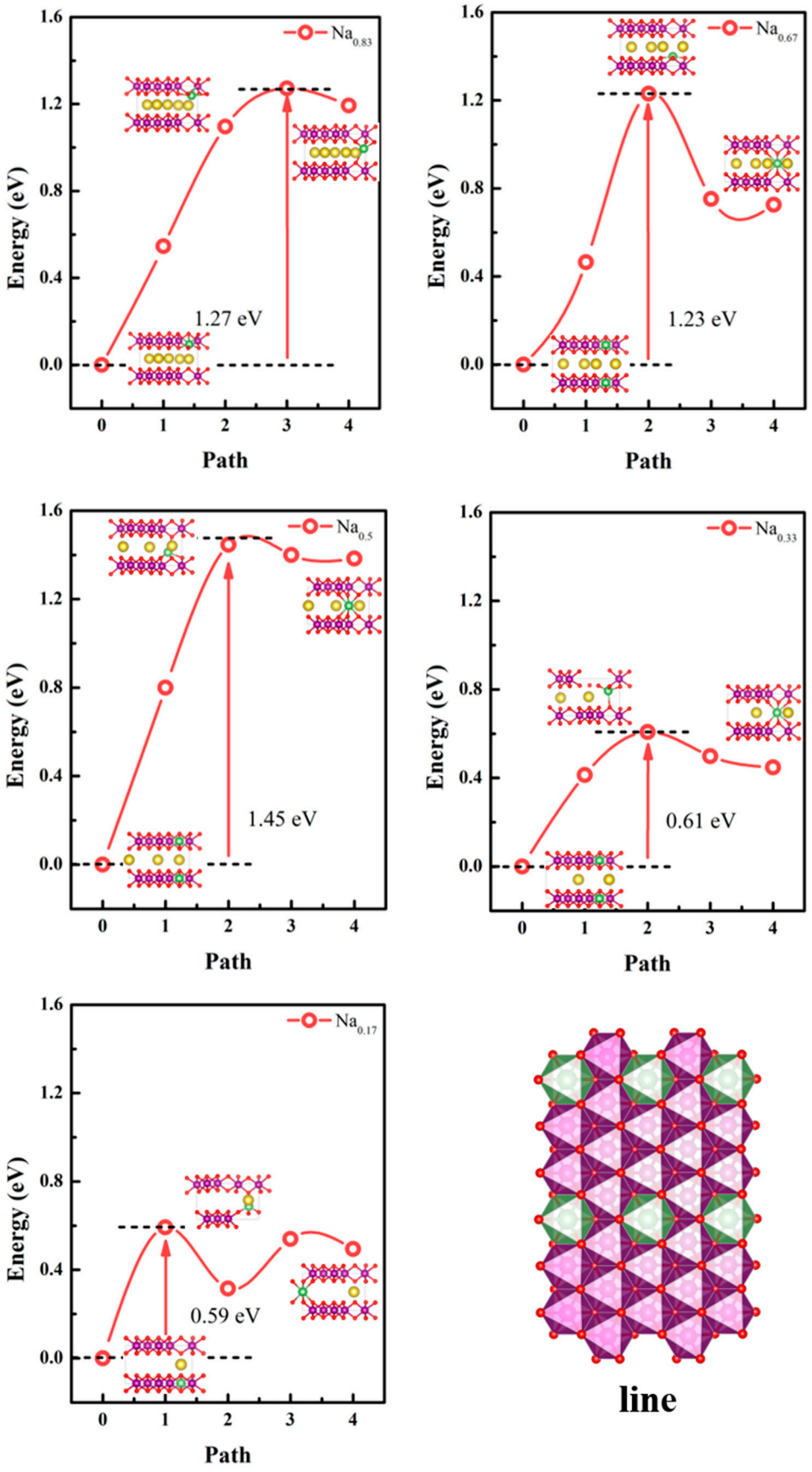


Figure S3. The energy profiles of Li-ion interlayer migration for the line configuration of $Na_xLi_{0.17}Mn_{0.83}O_2$ (x = 0.17, 0.33, 0.5, 0.67 and 0.83).

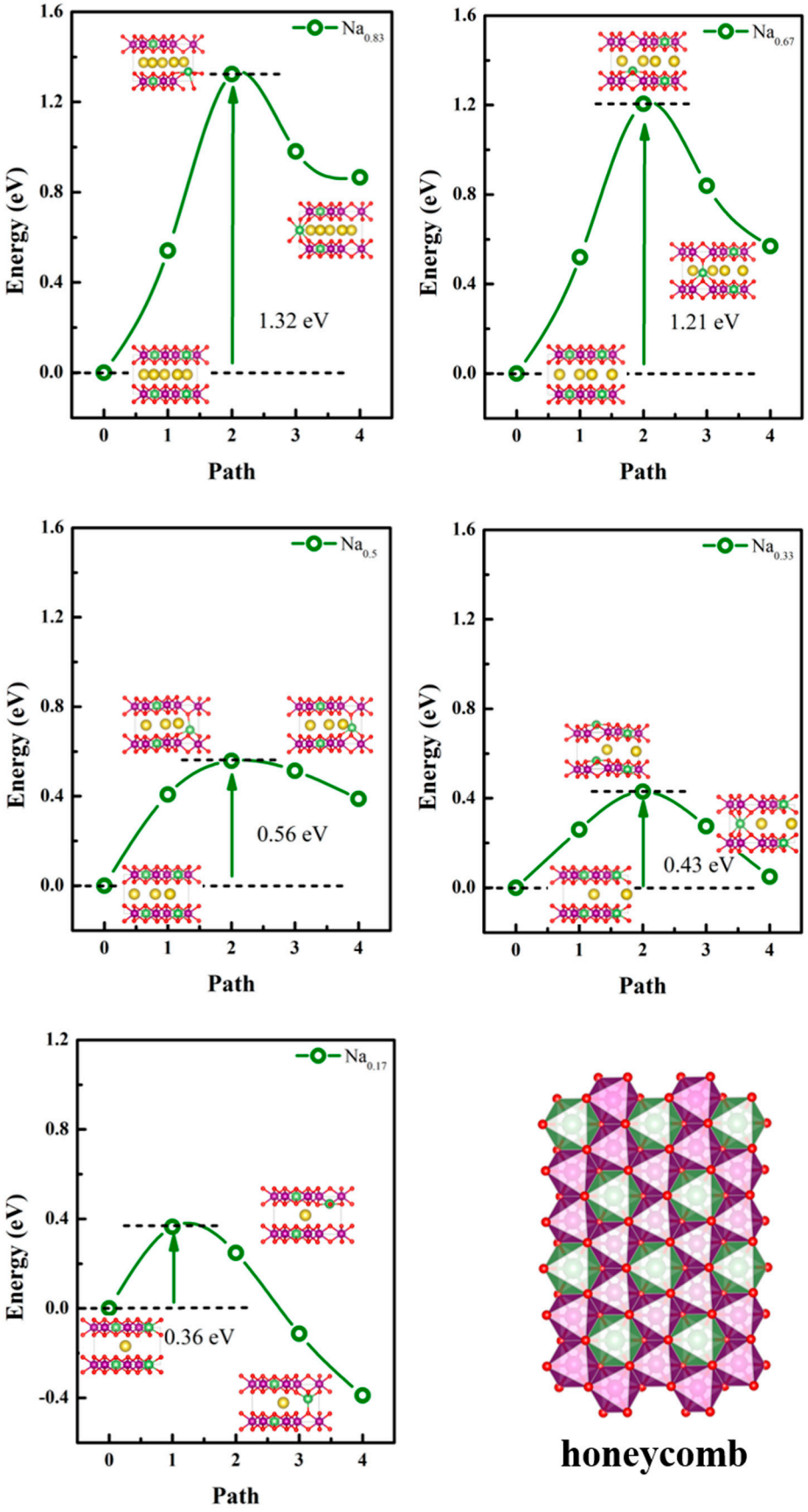


Figure S4. The energy profiles of Li-ion interlayer migration for the honeycomb configuration of $Na_xLi_{0.33}Mn_{0.67}O_2$ (x = 0.17, 0.33, 0.5, 0.67 and 0.83).

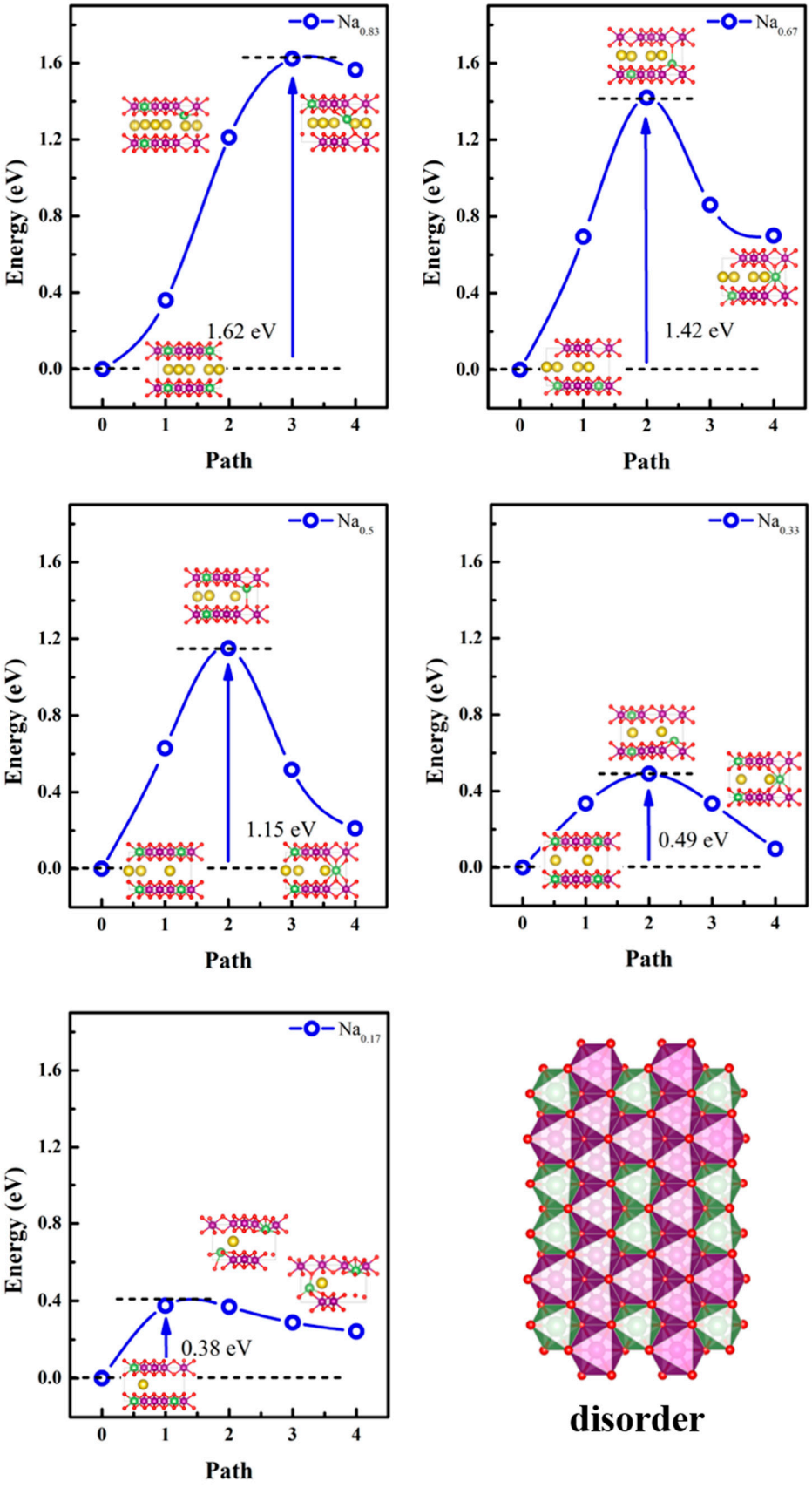


Figure S5. The energy profiles of Li-ion interlayer migration for the non-honeycomb configuration of $Na_xLi_{0.33}Mn_{0.67}O_2$ (x = 0.17, 0.33, 0.5, 0.67 and 0.83).