

## Aerobic Mukaiyama-type oxidation of essential oil of ortanic tangor from Reunion Island: an applied example of valorization on terpenes using salophen complexes

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### Supplementary Information

**Table S1:** *origin and purity of reagents and solvents used*

| Reagent                          | Provider        | purity                      |
|----------------------------------|-----------------|-----------------------------|
| (-)-Carvéol                      | Merck           | >99%                        |
| (+)-Carvone                      | Merck           | >99%                        |
| (R)-(+)-limonenelimonène         | Fluka           | >99%                        |
| (S)-(-)-limonenelimonène         | Merck           | >97%                        |
| Acétonitrile                     | SDS             | RPE for analysis            |
| Acide métachloroperbenzoïque     | Fluka           | >70%                        |
| Anisole                          | Fluka           | >99%                        |
| Co II acétate tétrahydrate       | Strem Chemicals | >98%                        |
| Dichlorométhane                  | Carlo Erba      | RPE for analysis            |
| Ethanol absolu                   | Carlo Erba      | RPE for analysis            |
| Gel de silice 60                 | Merck           |                             |
| Hydroxyde de sodium              | Carlo Erba      | pellets                     |
| Isobutyraldéhyde                 | Acros Organics  | >99%                        |
| Isohexane                        | Carlo Erba      | RPE for analysis            |
| Jacobsen (R-R)                   | Aldrich         | 98%                         |
| Jacobsen (S-S)                   | Aldrich         | 98%                         |
| Mn II acétate tétrahydrate       | Strem Chemicals | >99%                        |
| N-méthylimidazole                | Alta Aesar      | >99%                        |
| Oxyde de limonene limonène oxide | Aldrich         | >99% mixture of cis + trans |
| p-Xylène                         | Acros Organics  | >99%                        |
| Pentadécane                      | Sigma Aldrich   | >99%                        |
| Sulfate de sodium                | VWR             |                             |
| Toluène anhydre                  | SDS             | RPE for analysis            |

**Table S2:** Composition of the essential oil and fraction H of ortanic Tangor epicarps.

| Compounds                         | LRI <sup>a</sup> | % <sup>b</sup> |             |
|-----------------------------------|------------------|----------------|-------------|
|                                   |                  | Essential Oil  | Fraction H  |
| <b>Monoterpene hydrocarbons</b>   | <b>Total</b>     | <b>92.2</b>    | <b>99.0</b> |
| α-pinene                          | 979              | 1.5            | 0.9         |
| sabinene                          | 1010             | 0.6            | 0.3         |
| β-pinene                          | 1016             | tr             | 0.1         |
| γ-terpinene                       | 1025             | 0.5            | 0.3         |
| α-terpinene                       | 1035             | -              | 0.2         |
| myrcene                           | 1040             | 5.3            | 3.9         |
| α-phellandrene                    | 1064             | 0.3            | 0.2         |
| terpinolene                       | 1102             | -              | 0.2         |
| limonene                          | 1113             | 83.6           | 92.6        |
| menthatriene <1.3.8-p->           | 1115             | -              | tr          |
| (E)-β-ocimene                     | 1117             | 0.4            | 0.3         |
| <b>Oxygenated Monoterpenes</b>    | <b>Total</b>     | <b>4.1</b>     | <b>-</b>    |
| linalol                           | 1102             | 1.0            | -           |
| α-terpineol                       | 1198             | 2.4            | -           |
| citronellol                       | 1226             | 0.4            | -           |
| citronellyl propionate            | 1348             | 0.2            | -           |
| carvone                           | 1379             | 0.1            | -           |
| <b>Sesquiterpene hydrocarbons</b> | <b>Total</b>     | <b>0.9</b>     | <b>0.7</b>  |
| (-)-β-elemene                     | 1388             | tr             | tr          |
| α-copaene                         | 1401             | -              | tr          |
| β-humulene                        | 1409             | -              | tr          |
| (E)-caryophyllene                 | 1442             | -              | 0.1         |
| α-humulene                        | 1461             | -              | tr          |
| γ-murolene                        | 1470             | -              | tr          |
| α-selinene                        | 1496             | -              | 0.1         |
| valencene                         | 1500             | 0.8            | 0.4         |
| δ-guaiene                         | 1502             | tr             | tr          |
| γ-cadinene                        | 1507             | -              | 0.1         |
| isocaryophyllene                  | 1641             | 0.1            | -           |
| <b>Oxygenated sesquiterpenes</b>  | <b>Total</b>     | <b>0.6</b>     | <b>-</b>    |
| eudesmol acetate <β->             | 1848             | 0.6            | -           |
| <b>Alcohols</b>                   | <b>Total</b>     | <b>0.5</b>     | <b>-</b>    |
| 3-hexenol                         | 881              | 0.1            | -           |
| 1-octanol                         | 1070             | 0.3            | -           |
| arbozol <exo->                    | 1342             | 0.1            | -           |
| <b>Aldehydes</b>                  | <b>Total</b>     | <b>0.9</b>     | <b>-</b>    |
| furfural                          | 856              | 0.1            | -           |
| octanal                           | 1028             | 0.1            | -           |
| decanal                           | 1345             | 0.1            | -           |
| dodecanal                         | 1604             | tr             | -           |
| α-sinensal                        | 1756             | 0.6            | -           |
| <b>Carboxylic Acid</b>            | <b>Total</b>     | <b>0.1</b>     | <b>-</b>    |
| acetic acid                       | 1323             | 0.1            | -           |
| <b>Alkane</b>                     | <b>Total</b>     | <b>0.1</b>     | <b>-</b>    |
| nonane                            | 939              | 0.1            | -           |
| <b>Total Identified</b>           |                  | <b>99.4</b>    | <b>99.7</b> |

<sup>a</sup> LRI : Linear retention indices relative to C8-C22 n-alkanes on the apolar SPB-5 column

<sup>b</sup> tr : trace (<0.1%).

Composition of the essential oil from the tangor leaves. Extraction of the essential oil from the leaves of the ortanic tangor was performed in parallel of the tangor peels. The hydrocarbon fraction (Fraction H) of the leaves, which accounts for 45% of the crude essential oil was separated from oxidation fraction (Fraction O). As expected, the composition of this latter essential oil was totally different from that of the tangor peels. The most abundant component was sabinene (58.9%) while other main constituent were all monoterpenes: β-ocimene (11.4%), γ-terpinene (8.2 %), limonene (7.0%), β-pinene (4.3%), myrcene (2.9%), α-terpinene (2.7 %) and α-pinene (1.4 %), accounting for *ca.* 97% of all constituents from the essential oil, in accordance with the literature.<sup>1</sup>

**Table S3:** composition and limonene conversion of reaction blanks ( $t = 24h$ ,  $T=20^{\circ}C$ , ratio limonene:*N*-methylimidazole:cat = 83:5:1 in the Ref. Jacobsen test, 10 mL of toluene)

| -                                       | Blank 1 | Blank 2 | Blank 3 | Blank 4 | Ref. Jacobsen |
|-----------------------------------------|---------|---------|---------|---------|---------------|
| Essential Oil                           | x       | x       | x       | x       | x             |
| IBA                                     | x       | x       |         | x       | x             |
| <i>p</i> -Xylene                        | x       | x       | x       | x       | x             |
| Pentadecane                             | x       | x       | x       | x       | x             |
| <i>Rac</i> -Jacobsen Catalyst           | x       |         | x       |         | x             |
| <i>N</i> -methylimidazole               |         |         | x       | x       | x             |
| Solvent = Toluene                       | x       | x       | x       | x       | x             |
| Limonene conv./limonene oxide yield (%) | 80/45   | 2       | 0       | 2       | 99.5/57       |

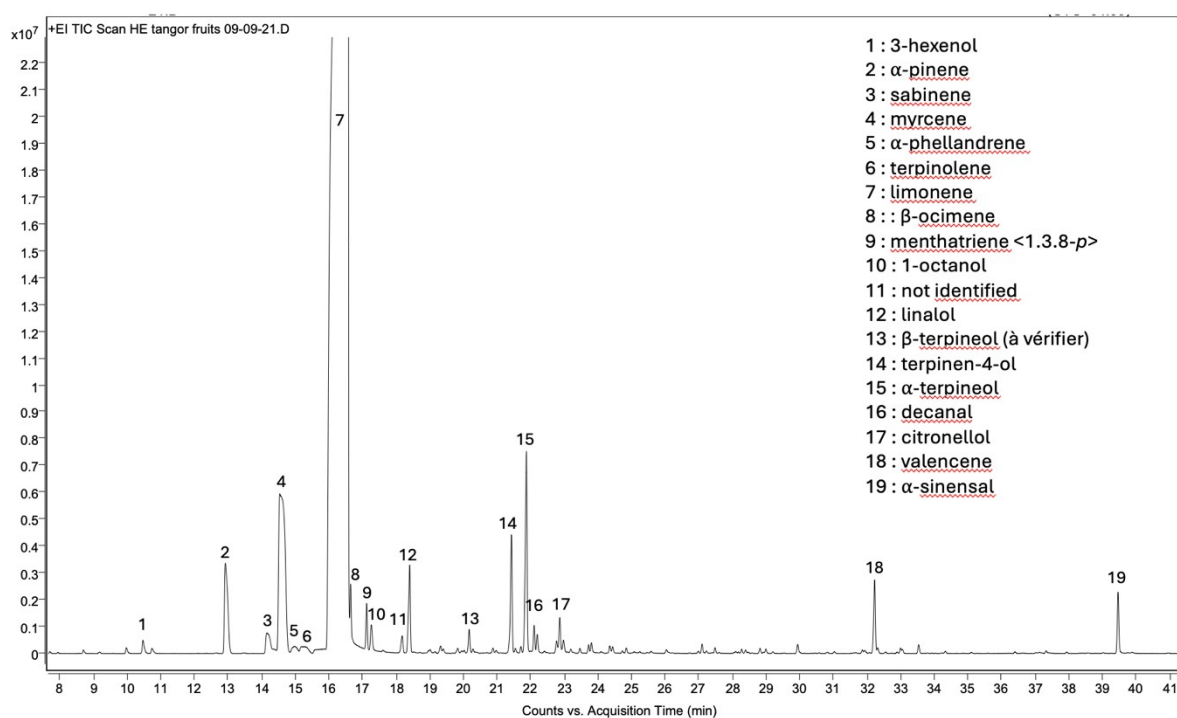


Figure S1: Essential oil GC-MS chromatogram of ortanic Tangor pericarps with major compounds identified

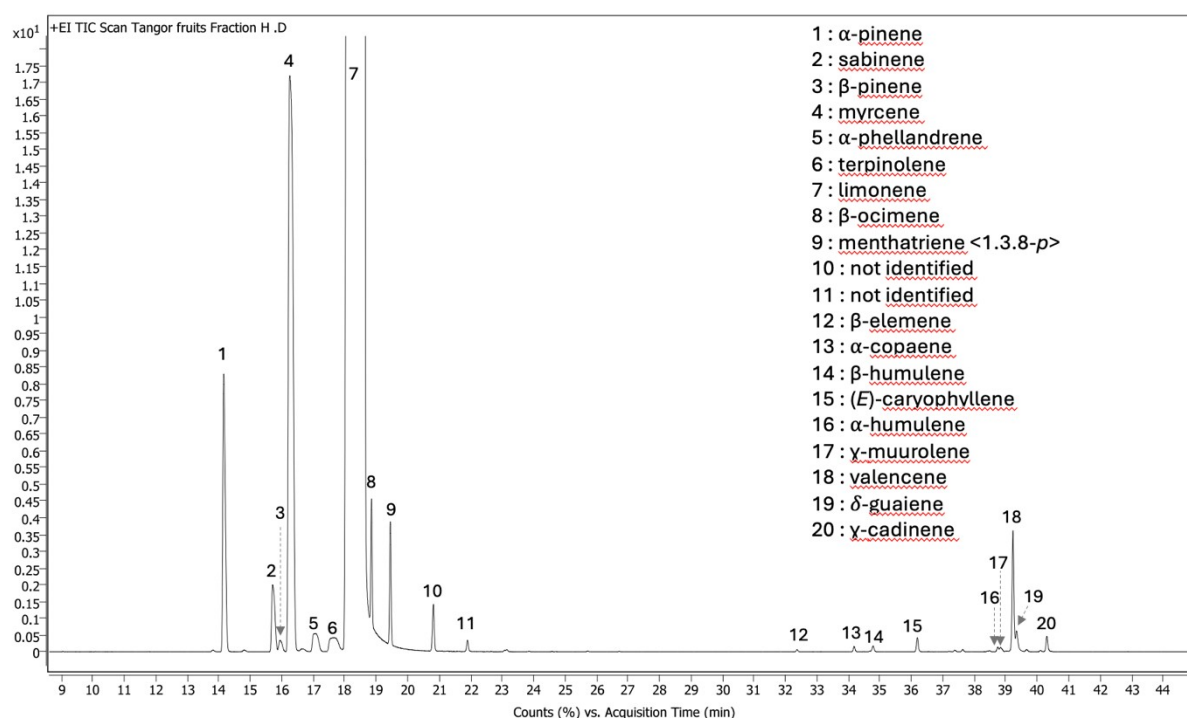


Figure S2: Fraction H GC-MS chromatogram of organic Tangor pericarps with peak identification

**Synthesis of the catalysts.** The synthesis of the Ni<sup>II</sup> and Cr<sup>III</sup> derivatives were described previously.<sup>2</sup>

**1.1 Synthesis of [Salophen{Mn<sup>III</sup>Cl}]:** [Salophen{Mn<sup>III</sup>Cl}] was prepared according to a slight modification of the preparation described by Hey-Hawkins and co-workers.<sup>3</sup> SalophenH<sub>2</sub> (0.8 g, 1.37 mmol) was poured in 20 mL of CH<sub>2</sub>Cl<sub>2</sub>, leading to a yellow suspension, then freshly distilled NEt<sub>3</sub> was added to the mixture (0.57 mL, 4.10 mmol, 3 eqv), leading to an orange suspension. In parallel, a solution of MnCl<sub>2</sub>·4H<sub>2</sub>O (0.85 g, 4.10 mmol, 3 eqv) in 20 mL of MeOH was added dropwise to the previous solution. The solution became darker, and a brownish precipitate was observed. The resulting suspension was stirred at room temperature during 7h under air. The solvent was removed under low pressure, and the brownish crude precipitate was purified by silica gel chromatography by elution using a CH<sub>2</sub>Cl<sub>2</sub>:CH<sub>3</sub>CH<sub>2</sub>OH:CH<sub>3</sub>COOCH<sub>2</sub>CH<sub>3</sub> 94:5:1 mixture. The combined organic fractions ( $\approx$  500 mL) were washed with water (3x500 mL), dried over Na<sub>2</sub>SO<sub>4</sub>, and the solvents were removed under vacuum, leading to [Salophen{Mn<sup>III</sup>Cl}] obtained as a pure dark brown powder (0.89 g, 96 %).

**1.2 Synthesis of [Salophen{Co<sup>II</sup>}]:** SalophenH<sub>2</sub> (0.78 g, 1.37 mmol) and Co(CH<sub>3</sub>COO)<sub>2</sub>·4H<sub>2</sub>O (0.33 g, 1.37 mmol, 1 eqv) were placed in a Schlenk tube under argon. Then, 30 mL of deoxygenated ethanol were added in the Schlenk tube. The mixture was heated at 80°C for 0.5 h and quickly turned dark red. The solvent was removed under vacuum. The brownish crude residue was purified by silica gel column chromatography by elution using CH<sub>2</sub>Cl<sub>2</sub>:CH<sub>3</sub>CH<sub>2</sub>OH:CH<sub>3</sub>COOCH<sub>2</sub>CH<sub>3</sub> mixtures with successive ratios equal to 100:0:0, 95:5:0, 94:5:1 and 89:10:1. [Salophen{Co<sup>II</sup>}] was thus obtained as a dark red powder, yielding 0.55 g (ca. 65%).

**1.3 Synthesis of [Salophen{V<sup>IV</sup>O}]:** (according to a slight modification of the preparation described by Hey-Hawkins and co-workers<sup>2</sup>) SalophenH<sub>2</sub> (0.12 g, 0.21 mmol) and VO(SO<sub>4</sub>)·5H<sub>2</sub>O (0.056 g, 0.24 mmol, > 1 eqv) were placed in a round flask topped with a reflux condenser. Methanol (15 mL) and NEt<sub>3</sub> (0.06 mL, 0.42 mmol) were successively added. The

solution was refluxed at air for 45 min. The solution was then placed at -20°C for several days, leading to the precipitation of a brown precipitate which was discarded through filtration. The resulting yellow filtrate was purified by silica gel column chromatography, by elution with CH<sub>2</sub>Cl<sub>2</sub>:CH<sub>3</sub>CH<sub>2</sub>OH mixtures with successive ratios equal to 100:0 and 95:5. After evaporation of the combined organic fractions, the targeted [Salophen{V<sup>IV</sup>O}] was obtained as a red brown powder (0.076 g, 55%).

1.4 Synthesis of [Salophen{Mo<sup>IV</sup>O}]: The synthesis was adapted from the literature:<sup>4</sup> SalophenH<sub>2</sub> (0.50 g, 0.86 mmol) was placed in a one-neck round-bottom flask open to air and topped with a reflux condenser. Tetrahydrofuran (THF, 50 mL) was added, leading to an orange solution. In parallel, a solution of [Mo(CO)<sub>6</sub>] (0.23 g, 0.86 mmol, 1 eqv) in 10 mL of THF was prepared, and added dropwise to the previous one. The resulting solution was refluxed for 94 h, then allowed to cool down to room temperature. The solvent was removed under vacuum and the brown crude residue was dissolved in 2 mL of CH<sub>2</sub>Cl<sub>2</sub>, then precipitated by adding 15 mL of *n*-hexane. The suspension was placed at -20°C overnight, then filtered on a glass frit. The brown precipitate of [Salophen{Mo<sup>IV</sup>O}] was dried at air (0.50 g, 82 %).

1.5 Synthesis of [Salophen{Mo<sup>VI</sup>O<sub>2</sub>}]: SalophenH<sub>2</sub> (0.20 g, 0.34 mmol) was introduced in a Schlenk tube topped with a reflux condenser under argon. Deaerated CHCl<sub>3</sub> (20 mL) was added and the resulting solution was heated up to 60°C, until the complete dissolution of the ligand. In parallel [MoO<sub>2</sub>(acac)<sub>2</sub>] (0.15 g, 0.38 mmol, > 1 eqv) was dispersed in 20 mL of deaerated CH<sub>3</sub>OH (under sonication). The suspension was quickly added in the solution of SalophenH<sub>2</sub> under an argon stream. The temperature was allowed to rise up to 80°C. The initial suspension turned slowly to a red solution, which was cooled down to room temperature after 16h of heating. The solvent was then removed under vacuum, leading to a red-brown crude solid. This solid was redissolved in 2 mL of CH<sub>2</sub>Cl<sub>2</sub> then precipitated with *n*-hexane (15 mL). The light yellow filtrate was discarded and the solid redissolved in 10 mL of CH<sub>3</sub>OH, leading to a red solution which was filtered to remove a few amount of remaining impurities. The solvent was removed under vacuum and the resulting brown solid identified as [Salophen{Mo<sup>VI</sup>O<sub>2</sub>}] was dried at air (0.23 g, 92 %).

Method for Identification of the volatile compounds. The IRR, formerly known as the Kovats index (KI). is a relative value determined from the retention time of the compound and those of a table of alkanes (C<sub>8</sub> to C<sub>22</sub>) injected under the same conditions as the analyzed samples. These indices are independent of the experimental conditions (oven temperature. choice of carrier gas. injection volume. etc.) and depend only on the nature of the column's stationary phase. The IRRs are calculated as follows (when the method is a gradient):

Calculation of IRR in gradient:

$$IRR = 100n + 100 \times \frac{Rt(X) - Rt(C_n)}{Rt(C_{n+1}) - Rt(C_n)}$$

Where Rt: Retention time

X: Compound to be analyzed

C: Hydrocarbons

n: Number of carbon atoms in the hydrocarbon preceding the compound

n+1 : Number of carbon atoms in the hydrocarbon following the compound

The IRRs of the compounds are then compared with the theoretical IRRs given in the literature<sup>5</sup> and with the compounds identified by the NIST 2020 chemical library.

Calculation of the composition of the mixtures after catalytic oxidation reactions: Prior to any calculation, 4 standard ranges were carried out at a constant *p*-xylene concentration (0.80 M, internal reference) and at terpene or terpene oxide concentrations of 0.80 M, 0.60 M, 0.40 M and 0.20 M in toluene:

DL-limonene / *p*-xylene  
 (+)-limonene oxide / *p*-xylene  
 (-)-carveol / *p*-xylene  
 (+)-carvone / *p*-xylene

The director coefficients of the different ranges are used to convert the peak area into a mole fraction for each product according to the following equation (it will be assumed that the response coefficients of homologous diastereomers of the pure commercial products listed above give an identical detection response):

$$Fm(x) = \frac{Area(x)}{Area(ref) \times coef(x)}$$

where :

Fm = molar fraction

x = oxidation product

ref = reference (*p*-xylene)

coef = directing coefficient of the standard range

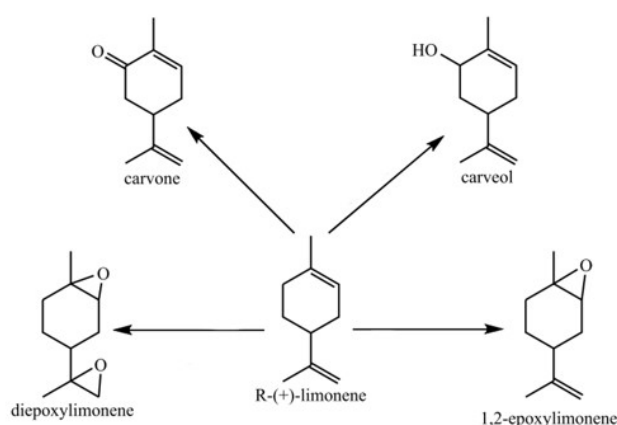


Figure S3: Main oxidation products of the limonene

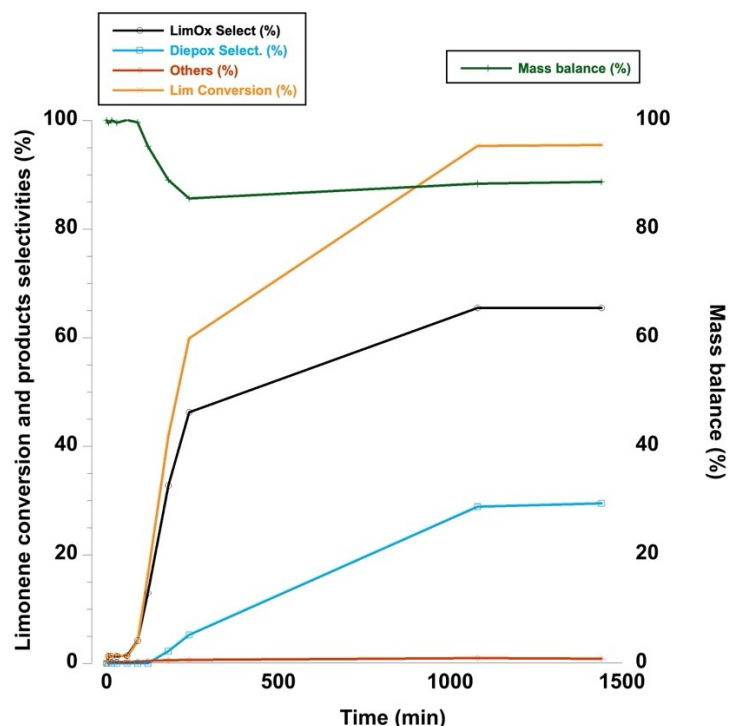


Figure S4: Catalytic performances of  $[\text{Salophen}-\{\text{Mo}^{\text{VI}}\text{O}_2\}]$  for the aerobic epoxidation of fraction H of the essential oil of ortanic tangor, showing the limonene conversion (orange line), the selectivity in limonene-1,2-oxide (black line) and in limonene diepoxide (blue line), the cumulative amount of other products detected (red line) and the corresponding mass balance (green line). Experimental conditions: 1 mL of Fraction H of the essential oil (corresponding to ca. 5.71 mmol of limonene), 30  $\mu\text{L}$  (0.35 mmol) of *N*-methylimidazole, 0.9 mL (7.3 mmol) of *p*-xylene (as internal standard), 5 mL of toluene, 0.07 mmol of  $[\text{Salophen}-\{\text{Mo}^{\text{VI}}\text{O}_2\}]$  (ca. 44 mg), 2 mL (21 mmol) of isobutyraldehyde (IBA), under a continuous  $\text{O}_2$  stream at  $T = 20^\circ\text{C}$ .

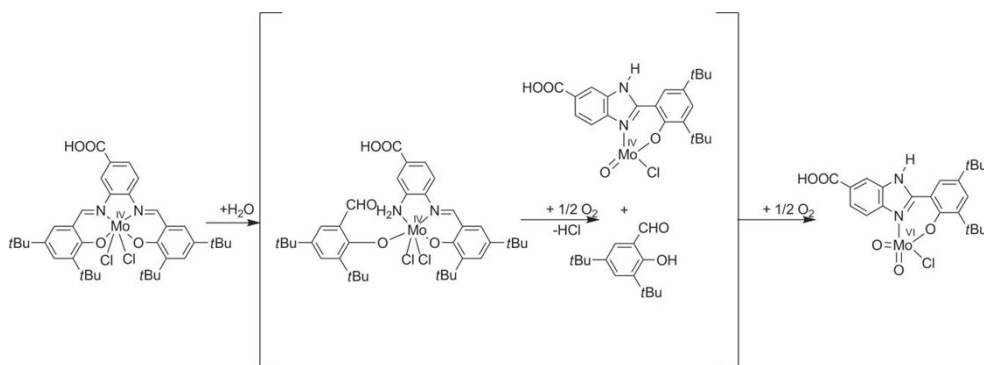
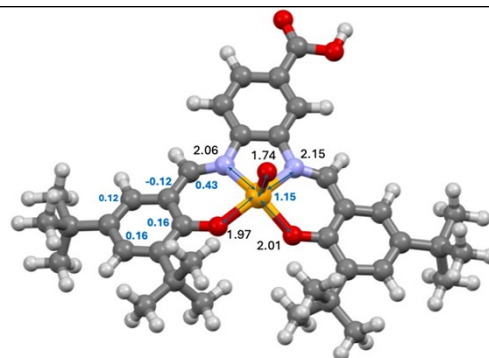
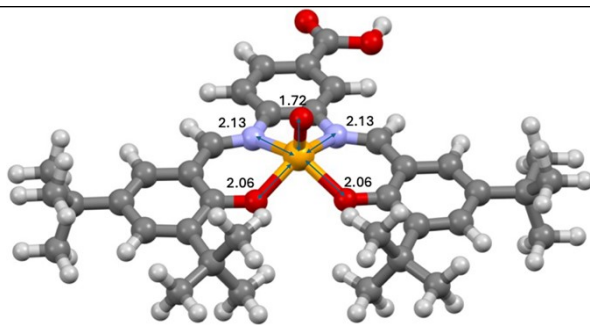


Figure S5: Proposed mechanism for the evolution of the  $[\text{Salophen}-\{\text{Mo}^{\text{IV}}\text{Cl}_2\}]$  complex in the presence of  $\text{H}_2\text{O}$  and  $\text{O}_2$  leading to a  $\{\text{Mo}^{\text{VI}}\text{O}_2\text{Cl}\}$  complex according to Hey-Hawkins and coworkers<sup>6</sup>

**Table S4:** structures and structural data for  $\text{Mo}^{\text{O}^{\text{S}}}$  and  $\text{Mo}^{\text{O}^{\text{T}}}$  with the corresponding distances (in black) and the Mulliken spin population (pop, in blue). C atoms are in grey, H atoms in white, Mo atoms in orange, O atoms in red and N atoms in violet

| $\text{Mo}^{\text{O}^{\text{S}}}$ | $\text{Mo}^{\text{O}^{\text{T}}}$ |
|-----------------------------------|-----------------------------------|
|-----------------------------------|-----------------------------------|



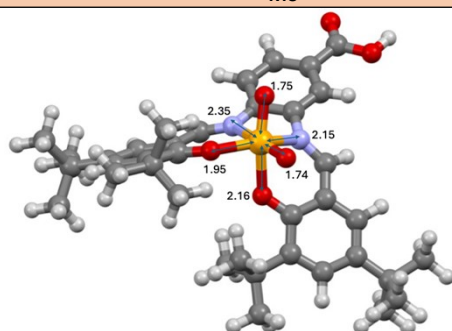
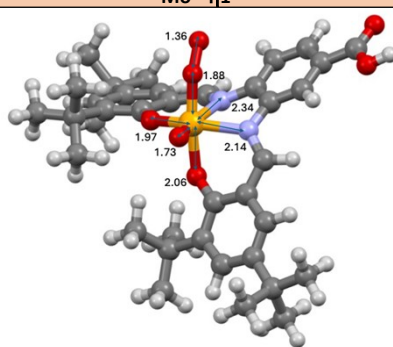
Nb atoms: 91  
E=- 1990.71574295 u.a. ; G=- 1990.046918 u.a.  
Esp = -1992.64516378 u.a. ; qh-G = -1990.018541 u.a.

Nb atoms: 91  
E=- 1990.7093209 u.a. ; G=- 1990.041597 u.a.  
Esp = -1992.63810187 u.a. ; qh-G = -1990.013834 u.a.  
pop: Mo=1.145344; C12=0.425961; C13=-0.118189;  
C14=0.156024; C16=0.155634; C18=0.120439

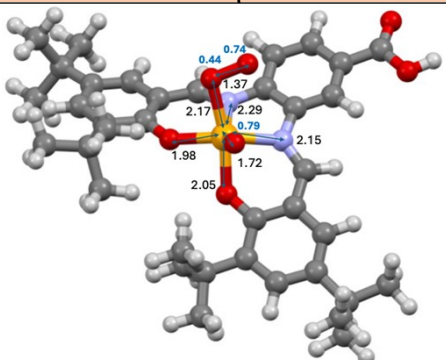
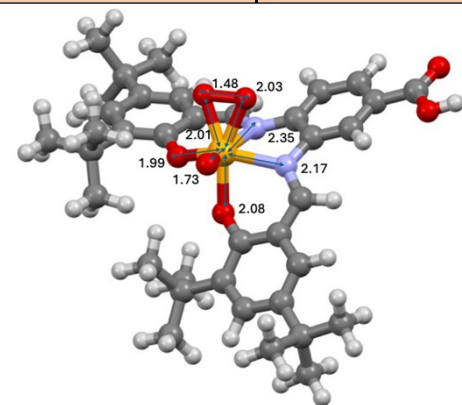
|    |            |            |            |
|----|------------|------------|------------|
| C  | 1.1115530  | 4.3390980  | 0.1320900  |
| C  | 0.5297420  | 3.0612580  | -0.0250740 |
| C  | -0.8863300 | 2.9318650  | -0.0318700 |
| C  | -1.6931020 | 4.0711520  | 0.1164280  |
| C  | -1.1023640 | 5.3435970  | 0.2515800  |
| C  | 0.3033120  | 5.4731150  | 0.2590810  |
| N  | 1.2436370  | 1.8336230  | -0.1777760 |
| C  | 2.5797890  | 1.7933050  | -0.0905540 |
| C  | 3.4251100  | 0.6478620  | -0.0176920 |
| C  | 2.9366280  | -0.7088990 | 0.0095820  |
| C  | 3.8807800  | -1.7895260 | 0.1634570  |
| C  | 5.2346280  | -1.4561640 | 0.2539930  |
| C  | 5.7538580  | -0.1222650 | 0.2148970  |
| C  | 4.8340180  | 0.9040490  | 0.0843710  |
| O  | 1.6301640  | -0.9834400 | -0.1113470 |
| Mo | 0.0899340  | 0.1760010  | -0.8402920 |
| O  | 0.0926400  | -0.0050610 | -2.5462870 |
| C  | 3.4031900  | -3.2606960 | 0.2120640  |
| C  | 2.6741450  | -3.6255950 | -1.1157640 |
| C  | 7.2777430  | 0.1037220  | 0.3197610  |
| C  | 7.9917790  | -0.6195770 | -0.8605560 |
| N  | -1.3606870 | 1.5870600  | -0.1901550 |
| C  | -2.6594720 | 1.2962030  | -0.0893670 |
| C  | -3.2726190 | 0.0078060  | -0.0323950 |
| C  | -4.7041420 | -0.0097700 | 0.0537440  |
| C  | -5.4121760 | -1.1938610 | 0.1778820  |
| C  | -4.6467450 | -2.4015140 | 0.2333160  |
| C  | -3.2521530 | -2.4681480 | 0.1641090  |
| C  | -2.5322120 | -1.2274570 | 0.0107720  |
| O  | -1.1925890 | -1.2459940 | -0.0819970 |
| C  | -2.5013300 | -3.8194130 | 0.2372420  |
| C  | -1.5412350 | -3.8162280 | 1.4637340  |
| C  | -6.9525220 | -1.2639260 | 0.2613600  |
| C  | -7.3732090 | -1.9244280 | 1.6076460  |
| C  | -1.9239130 | 6.5665470  | 0.3969160  |
| O  | -1.4887400 | 7.7251400  | 0.5280640  |
| C  | -7.6074040 | 0.1379060  | 0.1868200  |
| C  | -7.4981820 | -2.1169860 | -0.9220870 |
| C  | 7.6527550  | 1.6055150  | 0.2621110  |
| C  | 7.7997980  | -0.4748000 | 1.6683040  |
| O  | -3.2929630 | 6.3071840  | 0.3758920  |
| C  | 4.5825340  | -4.2535120 | 0.3871620  |
| C  | 2.4382010  | -3.4573300 | 1.4186480  |
| C  | -3.4704050 | -5.0197370 | 0.4025900  |
| C  | -1.6889260 | -4.0475160 | -1.0724730 |
| H  | -3.3613460 | 2.1280520  | -0.0077320 |
| H  | 5.1597190  | 1.9403070  | 0.0601520  |
| H  | -5.2219510 | 0.9447930  | 0.0199680  |
| H  | -2.7742660 | 4.0137500  | 0.1284920  |
| H  | 2.1879630  | 4.4661710  | 0.1558960  |
| H  | 5.9479010  | -2.2654810 | 0.3585810  |
| H  | 3.1125370  | 2.7441520  | -0.0375630 |

|    |            |            |            |
|----|------------|------------|------------|
| C  | 4.6940050  | -0.1228620 | -0.4443830 |
| C  | 3.2901420  | -0.0476280 | -0.1897470 |
| C  | 2.5200660  | -1.2501840 | -0.0580040 |
| C  | 3.1674260  | -2.5241190 | -0.1855610 |
| C  | 4.5511090  | -2.5181390 | -0.4105440 |
| C  | 5.3496050  | -1.3434690 | -0.5447480 |
| C  | 2.7095770  | 1.2646990  | -0.1405530 |
| N  | 1.4632080  | 1.5734830  | 0.1847670  |
| Mo | -0.0779510 | 0.2271720  | 0.8277290  |
| O  | 1.1792650  | -1.1879080 | 0.1453730  |
| N  | -1.1689520 | 1.8172630  | 0.1058040  |
| C  | -2.4771940 | 1.7825770  | -0.3110680 |
| C  | -3.3671850 | 0.6683700  | -0.2549120 |
| C  | -2.9878550 | -0.6425920 | 0.1945110  |
| C  | -3.9418800 | -1.7058870 | 0.2292900  |
| C  | -5.2459100 | -1.4136340 | -0.2015700 |
| C  | -5.6621810 | -0.1350930 | -0.6787360 |
| C  | -4.7137200 | 0.8801540  | -0.6985630 |
| O  | -1.6898970 | -0.8764640 | 0.5617530  |
| C  | -0.4431960 | 3.0367610  | 0.0880940  |
| C  | 0.9776930  | 2.9256660  | 0.1283240  |
| C  | 1.7828870  | 4.0687650  | 0.1330300  |
| C  | 1.1863980  | 5.3487660  | 0.0879630  |
| C  | -0.2213710 | 5.4655120  | 0.0496780  |
| C  | -1.0296120 | 4.3241120  | 0.0545080  |
| O  | 0.0939700  | 0.4559500  | 2.5404390  |
| C  | 6.8680220  | -1.4764230 | -0.7943520 |
| C  | -7.1242500 | 0.0645650  | -1.1384580 |
| C  | 1.9985850  | 6.5811050  | 0.0994470  |
| H  | 3.3763250  | 2.0780140  | -0.4344010 |
| H  | -4.9757120 | 1.8755690  | -1.0449010 |
| H  | 5.2400220  | 0.8103120  | -0.5477810 |
| C  | -3.5576590 | -3.1159280 | 0.7413690  |
| H  | 2.8631700  | 4.0080500  | 0.1952250  |
| H  | -2.1078500 | 4.4359090  | 0.0666540  |
| H  | -5.9830000 | -2.2067950 | -0.1701800 |
| C  | 2.3682950  | -3.8458810 | -0.0881850 |
| H  | -2.8656850 | 2.6858080  | -0.7767500 |
| H  | -0.6621170 | 6.4578680  | 0.0339590  |
| H  | 5.0514790  | -3.4755650 | -0.4947960 |
| C  | 3.2798940  | -5.0930000 | -0.2329900 |
| C  | 1.6587750  | -3.9439090 | 1.2950150  |
| C  | 1.3157770  | -3.8955190 | -1.2358890 |
| H  | 0.7244620  | -4.8179440 | -1.1599810 |
| H  | 0.6326530  | -3.0448940 | -1.1825120 |
| H  | 1.8112040  | -3.8879170 | -2.2165280 |
| H  | 1.1057650  | -4.8907220 | 1.3598890  |
| H  | 2.3931140  | -3.9227700 | 2.1117240  |
| H  | 0.9524800  | -3.1244800 | 1.4422890  |
| H  | 2.6594780  | -5.9947080 | -0.1597710 |
| H  | 3.7900480  | -5.1244480 | -1.2051630 |
| H  | 4.0354150  | -5.1483060 | 0.5624120  |

|   |            |            |            |   |            |            |            |
|---|------------|------------|------------|---|------------|------------|------------|
| H | 0.7397230  | 6.4609210  | 0.3699250  | C | 7.5641960  | -0.0980080 | -0.9165670 |
| H | -5.1917640 | -3.3327840 | 0.3354240  | C | 7.5222410  | -2.2471590 | 0.3904560  |
| H | -0.9964450 | -4.7691730 | 1.5126810  | C | 7.1121070  | -2.2620980 | -2.1167850 |
| H | -0.8142220 | -3.0052980 | 1.3867050  | H | 8.6008300  | -2.3619390 | 0.2183680  |
| H | -2.1060100 | -3.6988130 | 2.3990970  | H | 7.3807990  | -1.7062510 | 1.3349730  |
| H | -1.1626120 | -5.0105990 | -1.0183770 | H | 7.0951970  | -3.2502000 | 0.5108840  |
| H | -2.3558660 | -4.0730450 | -1.9451280 | H | 8.1895830  | -2.3752060 | -2.2966140 |
| H | -0.9503040 | -3.2575370 | -1.2217620 | H | 6.6729890  | -3.2665420 | -2.0827630 |
| H | -2.8821510 | -5.9442840 | 0.4563980  | H | 6.6749040  | -1.7326500 | -2.9732090 |
| H | -4.0614380 | -4.9517710 | 1.3261620  | H | 8.6377540  | -0.2428740 | -1.0907060 |
| H | -4.1584500 | -5.1174630 | -0.4481660 | H | 7.1688370  | 0.4856120  | -1.7585520 |
| H | -8.5929320 | -2.1875910 | -0.8663140 | H | 7.4553240  | 0.4964010  | 0.0001710  |
| H | -7.2313580 | -1.6641610 | -1.8856620 | C | -4.7589920 | -4.0975300 | 0.7154130  |
| H | -7.0969790 | -3.1376600 | -0.9087200 | C | -2.4390150 | -3.7179540 | -0.1580780 |
| H | -8.4675830 | -1.9946060 | 1.6704920  | C | -3.0607570 | -3.0232720 | 2.2152430  |
| H | -6.9678330 | -2.9384550 | 1.7092770  | H | -2.7836330 | -4.0227220 | 2.5783840  |
| H | -7.0171610 | -1.3331560 | 2.4612470  | H | -2.1885490 | -2.3709140 | 2.3023800  |
| H | -8.6985600 | 0.0379740  | 0.2467930  | H | -3.8527360 | -2.6317410 | 2.8679660  |
| H | -7.2878970 | 0.7814680  | 1.0172720  | H | -2.1639210 | -4.7171220 | 0.2079870  |
| H | -7.3727910 | 0.6469420  | -0.7573400 | H | -2.7866810 | -3.8192040 | -1.1955130 |
| H | 2.3406190  | -4.6719390 | -1.0791030 | H | -1.5469780 | -3.0888720 | -0.1492220 |
| H | 1.8010980  | -2.9896320 | -1.2751040 | H | -4.4274860 | -5.0741580 | 1.0903670  |
| H | 3.3501700  | -3.5141530 | -1.9746010 | H | -5.5825960 | -3.7597800 | 1.3584410  |
| H | 2.0888830  | -4.4985960 | 1.4497950  | H | -5.1474940 | -4.2488840 | -0.3006210 |
| H | 2.9515920  | -3.2423270 | 2.3662180  | C | -7.3966730 | 1.5092220  | -1.6270820 |
| H | 1.5684000  | -2.8031460 | 1.3324290  | C | -7.4381410 | -0.9095760 | -2.3124170 |
| H | 4.1827630  | -5.2745840 | 0.4230850  | C | -8.0881230 | -0.2338620 | 0.0480270  |
| H | 5.2910940  | -4.2078420 | -0.4510380 | H | -8.4433910 | 1.5988850  | -1.9447760 |
| H | 5.1332870  | -4.0827940 | 1.3223080  | H | -7.2274060 | 2.2466090  | -0.8313930 |
| H | 8.7419420  | 1.7158860  | 0.3371300  | H | -6.7653730 | 1.7742380  | -2.4854760 |
| H | 7.3373940  | 2.0677170  | -0.6826410 | H | -9.1323480 | -0.1077130 | -0.2692030 |
| H | 7.2051710  | 2.1692020  | 1.0914860  | H | -7.9724850 | -1.2594220 | 0.4187890  |
| H | 9.0789170  | -0.4793180 | -0.7899440 | H | -7.8982110 | 0.4491390  | 0.8860800  |
| H | 7.7938440  | -1.6983350 | -0.8581860 | H | -8.4803180 | -0.7890700 | -2.6384440 |
| H | 7.6551970  | -0.2188040 | -1.8253490 | H | -6.7838420 | -0.7095140 | -3.1708650 |
| H | 8.8864670  | -0.3348120 | 1.7461450  | H | -7.2975900 | -1.9573630 | -2.0208430 |
| H | 7.3262980  | 0.0306770  | 2.5199500  | O | 3.3709240  | 6.3279860  | 0.1514300  |
| H | 7.5947650  | -1.5484340 | 1.7589090  | O | 1.5621510  | 7.7474420  | 0.0640940  |
| H | -3.7985310 | 7.1451970  | 0.4729430  | H | 3.8679300  | 7.1764680  | 0.1636380  |

| Table S5: structural data for Mo <sup>1</sup> , Mo <sup>2</sup> <sub>η<sup>1</sup></sub> <sup>S</sup>                    |           |            |            |                                                                                                                      |           |            |            |
|--------------------------------------------------------------------------------------------------------------------------|-----------|------------|------------|----------------------------------------------------------------------------------------------------------------------|-----------|------------|------------|
| Mo <sup>1</sup>                                                                                                          |           |            |            | Mo <sup>2</sup> <sub>η<sup>1</sup></sub> <sup>S</sup>                                                                |           |            |            |
|                                       |           |            |            |                                  |           |            |            |
| Nb atoms = 92<br>E = - 2065.9319102 u.a.; G = - 2065.259845 u.a.<br>Esp = -2067.91713818 u.a. ; qh-G = -2065.231796 u.a. |           |            |            | Nb atoms = 93<br>E = - 2141.0391158 u.a.; G = -2140.367028 u.a.<br>Esp = -2143.04895656 u.a. ; G = -2140.337889 u.a. |           |            |            |
| C                                                                                                                        | 4.4210470 | -1.0818050 | -0.1402710 | C                                                                                                                    | 2.1288200 | -1.3926090 | -0.1961790 |
| C                                                                                                                        | 3.1013550 | -0.5586210 | 0.0381310  | C                                                                                                                    | 3.1876590 | -0.4270610 | -0.0497600 |
| C                                                                                                                        | 1.9361740 | -1.3850970 | -0.1938580 | C                                                                                                                    | 4.5505350 | -0.8032170 | -0.2501270 |
| C                                                                                                                        | 2.1412620 | -2.7293610 | -0.6900530 | C                                                                                                                    | 4.8982760 | -2.1067340 | -0.5741480 |
| C                                                                                                                        | 3.4574210 | -3.1787380 | -0.8058790 | C                                                                                                                    | 3.8257230 | -3.0343940 | -0.7517150 |

|    |            |            |            |    |            |            |            |
|----|------------|------------|------------|----|------------|------------|------------|
| C  | 4.6258860  | -2.3951210 | -0.5284830 | C  | 2.4673850  | -2.7326850 | -0.6027860 |
| C  | 2.9357770  | 0.8479890  | 0.1882320  | C  | 2.8860020  | 0.9679860  | 0.0605960  |
| N  | 1.8072930  | 1.4639190  | 0.5359820  | N  | 1.7192680  | 1.4885110  | 0.4108870  |
| Mo | 0.1364420  | 0.5248040  | 1.5190410  | Mo | 0.1411410  | 0.3703710  | 1.3252050  |
| O  | 0.9586790  | -0.5246310 | 2.6376620  | O  | -0.5242590 | 1.8282600  | 2.3025220  |
| O  | 0.7153200  | -0.8854930 | -0.0118180 | O  | -1.2993520 | 2.9412760  | 2.2607490  |
| N  | -0.6497910 | 1.8833310  | -0.2268810 | C  | 6.3557760  | -2.5802140 | -0.7617640 |
| C  | -1.7652990 | 1.7303000  | -0.8977190 | C  | 6.6672180  | -3.7091000 | 0.2653170  |
| C  | -2.8014030 | 0.7628060  | -0.6059200 | C  | 1.3747680  | -3.8068560 | -0.8144640 |
| C  | -2.7857110 | -0.0841910 | 0.5481700  | C  | 0.6612660  | -4.0758250 | 0.5422960  |
| C  | -3.9415410 | -0.8604510 | 0.8828940  | O  | 0.8424700  | -1.0323240 | -0.0137370 |
| C  | -5.0140500 | -0.8249370 | -0.0228210 | N  | -0.7468020 | 1.7459880  | -0.3470240 |
| C  | -5.0410840 | -0.0509060 | -1.2168650 | C  | -1.8995760 | 1.5857880  | -0.9602950 |
| C  | -3.9315340 | 0.7512510  | -1.4724900 | C  | -2.8795190 | 0.5646420  | -0.6909070 |
| O  | -1.6870340 | -0.1502290 | 1.3421380  | C  | -2.7719910 | -0.3687530 | 0.3932480  |
| C  | 0.2183790  | 2.9954380  | -0.3929950 | C  | -3.8909870 | -1.2036300 | 0.7274790  |
| C  | 1.5394710  | 2.8038940  | 0.0992260  | C  | -5.0124720 | -1.1295560 | -0.1105810 |
| C  | 2.4666360  | 3.8484860  | 0.0823650  | C  | -5.1294790 | -0.2673460 | -1.2395250 |
| C  | 2.0875260  | 5.1018350  | -0.4455400 | C  | -4.0591440 | 0.5833050  | -1.4928040 |
| C  | 0.7798640  | 5.2993640  | -0.9411870 | O  | -1.6244760 | -0.4673500 | 1.0997590  |
| C  | -0.1528120 | 4.2545570  | -0.9080860 | C  | -3.8754210 | -2.1222360 | 1.9738990  |
| O  | -0.0748260 | 1.9784450  | 2.4657360  | C  | -2.7445560 | -3.1826470 | 1.8527510  |
| C  | 6.0229560  | -3.0321310 | -0.6887260 | C  | -6.4162060 | -0.2949310 | -2.0930980 |
| C  | -6.2762990 | -0.1136090 | -2.1422840 | C  | -6.6136880 | -1.7224830 | -2.6838240 |
| C  | 3.0318840  | 6.2438870  | -0.4802510 | C  | 0.0268750  | 2.9283180  | -0.4714910 |
| H  | 3.7741250  | 1.4765070  | -0.1235140 | C  | -0.4341400 | 4.1792190  | -0.9332150 |
| H  | -3.9058090 | 1.3980450  | -2.3446810 | C  | 0.4227850  | 5.2857030  | -0.9275250 |
| H  | 5.2593140  | -0.4171530 | 0.0481860  | C  | 1.7437290  | 5.1658550  | -0.4397610 |
| C  | -4.0191460 | -1.6753220 | 2.1979210  | C  | 2.2109010  | 3.9248250  | 0.0419170  |
| H  | 3.4658810  | 3.7227210  | 0.4844000  | C  | 1.3579810  | 2.8193920  | 0.0121890  |
| H  | -1.1698350 | 4.4335720  | -1.2439270 | C  | 2.6038640  | 6.3722010  | -0.4303590 |
| H  | -5.8861890 | -1.4256610 | 0.2060140  | O  | 3.8767280  | 6.1240440  | 0.0762510  |
| C  | 0.9388340  | -3.6381010 | -1.0356540 | O  | 1.0729650  | -0.3454320 | 2.6001110  |
| H  | -1.9824470 | 2.4004090  | -1.7372090 | C  | -3.6540590 | -1.2572390 | 3.2517790  |
| H  | 0.5087330  | 6.2783350  | -1.3244510 | C  | -5.2150360 | -2.8846170 | 2.1536300  |
| H  | 3.6127510  | -4.1989980 | -1.1381000 | C  | -7.6420580 | 0.0678940  | -1.2034430 |
| C  | 1.3834830  | -5.0155160 | -1.5932960 | C  | -6.3598400 | 0.7107070  | -3.2701100 |
| C  | 0.0952420  | -3.8940790 | 0.2464440  | C  | 0.3438770  | -3.3219840 | -1.8765400 |
| C  | 0.0568780  | -2.9521810 | -2.1213830 | C  | 1.9616170  | -5.1529140 | -1.3151320 |
| H  | -0.8064820 | -3.5901170 | -2.3563530 | C  | 6.5470200  | -3.1313830 | -2.2058360 |
| H  | -0.3094460 | -1.9839160 | -1.7718210 | C  | 7.3750010  | -1.4344010 | -0.5434960 |
| H  | 0.6285200  | -2.7993850 | -3.0474270 | O  | 2.2733330  | 7.5040290  | -0.8267190 |
| H  | -0.7671180 | -4.5314860 | 0.0057060  | H  | 3.6610080  | 1.6583070  | -0.2811090 |
| H  | 0.6952850  | -4.4042930 | 1.0117830  | H  | -4.1013610 | 1.2971900  | -2.3102840 |
| H  | -0.2724720 | -2.9567440 | 0.6664030  | H  | 5.3101610  | -0.0354380 | -0.1355620 |
| H  | 0.4905970  | -5.6026140 | -1.8426650 | H  | 3.2153380  | 3.8550780  | 0.4449110  |
| H  | 1.9818150  | -4.9173300 | -2.5093510 | H  | -1.4645200 | 4.3044540  | -1.2495580 |
| H  | 1.9599820  | -5.5934750 | -0.8585260 | H  | -5.8563600 | -1.7683730 | 0.1210930  |
| C  | 7.1612510  | -2.0330370 | -0.3640060 | H  | -2.1911800 | 2.3049210  | -1.7328070 |
| C  | 6.1507930  | -4.2443570 | 0.2811520  | H  | 0.0801340  | 6.2576400  | -1.2692520 |
| C  | 6.2133050  | -3.5239880 | -2.1541870 | H  | 4.0864220  | -4.0494690 | -1.0277980 |
| H  | 7.1358850  | -4.7177430 | 0.1705720  | H  | -0.4323810 | -4.0871320 | -2.0144170 |
| H  | 6.0382760  | -3.9226370 | 1.3243580  | H  | -0.1349770 | -2.3914570 | -1.5644180 |
| H  | 5.3888210  | -5.0078270 | 0.0821020  | H  | 0.8337060  | -3.1574570 | -2.8464170 |
| H  | 7.1994420  | -3.9933400 | -2.2716180 | H  | -0.1422720 | -4.8122580 | 0.4055060  |
| H  | 5.4563130  | -4.2648300 | -2.4390240 | H  | 1.3698430  | -4.4740310 | 1.2808060  |
| H  | 6.1455840  | -2.6856520 | -2.8597110 | H  | 0.2239680  | -3.1603960 | 0.9438710  |
| H  | 8.1325600  | -2.5294610 | -0.4840840 | H  | 1.1388780  | -5.8627700 | -1.4665540 |
| H  | 7.1469760  | -1.1661800 | -1.0381210 | H  | 2.4856970  | -5.0442000 | -2.2744610 |
| H  | 7.0998730  | -1.6695150 | 0.6700590  | H  | 2.6503160  | -5.6031540 | -0.5877910 |
| C  | -5.3806350 | -2.4043210 | 2.3501890  | H  | 7.6979320  | -4.0658120 | 0.1360560  |
| C  | -2.9011770 | -2.7564730 | 2.2448940  | H  | 6.5562540  | -3.3431590 | 1.2940180  |
| C  | -3.8659430 | -0.7078040 | 3.4108720  | H  | 5.9982010  | -4.5693990 | 0.1414340  |
| H  | -3.9189370 | -1.2783410 | 4.3478070  | H  | 7.5778270  | -3.4848510 | -2.3425640 |
| H  | -2.9078110 | -0.1824640 | 3.3872930  | H  | 5.8761800  | -3.9735280 | -2.4150040 |
| H  | -4.6723870 | 0.0379980  | 3.4205230  | H  | 6.3500020  | -2.3508020 | -2.9521330 |
| H  | -2.9962900 | -3.3418910 | 3.1693890  | H  | 8.3940490  | -1.8176590 | -0.6799010 |
| H  | -2.9858550 | -3.4460760 | 1.3945500  | H  | 7.2302900  | -0.6173570 | -1.2629010 |
| H  | -1.9075070 | -2.3055810 | 2.2272520  | H  | 7.3100560  | -1.0198420 | 0.4710100  |
| H  | -5.3854860 | -2.9475190 | 3.3030770  | H  | -3.6433280 | -1.9046880 | 4.1388950  |
| H  | -6.2272010 | -1.7048540 | 2.3666430  | H  | -2.7054090 | -0.7163210 | 3.2146740  |

|                                                                                                                                                                     |            |            |            |                                                                                                                 |            |            |            |
|---------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------|------------|------------|-----------------------------------------------------------------------------------------------------------------|------------|------------|------------|
| H                                                                                                                                                                   | -5.5478630 | -3.1403470 | 1.5521150  | H                                                                                                               | -4.4639680 | -0.5256480 | 3.3736760  |
| C                                                                                                                                                                   | -6.1259270 | 0.8012370  | -3.3833580 | H                                                                                                               | -2.7569680 | -3.8392980 | 2.7332720  |
| C                                                                                                                                                                   | -6.4746720 | -1.5764480 | -2.6389800 | H                                                                                                               | -2.8855140 | -3.8062890 | 0.9593860  |
| C                                                                                                                                                                   | -7.5427350 | 0.3390380  | -1.3567750 | H                                                                                                               | -1.7643080 | -2.7089270 | 1.7917480  |
| H                                                                                                                                                                   | -7.0202740 | 0.7155460  | -4.0131510 | H                                                                                                               | -5.1511250 | -3.5015010 | 3.0584070  |
| H                                                                                                                                                                   | -6.0209640 | 1.8573920  | -3.1015270 | H                                                                                                               | -6.0666480 | -2.2028850 | 2.2801520  |
| H                                                                                                                                                                   | -5.2606310 | 0.5171050  | -3.9966340 | H                                                                                                               | -5.4255010 | -3.5580770 | 1.3117100  |
| H                                                                                                                                                                   | -8.4286530 | 0.2862200  | -2.0037000 | H                                                                                                               | -7.2875290 | 0.6469020  | -3.8525650 |
| H                                                                                                                                                                   | -7.7304880 | -0.2961050 | -0.4826270 | H                                                                                                               | -6.2623180 | 1.7460740  | -2.9175910 |
| H                                                                                                                                                                   | -7.4362360 | 1.3731070  | -1.0042780 | H                                                                                                               | -5.5256850 | 0.4949310  | -3.9507990 |
| H                                                                                                                                                                   | -7.3597380 | -1.6405110 | -3.2861150 | H                                                                                                               | -8.5641650 | 0.0401160  | -1.7994190 |
| H                                                                                                                                                                   | -5.6033940 | -1.9160390 | -3.2137140 | H                                                                                                               | -7.7634780 | -0.6334670 | -0.3690760 |
| H                                                                                                                                                                   | -6.6193090 | -2.2739630 | -1.8050490 | H                                                                                                               | -7.5353220 | 1.0758020  | -0.7822640 |
| O                                                                                                                                                                   | 4.2865500  | 5.9233760  | 0.0291520  | H                                                                                                               | -7.5353250 | -1.7621740 | -3.2798790 |
| O                                                                                                                                                                   | 2.7792810  | 7.3827020  | -0.9129030 | H                                                                                                               | -5.7723340 | -1.9970290 | -3.3331760 |
| H                                                                                                                                                                   | 4.8785420  | 6.7082760  | -0.0005650 | H                                                                                                               | -6.6921540 | -2.4822640 | -1.8967370 |
|                                                                                                                                                                     |            |            |            | H                                                                                                               | 4.4099760  | 6.9504790  | 0.0793800  |
| <b>Mo2<math>\eta_1^T</math></b>                                                                                                                                     |            |            |            | <b>Mo2<math>\eta_2^S</math></b>                                                                                 |            |            |            |
|                                                                                   |            |            |            |                              |            |            |            |
| Nb atoms = 93<br>E=-2141.0432879 u.a. ; G=-2140.372237 u.a.<br>Esp = -2143.04499829 u.a. ; qh-G = -2140.343699 u.a.<br>pop: Mo=0.788348; O10=0.436482; O11=0.735531 |            |            |            | Nb atoms = 93<br>E=-2141.0647821 u.a.; G=-2140.389549 u.a.<br>Esp = -2143.08098625 u.a. ; G = -2140.361428 u.a. |            |            |            |
| C                                                                                                                                                                   | 2.3838060  | -1.3027790 | -0.2556500 | C                                                                                                               | -0.7254300 | 4.2473800  | -0.7788860 |
| C                                                                                                                                                                   | 3.2645670  | -0.1700000 | -0.1591530 | C                                                                                                               | -0.2233280 | 2.9833550  | -0.3970350 |
| C                                                                                                                                                                   | 4.6744580  | -0.3322350 | -0.3268450 | C                                                                                                               | 1.1234550  | 2.8813730  | 0.0425640  |
| C                                                                                                                                                                   | 5.2342230  | -1.5802310 | -0.5602200 | C                                                                                                               | 1.9396170  | 4.0148570  | 0.1249130  |
| C                                                                                                                                                                   | 4.3327630  | -2.6852160 | -0.6630030 | C                                                                                                               | 1.4288020  | 5.2682420  | -0.2666050 |
| C                                                                                                                                                                   | 2.9420400  | -2.6000930 | -0.5372980 | C                                                                                                               | 0.0958830  | 5.3777140  | -0.7232840 |
| C                                                                                                                                                                   | 2.7591300  | 1.1652490  | -0.0673100 | N                                                                                                               | -0.9658260 | 1.7779970  | -0.3211160 |
| N                                                                                                                                                                   | 1.5198420  | 1.5330250  | 0.2278160  | C                                                                                                               | -2.1839110 | 1.6693130  | -0.8052290 |
| Mo                                                                                                                                                                  | 0.0387420  | 0.1994930  | 1.0443960  | C                                                                                                               | -3.1065090 | 0.5869640  | -0.5828130 |
| O                                                                                                                                                                   | -1.3088240 | 1.2927030  | 2.3545220  | C                                                                                                               | -2.7831540 | -0.6028540 | 0.1476260  |
| O                                                                                                                                                                   | -0.5330640 | 2.3855660  | 2.6317010  | C                                                                                                               | -3.8084850 | -1.5698780 | 0.4159410  |
| C                                                                                                                                                                   | 6.7518220  | -1.8185760 | -0.7155660 | C                                                                                                               | -5.0867640 | -1.3059450 | -0.0975440 |
| C                                                                                                                                                                   | 7.2361170  | -2.8020920 | 0.3910310  | C                                                                                                               | -5.4345990 | -0.1570420 | -0.8639290 |
| C                                                                                                                                                                   | 2.0393560  | -3.8506740 | -0.6500600 | C                                                                                                               | -4.4265090 | 0.7727820  | -1.0912270 |
| C                                                                                                                                                                   | 1.3399050  | -4.0990560 | 0.7198220  | O                                                                                                               | -1.5005150 | -0.8465350 | 0.5196430  |
| O                                                                                                                                                                   | 1.0536570  | -1.1413060 | -0.1300440 | Mo                                                                                                              | 0.0233100  | 0.2525090  | 1.1637410  |
| N                                                                                                                                                                   | -1.0244330 | 1.7559510  | -0.2541320 | O                                                                                                               | -0.2456730 | 1.9234020  | 2.2768620  |
| C                                                                                                                                                                   | -2.2573620 | 1.6547880  | -0.7241780 | C                                                                                                               | -3.5211550 | -2.8592430 | 1.2217690  |
| C                                                                                                                                                                   | -3.1712630 | 0.5635920  | -0.5540280 | C                                                                                                               | -2.9312140 | -2.5069840 | 2.6194500  |
| C                                                                                                                                                                   | -2.8508890 | -0.6313640 | 0.1720370  | C                                                                                                               | -6.8798560 | 0.0095130  | -1.3819820 |
| C                                                                                                                                                                   | -3.8650160 | -1.6251460 | 0.3888790  | C                                                                                                               | -7.8625000 | 0.0511370  | -0.1742370 |
| C                                                                                                                                                                   | -5.1261090 | -1.3807190 | -0.1688570 | N                                                                                                               | 1.5547160  | 1.5486580  | 0.3458210  |
| C                                                                                                                                                                   | -5.4720070 | -0.2227200 | -0.9291640 | C                                                                                                               | 2.7905270  | 1.1642910  | 0.0591040  |
| C                                                                                                                                                                   | -4.4802630 | 0.7329030  | -1.1035310 | C                                                                                                               | 3.2522850  | -0.1807480 | -0.0598680 |
| O                                                                                                                                                                   | -1.5929900 | -0.8409810 | 0.6088970  | C                                                                                                               | 4.6561250  | -0.4080900 | -0.1875520 |
| C                                                                                                                                                                   | -3.5726880 | -2.9109270 | 1.1982290  | C                                                                                                               | 5.1520460  | -1.6635620 | -0.5064380 |
| C                                                                                                                                                                   | -2.4853500 | -3.7497000 | 0.4643200  | C                                                                                                               | 4.1916640  | -2.6966520 | -0.7429720 |
| C                                                                                                                                                                   | -6.9005050 | -0.0829130 | -1.4989740 | C                                                                                                               | 2.8039130  | -2.5402120 | -0.6642190 |
| C                                                                                                                                                                   | -7.1940030 | -1.2721010 | -2.4611120 | C                                                                                                               | 2.3071280  | -1.2422410 | -0.2799700 |
| C                                                                                                                                                                   | -0.3057940 | 2.9796120  | -0.3169100 | O                                                                                                               | 0.9887250  | -0.9997390 | -0.1865240 |
| C                                                                                                                                                                   | -0.8584020 | 4.2526070  | -0.5790300 | C                                                                                                               | 1.8441570  | -3.7285450 | -0.8986270 |
| C                                                                                                                                                                   | -0.0436040 | 5.3886740  | -0.5545620 | C                                                                                                               | 0.7813270  | -3.3674890 | -1.9778550 |

|   |            |            |            |   |            |            |            |
|---|------------|------------|------------|---|------------|------------|------------|
| C | 1.3314070  | 5.2727260  | -0.2486030 | C | 6.6581260  | -1.9823520 | -0.6221480 |
| C | 1.8893520  | 4.0097210  | 0.0315330  | C | 6.9769850  | -2.4896960 | -2.0597240 |
| C | 1.0754320  | 2.8718910  | -0.0118680 | C | 2.2537120  | 6.4967450  | -0.1968770 |
| C | 2.1508930  | 6.5064880  | -0.2090760 | O | 1.8800840  | 7.6417720  | -0.5084060 |
| O | 3.4843900  | 6.2603890  | 0.1068390  | O | 0.9190720  | -0.5543590 | 2.4004460  |
| O | 0.8400480  | -0.2825950 | 2.4899460  | C | 7.5414440  | -0.7414060 | -0.3407260 |
| C | -3.0874780 | -2.5405650 | 2.6317150  | C | 7.0325550  | -3.0883520 | 0.4088990  |
| C | -4.8320220 | -3.8042880 | 1.3514360  | C | -7.0628330 | 1.3130860  | -2.1986780 |
| C | -7.9308800 | -0.0994460 | -0.3311600 | C | -7.2446310 | -1.1937400 | -2.3010570 |
| C | -7.0889320 | 1.2355220  | -2.2900640 | O | 3.5458060  | 6.2553880  | 0.2621290  |
| C | 0.9755990  | -3.6447720 | -1.7693240 | C | -4.8037190 | -3.6989290 | 1.4634490  |
| C | 2.8469580  | -5.1273820 | -1.0031570 | C | -2.5211560 | -3.7427160 | 0.4210750  |
| C | 7.0437070  | -2.4351330 | -2.1157520 | C | 2.5884140  | -4.9998800 | -1.3858360 |
| C | 7.5664590  | -0.5074950 | -0.5859790 | C | 1.1524620  | -4.0740520 | 0.4539060  |
| O | 1.7399920  | 7.6592270  | -0.4322600 | H | 3.5073410  | 1.9417460  | -0.2121080 |
| H | 3.4653700  | 1.9517270  | -0.3413090 | H | -4.6287170 | 1.6812950  | -1.6509590 |
| H | -4.6821180 | 1.6465470  | -1.6552000 | H | 5.3249350  | 0.4311720  | -0.0206540 |
| H | 5.2988280  | 0.5538430  | -0.2580770 | H | 2.9526620  | 3.9577470  | 0.5069850  |
| H | 2.9367390  | 3.9515990  | 0.3045900  | H | -1.7595400 | 4.3642270  | -1.0860740 |
| H | -1.9209740 | 4.3700310  | -0.7638790 | H | -5.8679150 | -2.0292370 | 0.1033310  |
| H | -5.8981380 | -2.1248670 | -0.0124120 | H | -2.5905280 | 2.4874970  | -1.4074460 |
| H | -2.6622530 | 2.4960510  | -1.2930030 | H | -0.2808410 | 6.3560530  | -1.0056340 |
| H | -0.4550980 | 6.3751290  | -0.7448450 | H | 4.5713380  | -3.6779030 | -1.0029270 |
| H | 4.7616930  | -3.6618170 | -0.8549330 | H | 0.0962640  | -4.2146680 | -2.1190790 |
| H | 0.3286690  | -4.5304860 | -1.8317480 | H | 0.1997170  | -2.4922260 | -1.6824170 |
| H | 0.3523030  | -2.7717730 | -1.5650920 | H | 1.2653900  | -3.1589230 | -2.9423190 |
| H | 1.4615820  | -3.5099380 | -2.7457380 | H | 0.4428420  | -4.8997770 | 0.3120870  |
| H | 0.6779150  | -4.9724460 | 0.6470650  | H | 1.8977770  | -4.3853400 | 1.1980850  |
| H | 2.0835280  | -4.2970480 | 1.5034420  | H | 0.6076350  | -3.2161780 | 0.8538120  |
| H | 0.7408370  | -3.2385730 | 1.0245010  | H | 1.8526020  | -5.7947360 | -1.5603070 |
| H | 2.1532070  | -5.9733040 | -1.0851740 | H | 3.1218600  | -4.8289500 | -2.3308840 |
| H | 3.3687190  | -5.0337140 | -1.9653490 | H | 3.3032720  | -5.3763670 | -0.6419890 |
| H | 3.5822090  | -5.3822450 | -0.2280990 | H | 8.1002450  | -3.3337440 | 0.3306190  |
| H | 8.3128260  | -2.9912390 | 0.2852700  | H | 6.8311910  | -2.7516300 | 1.4338610  |
| H | 7.0567150  | -2.3853530 | 1.3903550  | H | 6.4648360  | -4.0117770 | 0.2416000  |
| H | 6.7203520  | -3.7683060 | 0.3329640  | H | 8.0438950  | -2.7354660 | -2.1463220 |
| H | 8.1196580  | -2.6238240 | -2.2293040 | H | 6.4060720  | -3.3916740 | -2.3114500 |
| H | 6.5215540  | -3.3887840 | -2.2600110 | H | 6.7397070  | -1.7218860 | -2.8075260 |
| H | 6.7280980  | -1.7542540 | -2.9168940 | H | 8.6001100  | -1.0158460 | -0.4294090 |
| H | 8.6355870  | -0.7263880 | -0.6998660 | H | 7.3485790  | 0.0675240  | -1.0579750 |
| H | 7.2937690  | 0.2205560  | -1.3615900 | H | 7.3841850  | -0.3515300 | 0.6734740  |
| H | 7.4277570  | -0.0374450 | 0.3964570  | H | -2.7605710 | -3.4304420 | 3.1892200  |
| H | -2.8938260 | -3.4582190 | 3.2035750  | H | -1.9809590 | -1.9761970 | 2.5427700  |
| H | -2.1682570 | -1.9517640 | 2.6111810  | H | -3.6275870 | -1.8784740 | 3.1901670  |
| H | -3.8534580 | -1.9623060 | 3.1654850  | H | -2.2652830 | -4.6387330 | 1.0030200  |
| H | -2.2525250 | -4.6493670 | 1.0503710  | H | -2.9676470 | -4.0674780 | -0.5291380 |
| H | -2.8441470 | -4.0694250 | -0.5239860 | H | -1.6019230 | -3.1975000 | 0.2041090  |
| H | -1.5660520 | -3.1770820 | 0.3312250  | H | -4.5367700 | -4.5890070 | 2.0464750  |
| H | -4.5661860 | -4.6913660 | 1.9395220  | H | -5.5593340 | -3.1440010 | 2.0357440  |
| H | -5.6430940 | -3.2880150 | 1.8824840  | H | -5.2577280 | -4.0475310 | 0.5260680  |
| H | -5.2134080 | -4.1558840 | 0.3831590  | H | -8.0997590 | 1.3836320  | -2.5504380 |
| H | -8.1137240 | 1.2876300  | -2.6786770 | H | -6.8584510 | 2.2057680  | -1.5928640 |
| H | -6.9312010 | 2.1175740  | -1.6553900 | H | -6.4112610 | 1.3353140  | -3.0823360 |
| H | -6.4062010 | 1.2973150  | -3.1479210 | H | -8.8960800 | 0.1581660  | -0.5299750 |
| H | -8.9521970 | -0.0126590 | -0.7256590 | H | -7.8095590 | -0.8647470 | 0.4267170  |
| H | -7.8757540 | -1.0282400 | 0.2494590  | H | -7.6362390 | 0.8986820  | 0.4853470  |
| H | -7.7557950 | 0.7374240  | 0.3570230  | H | -8.2771280 | -1.0951670 | -2.6622310 |
| H | -8.2143590 | -1.1942420 | -2.8599620 | H | -6.5785340 | -1.2370060 | -3.1724940 |
| H | -6.4938610 | -1.2735470 | -3.3065460 | H | -7.1667930 | -2.1506940 | -1.7709600 |
| H | -7.1078940 | -2.2398400 | -1.9521760 | H | 4.0547440  | 7.0958320  | 0.3067020  |
| H | 3.9893640  | 7.1038890  | 0.1360700  | O | -1.4110660 | 1.0139930  | 2.3465210  |

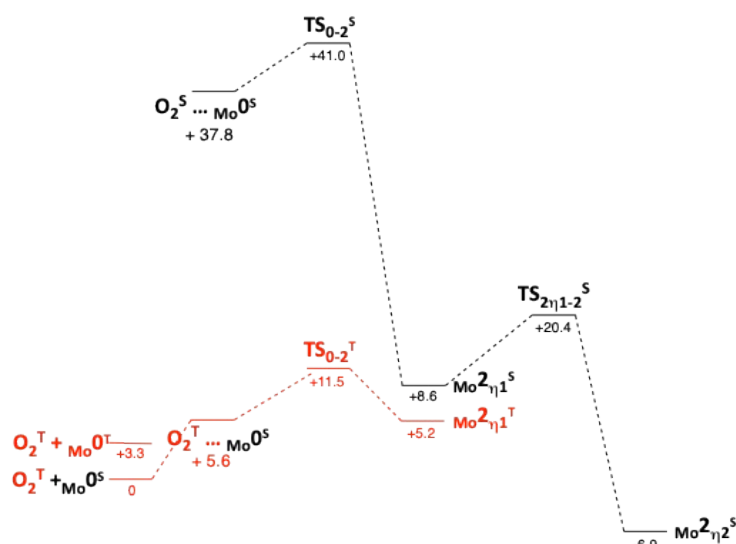
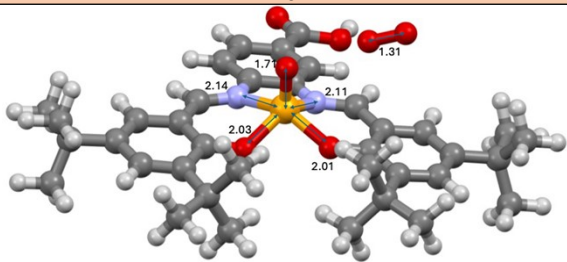
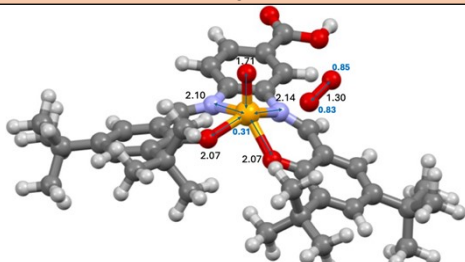
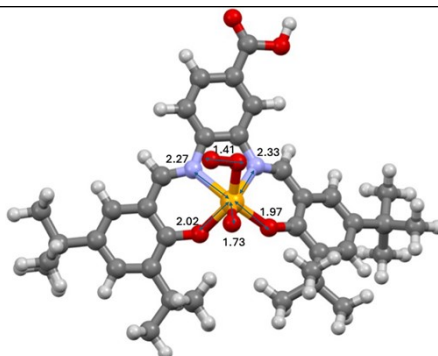


Figure S6: Gibbs energy profiles (in kcal.mol<sup>-1</sup>) for complete oxidation of Mo<sup>0</sup> with O<sub>2</sub> for the triplet (red) and singlet (black) spin states<sup>7</sup>

| Table S6: structural data for TS <sub>0-2</sub> <sup>S</sup> , TS <sub>0-2</sub> <sup>T</sup> and TS <sub>2η1-2</sub> <sup>S</sup>                             |            |                                                                                                                                                                                                                |            |
|----------------------------------------------------------------------------------------------------------------------------------------------------------------|------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------|
| TS <sub>0-2</sub> <sup>S</sup>                                                                                                                                 |            | TS <sub>0-2</sub> <sup>T</sup>                                                                                                                                                                                 |            |
|                                                                              |            |                                                                                                                             |            |
| Nb atoms = 93<br>E = -2140.9823112 u.a.; G = -2140.313132 u.a.<br>Esp = -2142.98152507 u.a.; qh-G = -2140.283574 u.a.<br>Freq min: -17.2448 cm <sup>-1</sup> . |            | Nb atoms = 93<br>E = -2141.0299322 u.a.; G = -2140.360188 u.a.<br>Esp = -2143.02900564 u.a.; qh-G = -2140.331174 u.a.<br>Freq min: -60.5057 cm <sup>-1</sup> .<br>pop: Mo=0.308149; O46=0.825796; O47=0.852584 |            |
| C                                                                                                                                                              | 2.3696110  | -1.3375230                                                                                                                                                                                                     | -0.1283810 |
| C                                                                                                                                                              | 3.1166050  | -0.1169740                                                                                                                                                                                                     | -0.1696560 |
| C                                                                                                                                                              | 4.5385750  | -0.1591620                                                                                                                                                                                                     | -0.2950890 |
| C                                                                                                                                                              | 5.2229720  | -1.3626790                                                                                                                                                                                                     | -0.4111670 |
| C                                                                                                                                                              | 4.4414840  | -2.5561670                                                                                                                                                                                                     | -0.3992260 |
| C                                                                                                                                                              | 3.0475810  | -2.5943860                                                                                                                                                                                                     | -0.2613990 |
| C                                                                                                                                                              | 2.5263920  | 1.1841460                                                                                                                                                                                                      | -0.0646260 |
| N                                                                                                                                                              | 1.2178270  | 1.4936110                                                                                                                                                                                                      | -0.0315720 |
| Mo                                                                                                                                                             | -0.2220070 | 0.1315540                                                                                                                                                                                                      | 0.6897890  |
| O                                                                                                                                                              | 2.7501850  | 1.4061330                                                                                                                                                                                                      | 2.3416480  |
| O                                                                                                                                                              | 3.8134690  | 0.9171680                                                                                                                                                                                                      | 2.9247200  |
| C                                                                                                                                                              | 6.7589820  | -1.4574180                                                                                                                                                                                                     | -0.5377150 |
| C                                                                                                                                                              | 7.4333870  | -0.0629070                                                                                                                                                                                                     | -0.5402640 |
| C                                                                                                                                                              | 2.2723650  | -3.9331320                                                                                                                                                                                                     | -0.2303290 |
| C                                                                                                                                                              | 3.2082160  | -5.1563650                                                                                                                                                                                                     | -0.4168350 |
| O                                                                                                                                                              | 1.0283350  | -1.3107540                                                                                                                                                                                                     | 0.0664390  |
| N                                                                                                                                                              | -1.3622860 | 1.8349920                                                                                                                                                                                                      | 0.0757530  |
| C                                                                                                                                                              | -2.6905620 | 1.8208570                                                                                                                                                                                                      | -0.0310170 |
| C                                                                                                                                                              | -3.5600910 | 0.6884670                                                                                                                                                                                                      | -0.0418640 |
| C                                                                                                                                                              | -3.0848380 | -0.6672460                                                                                                                                                                                                     | -0.1064290 |
| C                                                                                                                                                              | -4.0342460 | -1.7416250                                                                                                                                                                                                     | -0.2233110 |
| C                                                                                                                                                              | -5.3907970 | -1.3982280                                                                                                                                                                                                     | -0.2327440 |
| C                                                                                                                                                              | -5.8994700 | -0.0637160                                                                                                                                                                                                     | -0.1574440 |
| C                                                                                                                                                              | -4.9668660 | 0.9583690                                                                                                                                                                                                      | -0.0767220 |
| O                                                                                                                                                              | -1.7641700 | -0.9396500                                                                                                                                                                                                     | -0.0872990 |
| C                                                                                                                                                              | -3.5686640 | -3.2134650                                                                                                                                                                                                     | -0.3267330 |
| C                                                                                                                                                              | -4.7628620 | -4.1976360                                                                                                                                                                                                     | -0.4367890 |
| C                                                                                                                                                              | -0.9544750 | 4.3648190                                                                                                                                                                                                      | -0.2628500 |
| C                                                                                                                                                              | -0.3958890 | 3.0771330                                                                                                                                                                                                      | -0.1250550 |
| C                                                                                                                                                              | 1.0113990  | 2.9297910                                                                                                                                                                                                      | 0.0220610  |
| C                                                                                                                                                              | 1.8409810  | 4.0592220                                                                                                                                                                                                      | 0.0357060  |
| C                                                                                                                                                              | 1.2755890  | 5.3412770                                                                                                                                                                                                      | -0.1176360 |
| C                                                                                                                                                              | -0.1216520 | 5.4898830                                                                                                                                                                                                      | -0.2674850 |
| N                                                                                                                                                              | -1.1385870 | 1.8569440                                                                                                                                                                                                      | -0.0557710 |
| C                                                                                                                                                              | -2.3840690 | 1.7647270                                                                                                                                                                                                      | -0.5260770 |
| C                                                                                                                                                              | -3.2810930 | 0.6649040                                                                                                                                                                                                      | -0.4289930 |
| C                                                                                                                                                              | -2.9422990 | -0.6078810                                                                                                                                                                                                     | 0.1721810  |
| C                                                                                                                                                              | -3.9810070 | -1.6053610                                                                                                                                                                                                     | 0.3112610  |
| C                                                                                                                                                              | -5.2397420 | -1.3046880                                                                                                                                                                                                     | -0.2110600 |
| C                                                                                                                                                              | -5.5891550 | -0.0787620                                                                                                                                                                                                     | -0.8667420 |
| C                                                                                                                                                              | -4.6005320 | 0.8834430                                                                                                                                                                                                      | -0.9558620 |
| O                                                                                                                                                              | -1.6997950 | -0.8920900                                                                                                                                                                                                     | 0.5700060  |
| Mo                                                                                                                                                             | -0.1009630 | 0.3595680                                                                                                                                                                                                      | 0.9909450  |
| O                                                                                                                                                              | -0.4266100 | 0.7963850                                                                                                                                                                                                      | 2.6192190  |
| C                                                                                                                                                              | -3.7012870 | -2.9512240                                                                                                                                                                                                     | 1.0220340  |
| C                                                                                                                                                              | -2.6180600 | -3.7476790                                                                                                                                                                                                     | 0.2381850  |
| C                                                                                                                                                              | -7.0201760 | 0.1095070                                                                                                                                                                                                      | -1.4139560 |
| C                                                                                                                                                              | -7.3186900 | -0.9918900                                                                                                                                                                                                     | -2.4740560 |
| N                                                                                                                                                              | 1.4514930  | 1.5743770                                                                                                                                                                                                      | 0.1613210  |
| C                                                                                                                                                              | 2.6529860  | 1.1923360                                                                                                                                                                                                      | -0.2592620 |
| C                                                                                                                                                              | 3.1338340  | -0.1478580                                                                                                                                                                                                     | -0.3503110 |
| C                                                                                                                                                              | 4.5167800  | -0.3228740                                                                                                                                                                                                     | -0.6707730 |
| C                                                                                                                                                              | 5.0648360  | -1.5840060                                                                                                                                                                                                     | -0.8461140 |
| C                                                                                                                                                              | 4.1733800  | -2.6964820                                                                                                                                                                                                     | -0.7324430 |

$$\text{TS}_{2\eta 1-2}^{\text{S}}$$



Nb atoms = 93

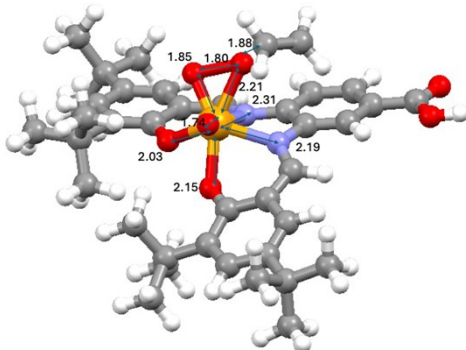
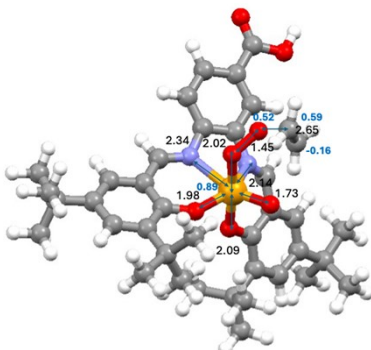
E=-2141.0205758 u.a.; G=-2140.345923 u.a.; Esp = -2143.02890473 u.a.; qh-G = -2140.317994 u.a. ; Freq min: -136.5468 cm<sup>-1</sup>.

|    |            |            |            |
|----|------------|------------|------------|
| C  | 0.8318820  | 2.8990230  | -0.0744280 |
| C  | -0.5772790 | 3.0371710  | -0.0124990 |
| C  | -1.1570790 | 4.3213840  | -0.0085110 |
| C  | -0.3441320 | 5.4562070  | -0.0959760 |
| C  | 1.0602390  | 5.3186150  | -0.1672070 |
| C  | 1.6475690  | 4.0388030  | -0.1403560 |
| N  | -1.2991270 | 1.8120970  | 0.0689810  |
| C  | -2.5624460 | 1.7467380  | -0.3065880 |
| C  | -3.3683900 | 0.5641070  | -0.3699340 |
| C  | -2.7838100 | -0.7341440 | -0.2401850 |
| C  | -3.5975610 | -1.9003730 | -0.4342150 |
| C  | -4.9559180 | -1.6863720 | -0.7058440 |
| C  | -5.5741840 | -0.4052190 | -0.8222550 |
| C  | -4.7547520 | 0.7073500  | -0.6656410 |
| O  | -1.4599390 | -0.8395900 | 0.0100190  |
| Mo | -0.1524650 | 0.1841880  | 1.1674740  |
| O  | -0.6186890 | -0.7557030 | 2.5468630  |
| C  | -3.0000330 | -3.3216670 | -0.3369140 |
| C  | -4.0602390 | -4.4236860 | -0.5975380 |
| C  | -7.0880030 | -0.3123240 | -1.1127360 |
| C  | -7.5757900 | 1.1545670  | -1.2121130 |
| C  | 1.8859070  | 6.5476080  | -0.2344680 |
| O  | 1.4534670  | 7.7135880  | -0.2550210 |
| N  | 1.3217720  | 1.5577790  | -0.0025890 |
| C  | 2.4876660  | 1.2502890  | -0.5343350 |
| C  | 3.1484080  | -0.0227640 | -0.4650660 |
| C  | 2.5530640  | -1.1455500 | 0.1822700  |
| C  | 3.2841070  | -2.3732340 | 0.3082140  |
| C  | 4.5582260  | -2.4080600 | -0.2763920 |
| C  | 5.1684850  | -1.3200120 | -0.9686730 |
| C  | 4.4455860  | -0.1337010 | -1.0436900 |
| O  | 1.2990360  | -1.0569190 | 0.6776530  |
| C  | 6.5759460  | -1.4978930 | -1.5790410 |
| C  | 7.0740100  | -0.2130180 | -2.2865300 |
| C  | 2.6994970  | -3.5913040 | 1.0610320  |
| C  | 3.7029920  | -4.7728110 | 1.1227770  |
| O  | 0.0937930  | 1.6962140  | 2.3030170  |
| O  | -1.1369550 | 1.9169910  | 2.9630120  |
| C  | 7.5906650  | -1.8564150 | -0.4530220 |
| C  | 6.5436200  | -2.6485800 | -2.6282820 |
| C  | 2.3484570  | -3.1919780 | 2.5262900  |
| C  | 1.4187720  | -4.0912350 | 0.3324710  |
| C  | -1.8805110 | -3.4849410 | -1.4082060 |
| C  | -2.4249260 | -3.5501100 | 1.0931820  |
| C  | -7.4056750 | -1.0220450 | -2.4622490 |
| C  | -7.8779490 | -1.0096810 | 0.0344060  |
| O  | 3.2518350  | 6.2805300  | -0.2827740 |

|   |            |            |            |
|---|------------|------------|------------|
| H | 3.0276320  | 2.0237920  | -1.0881700 |
| H | -5.1560170 | 1.7125500  | -0.7556270 |
| H | 4.8587870  | 0.7409390  | -1.5375960 |
| H | 2.7288180  | 3.9636650  | -0.1315050 |
| H | -2.2288460 | 4.4437960  | 0.1060950  |
| H | -5.5850890 | -2.5590290 | -0.8364770 |
| H | -3.0491930 | 2.6617600  | -0.6518940 |
| H | -0.7728070 | 6.4534760  | -0.0825880 |
| H | 5.1237900  | -3.3285410 | -0.1936640 |
| H | 1.0021710  | -4.9547440 | 0.8686770  |
| H | 0.6565310  | -3.3116550 | 0.2962740  |
| H | 1.6497870  | -4.4059930 | -0.6947630 |
| H | 1.9716190  | -4.0713860 | 3.0656950  |
| H | 3.2385900  | -2.8261090 | 3.0563870  |
| H | 1.5772920  | -2.4189520 | 2.5633110  |
| H | 3.2404310  | -5.6000570 | 1.6747980  |
| H | 3.9635390  | -5.1505200 | 0.1246660  |
| H | 4.6284420  | -4.5028020 | 1.6492290  |
| H | 8.5933450  | -1.9972130 | -0.8782690 |
| H | 7.6445580  | -1.0553330 | 0.2953650  |
| H | 7.3158170  | -2.7827290 | 0.0657890  |
| H | 7.5430530  | -2.7965810 | -3.0587090 |
| H | 6.2284870  | -3.5995510 | -2.1820200 |
| H | 5.8491830  | -2.4141110 | -3.4453950 |
| H | 8.0716360  | -0.3890350 | -2.7078680 |
| H | 6.4129760  | 0.0798530  | -3.1129700 |
| H | 7.1544960  | 0.6314920  | -1.5893790 |
| H | -1.9729130 | -4.5495160 | 1.1547480  |
| H | -1.6648790 | -2.8109200 | 1.3537980  |
| H | -3.2246920 | -3.4917860 | 1.8437110  |
| H | -1.4269230 | -4.4815390 | -1.3238860 |
| H | -2.2965100 | -3.3841020 | -2.4203650 |
| H | -1.0956110 | -2.7363830 | -1.2781250 |
| H | -3.5752300 | -5.4050060 | -0.5243170 |
| H | -4.8698970 | -4.4027720 | 0.1441860  |
| H | -4.5004010 | -4.3478420 | -1.6012030 |
| H | -8.6536680 | 1.1701360  | -1.4164720 |
| H | -7.4088150 | 1.7035710  | -0.2761320 |
| H | -7.0771780 | 1.6961120  | -2.0270810 |
| H | -8.9568900 | -0.9611670 | -0.1649190 |
| H | -7.6042480 | -2.0669750 | 0.1348110  |
| H | -7.6838420 | -0.5198680 | 0.9971060  |
| H | -8.4830280 | -0.9746360 | -2.6700020 |
| H | -6.8743430 | -0.5398560 | -3.2930700 |
| H | -7.1158740 | -2.0797050 | -2.4464220 |
| H | 3.7620200  | 7.1208890  | -0.3130480 |

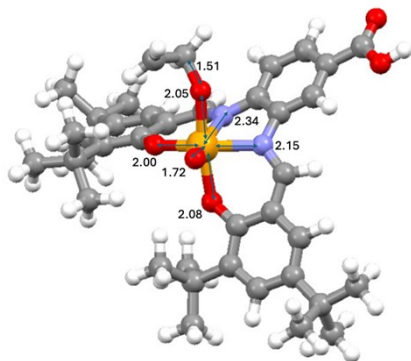
**Table S7:** structural data for transition states obtained for approach of ethylene at the different complexes.

| TS for $\text{Mo}2\eta_2^S + \text{C}_2\text{H}_4$ | TS for $\text{Mo}2\eta_1^T + \text{C}_2\text{H}_4$ |
|----------------------------------------------------|----------------------------------------------------|
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           |            |           |   |            |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |            |           |            |   |           |           |            |   |           |           |            |   |           |            |           |   |            |            |           |   |            |            |            |   |            |           |            |   |            |            |            |   |           |           |            |   |           |            |            |   |           |            |           |   |           |            |            |   |           |            |           |   |           |           |           |   |           |           |           |   |           |           |            |                                                                                               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| <p>Nb atoms = 99<br/>E = -2219.601815 u.a. ; G = -2218.879239 u.a.<br/>Esp = -2221.66323673 u.a.; qh-G = -2218.848933 u.a.<br/>Freq min : -559.6403 cm<sup>-1</sup>.</p>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               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|            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| <table><tr><td>C</td><td>4.5911150</td><td>-0.5462370</td><td>-0.1481820</td></tr><tr><td>C</td><td>3.1797670</td><td>-0.3123070</td><td>-0.1217450</td></tr><tr><td>C</td><td>2.2425580</td><td>-1.3811150</td><td>-0.3769060</td></tr><tr><td>C</td><td>2.7736630</td><td>-2.6879810</td><td>-0.7054600</td></tr><tr><td>C</td><td>4.1599760</td><td>-2.8465400</td><td>-0.6804520</td></tr><tr><td>C</td><td>5.1054080</td><td>-1.8069520</td><td>-0.3988030</td></tr><tr><td>C</td><td>2.7257220</td><td>1.0347340</td><td>-0.0867260</td></tr><tr><td>N</td><td>1.4733170</td><td>1.4466770</td><td>0.0973850</td></tr><tr><td>Mo</td><td>-0.0742980</td><td>0.1788070</td><td>0.9872460</td></tr><tr><td>O</td><td>-0.2318650</td><td>1.8568370</td><td>2.4145460</td></tr><tr><td>O</td><td>-1.5100780</td><td>0.6404720</td><td>2.0647110</td></tr><tr><td>O</td><td>0.9307280</td><td>-1.1424370</td><td>-0.3756400</td></tr><tr><td>C</td><td>1.8327430</td><td>-3.8767670</td><td>-1.0012740</td></tr><tr><td>C</td><td>0.8844320</td><td>-3.5260610</td><td>-2.1858470</td></tr><tr><td>C</td><td>6.6144640</td><td>-2.1310300</td><td>-0.4005710</td></tr><tr><td>C</td><td>7.0326530</td><td>-2.6745880</td><td>-1.7989760</td></tr><tr><td>N</td><td>-1.0678190</td><td>1.7394780</td><td>-0.3997790</td></tr><tr><td>C</td><td>-2.3330190</td><td>1.6601750</td><td>-0.7605030</td></tr><tr><td>C</td><td>-3.2362670</td><td>0.5675320</td><td>-0.5309450</td></tr><tr><td>C</td><td>-2.8330330</td><td>-0.7173770</td><td>-0.0321790</td></tr><tr><td>C</td><td>-3.8279050</td><td>-1.7342780</td><td>0.1722070</td></tr><tr><td>C</td><td>-5.1549290</td><td>-1.4069500</td><td>-0.1368630</td></tr><tr><td>C</td><td>-5.5854930</td><td>-0.1513230</td><td>-0.6557210</td></tr><tr><td>C</td><td>-4.6054300</td><td>0.8124730</td><td>-0.8534760</td></tr><tr><td>O</td><td>-1.5199560</td><td>-0.9852220</td><td>0.1690170</td></tr><tr><td>C</td><td>-3.4566640</td><td>-3.1441460</td><td>0.6893430</td></tr><tr><td>C</td><td>-2.7117730</td><td>-3.0500970</td><td>2.0532330</td></tr><tr><td>C</td><td>-7.0815420</td><td>0.0776790</td><td>-0.9616130</td></tr><tr><td>C</td><td>-7.9097720</td><td>-0.0948660</td><td>0.3461950</td></tr><tr><td>C</td><td>-0.2977300</td><td>2.9060830</td><td>-0.6069450</td></tr><tr><td>C</td><td>1.0807090</td><td>2.7605410</td><td>-0.3036770</td></tr><tr><td>C</td><td>1.9507860</td><td>3.8536900</td><td>-0.4408900</td></tr><tr><td>C</td><td>1.4556520</td><td>5.0955320</td><td>-0.8801900</td></tr><tr><td>C</td><td>0.0816750</td><td>5.2459780</td><td>-1.1788230</td></tr><tr><td>C</td><td>-0.7858130</td><td>4.1607550</td><td>-1.0396770</td></tr><tr><td>C</td><td>2.3431920</td><td>6.2715750</td><td>-1.0322200</td></tr><tr><td>O</td><td>3.6687040</td><td>5.9975300</td><td>-0.7015990</td></tr><tr><td>O</td><td>0.9888980</td><td>-0.5453670</td><td>2.1565450</td></tr><tr><td>C</td><td>-4.7056700</td><td>-4.0384270</td><td>0.9111690</td></tr><tr><td>C</td><td>-2.5592040</td><td>-3.8410880</td><td>-0.3742160</td></tr><tr><td>C</td><td>-7.3546360</td><td>1.4956030</td><td>-1.5224140</td></tr><tr><td>C</td><td>-7.5629890</td><td>-0.9620760</td><td>-2.0165590</td></tr><tr><td>O</td><td>1.9925880</td><td>7.4034850</td><td>-1.4111990</td></tr><tr><td>C</td><td>7.4804590</td><td>-0.8852020</td><td>-0.0887180</td></tr><tr><td>C</td><td>6.9120110</td><td>-3.2116840</td><td>0.6811840</td></tr><tr><td>C</td><td>2.6094270</td><td>-5.1622130</td><td>-1.3908940</td></tr><tr><td>C</td><td>1.0111230</td><td>-4.1929860</td><td>0.2834670</td></tr><tr><td>C</td><td>1.0364230</td><td>2.2904440</td><td>3.7303540</td></tr><tr><td>C</td><td>1.1484300</td><td>3.4927180</td><td>3.0475750</td></tr><tr><td>H</td><td>3.4758880</td><td>1.7871550</td><td>-0.3360580</td></tr></table> | C                                                                                                                                                                                                                                           | 4.5911150  | -0.5462370 | -0.1481820 | C | 3.1797670 | -0.3123070 | -0.1217450 | C | 2.2425580 | -1.3811150 | -0.3769060 | C | 2.7736630 | -2.6879810 | -0.7054600 | C | 4.1599760 | -2.8465400 | -0.6804520 | C | 5.1054080 | -1.8069520 | -0.3988030 | C | 2.7257220 | 1.0347340 | -0.0867260 | N | 1.4733170 | 1.4466770 | 0.0973850 | Mo | -0.0742980 | 0.1788070 | 0.9872460 | O | -0.2318650 | 1.8568370 | 2.4145460 | O | -1.5100780 | 0.6404720 | 2.0647110 | O | 0.9307280 | -1.1424370 | -0.3756400 | C | 1.8327430 | -3.8767670 | -1.0012740 | C | 0.8844320 | -3.5260610 | -2.1858470 | C | 6.6144640 | -2.1310300 | -0.4005710 | C | 7.0326530 | -2.6745880 | -1.7989760 | N | -1.0678190 | 1.7394780 | -0.3997790 | C | -2.3330190 | 1.6601750 | -0.7605030 | C | -3.2362670 | 0.5675320 | -0.5309450 | C | -2.8330330 | -0.7173770 | -0.0321790 | C | -3.8279050 | -1.7342780 | 0.1722070 | C | -5.1549290 | -1.4069500 | -0.1368630 | C | -5.5854930 | -0.1513230 | -0.6557210 | C | -4.6054300 | 0.8124730 | -0.8534760 | O | -1.5199560 | -0.9852220 | 0.1690170 | C | -3.4566640 | -3.1441460 | 0.6893430 | C | -2.7117730 | -3.0500970 | 2.0532330 | C | -7.0815420 | 0.0776790 | -0.9616130 | C | -7.9097720 | -0.0948660 | 0.3461950 | C | -0.2977300 | 2.9060830 | -0.6069450 | C | 1.0807090 | 2.7605410 | -0.3036770 | C | 1.9507860 | 3.8536900 | -0.4408900 | C | 1.4556520 | 5.0955320 | -0.8801900 | C | 0.0816750 | 5.2459780 | -1.1788230 | C | -0.7858130 | 4.1607550 | -1.0396770 | C | 2.3431920 | 6.2715750 | -1.0322200 | O | 3.6687040 | 5.9975300 | -0.7015990 | O | 0.9888980 | -0.5453670 | 2.1565450 | C | -4.7056700 | -4.0384270 | 0.9111690 | C | -2.5592040 | -3.8410880 | -0.3742160 | C | -7.3546360 | 1.4956030 | -1.5224140 | C | -7.5629890 | -0.9620760 | -2.0165590 | O | 1.9925880 | 7.4034850 | -1.4111990 | C | 7.4804590 | -0.8852020 | -0.0887180 | C | 6.9120110 | -3.2116840 | 0.6811840 | C | 2.6094270 | -5.1622130 | -1.3908940 | C | 1.0111230 | -4.1929860 | 0.2834670 | C | 1.0364230 | 2.2904440 | 3.7303540 | C | 1.1484300 | 3.4927180 | 3.0475750 | H | 3.4758880 | 1.7871550 | -0.3360580 | <table><tr><td>C</td><td>4.5832390</td><td>-0.7431330</td><td>-0.5153830</td></tr><tr><td>C</td><td>3.1959290</td><td>-0.4587600</td><td>-0.3184170</td></tr><tr><td>C</td><td>2.2222330</td><td>-1.5222500</td><td>-0.3184190</td></tr><tr><td>C</td><td>2.6751870</td><td>-2.8744310</td><td>-0.5424260</td></tr><tr><td>C</td><td>4.0492210</td><td>-3.0762550</td><td>-0.7007300</td></tr><tr><td>C</td><td>5.0374320</td><td>-2.0419270</td><td>-0.6880000</td></tr><tr><td>C</td><td>2.7958250</td><td>0.9145250</td><td>-0.3248700</td></tr><tr><td>N</td><td>1.5973630</td><td>1.3957200</td><td>-0.0215050</td></tr><tr><td>Mo</td><td>0.0700600</td><td>0.2796280</td><td>0.9822480</td></tr><tr><td>O</td><td>-0.8544240</td><td>1.7789810</td><td>1.9800700</td></tr><tr><td>O</td><td>0.0625920</td><td>2.7852860</td><td>2.4882210</td></tr><tr><td>O</td><td>0.9191990</td><td>-1.2503780</td><td>-0.1572970</td></tr><tr><td>C</td><td>1.6772500</td><td>-4.0559940</td><td>-0.5515650</td></tr><tr><td>C</td><td>0.5791980</td><td>-3.8269090</td><td>-1.6322330</td></tr><tr><td>C</td><td>6.5260780</td><td>-2.4082650</td><td>-0.8718300</td></tr><tr><td>C</td><td>6.7250140</td><td>-3.1203130</td><td>-2.2426410</td></tr><tr><td>N</td><td>-0.9035790</td><td>1.6070310</td><td>-0.6855490</td></tr><tr><td>C</td><td>-2.0980000</td><td>1.4494580</td><td>-1.2184200</td></tr><tr><td>C</td><td>-3.0572510</td><td>0.4263700</td><td>-0.8960940</td></tr><tr><td>C</td><td>-2.8639520</td><td>-0.5335740</td><td>0.1520990</td></tr><tr><td>C</td><td>-3.9406190</td><td>-1.4128890</td><td>0.5173180</td></tr><tr><td>C</td><td>-5.1204040</td><td>-1.3237040</td><td>-0.2325620</td></tr><tr><td>C</td><td>-5.3317890</td><td>-0.4155990</td><td>-1.3120720</td></tr><tr><td>C</td><td>-4.2912610</td><td>0.4550720</td><td>-1.6129920</td></tr><tr><td>O</td><td>-1.6773560</td><td>-0.6221210</td><td>0.7841050</td></tr><tr><td>C</td><td>-3.8024350</td><td>-2.4133300</td><td>1.6899630</td></tr><tr><td>C</td><td>-3.4762780</td><td>-1.6436560</td><td>3.0051780</td></tr><tr><td>C</td><td>-6.6786580</td><td>-0.4273510</td><td>-2.0677620</td></tr><tr><td>C</td><td>-7.8383690</td><td>-0.1234780</td><td>-1.0734650</td></tr><tr><td>C</td><td>-0.1478290</td><td>2.7935560</td><td>-0.8703700</td></tr><tr><td>C</td><td>1.2058630</td><td>2.7051760</td><td>-0.4516850</td></tr><tr><td>C</td><td>2.0493820</td><td>3.8189450</td><td>-0.5017280</td></tr><tr><td>C</td><td>1.5491620</td><td>5.0431530</td><td>-0.9896230</td></tr><tr><td>C</td><td>0.2028120</td><td>5.1424800</td><td>-1.4090480</td></tr><tr><td>C</td><td>-0.6422480</td><td>4.0298870</td><td>-1.3411600</td></tr><tr><td>C</td><td>2.3981750</td><td>6.2547930</td><td>-1.0558590</td></tr><tr><td>O</td><td>3.6997120</td><td>6.0296870</td><td>-0.6136010</td></tr><tr><td>O</td><td>0.9518470</td><td>-0.1977620</td><td>2.3924630</td></tr><tr><td>C</td><td>-5.1084410</td><td>-3.2151800</td><td>1.9329990</td></tr><tr><td>C</td><td>-2.6726820</td><td>-3.4367400</td><td>1.3772930</td></tr><tr><td>C</td><td>-6.7249380</td><td>0.6294380</td><td>-3.1994370</td></tr><tr><td>C</td><td>-6.9026520</td><td>-1.8306440</td><td>-2.7053510</td></tr><tr><td>O</td><td>2.0382260</td><td>7.3751080</td><td>-1.4605410</td></tr><tr><td>C</td><td>7.4438820</td><td>-1.1608230</td><td>-0.8387080</td></tr><tr><td>C</td><td>6.9686090</td><td>-3.3660390</td><td>0.2740730</td></tr><tr><td>C</td><td>2.3685750</td><td>-5.4081090</td><td>-0.8692470</td></tr><tr><td>C</td><td>1.0268920</td><td>-4.1774280</td><td>0.8585520</td></tr><tr><td>C</td><td>0.5812930</td><td>2.4015640</td><td>4.3021020</td></tr><tr><td>C</td><td>-0.4498570</td><td>2.9290590</td><td>5.0845500</td></tr><tr><td>H</td><td>3.5369240</td><td>1.6217510</td><td>-0.7052530</td></tr></table> | C | 4.5832390 | -0.7431330 | -0.5153830 | C | 3.1959290 | -0.4587600 | -0.3184170 | C | 2.2222330 | -1.5222500 | -0.3184190 | C | 2.6751870 | -2.8744310 | -0.5424260 | C | 4.0492210 | -3.0762550 | -0.7007300 | C | 5.0374320 | -2.0419270 | -0.6880000 | C | 2.7958250 | 0.9145250 | -0.3248700 | N | 1.5973630 | 1.3957200 | -0.0215050 | Mo | 0.0700600 | 0.2796280 | 0.9822480 | O | -0.8544240 | 1.7789810 | 1.9800700 | O | 0.0625920 | 2.7852860 | 2.4882210 | O | 0.9191990 | -1.2503780 | -0.1572970 | C | 1.6772500 | -4.0559940 | -0.5515650 | C | 0.5791980 | -3.8269090 | -1.6322330 | C | 6.5260780 | -2.4082650 | -0.8718300 | C | 6.7250140 | -3.1203130 | -2.2426410 | N | -0.9035790 | 1.6070310 | -0.6855490 | C | -2.0980000 | 1.4494580 | -1.2184200 | C | -3.0572510 | 0.4263700 | -0.8960940 | C | -2.8639520 | -0.5335740 | 0.1520990 | C | -3.9406190 | -1.4128890 | 0.5173180 | C | -5.1204040 | -1.3237040 | -0.2325620 | C | -5.3317890 | -0.4155990 | -1.3120720 | C | -4.2912610 | 0.4550720 | -1.6129920 | O | -1.6773560 | -0.6221210 | 0.7841050 | C | -3.8024350 | -2.4133300 | 1.6899630 | C | -3.4762780 | -1.6436560 | 3.0051780 | C | -6.6786580 | -0.4273510 | -2.0677620 | C | -7.8383690 | -0.1234780 | -1.0734650 | C | -0.1478290 | 2.7935560 | -0.8703700 | C | 1.2058630 | 2.7051760 | -0.4516850 | C | 2.0493820 | 3.8189450 | -0.5017280 | C | 1.5491620 | 5.0431530 | -0.9896230 | C | 0.2028120 | 5.1424800 | -1.4090480 | C | -0.6422480 | 4.0298870 | -1.3411600 | C | 2.3981750 | 6.2547930 | -1.0558590 | O | 3.6997120 | 6.0296870 | -0.6136010 | O | 0.9518470 | -0.1977620 | 2.3924630 | C | -5.1084410 | -3.2151800 | 1.9329990 | C | -2.6726820 | -3.4367400 | 1.3772930 | C | -6.7249380 | 0.6294380 | -3.1994370 | C | -6.9026520 | -1.8306440 | -2.7053510 | O | 2.0382260 | 7.3751080 | -1.4605410 | C | 7.4438820 | -1.1608230 | -0.8387080 | C | 6.9686090 | -3.3660390 | 0.2740730 | C | 2.3685750 | -5.4081090 | -0.8692470 | C | 1.0268920 | -4.1774280 | 0.8585520 | C | 0.5812930 | 2.4015640 | 4.3021020 | C | -0.4498570 | 2.9290590 | 5.0845500 | H | 3.5369240 | 1.6217510 | -0.7052530 |
| C                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      | 4.5911150                                                                                                                                                                                                                                   | -0.5462370 | -0.1481820 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |            |            |           |   |            |            |           |   |            |            |           |   |            |           |            |   |            |            |           |   |            |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |            |           |            |   |           |           |            |   |           |           |            |   |           |            |           |   |            |            |           |   |            |            |            |   |            |           |            |   |            |            |            |   |           |           |            |   |           |            |            |   |           |            |           |   |           |            |            |   |           |            |           |   |           |           |           |   |         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|            |            |   |           |            |            |   |           |           |            |   |           |           |            |    |           |           |           |   |            |           |           |   |           |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |           |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |            |            |           |   |            |            |           |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |            |           |            |   |           |           |            |   |           |           |            |   |           |            |           |   |            |            |           |   |            |            |           |   |            |           |            |   |            |            |            |   |           |           |            |   |           |            |            |   |           |            |           |   |           |            |            |   |           |            |           |   |           |           |           |   |            |           |           |   |           |           |            |
| C                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      | 3.1797670                                                                                                                                                                                                                                   | -0.3123070 | -0.1217450 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| -0.6804520 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| -0.0867260 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| 0.1690170  |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| 0.6811840  |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| -1.3908940 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| 0.2834670  |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| -0.5153830 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| -0.3184170 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| -0.3184190 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| -0.5424260 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| -0.7007300 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| -0.6880000 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| 3.0051780  |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| -2.7053510 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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| -0.8387080 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |      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                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              | -0.4498570                                                                                                                                                                                                                                  | 2.9290590  | 5.0845500  |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |            |            |           |   |            |            |           |   |            |            |           |   |            |           |            |   |            |            |           |   |            |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |            |           |            |   |           |           |            |   |           |           |            |   |           |            |           |   |            |            |           |   |            |            |            |   |            |           |            |   |            |            |            |   |           |           |            |   |           |            |            |   |           |            |           |   |           |            |            |   |           |            |           |   |           |           |           |   |         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|            |            |   |           |            |            |   |           |           |            |   |           |           |            |    |           |           |           |   |            |           |           |   |           |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |           |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |            |            |           |   |            |            |           |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |            |           |            |   |           |           |            |   |           |           |            |   |           |            |           |   |            |            |           |   |            |            |           |   |            |           |            |   |            |            |            |   |           |           |            |   |           |            |            |   |           |            |           |   |           |            |            |   |           |            |           |   |           |           |           |   |            |           |           |   |           |           |            |
| H                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      | 3.5369240                                                                                                                                                                                                                                   | 1.6217510  | -0.7052530 |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |           |            |   |           |           |           |    |            |           |           |   |            |           |           |   |            |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |            |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |            |            |           |   |            |            |           |   |            |            |           |   |            |           |            |   |            |            |           |   |            |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |           |           |            |   |            |           |            |   |           |           |            |   |           |           |            |   |           |            |           |   |            |            |           |   |            |            |            |   |            |           |            |   |            |            |            |   |           |           |            |   |           |            |            |   |           |            |           |   |           |            |            |   |           |            |           |   |           |           |           |   |         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|            |            |   |           |            |            |   |           |           |            |   |           |           |            |    |           |           |           |   |            |           |           |   |           |           |           |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |           |            |            |   |            |           |            |   |            |           |            |   |            |           |            |   |            |            |           |   |            |            |           |   |            |            |            |   |            |            |            |   |            |           |            |   |            |            |           |   |            |            |           |   |            |            |           |   |            |            |            |   |       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|   |            |            |            |   |            |            |            |
|---|------------|------------|------------|---|------------|------------|------------|
| H | -4.8651720 | 1.7924750  | -1.2430610 | H | -4.3975390 | 1.1921400  | -2.4038040 |
| H | 5.2487290  | 0.2971990  | 0.0421910  | H | 5.2757200  | 0.0938140  | -0.5200920 |
| H | 3.0056400  | 3.7793420  | -0.2039210 | H | 3.0728240  | 3.7704320  | -0.1470580 |
| H | -1.8404570 | 4.3034400  | -1.2499240 | H | -1.6861040 | 4.1395580  | -1.6169880 |
| H | -5.9112420 | -2.1649320 | 0.0278540  | H | -5.9342970 | -1.9899970 | 0.0279430  |
| H | -2.7859330 | 2.5102030  | -1.2776630 | H | -2.4431190 | 2.1757460  | -1.9612900 |
| H | -0.2810480 | 6.2158340  | -1.5049340 | H | -0.1649560 | 6.1019960  | -1.7596220 |
| H | 4.5563000  | -3.8330290 | -0.8917990 | H | 4.3949050  | -4.0930460 | -0.8475170 |
| H | 0.1969830  | -4.3626430 | -2.3717420 | H | -0.1282890 | -4.6675580 | -1.6243460 |
| H | 0.2980130  | -2.6311930 | -1.9679810 | H | 0.0279590  | -2.9036390 | -1.4426950 |
| H | 1.4635590  | -3.3526450 | -3.1038130 | H | 1.0271150  | -3.7699100 | -2.6342000 |
| H | 0.3315290  | -5.0346270 | 0.0957580  | H | 0.2790750  | -4.9821730 | 0.8591000  |
| H | 1.6807710  | -4.4696730 | 1.1089710  | H | 1.7867100  | -4.4141020 | 1.6155680  |
| H | 0.4165010  | -3.3322530 | 0.5955070  | H | 0.5327790  | -3.2477630 | 1.1474170  |
| H | 1.8869900  | -5.9566780 | -1.6163120 | H | 1.6096800  | -6.2005130 | -0.8768800 |
| H | 3.2293000  | -5.0149100 | -2.2858760 | H | 2.8517430  | -5.4030180 | -1.8558650 |
| H | 3.2499480  | -5.5256290 | -0.5759610 | H | 3.1171690  | -5.6807500 | -0.1132890 |
| H | 7.9813920  | -3.4627230 | 0.6831480  | H | 8.0235490  | -3.6453970 | 0.1493140  |
| H | 6.6417240  | -2.8481670 | 1.6808650  | H | 6.8539340  | -2.8831220 | 1.2529670  |
| H | 6.3520870  | -4.1369070 | 0.4984470  | H | 6.3776660  | -4.2899590 | 0.2847660  |
| H | 8.1023140  | -2.9241460 | -1.8042020 | H | 7.7789420  | -3.3997430 | -2.3752250 |
| H | 6.4777770  | -3.5815040 | -2.0681300 | H | 6.1256330  | -4.0356380 | -2.3188000 |
| H | 6.8507700  | -1.9251090 | -2.5801670 | H | 6.4376860  | -2.4603920 | -3.0714840 |
| H | 8.5419330  | -1.1635110 | -0.0954070 | H | 8.4889620  | -1.4692870 | -0.9683580 |
| H | 7.3408760  | -0.0945030 | -0.8379260 | H | 7.2041260  | -0.4569710 | -1.6468280 |
| H | 7.2526850  | -0.4694550 | 0.9015670  | H | 7.3718820  | -0.6281220 | 0.1186140  |
| H | -2.4742280 | -4.0606320 | 2.4128550  | H | -3.3784230 | -2.3565170 | 3.8352870  |
| H | -1.7797370 | -2.4894120 | 1.9717830  | H | -2.5430540 | -1.0822090 | 2.9237650  |
| H | -3.3401800 | -2.5575050 | 2.8069630  | H | -4.2820210 | -0.9396720 | 3.2537290  |
| H | -2.2504880 | -4.8312170 | -0.0123990 | H | -2.5580620 | -4.1338770 | 2.2187170  |
| H | -3.1119520 | -3.9775950 | -1.3142090 | H | -2.9156980 | -4.0219660 | 0.4795470  |
| H | -1.6639190 | -3.2525680 | -0.5787650 | H | -1.7185500 | -2.9344540 | 1.2134650  |
| H | -4.3763150 | -5.0164530 | 1.2834740  | H | -4.9531670 | -3.8945650 | 2.7802980  |
| H | -5.3902440 | -3.6144940 | 1.6582870  | H | -5.9545490 | -2.5617910 | 2.1850600  |
| H | -5.2639070 | -4.2147500 | -0.0181520 | H | -5.3856160 | -3.8304700 | 1.0663460  |
| H | -8.4267700 | 1.6100430  | -1.7261340 | H | -7.6945970 | 0.5785100  | -3.7105610 |
| H | -7.0683920 | 2.2782380  | -0.8075080 | H | -6.6102970 | 1.6494440  | -2.8092900 |
| H | -6.8181530 | 1.6715300  | -2.4643180 | H | -5.9443130 | 0.4551980  | -3.9517320 |
| H | -8.9784190 | 0.0543600  | 0.1407970  | H | -8.8028010 | -0.1411470 | -1.5988130 |
| H | -7.7890510 | -1.0964370 | 0.7764380  | H | -7.8863990 | -0.8616210 | -0.2637140 |
| H | -7.5996000 | 0.6353640  | 1.1045310  | H | -7.7132600 | 0.8671930  | -0.6177320 |
| H | -8.6312410 | -0.8199450 | -2.2287020 | H | -7.8660570 | -1.8589780 | -3.2321300 |
| H | -7.0081310 | -0.8510270 | -2.9572010 | H | -6.1091330 | -2.0632970 | -3.4272190 |
| H | -7.4268720 | -1.9923300 | -1.6660310 | H | -6.9119410 | -2.6251310 | -1.9492980 |
| H | 4.2210820  | 6.8034960  | -0.8140250 | H | 4.2240850  | 6.8602230  | -0.6644780 |
| H | 1.7561410  | 1.4872400  | 3.5970300  | H | 1.5098730  | 2.9607820  | 4.1970300  |
| H | 1.8862740  | 3.6344990  | 2.2667950  | H | -0.4531160 | 3.9740810  | 5.3861580  |
| H | 0.4792030  | 4.3216060  | 3.2571320  | H | -1.3372580 | 2.3421680  | 5.3084640  |
| H | 0.3838070  | 2.2018910  | 4.5937190  | H | 0.6647970  | 1.3295460  | 4.1464400  |

### TS for Mo<sub>1</sub> + C<sub>2</sub>H<sub>4</sub>



Nb atoms = 98

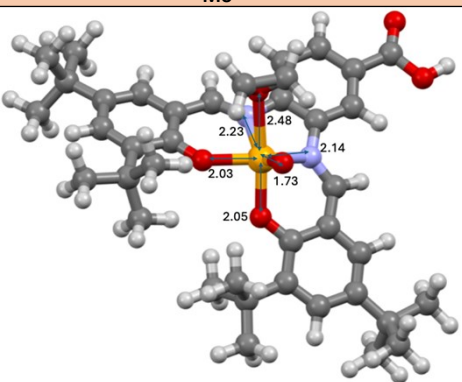
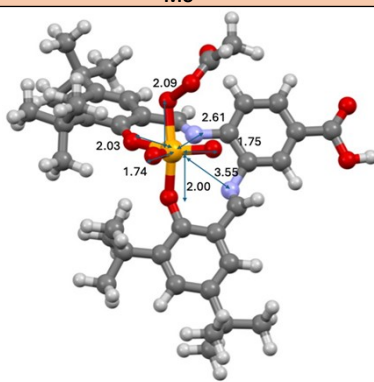
E = -2144.4496912 u.a.; G = -2143.729604 u.a.

Esp = -2146.47934023 u.a. ; qh-G = -2143.699385 u.a.

Freq min: -273.9864

|   |            |           |            |   |           |           |           |
|---|------------|-----------|------------|---|-----------|-----------|-----------|
| C | -0.4676230 | 4.1544650 | -1.0146360 | H | 3.2229760 | 3.8359420 | 0.2430550 |
|---|------------|-----------|------------|---|-----------|-----------|-----------|

|    |            |            |            |   |            |            |            |
|----|------------|------------|------------|---|------------|------------|------------|
| C  | -0.0000580 | 2.9085990  | -0.5378710 | H | -1.5003980 | 4.2730230  | -1.3285230 |
| C  | 1.3471140  | 2.8007160  | -0.0929470 | H | -5.7952900 | -1.9452400 | -0.1402810 |
| C  | 2.2035110  | 3.9033740  | -0.1205270 | H | -2.1951830 | 2.2777440  | -1.8150770 |
| C  | 1.7249980  | 5.1437350  | -0.5998390 | H | 0.0421870  | 6.2262460  | -1.4035120 |
| C  | 0.3906490  | 5.2605070  | -1.0497460 | H | 4.3059590  | -4.0351930 | -0.9142810 |
| N  | -0.7732890 | 1.7296590  | -0.3890810 | H | -0.2410950 | -4.4056420 | -1.6734320 |
| C  | -1.9237720 | 1.5638130  | -1.0306480 | H | -0.0232150 | -2.6651700 | -1.3652130 |
| C  | -2.8934010 | 0.5379150  | -0.7879540 | H | 0.9317840  | -3.4736740 | -2.6310840 |
| C  | -2.7903620 | -0.4037210 | 0.2986630  | H | 0.1826850  | -4.9122500 | 0.7740020  |
| C  | -3.8836040 | -1.3118200 | 0.5535620  | H | 1.7206570  | -4.4577190 | 1.5466540  |
| C  | -4.9760310 | -1.2581180 | -0.3158080 | H | 0.5066030  | -3.2133200 | 1.1867570  |
| C  | -5.0944420 | -0.3632790 | -1.4264740 | H | 1.4550970  | -6.0421840 | -1.0680710 |
| C  | -4.0499610 | 0.5245680  | -1.6316910 | H | 2.7126010  | -5.2131770 | -2.0008240 |
| O  | -1.6953930 | -0.4295960 | 1.0701370  | H | 2.9870970  | -5.6294070 | -0.2877090 |
| Mo | 0.1383710  | 0.3386860  | 1.2519150  | H | 7.9558700  | -3.7851870 | 0.0807480  |
| O  | -0.4470030 | 1.9796910  | 2.3323270  | H | 6.8192290  | -3.0680630 | 1.2478660  |
| C  | -3.8352150 | -2.3260990 | 1.7224590  | H | 6.2891260  | -4.3819580 | 0.1822220  |
| C  | -3.6405080 | -1.5837050 | 3.0776110  | H | 7.7051130  | -3.3431660 | -2.4160160 |
| C  | -6.3533120 | -0.4220090 | -2.3189320 | H | 6.0312310  | -3.9262710 | -2.3927010 |
| C  | -7.6196680 | -0.1403520 | -1.4569880 | H | 6.3914590  | -2.3105290 | -3.0286620 |
| N  | 1.7068940  | 1.4778420  | 0.3191850  | H | 8.4867720  | -1.5468650 | -0.8740890 |
| C  | 2.8817730  | 0.9744390  | -0.0293180 | H | 7.2320220  | -0.4457190 | -1.4659440 |
| C  | 3.2334340  | -0.4130280 | -0.1097000 | H | 7.4029080  | -0.7526520 | 0.2804260  |
| C  | 4.6078610  | -0.7284630 | -0.3381770 | H | -3.6528720 | -2.3090710 | 3.9021740  |
| C  | 5.0192890  | -2.0256790 | -0.6091780 | H | -2.6825740 | -1.0594350 | 3.1013430  |
| C  | 3.9971350  | -3.0207450 | -0.6893360 | H | -4.4506560 | -0.8609470 | 3.2488440  |
| C  | 2.6315170  | -2.7842800 | -0.5009640 | H | -2.6087620 | -4.0339280 | 2.3328320  |
| C  | 2.2237160  | -1.4386360 | -0.1727810 | H | -2.7985640 | -3.8872920 | 0.5699960  |
| O  | 0.9278890  | -1.1548180 | 0.0375060  | H | -1.7040100 | -2.7936810 | 1.4409800  |
| C  | 1.5966050  | -3.9310960 | -0.5843140 | H | -5.0512500 | -3.8467460 | 2.6808740  |
| C  | 0.4938330  | -3.5897000 | -1.6297320 | H | -6.0208550 | -2.5238890 | 2.0121840  |
| C  | 6.4939280  | -2.4257690 | -0.8323880 | H | -5.3230500 | -3.7637020 | 0.9358140  |
| C  | 6.6618490  | -3.0397060 | -2.2538570 | H | -7.2189350 | 0.5377580  | -4.0721900 |
| C  | 2.5859060  | 6.3460260  | -0.6322650 | H | -6.2595530 | 1.6469240  | -3.0793330 |
| O  | 2.2452230  | 7.4777280  | -1.0250540 | H | -5.4504720 | 0.4567900  | -4.1294660 |
| O  | 0.9884490  | -0.2450530 | 2.6305740  | H | -8.5230000 | -0.1967070 | -2.0793620 |
| C  | 7.4529740  | -1.2150540 | -0.7140900 | H | -7.7298650 | -0.8672210 | -0.6431000 |
| C  | 6.9117170  | -3.4804910 | 0.2349620  | H | -7.5729060 | 0.8608910  | -1.0092890 |
| C  | -6.3088410 | 0.6197470  | -3.4645460 | H | -7.3712550 | -1.8991410 | -3.5834840 |
| C  | -6.4710940 | -1.8381410 | -2.9569130 | H | -5.5985780 | -2.0565860 | -3.5859920 |
| O  | 3.8776170  | 6.1015710  | -0.1702140 | H | -6.5405460 | -2.6242240 | -2.1952270 |
| C  | -5.1411500 | -3.1567770 | 1.8326540  | H | 4.4113440  | 6.9269860  | -0.2033840 |
| C  | -2.6582050 | -3.3193180 | 1.4997040  | C | -1.9862480 | 2.0836710  | 3.6762460  |
| C  | 2.2397340  | -5.2771050 | -1.0112450 | C | -1.7319390 | 2.7737070  | 2.4156500  |
| C  | 0.9598990  | -4.1369860 | 0.8221640  | H | -1.5182990 | 2.4336560  | 4.5901570  |
| H  | 3.6413370  | 1.6733860  | -0.3894000 | H | -1.4847500 | 3.8352390  | 2.4722560  |
| H  | -4.0845580 | 1.2429280  | -2.4459770 | H | -2.4090570 | 2.5482290  | 1.5896270  |
| H  | 5.3268690  | 0.0847380  | -0.2913800 | H | -2.4644680 | 1.1107290  | 3.6759550  |

| Table S8: structural data for $\text{Mo}3^{\text{S}}$ and $\text{Mo}4^{\text{S}}$   |                                                                                      |
|-------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------|
| $\text{Mo}3^{\text{S}}$                                                             | $\text{Mo}4^{\text{S}}$                                                              |
|  |  |
| Nb atoms = 98                                                                       | 100                                                                                  |

|                                                                                                |            |            |            |                                                                                               |            |            |            |
|------------------------------------------------------------------------------------------------|------------|------------|------------|-----------------------------------------------------------------------------------------------|------------|------------|------------|
| E=-2144.4803845 u.a. ; G=-2143.758316 u.a.<br>Esp=-2146.51148752 u.a.; qh-G= -2143.727661 u.a. |            |            |            | E=-2369.5986934 u.a. ; G=-2368.886387 u.a.<br>Esp=-2371.76171838 u.a.; qh-G=-2368.854198 u.a. |            |            |            |
| C                                                                                              | 2.4766360  | -1.3383940 | -0.2268680 | C                                                                                             | 2.6399050  | -1.1891820 | -0.1629590 |
| C                                                                                              | 3.3210560  | -0.1747030 | -0.2217920 | C                                                                                             | 3.4824470  | -0.0564320 | -0.3628640 |
| C                                                                                              | 4.7311460  | -0.3042550 | -0.4224570 | C                                                                                             | 4.8668580  | -0.2095300 | -0.5743700 |
| C                                                                                              | 5.3286970  | -1.5425140 | -0.6086300 | C                                                                                             | 5.4520600  | -1.4790290 | -0.6852840 |
| C                                                                                              | 4.4639590  | -2.6801260 | -0.6215620 | C                                                                                             | 4.5842100  | -2.5927540 | -0.5778120 |
| C                                                                                              | 3.0763280  | -2.6307250 | -0.4533020 | C                                                                                             | 3.2029370  | -2.5014550 | -0.3226200 |
| C                                                                                              | 2.7961080  | 1.1567250  | -0.1592090 | C                                                                                             | 2.8953630  | 1.3000240  | -0.3363650 |
| N                                                                                              | 1.5491530  | 1.5189710  | 0.1139160  | N                                                                                             | 1.8655430  | 1.5769490  | -1.0636230 |
| Mo                                                                                             | 0.0712770  | 0.2012250  | 0.9246330  | Mo                                                                                            | 0.0583470  | -0.1509190 | 1.4607870  |
| O                                                                                              | 0.6364320  | -0.1218880 | 2.5305040  | O                                                                                             | -1.6939290 | 0.6938220  | 2.2297160  |
| C                                                                                              | 6.8480900  | -1.7370720 | -0.8048630 | O                                                                                             | 0.9791560  | 1.2836980  | 1.8452860  |
| C                                                                                              | 7.4055430  | -2.6447580 | 0.3315700  | C                                                                                             | 6.9631660  | -1.6978450 | -0.9280160 |
| C                                                                                              | 2.2113010  | -3.9133150 | -0.4812040 | C                                                                                             | 7.5653560  | -2.5377100 | 0.2373640  |
| C                                                                                              | 1.5148740  | -4.1018810 | 0.8998990  | C                                                                                             | 2.3284830  | -3.7732220 | -0.2115300 |
| O                                                                                              | 1.1465670  | -1.2486380 | -0.0481230 | C                                                                                             | 1.8128310  | -3.9326080 | 1.2492110  |
| N                                                                                              | -0.9940520 | 1.7103170  | -0.3314260 | O                                                                                             | 1.3330640  | -1.0308560 | 0.1948980  |
| C                                                                                              | -2.2556570 | 1.6249300  | -0.7663560 | N                                                                                             | -0.8134340 | 1.4208580  | -0.4259750 |
| C                                                                                              | -3.1518890 | 0.5245460  | -0.6529000 | C                                                                                             | -1.9406370 | 1.2121640  | -1.0592520 |
| C                                                                                              | -2.8333860 | -0.6839260 | 0.0650510  | C                                                                                             | -2.8320350 | 0.0680010  | -0.9723450 |
| C                                                                                              | -3.8461700 | -1.7066080 | 0.2141440  | C                                                                                             | -2.6021710 | -1.0556770 | -0.1228240 |
| C                                                                                              | -5.0826690 | -1.4781890 | -0.3885070 | C                                                                                             | -3.6052870 | -2.0853300 | -0.0278580 |
| C                                                                                              | -5.4221790 | -0.3021470 | -1.1375210 | C                                                                                             | -4.7440110 | -1.9571940 | -0.8351470 |
| C                                                                                              | -4.4498830 | 0.6761570  | -1.2478540 | C                                                                                             | -4.9798920 | -0.8710650 | -1.7228440 |
| O                                                                                              | -1.6179940 | -0.8650940 | 0.5784510  | C                                                                                             | -4.0120570 | 0.1317740  | -1.7616820 |
| C                                                                                              | -3.5508290 | -3.0089120 | 0.9961690  | O                                                                                             | -1.4601330 | -1.1597560 | 0.5705870  |
| C                                                                                              | -2.4030970 | -3.7922840 | 0.2928400  | C                                                                                             | -3.4444520 | -3.2678190 | 0.9577490  |
| C                                                                                              | -6.8275630 | -0.1851030 | -1.7668620 | C                                                                                             | -2.1713630 | -4.0911900 | 0.6092730  |
| C                                                                                              | -7.0487690 | -1.3572370 | -2.7685610 | C                                                                                             | -6.2731430 | -0.8382780 | -2.5680780 |
| C                                                                                              | -0.2937310 | 2.9402160  | -0.4055380 | C                                                                                             | -6.3304140 | -2.0944410 | -3.4871350 |
| C                                                                                              | -0.8618950 | 4.2109270  | -0.6652830 | C                                                                                             | -0.2144750 | 2.7076600  | -0.6060920 |
| C                                                                                              | -0.0568770 | 5.3548240  | -0.6774290 | C                                                                                             | -0.9635190 | 3.8935600  | -0.4339240 |
| C                                                                                              | 1.3307030  | 5.2576380  | -0.4270830 | C                                                                                             | -0.3698560 | 5.1475180  | -0.6312050 |
| C                                                                                              | 1.9093040  | 3.9951300  | -0.1652980 | C                                                                                             | 0.9931100  | 5.2357020  | -0.9908160 |
| C                                                                                              | 1.1028510  | 2.8530730  | -0.1502670 | C                                                                                             | 1.7624370  | 4.0555960  | -1.1204100 |
| C                                                                                              | 2.1375890  | 6.4935210  | -0.4334100 | C                                                                                             | 1.1727690  | 2.7977970  | -0.9199610 |
| O                                                                                              | 3.4889260  | 6.2642930  | -0.1669120 | C                                                                                             | 1.5808800  | 6.5687660  | -1.2055910 |
| O                                                                                              | -1.3369080 | 1.7048210  | 2.3126460  | O                                                                                             | 2.9389160  | 6.5314120  | -1.5504670 |
| C                                                                                              | -1.9485890 | 1.1399330  | 3.5751760  | O                                                                                             | 0.3669460  | -1.1988310 | 2.8174020  |
| C                                                                                              | -0.8552270 | 2.1447970  | 3.6768040  | C                                                                                             | -3.3327980 | -2.7201360 | 2.4126710  |
| C                                                                                              | -3.1355450 | -2.6732280 | 2.4598620  | C                                                                                             | -4.6553290 | -4.2382940 | 0.9137440  |
| C                                                                                              | -4.7871180 | -3.9435980 | 1.0660830  | C                                                                                             | -7.5151440 | -0.8366260 | -1.6279710 |
| C                                                                                              | -7.9091310 | -0.2557620 | -0.6482030 | C                                                                                             | -6.3542630 | 0.4191530  | -3.4698430 |
| C                                                                                              | -7.0171450 | 1.1463250  | -2.5355830 | C                                                                                             | 1.1315050  | -3.6735620 | -1.2029190 |
| C                                                                                              | 1.1426950  | -3.8064770 | -1.6099330 | C                                                                                             | 3.1140700  | -5.0645130 | -0.5692030 |
| C                                                                                              | 3.0557360  | -5.1854370 | -0.7572880 | C                                                                                             | 7.1775350  | -2.4606850 | -2.2689550 |
| C                                                                                              | 7.1158170  | -2.4151900 | -2.1811860 | C                                                                                             | 7.7450780  | -0.3622190 | -1.0074770 |
| C                                                                                              | 7.6199440  | -0.3944340 | -0.7714080 | O                                                                                             | 0.9964890  | 7.6693120  | -1.1105720 |
| O                                                                                              | 1.7145910  | 7.6456520  | -0.6503200 | H                                                                                             | 3.3209460  | 2.0278310  | 0.3720240  |
| H                                                                                              | 3.5043760  | 1.9423880  | -0.4323650 | H                                                                                             | -4.1433890 | 1.0037120  | -2.3969820 |
| H                                                                                              | -4.6465030 | 1.5950970  | -1.7933790 | H                                                                                             | 5.4672480  | 0.6903860  | -0.6760620 |
| H                                                                                              | 5.3256950  | 0.6052120  | -0.4240600 | H                                                                                             | 2.8097930  | 4.1234890  | -1.3952270 |
| H                                                                                              | 2.9716240  | 3.9463170  | 0.0446550  | H                                                                                             | -1.9863120 | 3.8165210  | -0.0790530 |
| H                                                                                              | -1.9283570 | 4.3198000  | -0.8297950 | H                                                                                             | -5.5004300 | -2.7320630 | -0.7738760 |
| H                                                                                              | -5.8448650 | -2.2423260 | -0.2881530 | H                                                                                             | -2.2966550 | 1.9906730  | -1.7476110 |
| H                                                                                              | -2.6751520 | 2.4931940  | -1.2789100 | H                                                                                             | -0.9418750 | 6.0612850  | -0.5002740 |
| H                                                                                              | -0.4855370 | 6.3348050  | -0.8650960 | H                                                                                             | 5.0134610  | -3.5825600 | -0.6890860 |
| H                                                                                              | 4.9195180  | -3.6517140 | -0.7746390 | H                                                                                             | 0.4931560  | -4.5635140 | -1.1105620 |
| H                                                                                              | 0.5179790  | -4.7104870 | -1.6173200 | H                                                                                             | 0.5267370  | -2.7873580 | -1.0005000 |
| H                                                                                              | 0.4979560  | -2.9386050 | -1.4564350 | H                                                                                             | 1.4965690  | -3.6174600 | -2.2386440 |
| H                                                                                              | 1.6254130  | -3.7184880 | -2.5933800 | H                                                                                             | 1.1672340  | -4.8200620 | 1.3245120  |
| H                                                                                              | 0.8841210  | -5.0014360 | 0.8782530  | H                                                                                             | 2.6575860  | -4.0637630 | 1.9405550  |
| H                                                                                              | 2.2620720  | -4.2272390 | 1.6955620  | H                                                                                             | 1.2416800  | -3.0642790 | 1.5811440  |
| H                                                                                              | 0.8872750  | -3.2433000 | 1.1465830  | H                                                                                             | 2.4292120  | -5.9210700 | -0.5113230 |
| H                                                                                              | 2.3882920  | -6.0562060 | -0.7755200 | H                                                                                             | 3.5210050  | -5.0295850 | -1.5893800 |
| H                                                                                              | 3.5662730  | -5.1406770 | -1.7290380 | H                                                                                             | 3.9393130  | -5.2556430 | 0.1305140  |
| H                                                                                              | 3.8058310  | -5.3634530 | 0.0252220  | H                                                                                             | 8.6386690  | -2.7133370 | 0.0722330  |
| H                                                                                              | 8.4839610  | -2.8048480 | 0.1964770  | H                                                                                             | 7.4425590  | -2.0141570 | 1.1943360  |
| H                                                                                              | 7.2465350  | -2.1818250 | 1.3140210  | H                                                                                             | 7.0722320  | -3.5131590 | 0.3264970  |
| H                                                                                              | 6.9202910  | -3.6283210 | 0.3422250  | H                                                                                             | 8.2490830  | -2.6335430 | -2.4478450 |

|   |            |            |            |   |            |            |            |
|---|------------|------------|------------|---|------------|------------|------------|
| H | 8.1935120  | -2.5732620 | -2.3239490 | H | 6.6741230  | -3.4350130 | -2.2618980 |
| H | 6.6224010  | -3.3917070 | -2.2578890 | H | 6.7741850  | -1.8828520 | -3.1107580 |
| H | 6.7482970  | -1.7878700 | -3.0036440 | H | 8.8122430  | -0.5660800 | -1.1713190 |
| H | 8.6921390  | -0.5824320 | -0.9110400 | H | 7.3922680  | 0.2640790  | -1.8372860 |
| H | 7.2951050  | 0.2819290  | -1.5732000 | H | 7.6522630  | 0.2154140  | -0.0785640 |
| H | 7.4960540  | 0.1219880  | 0.1895810  | H | -3.2050530 | -3.5572180 | 3.1146980  |
| H | -2.9498710 | -3.6024970 | 3.0157410  | H | -2.4826550 | -2.0447770 | 2.5242850  |
| H | -2.2221230 | -2.0741570 | 2.4798090  | H | -4.2438010 | -2.1720430 | 2.6906420  |
| H | -3.9359930 | -2.1233600 | 2.9751850  | H | -2.0668470 | -4.9301520 | 1.3127780  |
| H | -2.1818020 | -4.7116240 | 0.8529950  | H | -2.2385840 | -4.5011650 | -0.4088220 |
| H | -2.6973290 | -4.0767150 | -0.7268710 | H | -1.2781380 | -3.4697060 | 0.6750150  |
| H | -1.4926010 | -3.1928190 | 0.2376940  | H | -4.4860700 | -5.0467280 | 1.6372270  |
| H | -4.5213430 | -4.8424280 | 1.6366480  | H | -5.5944810 | -3.7388400 | 1.1889920  |
| H | -5.6373660 | -3.4680700 | 1.5738660  | H | -4.7806660 | -4.6996380 | -0.0760820 |
| H | -5.1136550 | -4.2720470 | 0.0703270  | H | -7.2834630 | 0.3958130  | -4.0553880 |
| H | -8.0259930 | 1.1829660  | -2.9662150 | H | -6.3558640 | 1.3427080  | -2.8764820 |
| H | -6.9066940 | 2.0166790  | -1.8751910 | H | -5.5143930 | 0.4655990  | -4.1754160 |
| H | -6.3000110 | 1.2444600  | -3.3611510 | H | -8.4437170 | -0.8237700 | -2.2171970 |
| H | -8.9145770 | -0.1915440 | -1.0861930 | H | -7.5358130 | -1.7248340 | -0.9848390 |
| H | -7.8508300 | -1.1937840 | -0.0828150 | H | -7.5053050 | 0.0466340  | -0.9764800 |
| H | -7.7899860 | 0.5726850  | 0.0621940  | H | -7.2553780 | -2.0944110 | -4.0821200 |
| H | -8.0518890 | -1.2950210 | -3.2121950 | H | -5.4756050 | -2.1092550 | -4.1758230 |
| H | -6.3106550 | -1.3207730 | -3.5801520 | H | -6.3043400 | -3.0225920 | -2.9033390 |
| H | -6.9601230 | -2.3345280 | -2.2786930 | H | 3.2623670  | 7.4517980  | -1.6736900 |
| H | 3.9822500  | 7.1148780  | -0.1830000 | O | -1.4682700 | 1.6729400  | 3.3281640  |
| H | -1.0869630 | 3.1840440  | 3.8917410  | C | -2.0604320 | 2.8995900  | 3.1076190  |
| H | -2.9833390 | 1.4389610  | 3.7160210  | O | -2.7266440 | 3.2283930  | 2.1137070  |
| H | -1.7059540 | 0.0951280  | 3.7396700  | C | -1.7473640 | 3.8068200  | 4.2882210  |
| H | 0.1441470  | 1.7886210  | 3.9063900  | H | -1.3229990 | 3.2405690  | 5.1202780  |
|   |            |            |            | H | -1.0229330 | 4.5638610  | 3.9640660  |
|   |            |            |            | H | -2.6634160 | 4.3191660  | 4.6014730  |

| Table S9: structural data for the different TS involving Mo <sup>1S</sup> and peracetic acid                                                             |            |            |            |                                                                                                                                                           |            |            |            |
|----------------------------------------------------------------------------------------------------------------------------------------------------------|------------|------------|------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------|------------|------------|------------|
| TS for Mo <sup>1</sup> with peracetic acid (direct interaction)                                                                                          |            |            |            | TS for Mo <sup>1</sup> -catalyzed epoxidation of ethylene with peracetic acid involving hydrogen bonds                                                    |            |            |            |
| Nb atoms: 101<br>E= -2370.0765296 u.a.; G=-2369.349493 u.a.<br>Esp=-2372.23183347 u.a.; qh-G=-2369.317917 u.a.<br>Freq min: -378.0227 cm <sup>-1</sup> . |            |            |            | Nb atoms: 107<br>E= -2448.6807303 u.a.; G= -2447.909718 u.a.<br>Esp=-2450.88868103 u.a.; qh-G=-2447.874158 u.a.<br>Freq min: -367.6059 cm <sup>-1</sup> . |            |            |            |
| C                                                                                                                                                        | 1.3333760  | 4.3486910  | -0.0207960 | C                                                                                                                                                         | -1.4913450 | -2.0284780 | -1.0640220 |
| C                                                                                                                                                        | 0.8367190  | 3.0343110  | 0.0773250  | C                                                                                                                                                         | -0.1623940 | -2.3271340 | -1.4704260 |
| C                                                                                                                                                        | -0.5221100 | 2.8068140  | 0.4145580  | C                                                                                                                                                         | 0.0847310  | -3.4790450 | -2.2468990 |
| C                                                                                                                                                        | -1.3738610 | 3.9011150  | 0.6464240  | C                                                                                                                                                         | -0.9811420 | -4.3072480 | -2.6193650 |
| C                                                                                                                                                        | -0.8638510 | 5.2108860  | 0.5790010  | C                                                                                                                                                         | -2.2976680 | -4.0069100 | -2.1992990 |
| C                                                                                                                                                        | 0.4900960  | 5.4333900  | 0.2395700  | C                                                                                                                                                         | -2.5566800 | -2.8701800 | -1.4038710 |
| N                                                                                                                                                        | 1.6179790  | 1.8555810  | -0.1333890 | N                                                                                                                                                         | 0.8326280  | -1.4476940 | -0.9591550 |
| C                                                                                                                                                        | 2.9097110  | 1.8525500  | 0.1468760  | C                                                                                                                                                         | 2.0354820  | -1.3452310 | -1.4676150 |
| C                                                                                                                                                        | 3.7539930  | 0.7002720  | 0.1889430  | C                                                                                                                                                         | 3.1509930  | -0.6467320 | -0.8617110 |
| C                                                                                                                                                        | 3.1678390  | -0.6034170 | 0.2110430  | C                                                                                                                                                         | 3.0990780  | -0.0692900 | 0.4454830  |
| C                                                                                                                                                        | 4.0046350  | -1.7482100 | 0.4427640  | C                                                                                                                                                         | 4.2907340  | 0.4245110  | 1.0634590  |
| C                                                                                                                                                        | 5.3778190  | -1.5076830 | 0.5817700  | C                                                                                                                                                         | 5.4610730  | 0.4043500  | 0.2862550  |
| C                                                                                                                                                        | 5.9950170  | -0.2197030 | 0.5391890  | C                                                                                                                                                         | 5.5416840  | -0.0989730 | -1.0418540 |
| C                                                                                                                                                        | 5.1578580  | 0.8748370  | 0.3595050  | C                                                                                                                                                         | 4.3767350  | -0.6390240 | -1.5843100 |
| O                                                                                                                                                        | 1.8313790  | -0.7240710 | 0.0750330  | O                                                                                                                                                         | 1.9202230  | -0.0002460 | 1.1201790  |
| Mo                                                                                                                                                       | 0.4190390  | 0.1705460  | -1.0805110 | Mo                                                                                                                                                        | 0.0150460  | -0.3418040 | 0.9469480  |
| O                                                                                                                                                        | -1.3926120 | 0.7350820  | -2.4505400 | O                                                                                                                                                         | -0.7659300 | 0.5885830  | 2.1864030  |
| O                                                                                                                                                        | -3.1286710 | -0.0930500 | -2.6817550 | C                                                                                                                                                         | 4.2991830  | 0.9269730  | 2.5286920  |
| C                                                                                                                                                        | -3.9490010 | 0.8480760  | -2.2377180 | C                                                                                                                                                         | 5.7126160  | 1.3845100  | 2.9772370  |
| C                                                                                                                                                        | -5.4333930 | 0.5549860  | -2.3296100 | C                                                                                                                                                         | 6.8863890  | -0.0474320 | -1.8003790 |
| C                                                                                                                                                        | 3.4177280  | -3.1751760 | 0.5056330  | C                                                                                                                                                         | 6.7794280  | -0.6468550 | -3.2248180 |
| C                                                                                                                                                        | 2.3478670  | -3.2509530 | 1.6355240  | C                                                                                                                                                         | -3.3824550 | -4.9359840 | -2.6034070 |
| C                                                                                                                                                        | 7.5263660  | -0.1033950 | 0.6992920  | O                                                                                                                                                         | -3.2262610 | -5.9948110 | -3.2405460 |
| C                                                                                                                                                        | 7.9545730  | -0.6962430 | 2.0744260  | N                                                                                                                                                         | -1.6112410 | -0.7839790 | -0.3676120 |
| N                                                                                                                                                        | -0.9348400 | 1.4448890  | 0.3933090  | C                                                                                                                                                         | -2.6177570 | 0.0448020  | -0.6088380 |
| C                                                                                                                                                        | -2.0280120 | 1.0559060  | 1.0118220  | C                                                                                                                                                         | -2.5579250 | 1.4642870  | -0.4467370 |
| C                                                                                                                                                        | -2.6343690 | -0.2461000 | 0.9173860  | C                                                                                                                                                         | -1.2700000 | 2.1142470  | -0.4304880 |

|   |            |            |            |   |            |            |            |
|---|------------|------------|------------|---|------------|------------|------------|
| C | -3.8868760 | -0.4321330 | 1.5671610  | C | -1.2146490 | 3.5383410  | -0.6478830 |
| C | -4.5826870 | -1.6335240 | 1.4633280  | C | -2.4325420 | 4.2200420  | -0.7372980 |
| C | -3.9910560 | -2.6583750 | 0.6688350  | C | -3.7243450 | 3.6069500  | -0.6872260 |
| C | -2.7563760 | -2.5510980 | 0.0112840  | C | -3.7589830 | 2.2242940  | -0.5755990 |
| C | -2.0529010 | -1.3127090 | 0.1688980  | O | -0.1546820 | 1.3847520  | -0.2702150 |
| O | -0.8066840 | -1.1670130 | -0.3443100 | C | -4.9893090 | 4.4875940  | -0.7798230 |
| C | -2.1829320 | -3.7174620 | -0.8272800 | C | -6.2901280 | 3.6487600  | -0.7205020 |
| C | -1.8616800 | -3.2422700 | -2.2756140 | C | 0.1403210  | 4.2783060  | -0.7410600 |
| C | -5.9426310 | -1.8885330 | 2.1500980  | C | -0.0350280 | 5.7916690  | -1.0321780 |
| C | -7.0230910 | -2.1950420 | 1.0710320  | O | -0.1788490 | -1.9845020 | 1.5881550  |
| C | -1.7281150 | 6.3923890  | 0.8203080  | C | -5.0058510 | 5.4928350  | 0.4099610  |
| O | -1.3675550 | 7.5806300  | 0.7436640  | C | -4.9830720 | 5.2788870  | -2.1212780 |
| O | 0.1444230  | 1.6956750  | -2.1157230 | C | 0.9064020  | 4.1387270  | 0.6058610  |
| O | 1.0068440  | -0.7800770 | -2.4033960 | C | 0.9898420  | 3.6705660  | -1.8969850 |
| C | -0.8883270 | -4.2471300 | -0.1448460 | C | 3.3372890  | 2.1374500  | 2.7047060  |
| C | -3.1813270 | -4.9006400 | -0.9341690 | C | 3.8594800  | -0.2343990 | 3.4715050  |
| C | -5.8166420 | -3.1076320 | 3.1113430  | C | 7.3478850  | 1.4344720  | -1.9309600 |
| C | -6.4191750 | -0.6690120 | 2.9780070  | C | 7.9619790  | -0.8539810 | -1.0140160 |
| C | 2.7850870  | -3.5300490 | -0.8733060 | O | -4.6387990 | -4.5056060 | -2.2052180 |
| C | 4.5000300  | -4.2424820 | 0.8152460  | O | -1.9152970 | -3.6223700 | 2.8072940  |
| C | 8.2270180  | -0.8960710 | -0.4439720 | C | -1.5567790 | -4.9912670 | 1.7286970  |
| C | 8.0110580  | 1.3664670  | 0.6352000  | C | -0.3965770 | -5.1077190 | 2.5130210  |
| O | -3.0299210 | 6.0491420  | 1.1654610  | O | -3.0545010 | -2.1915020 | 3.4004500  |
| O | -3.4982950 | 1.9411160  | -1.7359830 | C | -3.8616280 | -1.7252150 | 2.4529790  |
| H | -2.5666740 | 1.7716840  | 1.6394160  | O | -3.8540360 | -2.0481330 | 1.2283020  |
| H | 5.5539280  | 1.8859420  | 0.3401850  | C | -4.8280910 | -0.6589910 | 2.9872800  |
| H | -4.2927000 | 0.3997760  | 2.1349250  | H | -3.5107980 | -0.3696980 | -1.0763010 |
| H | -2.4325720 | 3.7628940  | 0.8303030  | H | 4.3803510  | -1.0758070 | -2.5786600 |
| H | 2.3568460  | 4.5298830  | -0.3339630 | H | -4.7010080 | 1.6842180  | -0.5771890 |
| H | 6.0230530  | -2.3648540 | 0.7345350  | H | -3.5510180 | -2.6799010 | -1.0178980 |
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| H | 0.8493710  | 6.4546700  | 0.1598180  | H | 6.3676710  | 0.7961560  | 0.7314470  |
| H | -4.5367040 | -3.5883330 | 0.5634970  | H | 2.2642110  | -1.8474550 | -2.4141520 |
| H | -0.4615710 | -5.0623210 | -0.7449860 | H | -0.8140710 | -5.2006350 | -3.2133740 |
| H | -0.1401530 | -3.4576970 | -0.0589300 | H | -2.3976700 | 5.2967820  | -0.8587380 |
| H | -1.1049870 | -4.6377890 | 0.8595010  | H | 1.9606790  | 4.1822560  | -1.9539190 |
| H | -1.5392680 | -4.1046620 | -2.8751040 | H | 1.1665740  | 2.6045680  | -1.7329110 |
| H | -2.7417880 | -2.7945610 | -2.7513920 | H | 0.4810130  | 3.7983450  | -2.8624710 |
| H | -1.0631120 | -2.4987670 | -2.2965190 | H | 1.8712770  | 4.6613340  | 0.5428160  |
| H | -2.7273480 | -5.6891840 | -1.5465970 | H | 0.3305260  | 4.5812430  | 1.4296190  |
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| H | -7.3826880 | -0.8992110 | 3.4497930  | H | -4.1006530 | 5.9241150  | -2.2118450 |
| H | -5.7102500 | -0.4179540 | 3.7779920  | H | -4.9885000 | 4.5938080  | -2.9790420 |
| H | -6.5629670 | 0.2201780  | 2.3501380  | H | -7.1606450 | 4.3143660  | -0.7794760 |
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| H | 1.9096310  | -4.2573570 | 1.6647440  | H | 2.8511010  | -0.5837760 | 3.2347830  |
| H | 2.8055450  | -3.0505280 | 2.6143930  | H | 4.5515740  | -1.0840660 | 3.3944890  |
| H | 1.5450730  | -2.5286660 | 1.4713940  | H | 3.3887010  | 2.4976430  | 3.7409540  |
| H | 4.0201620  | -5.2277340 | 0.8646220  | H | 3.6221720  | 2.9651630  | 2.0419120  |
| H | 5.2692070  | -4.2932070 | 0.0329440  | H | 2.3026510  | 1.8630940  | 2.4911910  |
| H | 4.9919880  | -4.0660130 | 1.7815390  | H | 5.6623370  | 1.7119130  | 4.0227930  |
| H | 9.1017940  | 1.3989400  | 0.7492620  | H | 6.4497460  | 0.5721970  | 2.9227860  |
| H | 7.7651050  | 1.8349940  | -0.3268150 | H | 6.0802590  | 2.2334310  | 2.3850700  |
| H | 7.5766960  | 1.9747600  | 1.4397650  | H | 7.7533430  | -0.5792070 | -3.7254270 |
| H | 9.3179950  | -0.8330450 | -0.3349480 | H | 6.4937980  | -1.7069550 | -3.2015180 |
| H | 7.9514500  | -1.9575870 | -0.4335950 | H | 6.0517630  | -0.1038550 | -3.8422230 |
| H | 7.9557370  | -0.4884550 | -1.4262180 | H | 8.9252280  | -0.8124140 | -1.5397270 |
| H | 9.0446380  | -0.6316950 | 2.1910230  | H | 8.1161640  | -0.4540720 | -0.0045420 |
| H | 7.4877200  | -0.1451840 | 2.9010940  | H | 7.6690460  | -1.9074590 | -0.9175040 |
| H | 7.6714300  | -1.7513440 | 2.1713610  | H | 8.3113140  | 1.4862880  | -2.4555090 |
| H | -3.5753700 | 6.8573660  | 1.2942050  | H | 6.6159740  | 2.0239540  | -2.4979990 |
| H | -1.9684680 | 1.5147210  | -2.0779380 | H | 7.4752910  | 1.9099990  | -0.9509240 |

|                                                                                                                                                      |            |            |            |   |            |            |            |
|------------------------------------------------------------------------------------------------------------------------------------------------------|------------|------------|------------|---|------------|------------|------------|
| H                                                                                                                                                    | -5.9971300 | 1.4894060  | -2.2696070 | H | -5.3288100 | -5.1535820 | -2.4725270 |
| H                                                                                                                                                    | -5.6601890 | 0.0305510  | -3.2624910 | H | -1.2887890 | -2.9046230 | 2.4297410  |
| H                                                                                                                                                    | -5.7245700 | -0.0950430 | -1.4947660 | H | -4.3141760 | 0.3107630  | 2.9830090  |
|                                                                                                                                                      |            |            |            | H | -5.7065680 | -0.6041050 | 2.3367650  |
|                                                                                                                                                      |            |            |            | H | -5.1251520 | -0.8831500 | 4.0154230  |
|                                                                                                                                                      |            |            |            | H | -0.4083380 | -5.6724840 | 3.4396880  |
|                                                                                                                                                      |            |            |            | H | -2.4137230 | -5.6279080 | 1.9252170  |
|                                                                                                                                                      |            |            |            | H | -1.5145490 | -4.5533180 | 0.7364980  |
|                                                                                                                                                      |            |            |            | H | 0.5102690  | -4.5706960 | 2.2514190  |
| TS for Mo1-catalyzed epoxidation of ethylene<br>with peracetic acid with no interaction                                                              |            |            |            |   |            |            |            |
| Nb atoms: 107<br>E= -2448.6878364 u.a.; G=-2447.916910 u.a.; Esp=-2450.89560985 u.a.; qh-G=-2447.881363 u.a.; Freq min: -309.1741 cm <sup>-1</sup> . |            |            |            |   |            |            |            |
| C                                                                                                                                                    | 0.0456500  | -3.8978890 | -1.5209350 | H | -3.2387580 | -0.3119250 | -0.7573990 |
| C                                                                                                                                                    | -0.0821420 | -2.6229460 | -0.9325520 | H | 4.5042730  | -1.9385900 | -2.3091550 |
| C                                                                                                                                                    | -1.3701300 | -2.1478340 | -0.5625150 | H | -4.2205630 | 1.8608950  | -0.5258560 |
| C                                                                                                                                                    | -2.5123200 | -2.9275600 | -0.7726810 | H | -3.4895810 | -2.5703500 | -0.4667250 |
| C                                                                                                                                                    | -2.3711750 | -4.1996170 | -1.3690400 | H | 1.0253950  | -4.2967170 | -1.7665650 |
| C                                                                                                                                                    | -1.0949000 | -4.6791280 | -1.7428420 | H | 6.7613670  | 0.1872920  | 0.6558460  |
| N                                                                                                                                                    | 0.9971800  | -1.7664530 | -0.5791960 | H | 2.3442670  | -2.4972910 | -1.9837250 |
| C                                                                                                                                                    | 2.1886470  | -1.8445460 | -1.1175430 | H | -1.0167830 | -5.6691400 | -2.1816160 |
| C                                                                                                                                                    | 3.3779280  | -1.1678990 | -0.6416480 | H | -1.6400610 | 5.2033870  | -1.3201600 |
| C                                                                                                                                                    | 3.4207570  | -0.4125740 | 0.5727940  | H | 2.5819390  | 3.5942940  | -2.3267990 |
| C                                                                                                                                                    | 4.6738150  | 0.0515400  | 1.0856830  | H | 1.6661680  | 2.1319330  | -1.8883410 |
| C                                                                                                                                                    | 5.8090130  | -0.1787810 | 0.2913230  | H | 1.0515050  | 3.2239720  | -3.1536890 |
| C                                                                                                                                                    | 5.7988630  | -0.8650820 | -0.9549040 | H | 2.6004380  | 4.3947580  | 0.0859130  |
| C                                                                                                                                                    | 4.5738200  | -1.3692110 | -1.3868680 | H | 1.0829410  | 4.5657670  | 0.9998390  |
| O                                                                                                                                                    | 2.2792590  | -0.1444080 | 1.2587680  | H | 1.7013380  | 2.9495710  | 0.6014420  |
| Mo                                                                                                                                                   | 0.3437040  | -0.3653400 | 1.1778670  | H | 1.7729080  | 5.8362670  | -1.7492750 |
| O                                                                                                                                                    | 0.0795540  | -1.8856490 | 2.0130080  | H | 0.2115950  | 5.5737800  | -2.5441980 |
| C                                                                                                                                                    | 4.7792750  | 0.7450130  | 2.4668110  | H | 0.2777840  | 6.1331660  | -0.8517500 |
| C                                                                                                                                                    | 6.2422680  | 1.1291940  | 2.8139740  | H | -5.0251080 | 6.4851010  | -0.1774930 |
| C                                                                                                                                                    | 7.1149780  | -1.0381310 | -1.7449550 | H | -4.2353430 | 5.3806720  | 0.9735870  |
| C                                                                                                                                                    | 6.9060670  | -1.8142710 | -3.0692640 | H | -3.2572600 | 6.3469610  | -0.1457560 |
| N                                                                                                                                                    | -1.3607030 | -0.8157600 | -0.0339540 | H | -5.0846520 | 5.9418000  | -2.6659720 |
| C                                                                                                                                                    | -2.2919790 | 0.0698630  | -0.3774050 | H | -3.3190170 | 5.7941180  | -2.7144620 |
| C                                                                                                                                                    | -2.1012950 | 1.4840430  | -0.4052940 | H | -4.3303350 | 4.4553970  | -3.2896730 |
| C                                                                                                                                                    | -3.2407390 | 2.3200520  | -0.6138900 | H | -6.4640780 | 4.6442270  | -1.0432710 |
| C                                                                                                                                                    | -3.0960080 | 3.6675460  | -0.9069150 | H | -5.7922520 | 3.1197010  | -1.6471530 |
| C                                                                                                                                                    | -1.7589500 | 4.1582670  | -1.0568120 | H | -5.7476230 | 3.4880990  | 0.0935350  |
| C                                                                                                                                                    | -0.5993430 | 3.3947810  | -0.8972350 | H | 4.3452160  | 0.2507750  | 4.5543790  |
| C                                                                                                                                                    | -0.7621350 | 2.0179270  | -0.4922610 | H | 3.2346240  | -0.5226540 | 3.4069610  |
| O                                                                                                                                                    | 0.2903210  | 1.2277550  | -0.2536460 | H | 4.8903750  | -1.1440180 | 3.5962750  |
| C                                                                                                                                                    | 0.8081430  | 4.0001040  | -1.1081060 | H | 4.0498190  | 2.5398410  | 3.4736470  |
| C                                                                                                                                                    | 0.7494810  | 5.4727320  | -1.5917700 | H | 4.2661250  | 2.7526490  | 1.7219580  |
| C                                                                                                                                                    | -4.2860130 | 4.6329720  | -1.0958760 | H | 2.8722560  | 1.8436930  | 2.3437380  |
| C                                                                                                                                                    | -5.6491270 | 3.9211150  | -0.9102740 | H | 6.2584470  | 1.5982030  | 3.8054060  |
| C                                                                                                                                                    | -3.5359060 | -5.0862600 | -1.6002490 | H | 6.9037520  | 0.2534160  | 2.8541110  |
| O                                                                                                                                                    | -3.4907500 | -6.2319640 | -2.0850470 | H | 6.6602880  | 1.8532360  | 2.1015460  |
| O                                                                                                                                                    | -0.3755350 | 0.7628130  | 2.2998410  | H | 7.8632790  | -1.9020610 | -3.5983350 |
| C                                                                                                                                                    | 1.5968360  | 3.9710470  | 0.2326400  | H | 6.5328650  | -2.8316590 | -2.8916780 |
| C                                                                                                                                                    | 1.5739970  | 3.1788430  | -2.1880330 | H | 6.2043780  | -1.2981370 | -3.7376720 |
| C                                                                                                                                                    | -4.1918100 | 5.7810800  | -0.0473950 | H | 9.0893660  | -1.9402690 | -1.4291940 |
| C                                                                                                                                                    | -4.2486790 | 5.2424120  | -2.5286920 | H | 8.3691790  | -1.3043760 | 0.0599710  |
| C                                                                                                                                                    | 3.9328740  | 2.0502700  | 2.4973100  | H | 7.7667750  | -2.8226080 | -0.6308270 |
| C                                                                                                                                                    | 4.2759490  | -0.2337860 | 3.5711520  | H | 8.6421280  | 0.2580850  | -2.6401090 |
| C                                                                                                                                                    | 7.6966070  | 0.3645610  | -2.0915800 | H | 6.9979750  | 0.9337490  | -2.7182310 |
| C                                                                                                                                                    | 8.1454670  | -1.8229410 | -0.8801530 | H | 7.8979000  | 0.9565270  | -1.1905090 |
| O                                                                                                                                                    | -4.7500490 | -4.5191390 | -1.2172680 | H | -5.4768530 | -5.1678600 | -1.3565370 |
| O                                                                                                                                                    | -4.1711450 | -1.0495030 | 1.8500160  | H | -4.9894790 | -1.4164460 | 2.3300520  |
| O                                                                                                                                                    | -5.1152760 | -1.1106060 | 0.3017770  | H | -3.5453160 | -2.4228010 | 4.0940380  |
| C                                                                                                                                                    | -6.3204460 | -1.6424540 | 0.6060780  | H | -2.0328850 | -1.9986780 | 3.0953930  |
| C                                                                                                                                                    | -2.9376250 | -1.6675770 | 3.5978260  | H | -2.5810260 | 0.4079550  | 3.2231120  |
| C                                                                                                                                                    | -3.2505620 | -0.3266140 | 3.6601620  | H | -4.1069380 | 0.0392950  | 4.2232540  |
| O                                                                                                                                                    | -6.6241480 | -1.9507620 | 1.7925770  | C | -7.2341080 | -1.8265370 | -0.5882220 |
|                                                                                                                                                      |            |            |            | H | -8.1614020 | -2.3059630 | -0.2664360 |
|                                                                                                                                                      |            |            |            | H | -7.4605750 | -0.8546630 | -1.0413450 |
|                                                                                                                                                      |            |            |            | H | -6.7297930 | -2.4412400 | -1.3417430 |

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- <sup>6</sup> Soluble monometallic salen complexes derived from O-functionalised diamines as metalloligands for the synthesis of heterobimetallic complexes. M. Schley, S. Fritzsche, P. Lönnecke, E. Hey-Hawkins, *Dalton Trans.* 39 (2010) 4090–4106, doi: 10.1039/b920706f.
- <sup>7</sup> A TS for formation of  $\text{Mo}_2\mathbf{2}_{\eta^1}$  from the separated reactant could be obtained ( $\text{TS}_{0-2}^5$ ), but it is 41.0 kcal mol<sup>-1</sup> above the reactants so that direct formation of the singlet adduct was not considered.