

## Supporting Information

### Structural Stability and Bonding Nature of Li-Sn-Carbon Nanocomposite for Anode of Li-Ion Battery: First Principles Approach

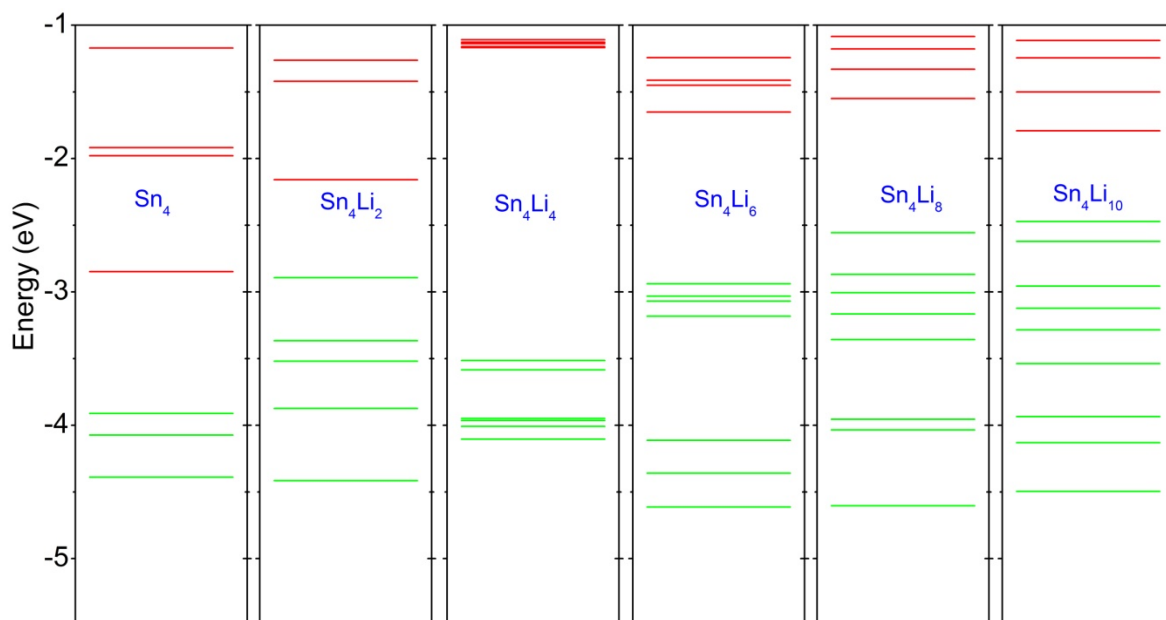
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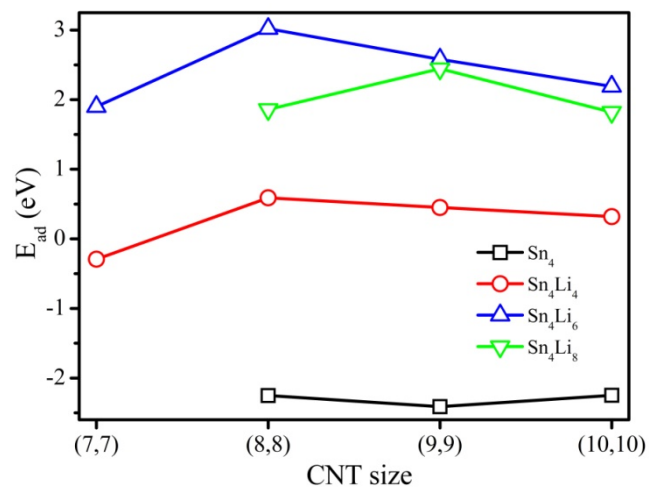
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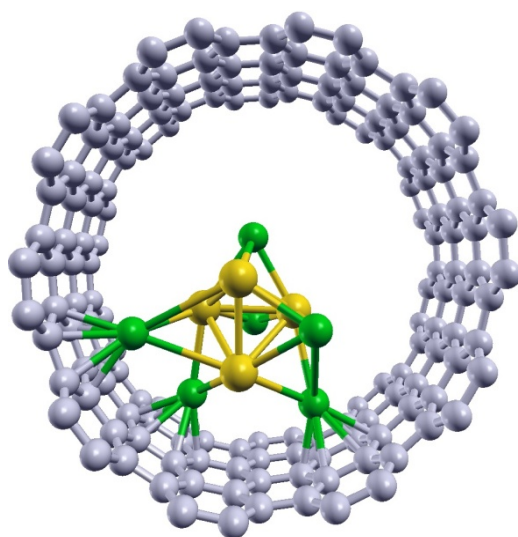
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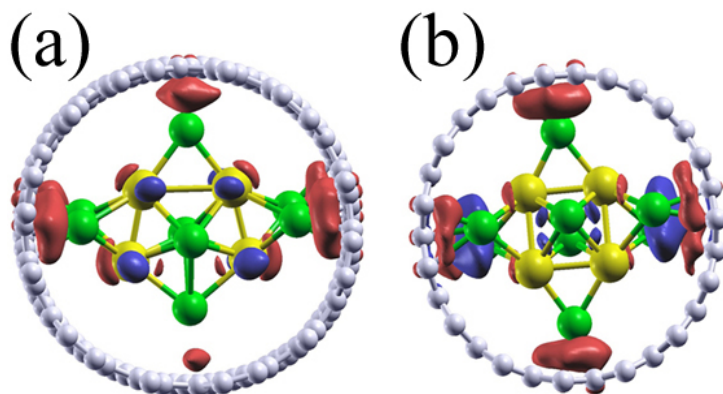
**Fig. S1** Kohn-Sham energy levels of various clusters are given. The green colored energy levels are occupied while red colored levels are empty.



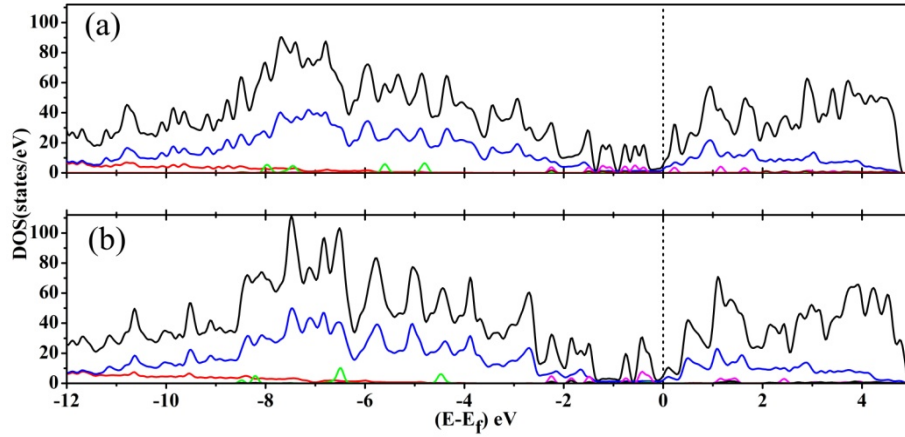
**Fig. S2** Adsorption energy ( $E_{ad}$ ) of cluster with various sized arm chair CNT. It shows that the  $E_{ad}$  value is negative for the insertion of  $\text{Sn}_4$  cluster into all CNTs



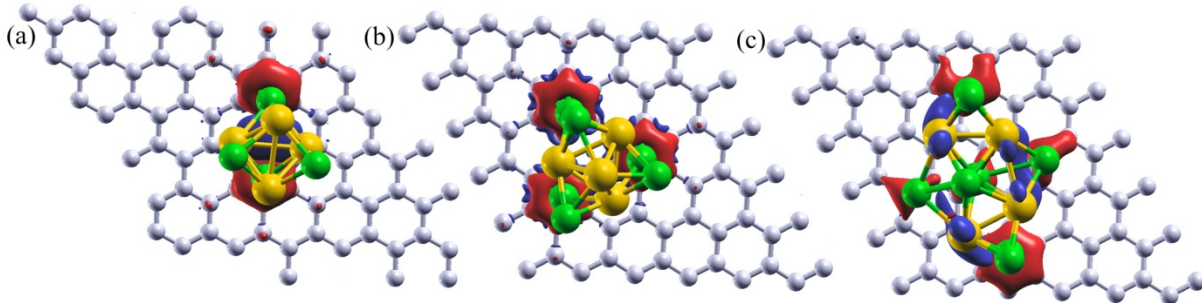
**Fig. S3** The ball and stick model of optimized structure of  $\text{Li}_6\text{Sn}_4/(10,10)$  CNT system. It shows that cluster prefers to interact with one side of CNT's wall. Grey, green, and yellow balls correspond to C, Li, and Sn atoms, respectively.



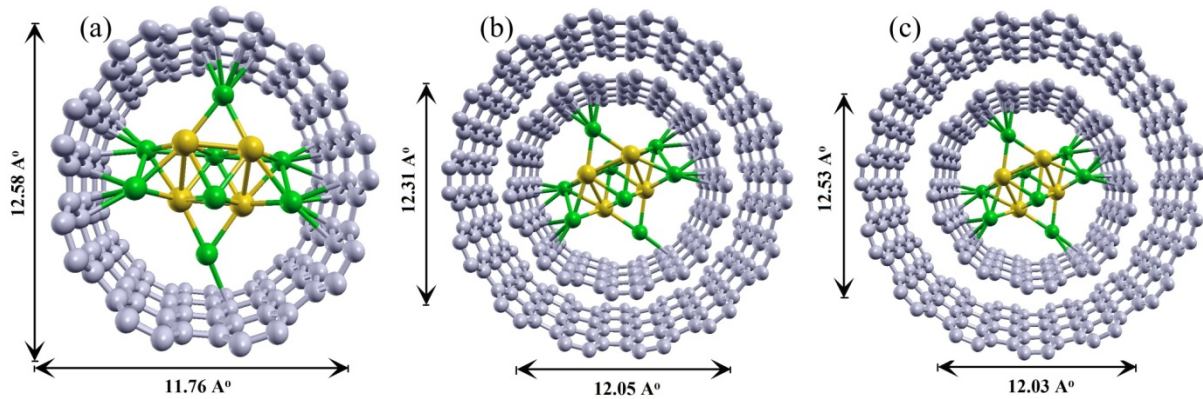
**Fig. S4** Excess and depletion charge density diagram of a)  $\text{Li}_8\text{Sn}_4/(12,6)$  CNT and b)  $\text{Li}_8\text{Sn}_4/(15,0)$  CNT system. Further details, refer figure caption of Fig. 3 (main text).



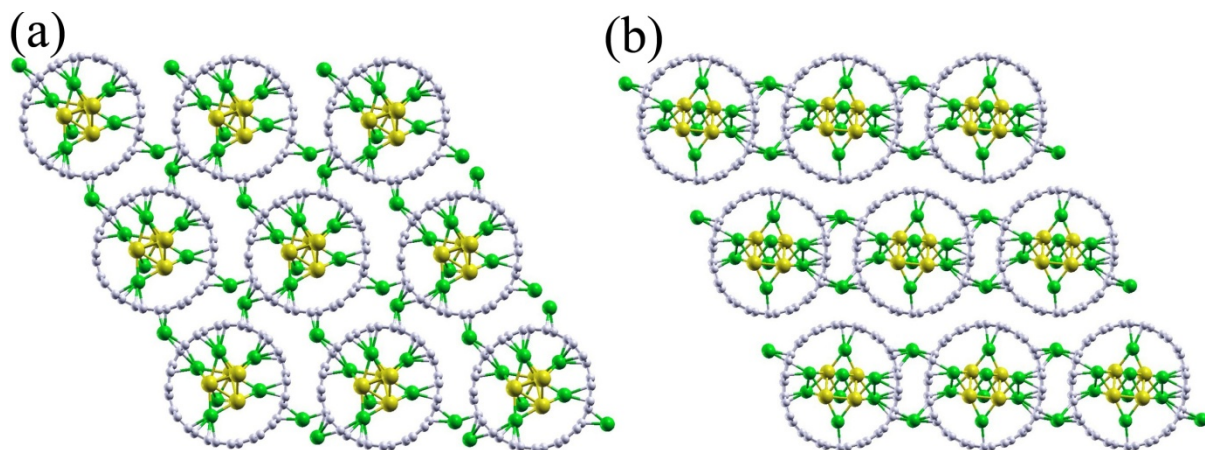
**Fig. S5** Electronic density of states of a)  $\text{Li}_8\text{Sn}_4/(12,6)$  CNT and b)  $\text{Li}_8\text{Sn}_4/(15,0)$  CNT system. Total, C-2s, C-2p, Sn- 5s and Sn-5p states are represented by black, red, blue, green, and pink solid lines, respectively.



**Fig. S6** Excess and depletion charge densities diagram of a)  $\text{Li}_4\text{Sn}_4/\text{Gr}$ , b)  $\text{Li}_6\text{Sn}_4/\text{Gr}$  and c)  $\text{Li}_8\text{Sn}_4/\text{Gr}$  system. Further details, refer figure caption of Fig.3 (main text).



**Fig. S7**  $\text{Li}_8\text{Sn}_4$  is inserted into a) (9,9) CNT and (b - c) double walled CNTs (DWCNT) with inter wall distance of 3.47 Å and 3.94 Å , respectively. As discussed in the main text, the SWCNTs undergo deformation from their regular cylindrical shape to elliptical one, when  $\text{Li}_8\text{Sn}_4$  cluster was inserted. Hence, similar study was performed on DWCNT modelled by inserting (9,9) CNT into both (14,14) and (15,15) CNTs. These DWCNTs could explain the reduced structural deformation observed in the MWCNT/Sn composites. Note that the actual construction of DWCNT will lead to different chiral vectors for inner and outer walls, due to AB stacking of graphite. However, the AA stacking of bilayer graphene could lead to same chirality for both the walls. Note that for bilayer graphene, AA stacking is energetically more favourable than AB stacking. It enforces us to design the DWCNT with inner CNT as (9,9) armchair which is rolled by outer wall dimension of, either (14,14) or (15,15). This study is essential as the interwall separation of the constructed DWCNT are lied near to the actual inter wall distance of 3.35 Å . Based on our first principles calculations, the deformation parameters ( $a/b$  ratio,  $|a - b|$ ) of inner wall of DWCNT shown in (b) and (c) are (1.02, 0.26 Å ) and (1.04, 0.50 Å ) , respectively. Thus, the insertion of Sn-Li cluster into rigid CNT is found to be an ingenious way to control the deformation of CNT.



**Fig.S8** Li ion intercalation in hexagonal assembly of a)  $\text{Li}_6\text{Sn}_4/(8,8)$  CNT and b)  $\text{Li}_8\text{Sn}_4/(9,9)$  CNT. In latter case, Li ions prefer to interact with two CNTs, rather three CNTs due to its deformation, from regular CNT to elliptical CNT.