Electronic Supplementary Information

Nanoscale iron (oxyhydr) oxides modified carbon nanotube filter for rapid and effective Sb(III) removal

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Bed volume calculations

To quantitatively evaluate filtration capabilities, single-pass filtration was conducted under the experimental condition that a 100 or 200 μg/L of Sb(III) solution was pumped though a iron (oxyhydr) oxides/CNT filter at pH 7, and the effluent was no longer returned, the filter bed volume was 0.113 mL, and empty bed contact time (EBCT) was 4.5 s at a fixed flow rate of 1.5 mL/min. The number of bed volume was calculated by following equation:

$$Bed\ Volume = \frac{1.5 \times t}{0.113}$$

Where t is filtration time (min).

Method of DFT calculations

Density function theory calculation were p erformed by using the CP2K package.[1] PBE functional[2] with Grimme D3 correction[3] was used to describe the systemm. Unrestricted Kohn-Sham DFT has been used as the electronic structure method in the framework of the Gaussian and plane waves method.[4, 5] The Goedecker-Teter-Hutter (GTH) pseudopotentials,[6,7] DZVPMOLOPT-GTH basis sets[4] were utilized to describe the molecules. A plane-wave energy cut-off of 500 Ry has been employed. It's known that the generalized gradient functional fails to reproduce the correlated elements, and the "+U" Hubbard correction[8] improve the electronic descriptions. Accordingly, we adopt DFT+U method with effective U value is equal to 5 eV for Fe 3d states.[9]

A six atomic layer (4×4) hematite Fe_2O_3 (001) surface and six-layer αFeOOH (100) surface have been used in the simulation. As the Fe_2O_3 is anti-ferromagnetic, we initially set the magnetic moments layer by layer as the spin polarization of neighboring layers are opposite. A vacuum of 15 Å has been added along Z-direction to decouple the interaction between images. The adsorption energies (E_{ad}) is calculated using:

$$E_{ad}=(E_{sur}+E_{mol})-E_{mol/sur}$$

Where the $E_{mol/sur}$ is the total energy of the adsorbate binding with surface, E_{sur} and E_{mol} are the isolated energy of surface and molecule, respectively.

Reference

1. J. Hutter, M. Iannuzzi, F. Schiffmann and J. VandeVondele, *Wires. Comput. Mol. Sci.*, 2014, **4**, 15-25.
2. J. Perdew, K. Burke and M. Ernzerhof, *phys. Rev. lett.,* 1996, **77**, 3865.
3. S.Grimme, *J. comput. Chem.,* 2006, **27**,1787–1799.
4. J. VandeVondele and J. Hutter, *J. chem.. phys.*, 2007, **127**, 114105,
5. J. VandeVondele, M. Krack, F. Mohamed, M. Parrinello, T. Chassaing,and J. Hutter, *Comput. Phys. Commun.*, 2005, **167**, 103–128.
6. S. Goedecker, M. Teter, and J. Hutter, *Phys. Rev. B*, 1996, **54**, 1703.
7. C. Hartwigsen, S. Gœdecker, and J. Hutter, *Phys. Rev. B*, 1998, **58**, 3641.
8. S. Dudarev, G. Botton, S. Savrasov, C. Humphreys, and A. Sutton, *Phys. Rev. B*, 1998, **57**, 1505.
9. L. Wang, T. Maxisch, and G. Ceder, *Phys. Rev. B*, 2006, **73**, 195107.

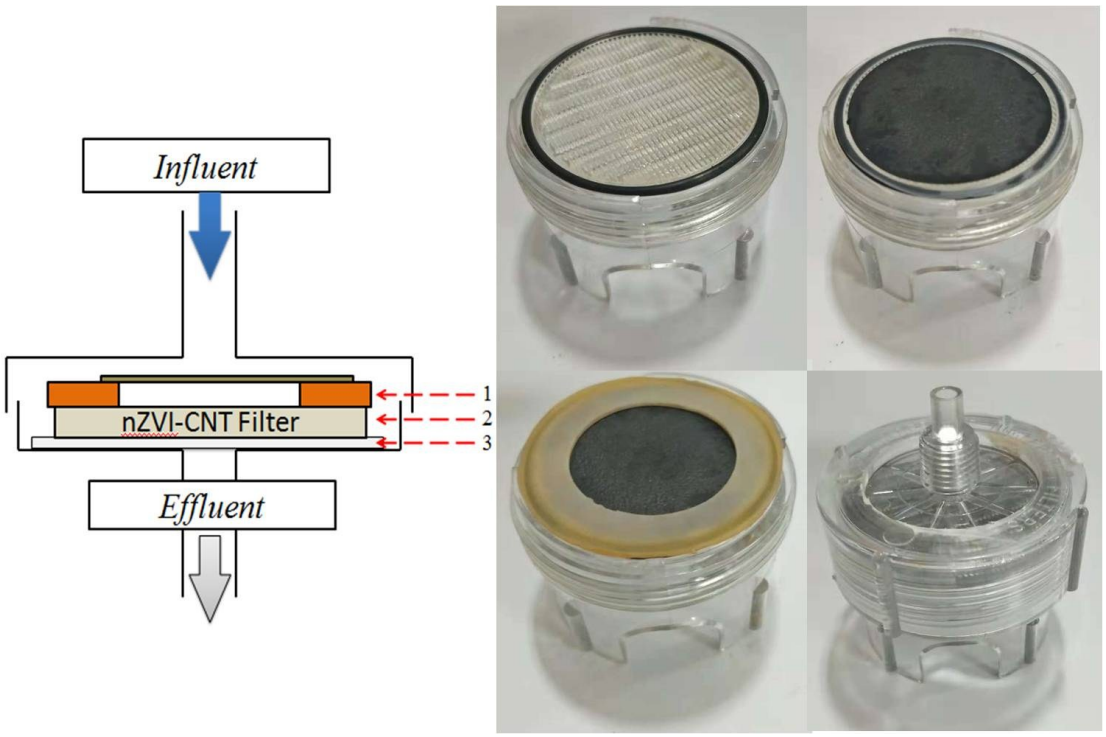

Fig. S1 Schematics of filtration apparatus: (1) a ring-shaped silicone rubber separator and seal; (2) a iron (oxyhydr) oxides/CNT Filter; (3) a PTFE membrane support.

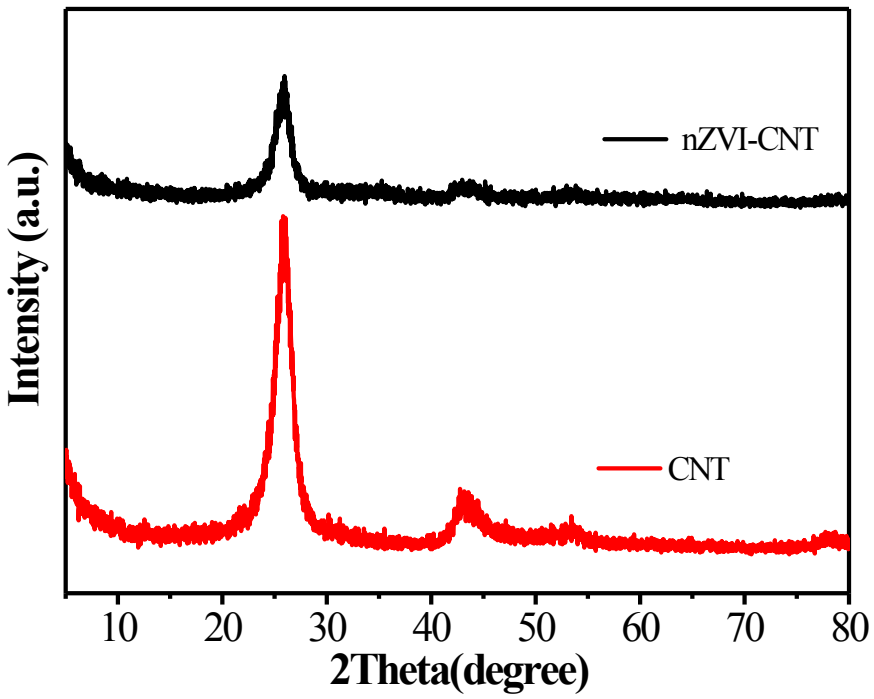

Fig. S2 XRD patterns of the CNT and iron (oxyhydr) oxides/CNT.

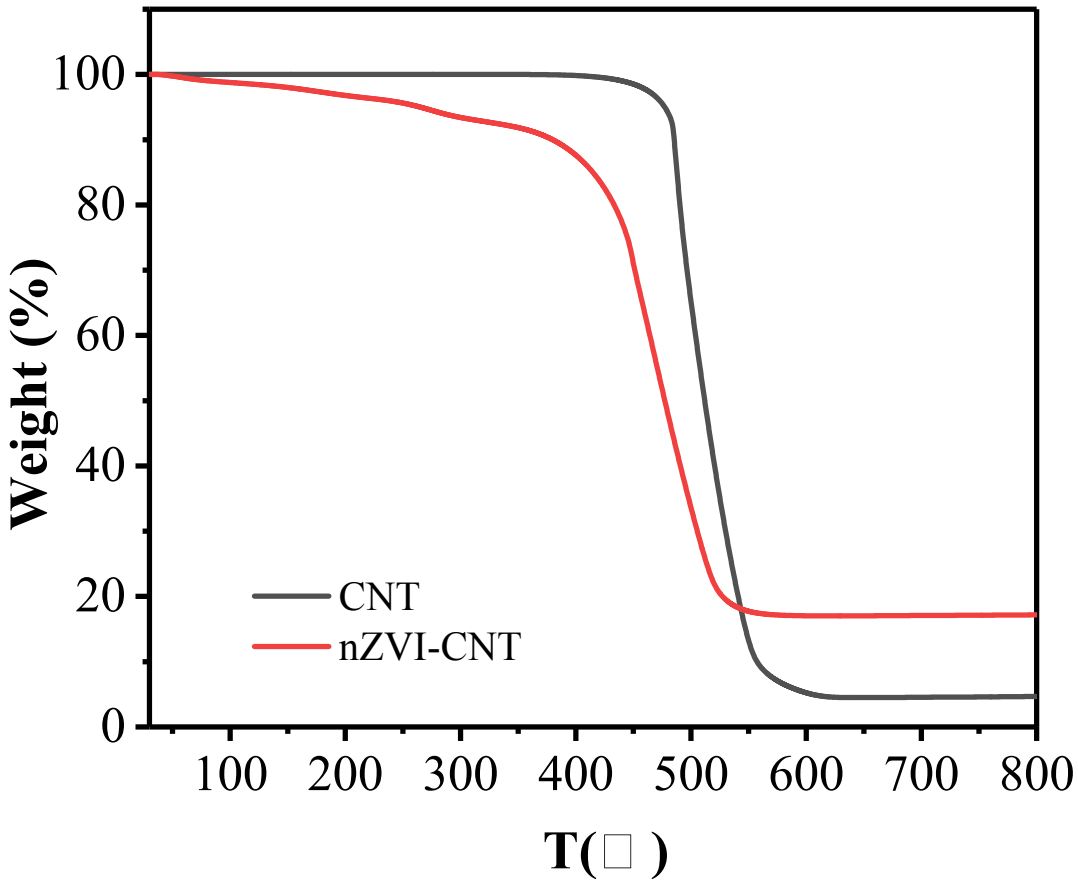

Fig. S3 TGA image of the CNT and iron (oxyhydr) oxides/CNT.

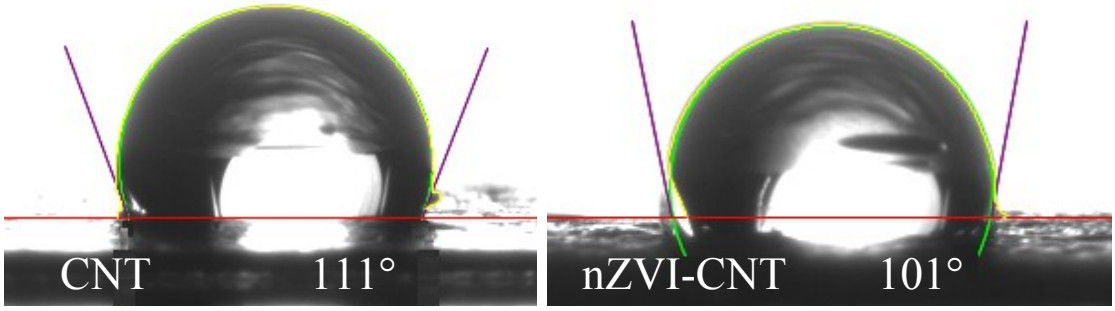

Fig. S4 CNT and iron (oxyhydr) oxides/CNT network contact angle measurement after 30s. Both CNT network and iron (oxyhydr) oxides/CNT network were hydrophobic with contact angle of 111° and 101° respectively.

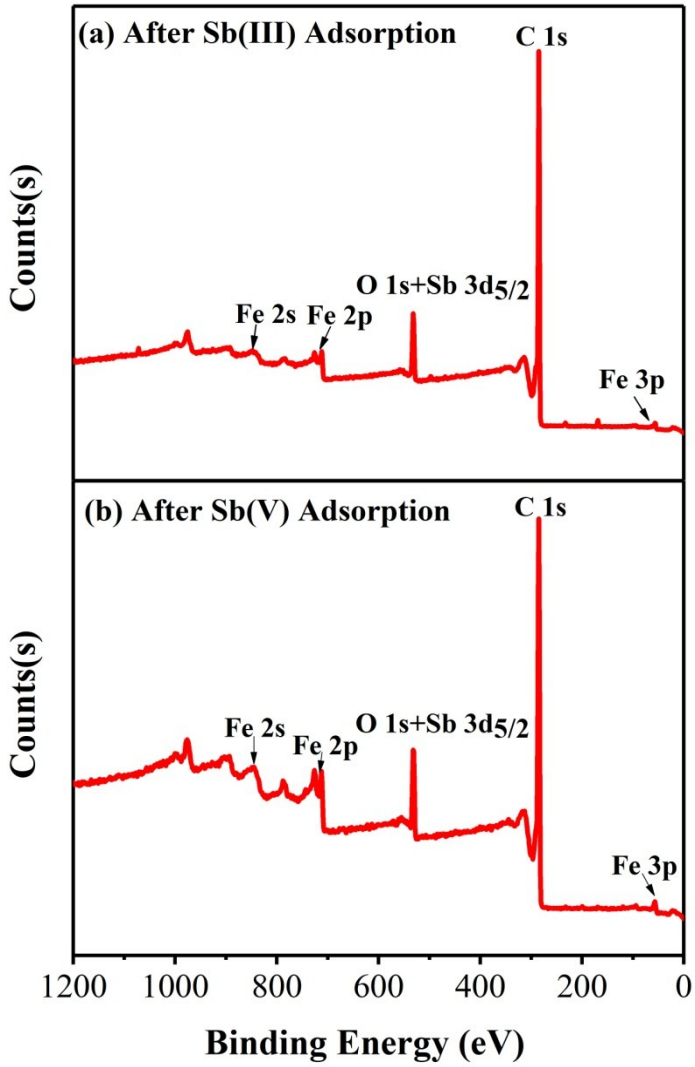

Fig. S5 XPS Survey scan for iron (oxyhydr) oxides/CNT filter (a) After Sb(III) adsorption and (b)after Sb(V) adsorption.

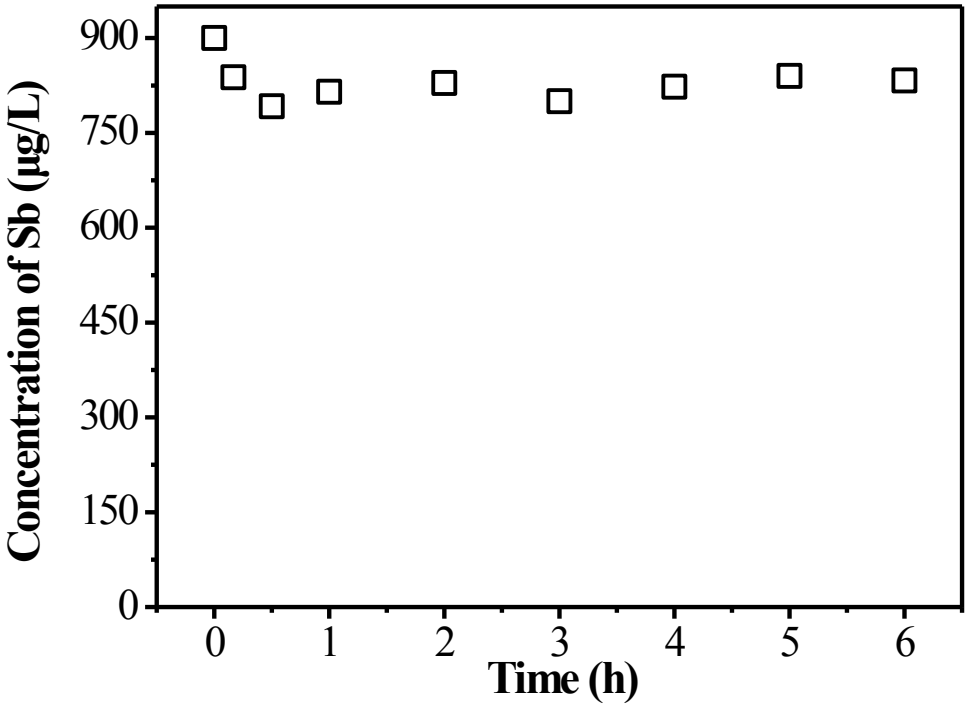

Fig. S6 Sb(III) sorption on the pure CNT filter in recirculation mode at a flow rate of 3 mL/min.

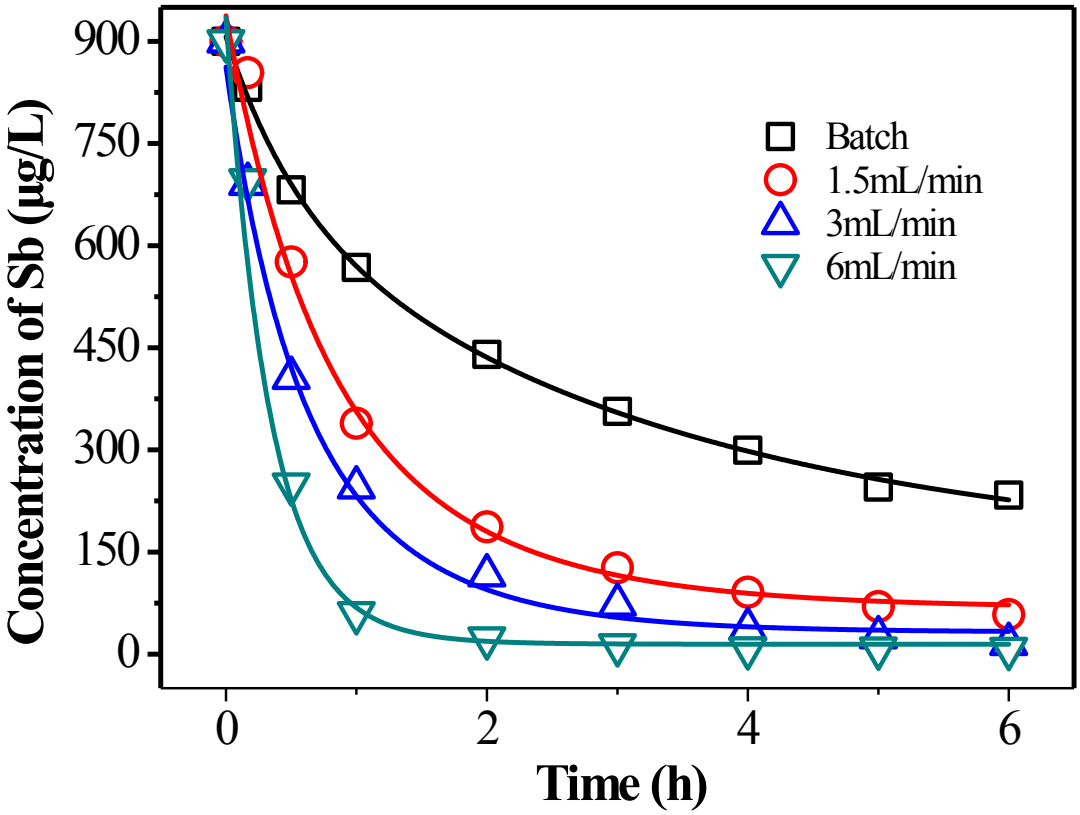

Fig. S7 The change in concentration of Sb with time when 100mL 900µg/L Sb solution was reacted with a iron (oxyhydr) oxides/CNT filter, and experiment was conducted in batch mode and recirculation mode (flow rate of 1.5mL/min, 3mL/min and 6 mL/min).

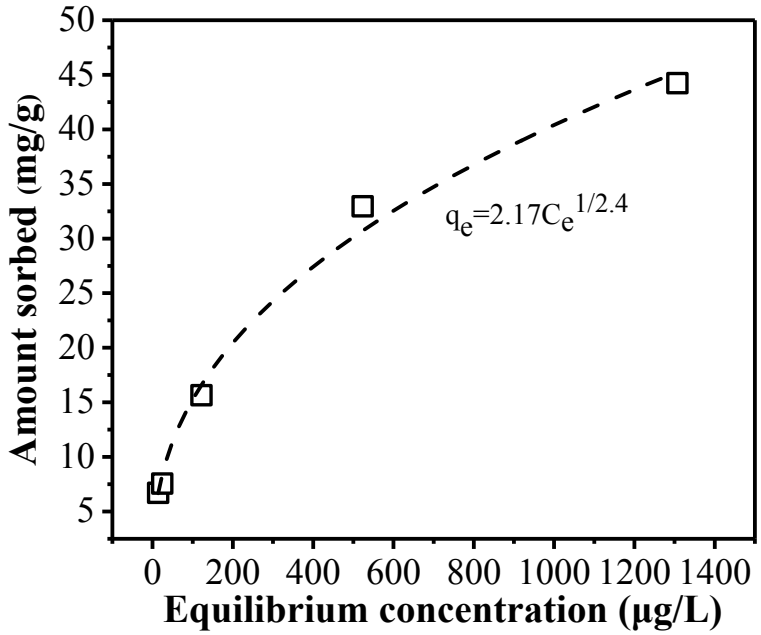

Fig. S8 Iron (oxyhydr) oxides/CNT Sb sorption isotherms. Experiments were conducted in recirculation mode for 12 h using a 100 mL reservoir, flow rate was 3ml/min. Experimental data are fittings to the Freundlich isotherm model, and correlation coefficients>99%.

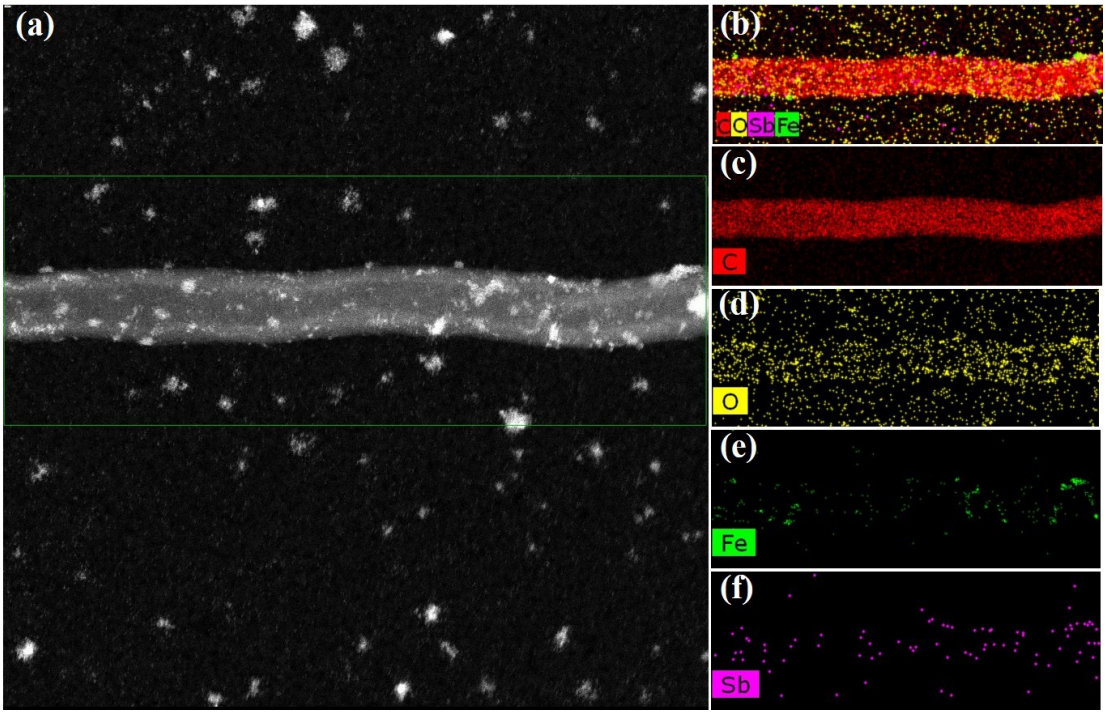

Fig. S9 HAADF image (a); EDS mapping (b) of Sb-loaded iron (oxyhydr) oxides/CNT; and the corresponding EDS elemental C(c), O(d), Fe(e) and Sb(f) maps.

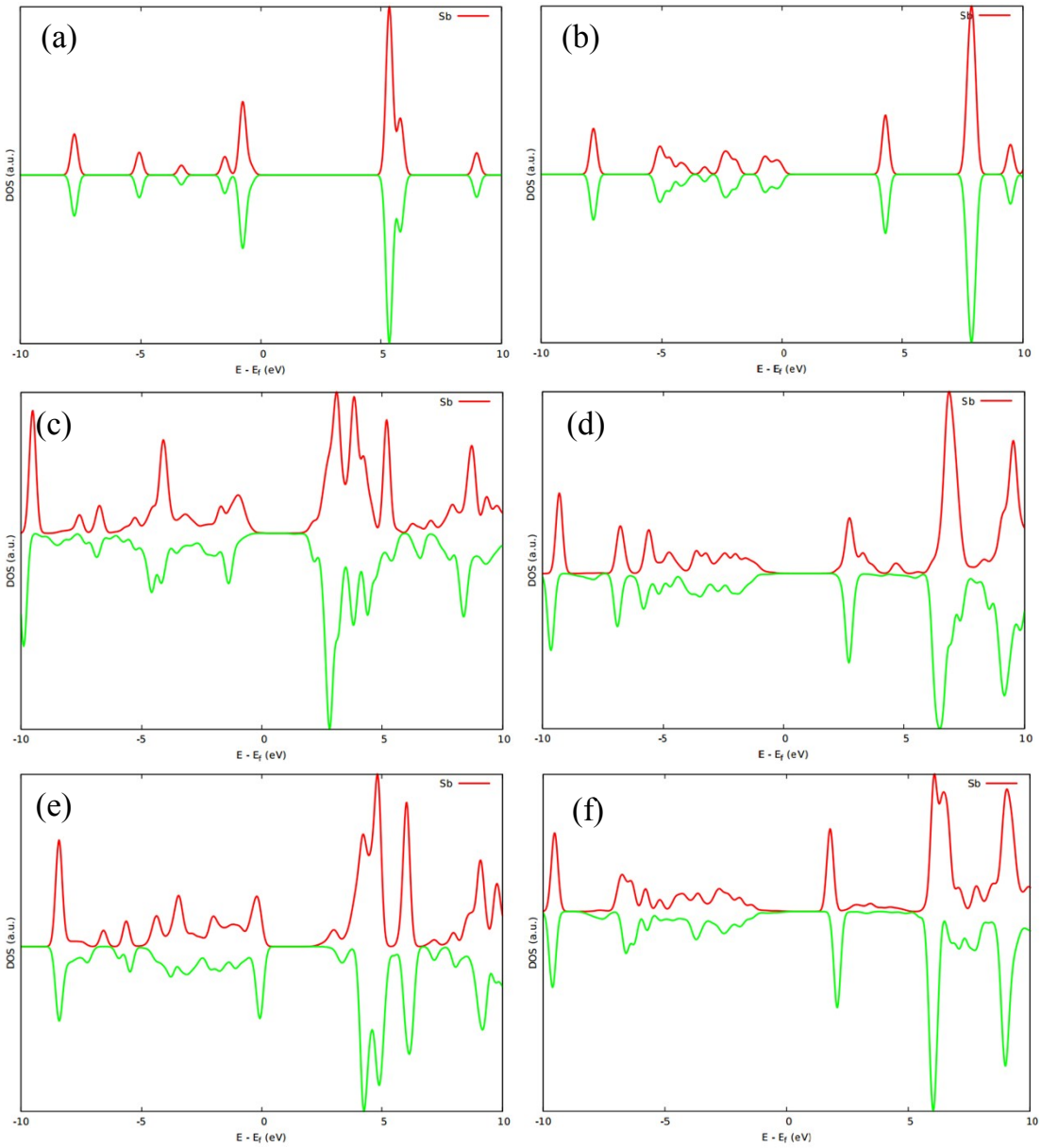

Fig. S10 The projected density of state for Sb adsobed on FeOOH and Fe_2O_3 surface. Dos of isolated Sb(V) (a), isolated Sb(III)(b), Sb(III) on FeOOH(c), Sb(V) on FeOOH(d), Sb(III) on Fe_2O_3(e), Sb(V) on Fe_2O_3(f).