Supporting information for

Observation of Inserted Oxocarbonyl Species in the Tantalum Cation-Mediated Activation of Carbon Dioxide Dictated by Two-State Reactivity

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PBE0-D3(BJ)/def2-TZVP level of theory (bond lengths in Ångströms and bond angles in degrees).

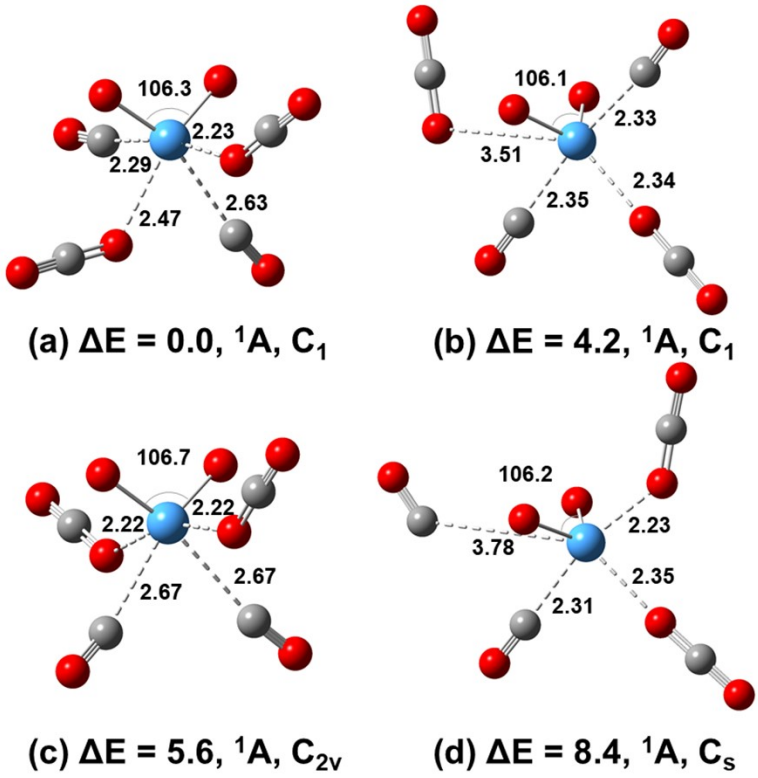

Figure S1. Optimized geometries of isomers for the $[TaO_2(CO)_2(CO_2)_2]^+$ complex in the singlet state and their relative energies (in kcal·mol^{-1}) calculated at the PBE0-D3(BJ)/def2-TZVP level of theory (bond lengths in Ångströms and bond angles in degrees).

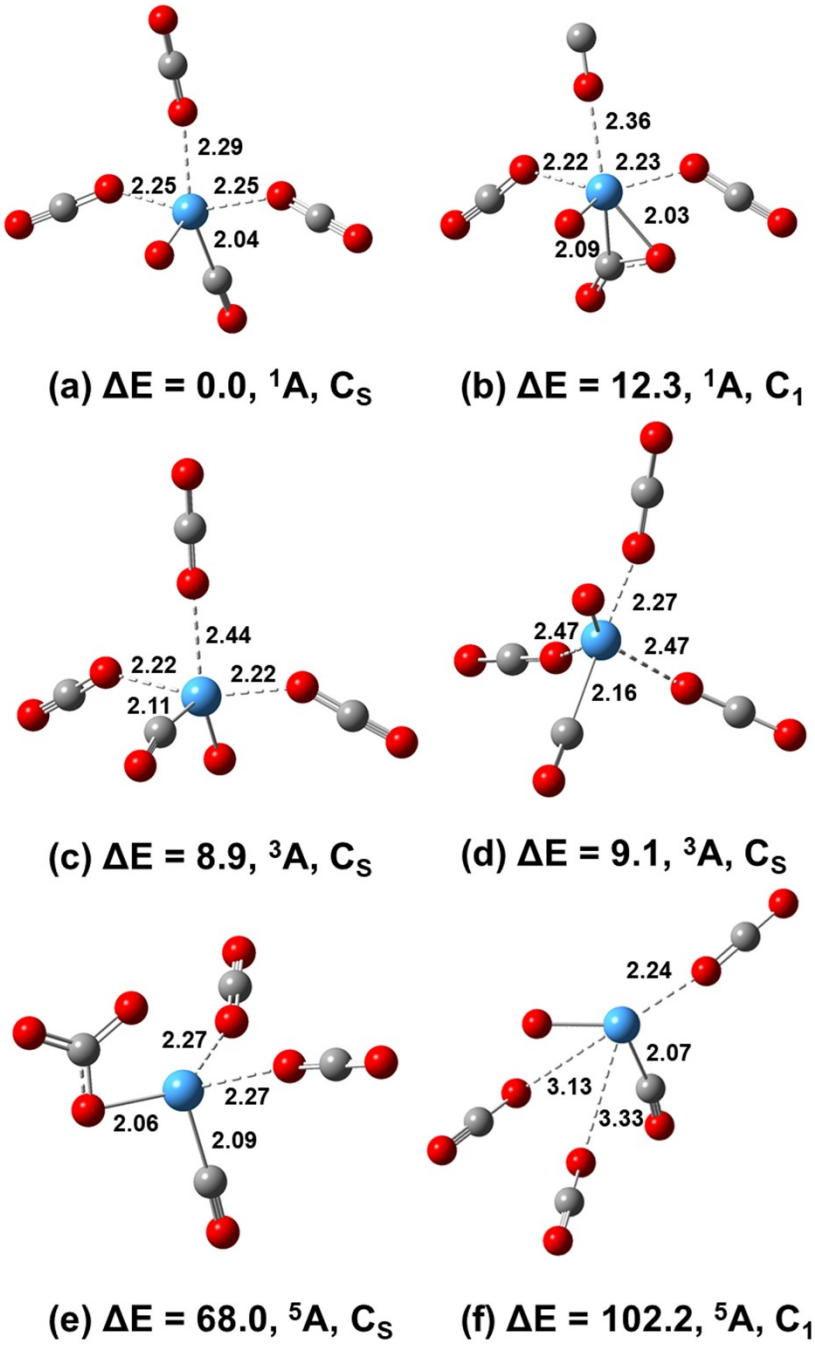

Figure S2. Optimized geometries of isomers for the $[TaO(CO)(CO_2)_3]^+$ complex in different spin states and their relative energies (in kcal·mol^{-1}) calculated at the PBE0-D3(BJ)/def2-TZVP level of theory (bond lengths in Ångströms and bond angles in degrees).

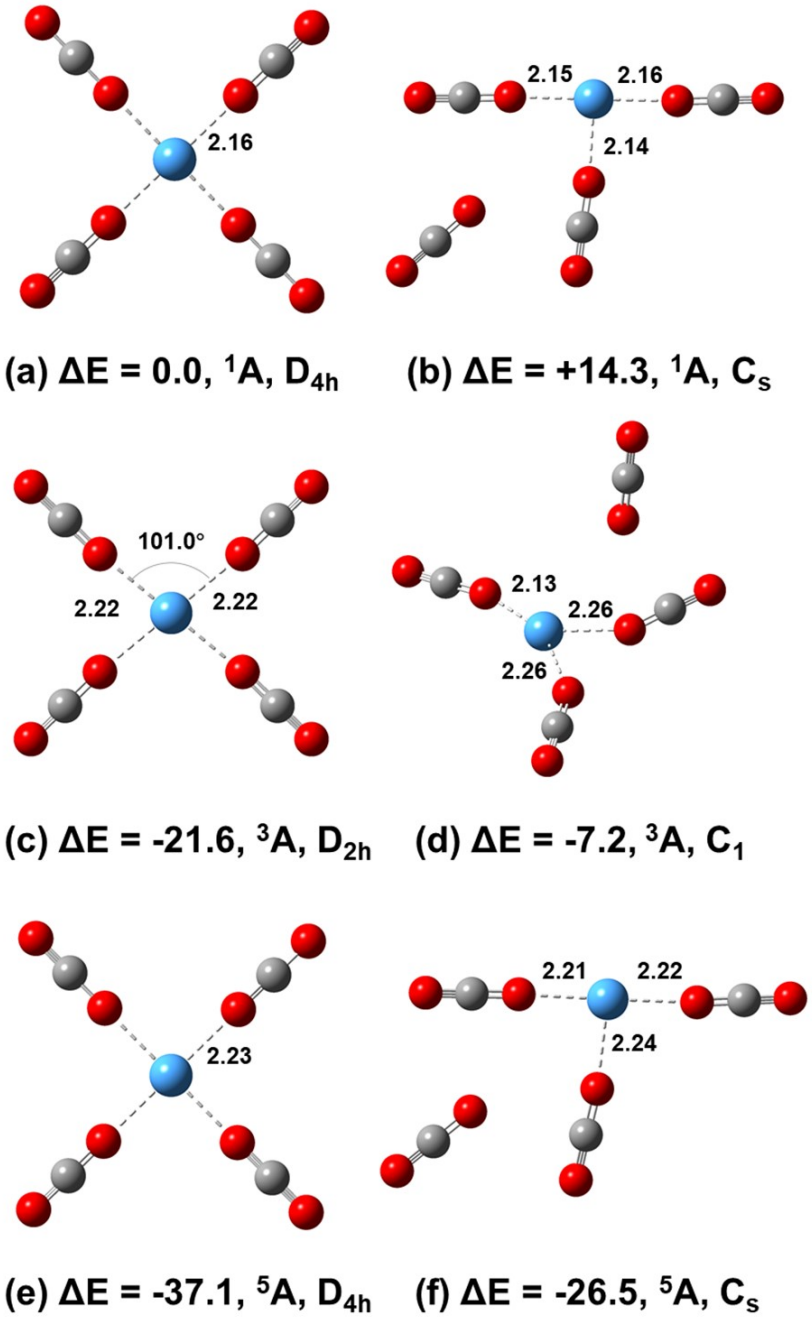

Figure S3. Optimized geometries of isomers for the $[Ta(CO_2)_4]^+$ complex in different spin states and their relative energies (in kcal·mol^{-1}) calculated at the PBE0-D3(BJ)/def2-TZVP level of theory (bond lengths in Ångströms and bond angles in degrees).