

Fig.S1 depicts the structures of intermediates, transition states, and reaction products involved in the C-H and C-S bond cleavage reaction pathways of $\text{C}_2\text{H}_6\text{OS}$ upon adsorption onto the pure $\text{CuO}(111)$ surface. IM(I) is the most stable adsorption structure formed by $\text{C}_2\text{H}_6\text{OS}$ on the pure $\text{CuO}(111)$ surface, TS1 is the transition state formed when IM(I) breaks the C-H bond, P1 is the decomposition product corresponding to TS1. TS2 is the transition state formed when IM(I) breaks the C-S bond and CH_3 bonds with the surface O atom, P2 is the decomposition product corresponding to TS2. TS3 is the transition state formed when IM(I) breaks the C-S bond and CH_3 is bonded to the surface Cu atom, P3 is the decomposition product corresponding to TS3.

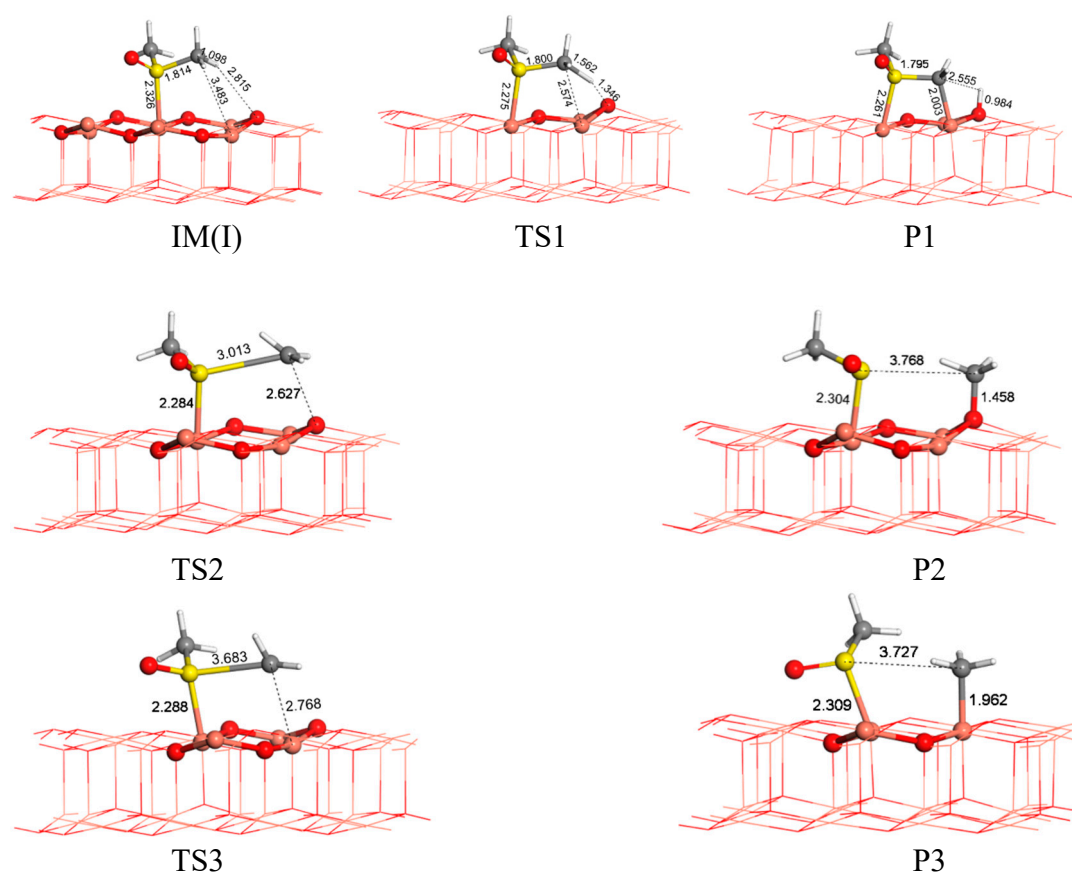


Fig.S1 The intermediates, transition states and product structures involved in the three main reaction paths of $\text{C}_2\text{H}_6\text{OS}$ on the pure $\text{CuO}(111)$ surface.

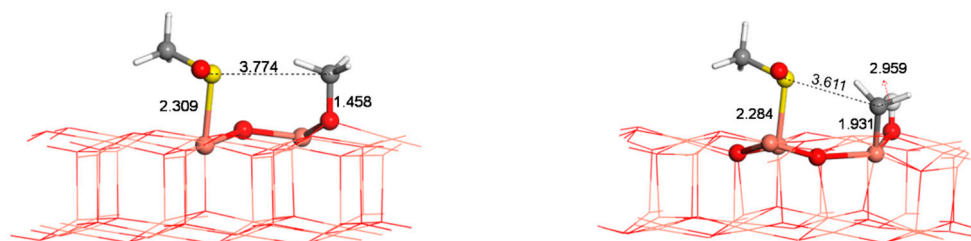
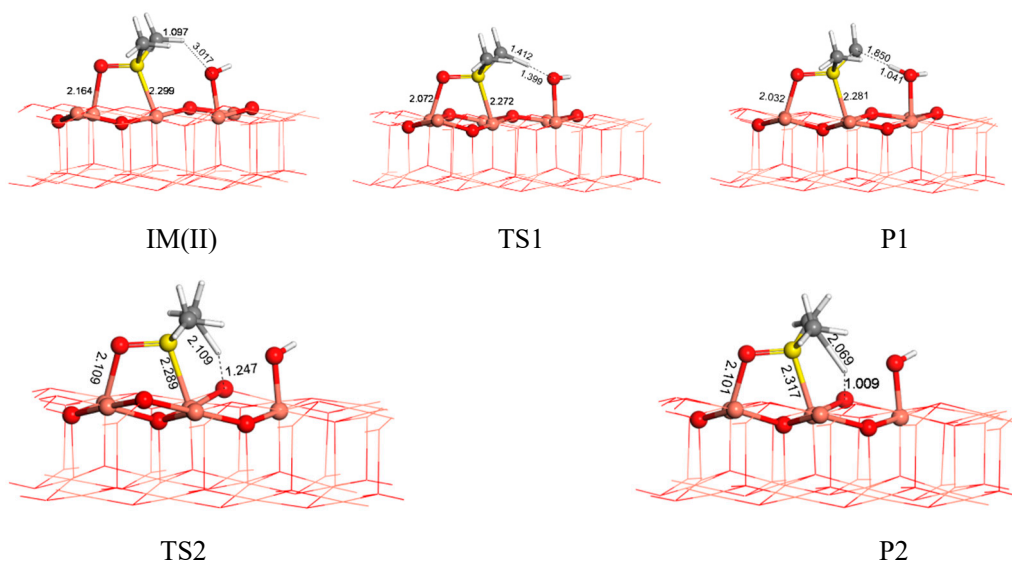
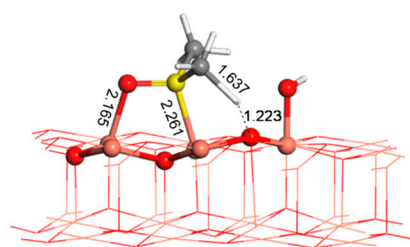


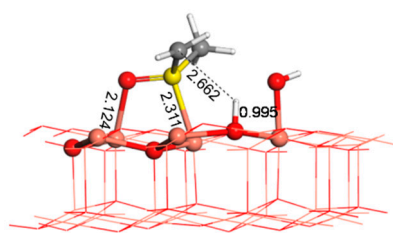
Fig.S2 Structures of C_2H_5OS (i.e., IM2 in Fig.5) breaking C-S bond on the perfect CuO (111) surface.

Fig.S3 depicts the structures of intermediates, transition states, and reaction products involved in the C-H and C-S bond cleavage reaction pathways of C_2H_6OS , i.e. DS-OH-4 in Fig.3 upon adsorption onto the OH pre-adsorbed CuO(111) surface. IM(II) is the most stable adsorption structure formed by C_2H_6OS on the OH pre-adsorbed CuO(111) surface. TS1 is the transition state formed when IM(II) breaks C-H bond and bonds with the O atom in the surface OH, P1 is the decomposition product corresponding to TS1. TS2 and TS3 are the transition states when IM(II) breaks different C-H bonds and bonds with the surface O atoms, P2 and P3 are the decomposition products corresponding to TS2 and TS3, respectively. TS4 is the transition state formed when IM(II) breaks C-S bonds and CH_3 bonds with the surface Cu atoms, P4 is the decomposition product corresponding to TS4. TS5 is the transition state formed when IM(II) breaks the C-S bond and CH_3 bonds with the the surface O atom, and P5 is the decomposition product corresponding to TS5.

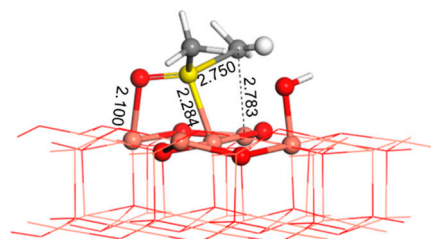




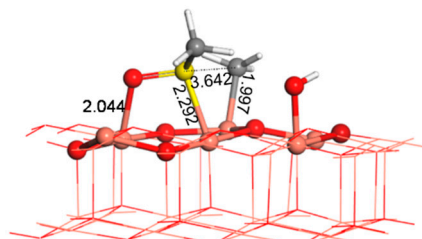
TS3



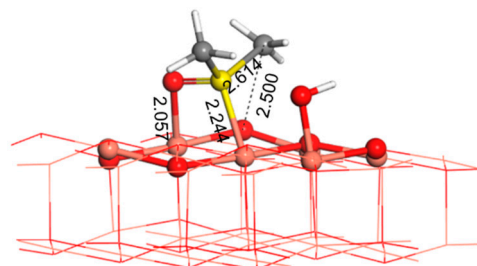
P3



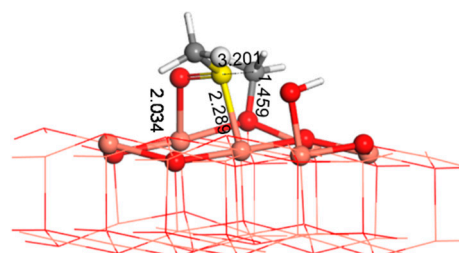
TS4



P4



TS5



P5

Fig.S3 Structures of the intermediates, transition states and product involved in various reaction pathways for DS-OH-4 to break C-H or C-S bond.