Supporting Information

Mg-doped cathodic properties and solid state ionic conduction in P2-Type layered material for Na-ion batteries and supercapacitors

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Table S1. Buckingham Potential parameter used in this study for $Na_{0.67}MnO_2$ and $Na_{0.67}Mn_{0.9}Mg_{0.1}O_2$. for two-body interactions can be written as, $\phi_{ij}^{(rij)} = A_{ij} \exp\left(\frac{-r_{ij}}{\rho_{ij}}\right) - \frac{Cij}{rij^6}$, where A is energy in and ρ is the distance between the pair of atoms. These parameters should be adopted purposefully to replicate the experimental data.

Interaction	A (eV)	ρ (Å)	C (eV Å^{-6})
Na^{2+} - O^{2-}	1226.84	0.3065	0.000
Mn^{3+} - O^{2-}	1267.5	0.3214	0.00
Mg^{2+} - O^{2-}	946.627	0.318	0.00
O^{2-} - O^{2-}	22764.3	0.149	27.88

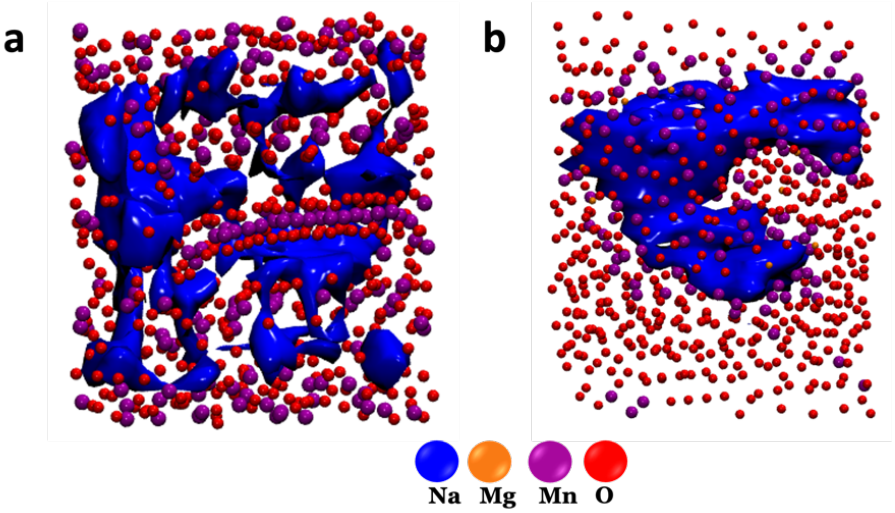

Figure S1. The reference position $r_i(0)$ for (a) $Na_{0.67}MnO_2$, (b) $Na_{0.67}Mn_{0.9}Mg_{0.1}O_2$ at 600 K.

We conducted an equilibration run of 20 ns with 1 fs timestep at 300 K for $Na_{0.67}MnO_2$, desodiated MnO_2, $Na_{0.67}Mn_{0.9}Mg_{0.1}O_2$, and desodiated $Mn_{0.9}Mg_{0.1}O_2$ to verify the structural stability dynamically. We reported the temperature and energy changes during the simulation period for the aforementioned system in Figure S2. We analyzed that during the sodiation and desodiation process in $Na_{0.67}MnO_2$, the structure is highly destabilized at the end of fully discharged. But in $Na_{0.67}Mn_{0.9}Mg_{0.1}O_2$, it is less destabilized compared to $Na_{0.67}MnO_2$. We hope that at higher temperatures, it will be stabilized during full discharge.

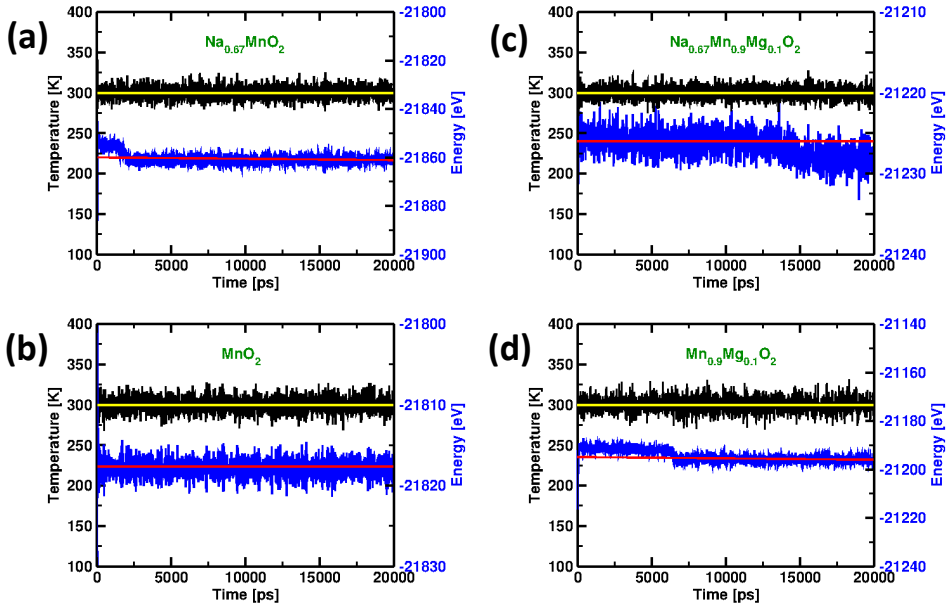

Figure S2. The fluctuation of temperature and energy during MD simulation at 300 K of (a) $Na_{0.67}MnO_2$, (c) $Na_{0.67}Mn_{0.9}Mg_{0.1}O_2$ and their respective desodiated materials in (b), (d).

In Figure S3, we additionally present the variation in bond length between Na-O and Mn-O in sodiated materials and just Mn-O in desodiated materials. The Na-O bond fluctuated more in the Mg-doped material during the simulation than in the undoped one. The bond strength of Na-O is reduced in $Na_{0.67}Mn_{0.9}Mg_{0.1}O_2$, which allows for easier Na-ion diffusion.

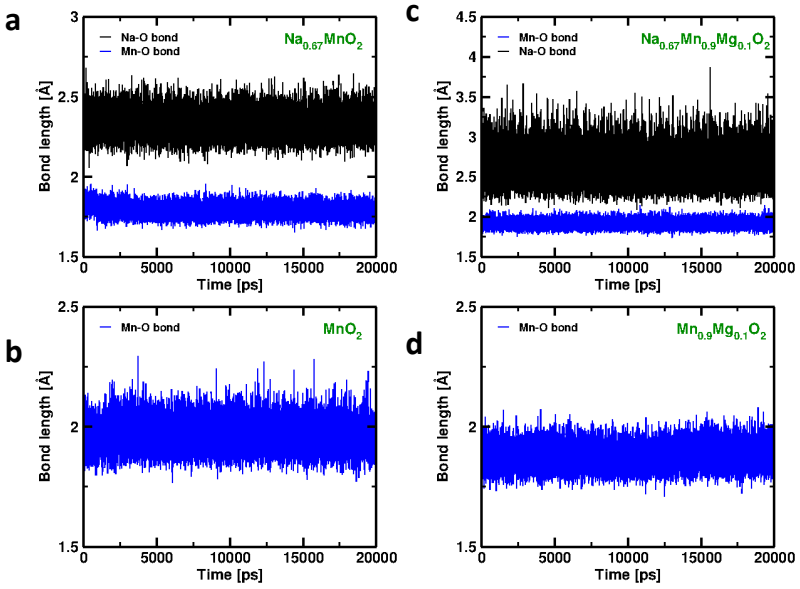

Figure S3. The fluctuation of bond length of both Mn-O and Na-O during MD simulation at 300 K of (a) $Na_{0.67}MnO_2$, (c) $Na_{0.67}Mn_{0.9}Mg_{0.1}O_2$ and only Mn-O bond length in their respective desodiated materials in (b), (d).

Table S2. The optimized coordinate file in POSCAR format for $Na_{0.67}MnO_2$

$Na_{0.67}MnO_2$
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0.0000000000000000 0.0000000000000000 11.2480001450000007
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Table S3. The optimized coordinate file in POSCAR format for $Na_{0.66}Mg_{0.1}Mn_{0.9}O_2$

$Na_{0.66}Mg_{0.1}Mn_{0.9}O_2$
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0.0000000000000000 0.0000000000000000 11.2480001450000007
Na Mn O Mg
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Direct
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