

*Electronic Supporting Information for*

**Amorphous  $\text{Si}_{1-y}\text{C}_y$  composite anode materials: *ab initio*  
molecular dynamics for behaviors of Li and Na in the  
framework**

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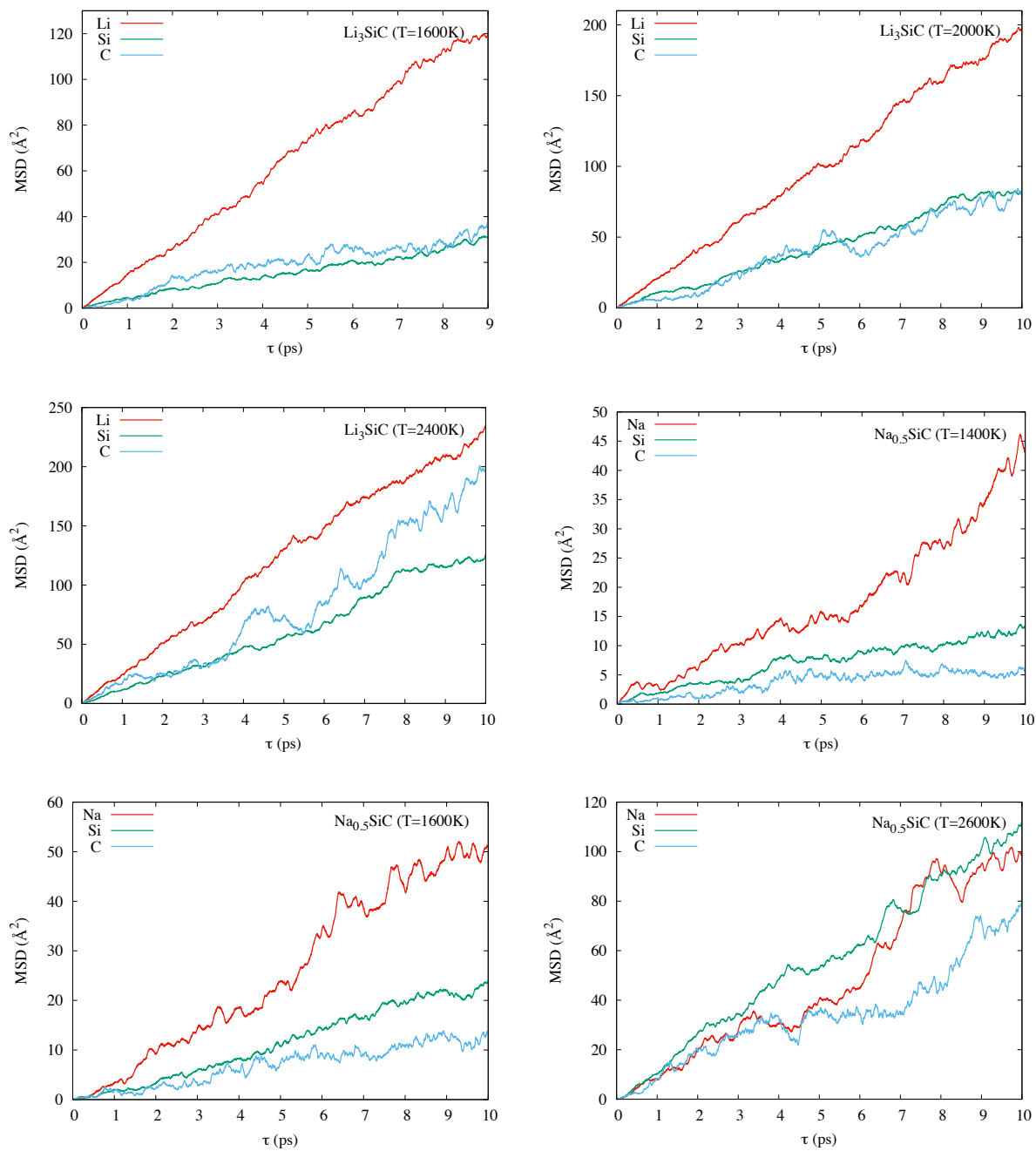
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● **Diffusion coefficient calculations**

The diffusion coefficients of Li and Na ( $D_{Li}$  and  $D_{Na}$ ) are obtained from the *ab-initio* MD simulations of a- $M_xSi_{1-y}C_y$  phase, as predicted for the diffusivity at 300 K. The Einstein relation  $D = \langle MSD \rangle / 6t$  with random walk patterns is used to estimate  $D_{Li}$  and  $D_{Na}$  values at given temperatures that are 1400, 1600, 2000, 2400, and 2600 K respectively as sampling points, and the temperatures are controlled by the Nosé-Hoover thermostat. The MSD<sup>1</sup> is defined as

$$\langle MSD \rangle \equiv \langle |\vec{r}_i(t) - \vec{r}_i(0)|^2 \rangle = \frac{1}{N} \sum_{i=1}^N |\vec{r}_i(t) - \vec{r}_i(0)|^2 \quad \text{S.1}$$

where  $N$  is the number of particles to be averaged in a selected configuration and the angle brackets mean the ensemble average over the MD time period. In this MSD equation,  $\vec{r}_i(t)$  represents a specific atom position at time  $t$ . For each alloy configuration, total time period for the AIMD simulations is up to 9 ~ 10 ps which is appropriately enough to confirm the slopes of the random walk profiles. The estimations of  $D_{Li}$  and  $D_{Na}$  values are conducted by using linear fits over the total time 9 ~ 10 ps, and an Arrhenius plot based on  $\ln(D_{Li})$  and  $\ln(D_{Na})$  as a function of reciprocal temperature ( $1000/T$ ) is used to predict  $D_{Li}$  and  $D_{Na}$  values at the three different temperature sampling points for a- $Li_xSi_{1-y}C_y$  and a- $Na_xSi_{1-y}C_y$  phases, respectively.



**Fig. S1** Mean Squared Displacement (MSD) plots extrapolated to predict the diffusion coefficients ( $D_M$  at 300 K;  $M=\text{Li}$  and  $\text{Na}$ ) of **a)** Li atoms at 1600 K, 2000 K, 2400 K and **b)** Na atoms at 1400 K, 1600 K, 2600 K in  $\text{a-Si}_{1-y}\text{C}_y$  composite structure.

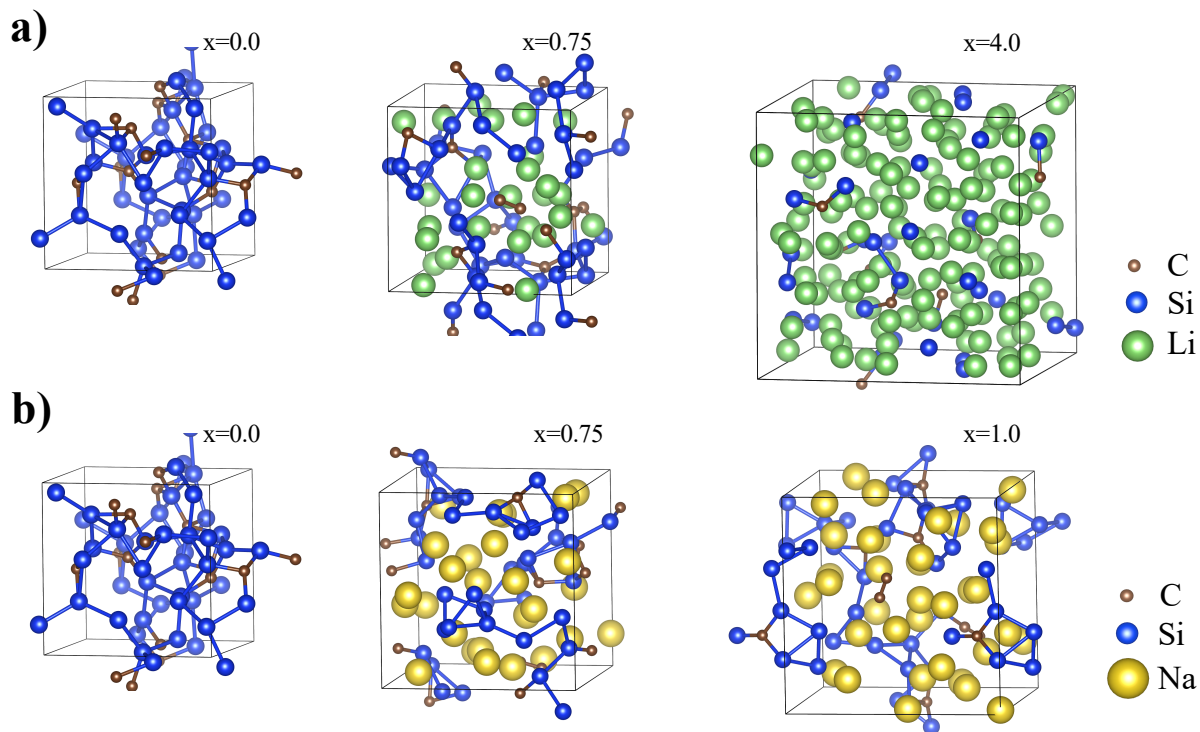
● **Theoretical capacity calculations**

The theoretical specific capacity of  $a\text{-M}_x\text{Si}_{1-y}\text{C}_y$  (M: Li and Na) is given by:

$$Q_{\text{theoretical}} = \frac{nF}{3.6 \times Mw} \quad \text{S.2}$$

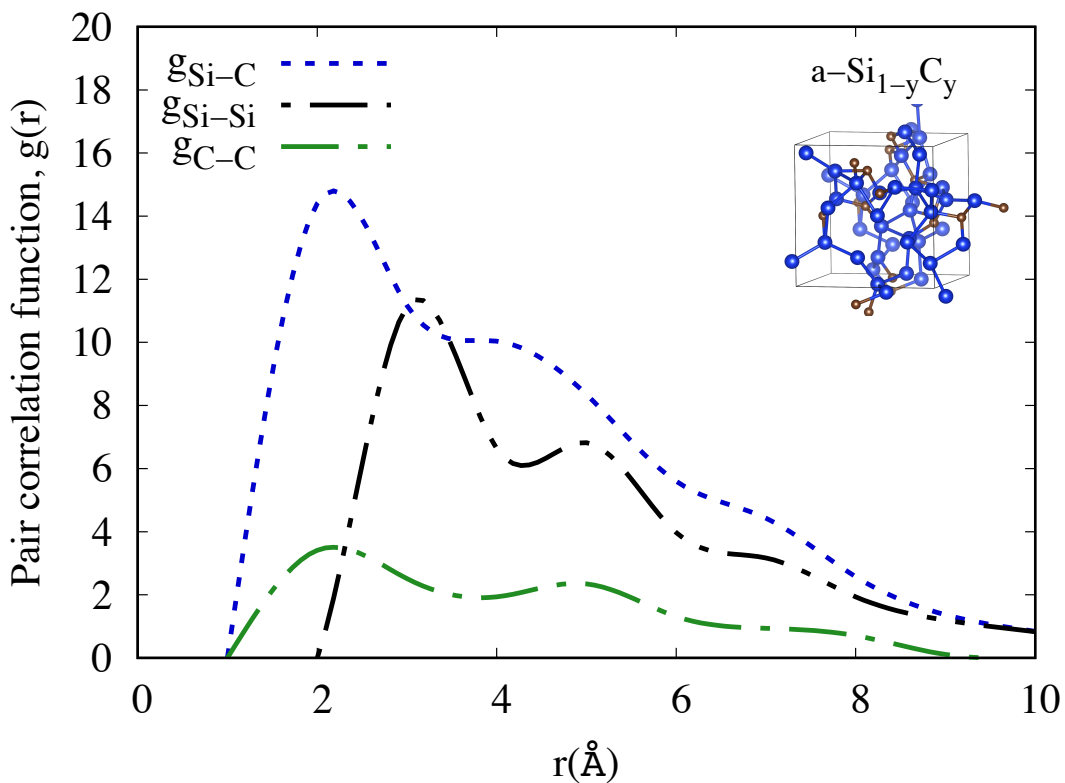
where  $n$  is the number of charge carrier,  $F$  is Faraday's constant,  $Mw$  is the molecular weight of the amorphous active materials used as the anode materials in this work.

● **Structural evolutions of  $a\text{-M}_x\text{Si}_{1-y}\text{C}_y$  ( $\text{M}_x$ :  $\text{Li}_x$  and  $\text{Na}_x$ ) phases**



**Fig. S2** Structural evolutions of  $a\text{-M}_x\text{Si}_{1-y}\text{C}_y$  ( $\text{M}_x$ : **a)**  $\text{Li}_x$  and **b)**  $\text{Na}_x$ ) phases with  $x$  ratio: 0.0, 0.75, 1.0, and 4.0.

● **Pair correlation functions of a-Si<sub>1-y</sub>C<sub>y</sub> phase**



**Fig. S3** Pair correlation functions of a-Si<sub>1-y</sub>C<sub>y</sub> phase for Si-C, Si-Si, and C-C pair interactions.

The two-body structural correlations of the amorphous phases such as amorphous silicon carbide (a-SiC) and the a-Si<sub>1-y</sub>C<sub>y</sub> composite phase are generally comparable to one another, although the unit system size, its density, and composition ratio are differently used each other. In the case of Si-C pair bonds, the position of the  $g_{\text{Si-C}}(r)$  displays the bond distance of  $\sim 2$  Å in the a-Si<sub>1-y</sub>C<sub>y</sub> phase, as shown in Fig. S3, which is close to the Si-C bond length of  $\sim 1.9$  Å in the a-SiC, and its bond length is the same as zinc-blend structure, demonstrated in previous work.<sup>2</sup> Also, the Si-Si pair bonds at 1<sup>st</sup> and 2<sup>nd</sup> coordination shells included in the a-Si<sub>1-y</sub>C<sub>y</sub> phase exhibit a similar pattern to those of the a-SiC shown in the previous work<sup>2</sup>. In the meantime, the case of the C-C pair bonds resulted from the a-Si<sub>1-y</sub>C<sub>y</sub> phase reveals that whereas the  $g_{\text{C-C}}(r)$  peak of the a-SiC which is  $\sim 3$  Å, and this might stem from the homonuclear correlations shown in the previous work<sup>2</sup>, the peak

of the  $g_{C-C}(r)$ , which is  $\sim 2$  Å in the  $a\text{-Si}_{1-y}\text{C}_y$  phase, might indicate the presence of the partial heteronuclear correlations. However, most of the C-C bond pairs shown in Fig. S3 represent that the C-C bond pair distances are larger than the nearest neighboring distances (*i.e.*, 1.43 and 1.55 Å for each) of typical graphite or diamond structures. Thus, those results mean that the  $a\text{-Si}_{1-y}\text{C}_y$  phase is relatively well randomized to be used as an amorphous structure.

### ● Reference

- 1 D. C. Rapaport, *The art of molecular dynamics simulation*, Cambridge university press, 2004.
- 2 J. P. Rino, I. Ebbsjö, P. S. Branicio, R. K. Kalia, A. Nakano, F. Shimojo, and P. Vashishta, *Phys. Rev. B*, 2004, **70**, 045207.