Improving Energy Materials with Aberration-corrected STEM and Theoretical Calculations

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Today's aberration-corrected scanning transmission electron microscope (STEM) allows direct, real space imaging at atomic resolution with low accelerating voltages to minimize damage [1,2]. Combined with theoretical calculations, such capabilities represent an unprecedented opportunity to unravel the functionality of energy materials and improve their performance. Three case studies will be presented. The first shows how atomic-resolution imaging and electron energy loss spectroscopy (EELS) reveal nanoporous carbons to comprise defective graphene sheets with a high density of 5- and 7-membered ring defects, as shown in Fig. 1 [3]. The sheets become rumpled, and therefore unable to stack into graphite, leaving nanopores between the sheets that are the origin of the enhanced gas absorption.

The second example shows how single Nb atoms are identified as the sites of high catalytic activity in niobium–carbon catalysts for the oxygen-reduction reaction in fuel cells. The single atoms are trapped within the graphitic carbon sheets (Fig. 2), and increase the catalytic efficiency two orders of magnitude, at the same time giving the catalyst high stability [4].

The final example shows how Z-contrast STEM, EELS and density functional calculations can be used to determine the electrical activity of defects in CdTe solar cells. Dislocations are commonly assumed to act as recombination sites for electrons and holes, reducing cell efficiency, however, these results show that partial dislocation pairs induce band bending that may instead enhance carrier separation and improve cell efficiency [5]. Similarly, grain boundaries are also commonly assumed to be recombination sites, and electron beam induced current (EBIC) imaging shows that this is indeed the case in as-grown material. However, after a heat treatment in $CdCl_2$, EBIC imaging shows the grain boundaries are sites of high current generation. Z-contrast imaging of the structure, combined with EELS and density functional calculations reveals that the Cl substitutes for a high fraction of the Te sites at the grain boundary, turning the material n-type, and making each grain boundary a p-n junction that assists carrier separation. This result provides an explanation of why polycrystalline solar cells are more efficient than single crystal cells [6].

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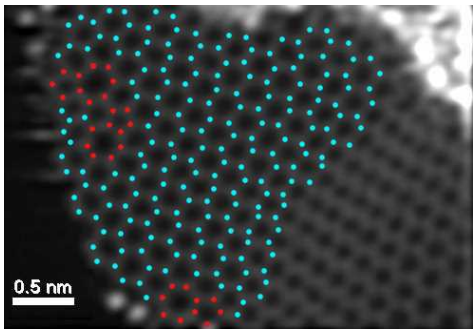

FIG. 1. Annular dark-field STEM image of ultramicroporous carbon showing a graphene layer with 5- and 7- membered defects. [3].

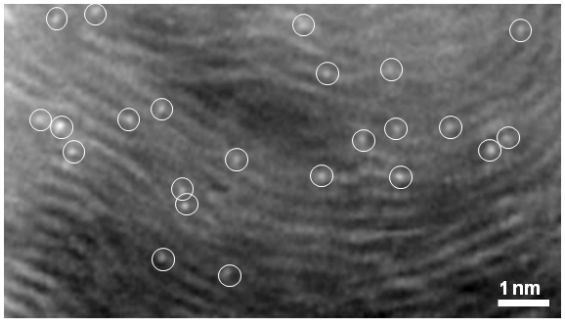

FIG. 2. Annular dark-field STEM image of a Nb-C catalyst showing single Nb atoms trapped in graphitic carbon [4].

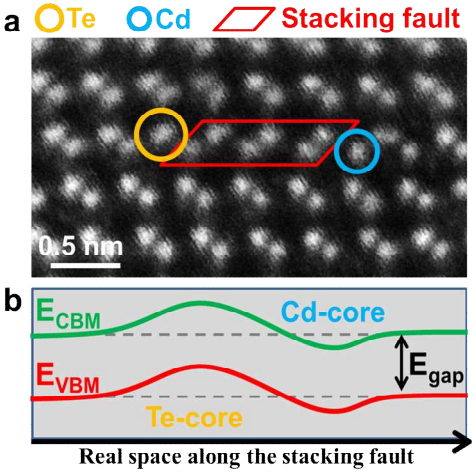

FIG. 3. (a) Z-contrast image shows Cd and Te single atom columns (blue and yellow circles) at two ends of an intrinsic stacking fault (red box). (b) Sketch map shows band bending along the stacking fault. [5].

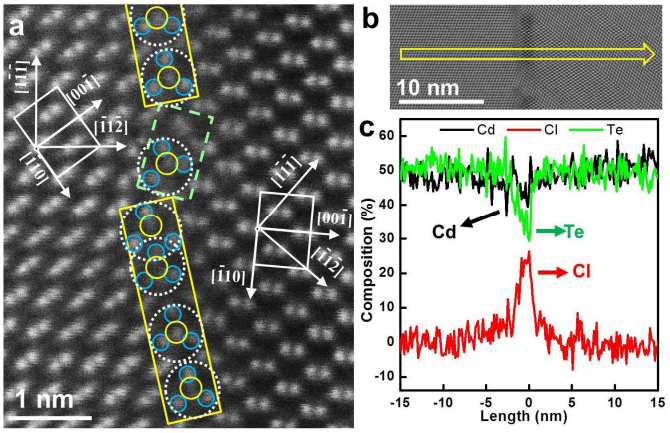

FIG. 4. (a) Structure of a Σ9 GB. (b) Composition profile showing significant Cl enrichment and Te depletion at the GB plane in (c). Blue and yellow circles indicate Cd and Te columns, respectively [6].