S1: The effect of OH on g-C_3N_4

In order to estimate the effect from OH after the water has been split into H and OH group, we calculated the model shown in Figure S1 (namely $(OH)_1$/hole). The calculations were performed in the same scheme as those in the main body of the present work. Our calculation shows that the OH is adsorbed on g-C_3N_4 via a chemical bond (C-O bond), which is different to the adsorption of H_2O. The quasiparticle band gap is 5.033 eV, which is 0.4 eV larger than that of pure bg-C_3N_4. The optical response of $(OH)_1$/hole, obtained by BSE, was also plotted in Figure S1. It shows the first and second bright exciton (I^1 and I^2) is located at 2.98 eV and 3.11 eV, respectively. This indicates the adsorption of OH group hardly helps the visible light absorption for the reaction.

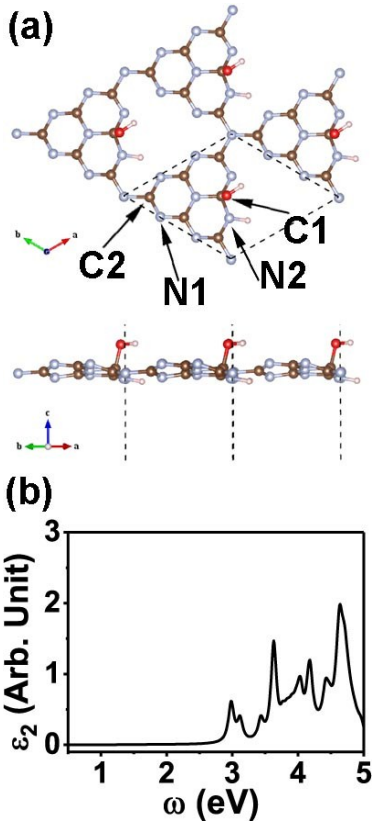

Figure S1 (a) The geometry structure of OH adsorbed g-C_3N_4 ($(OH)_1$/hole). The silver, brown, white and red balls represent for C, N, H and O atoms, respectively. (b) The optical response of $(OH)_1$/hole by BSE.

The three-dimensional electron probability distribution of I^1 in real space as a function of electron position ($|\psi(r_e; r_h)|^2$) were plotted in Figure S2, as well as the partial density of states (PDOS). The excitons distribute on both g-C_3N_4 and OH, but hardly on the H atom connected to N atom. The PDOS (Figure S2c) indicates the 2p orbitals of C and N atom connected by OH and H locate farther away from Fermi level, which contribute less to the visible light absorption. In general, we conclude

that adsorption of OH group has opposite influence on the excitonic properties, compared to the adsorption of H_2O molecules.

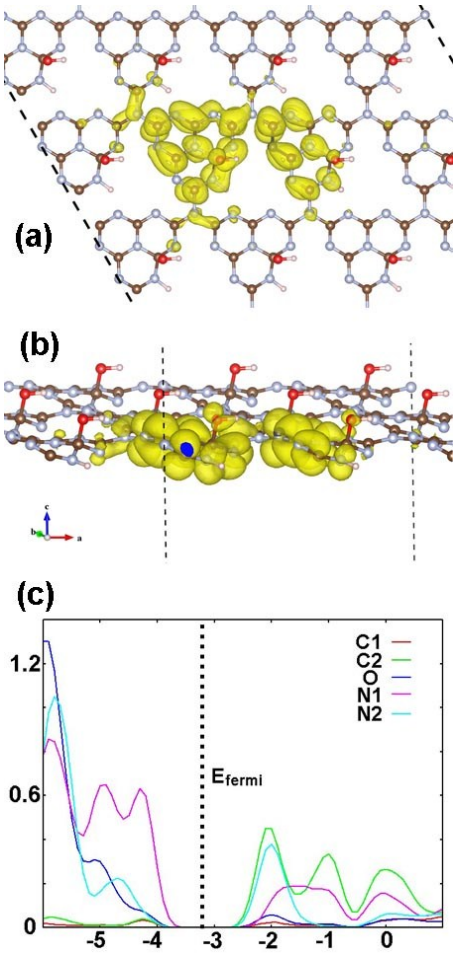

Figure S2 (a) Top view and (b) zoom in view of the three-dimensional electron probability distribution of I^1 in real space as a function of electron position ($|\psi(r_e; r_h)|^2$) for $(OH)_1$/hole. (c) The PDOS of $(OH)_1$/hole, where different atoms are labeled in Figure S1.