

### **Description of Additional Supplementary Files**

**Supplementary Data 1:** The atomic coordinates of the optimized computational models of different ketone molecules and the corresponding imine molecules.

**Supplementary Data 2:** The atomic coordinates of the optimized computational models of various intermediates generated during N-N coupling on the surface of  $\text{WO}_3$  with and without  $\text{CH}_3\text{CN}$ .