

# Tuned Hydrogen Bonding in Rare Earth MOFs for Design of Optical and Electronic Properties: An Exemplar Study of Y-DOBDC

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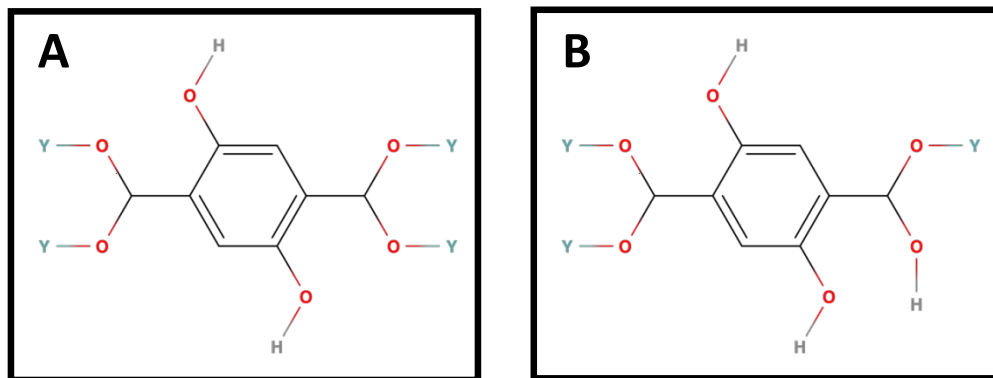
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**Table S1.** Total number of H-bonds for each H-bonding configuration % and on which type of ligand the H-bonds are located.

H-Bonding %	Number of H-bonds	Monodentate H-Bonds	Bidentate H-Bonds
0	0	0	0
20	4	4	0
40	8	0	8
60	12	4	8
80	16	0	16
100	20	4	16

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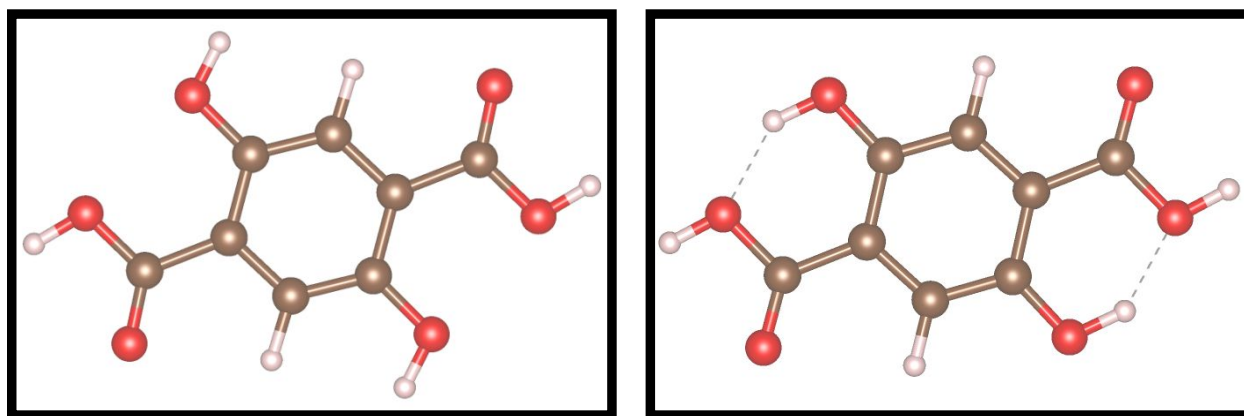
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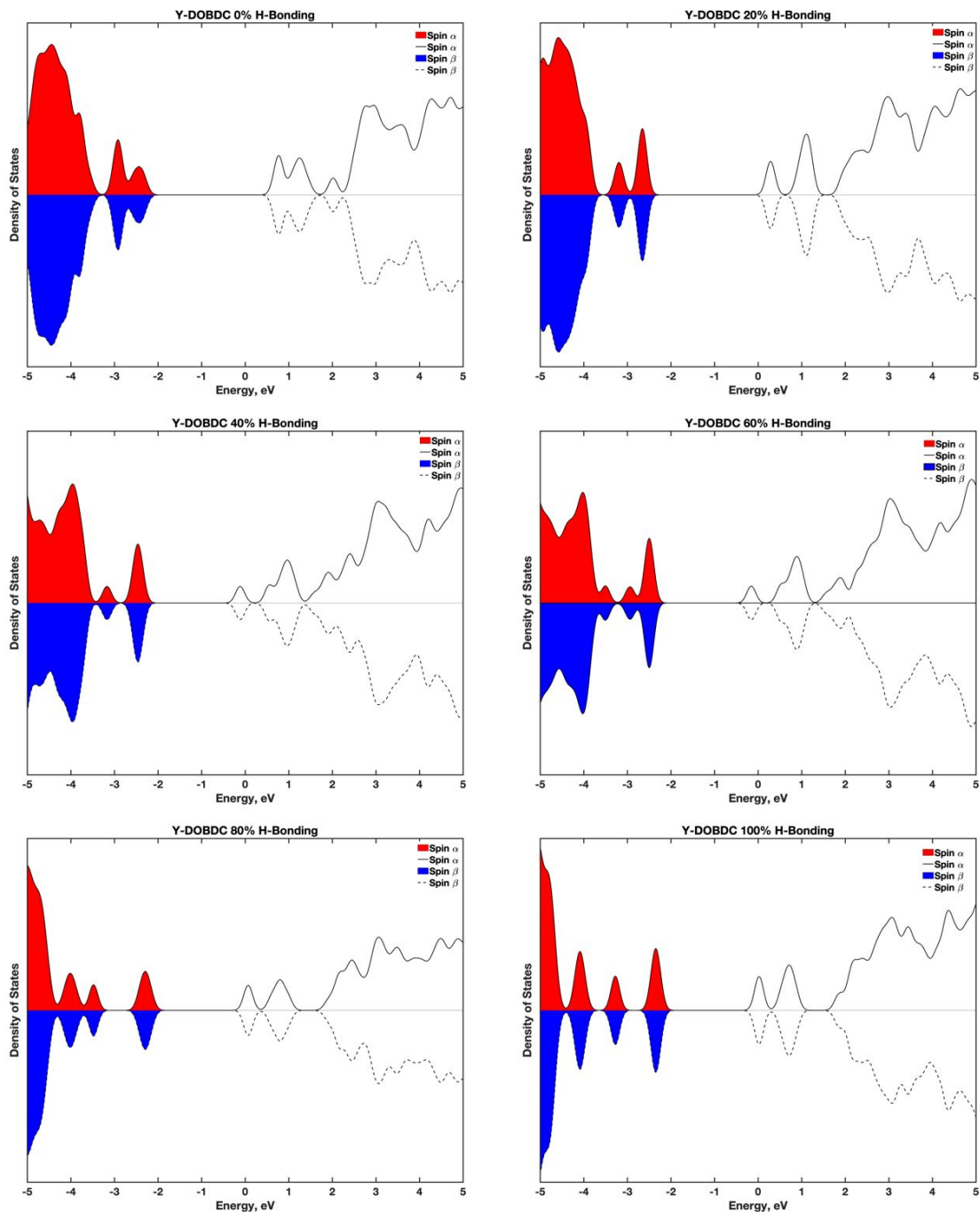
**Figure S1.** Two binding structures of the DOBDC linkers to the metal clusters in a (A) bidentate and (B) monodentate fashion.

**Table S2.** Calculated total energies and calculated average H-bond strength of isolated H<sub>4</sub>-DOBDC.

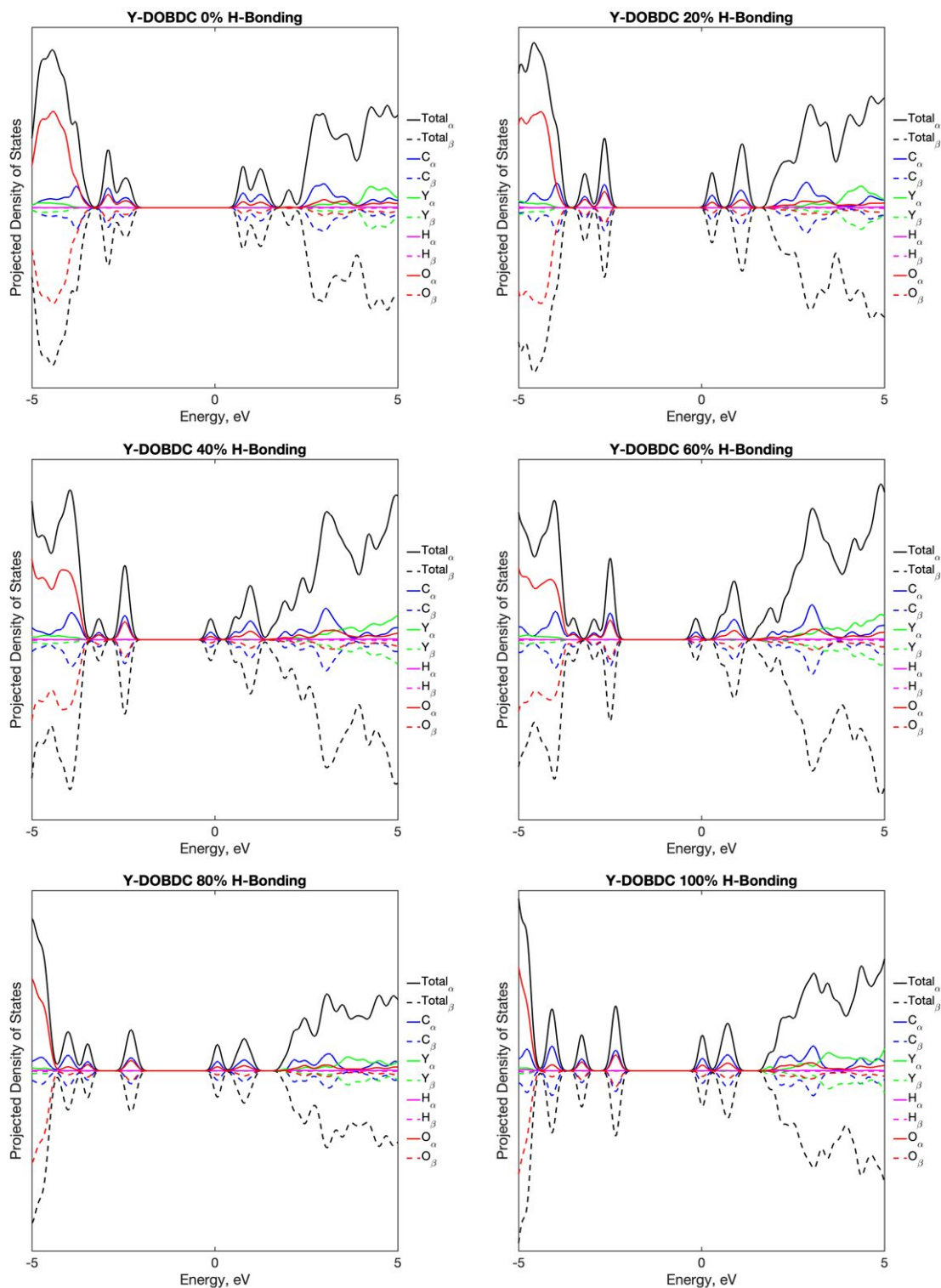
H-Bonding Configuration	Total Energy (eV)	H-Bond Strength	kJ/mol
Interacting	-140.02	$E_{IHB}$	27.75
Not Interacting	-139.44		



**Figure S2.** Calculated optimized geometries of isolated H<sub>4</sub>-DOBDC molecules with no H-bonding (left) and H-bonding (right). Colors: H (white), O (red), C (grey)



**Figure S3.** Calculated density of states for Y-DOBDC with H-bonding configurations from 0-100% using HSE06. Occupied (shaded) and unoccupied (unshaded) electronic states are plotted for both the spin alpha (red/solid line) and spin beta (blue/dotted line) projections.



**Figure S4.** Projected density of states (PDOS) for Y-DOBDC with H-bonding configurations from 0-100%, calculated with the HSE06 hybrid functional. Each PDOS plot provides the total DOS (black) and the contribution of the individual elements: C (blue), Y (green), O (red), H (pink). Each PDOS is plotted for both the spin up (solid lines) and spin down (dashed lines) projection.

### Calculated optical spectra.

The DFT calculated ground state electronic structures provide solutions to solving the one electron Kohn-Sham (KS) equation in the form of Kohn-Sham orbitals (KSOs),  $\varphi_{\sigma,i}^{KS}$ , and their respective energies,  $\varepsilon_{i,\sigma}$ . The use of spin unrestricted DFT provides solutions for both spin up ( $\alpha$ ) and spin down ( $\beta$ ) projections, indicated by  $\sigma$ . From the calculated KSOs the transition dipole moment can be calculated with spin conservation in the independent orbital approximation between two states  $i$  and  $j$ ,  $\vec{D}_{\sigma,ij} = e \int \varphi_{\sigma,i}^{KS*} \vec{r} \varphi_{\sigma,j}^{KS} d\vec{r}$ . The transition dipole moment is then used to calculate the oscillator strength,  $f_{\sigma,ij}$ , for the transition  $ij$ , as  $f_{\sigma,ij} = |\vec{D}_{\sigma,ij}|^2 \frac{24\pi m_e \omega_{ij}}{3\hbar e^2}$ , where  $\omega_{ij}$  is the energy of the transition. The oscillator strength is used to weight the absorption of the transition,  $a_{\sigma,ij}$ , allowing for the calculation of the absorption spectrum as  $a_{\sigma}(\varepsilon) = \sum_{\sigma,ij} f_{\sigma,ij} \delta(\varepsilon - \Delta\varepsilon_{\sigma,ij})$ ;  $a(\varepsilon) = a_{\alpha} + a_{\beta}$ .