

Supporting Information: Intersystem Crossing Enables
4-Thiothymidine to Act as a Photosensitizer in Photodynamic
Therapy: An Ab Initio QM/MM Study

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Table of Contents

1. System Setup
2. QM/MM Computational Protocol
3. Full QM/MM Conical Intersection Optimizations
4. Convergence Criteria
5. Energy Gaps at Intersection Points
6. Geometric Parameters of S1S0-MIN
7. Single-Point QM Energies
8. Spin-Orbit Couplings
9. Local Hydrogen-Bonding Networks
10. References
11. Force field
12. Cartesian Coordinates of All Optimized Structures

1 System Setup

Initial Cartesian coordinates of 4-thiothymidine (an RNA monomer analogue) were prepared using GaussView 4.0. To explicitly consider solvation effects, 4-thiothymidine was solvated in a spherical water box with a radius of 25 Å. This solvated system was relaxed by performing constrained molecular mechanics (MM) minimizations (1000 steps) and 5-ps molecular dynamics (MD) runs, in which the outermost water molecules were frozen (i.e., those more than 20 Å away from the center of mass of 4-thiothymidine). In the MM minimizations and dynamics runs, 4-thiothymidine was modeled by the CHARMM22 force field [1] (modified based on the parameters of thymidine, see section 11) and water molecules were represented by the TIP3P model. [2] All MM computations were carried out using the CHARMM package (version c31b1). [3] Removal of the frozen water molecules in the final MD snapshot generated the starting structure for the following QM/MM computations.

2 QM/MM Computational Protocol

The QM subsystem contained the 4-thiothymine chromophore of 4-thiothymidine; the MM subsystem consisted of the ribose group and the solvent water molecules (see Fig. 1).

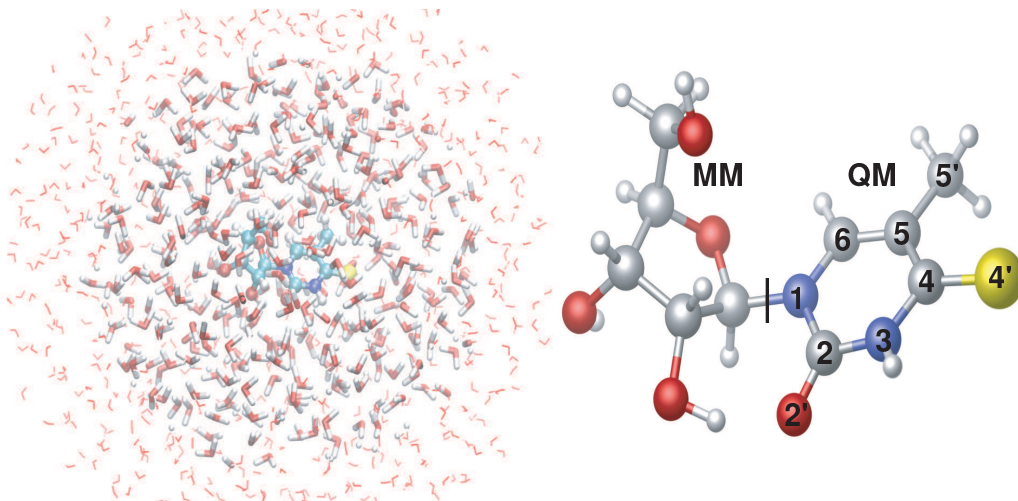


Figure 1: The QM/MM computational protocol used in this work. 4-thiothymidine was solvated in a spherical water box of 20 Å. All atoms up to 10 Å away from any atom of 4-thiothymidine were allowed to move in the geometry optimizations (1133 atoms), and the remaining 2196 atoms were frozen at the positions adopted in the final MD snapshot. The QM subsystem (4-thiothymine chromophore) was described using the ab initio CASPT2//CASSCF method, while the MM subsystem (the remaining part) was treated by the CHARMM force field (ribose) and TIP3P model (water). A hydrogen link atom was used at the QM/MM boundary (black line). Color code: Sulfur in yellow; nitrogen in blue; oxygen in red; carbon in gray; and hydrogen in white. Also shown is the atomic numbering.

In all QM/MM geometry optimizations including those for intersection points (see below), all water molecules within 10 Å from any atom of the 4-thiothymidine monomer were allowed to move (active region, 1133 atoms) while the other atoms were kept fixed (2196 atoms).

All QM/MM computations were done with the locally modified ChemShell version 3.5 [4]. An electronic embedding scheme [5] was adopted in the QM/MM calculations with the MM point charges being incorporated into the one-electron Hamiltonian during the QM calculations. A hydrogen link atom with the charge-shift scheme [6, 7] was employed to treat the QM/MM boundary. No cutoffs were introduced for the nonbonding MM and QM/MM interactions (electrostatic and van der Waals). The built-in DL_POLY module in ChemShell [8] was used to compute the MM energies and gradients. The MOLPRO2010 package [9] was employed to compute energies, gradients, derivative coupling vectors, and spin-orbit couplings of the QM subsystem.

The state-averaged complete-active-space self-consistent field (SA-CASSCF) method was chosen as the QM electronic structure method. At the SA-CASSCF level, full QM/MM geometry optimizations are expensive for higher excited states, and in particular for intersection points (see the following section). To balance computational accuracy and efficiency, we adopted an active space of 10 electrons in 8 orbitals. In all CASSCF computations, equal state weights were used for the S_0 , S_1 , and S_2 states, and for the T_1 and T_2 states, respectively. To account for dynamic correlation, the second-order CASSCF perturbation approach (CASPT2) [10–12] was employed to re-evaluate the energies of all optimized structures. To minimize intruder-state issues in the CASPT2 computations, an energy-level shift of 0.2 a.u. was applied. [13] The 6-31G* basis set [14, 15] was used in all QM computations.

3 Full QM/MM Conical Intersection Optimizations

Gradient difference and derivative coupling vectors are required for optimizing minimum-energy conical intersections in the gradient projection method of Bearpark et al. [16] and the Lagrange-Newton method of Manaa and Yarkony. [17]

However, rigorous computation of these vectors remains expensive at the QM/MM level. The derivative coupling vector between electronic states I and J with same spin can be partitioned into QM and MM terms:

$$\mathbf{h}_{IJ}(\mathbf{r}_{qm}, \mathbf{r}_{mm}) = \langle \Psi_I(\mathbf{r}_{qm}, \mathbf{r}_{mm}) | \frac{\partial}{\partial \mathbf{r}_{qm}} | \Psi_J(\mathbf{r}_{qm}, \mathbf{r}_{mm}) \rangle \quad (1)$$

and

$$\mathbf{h}_{IJ}(\mathbf{r}_{qm}, \mathbf{r}_{mm}) = \langle \Psi_I(\mathbf{r}_{qm}, \mathbf{r}_{mm}) | \frac{\partial}{\partial \mathbf{r}_{mm}} | \Psi_J(\mathbf{r}_{qm}, \mathbf{r}_{mm}) \rangle . \quad (2)$$

The computation of the QM contribution is affordable at the SA-CASSCF level as long as the QM region is small (tens of atoms), whereas the explicit computation of the MM contributions is very expensive (involving usually thousands of MM atoms). Thus, it is impractical to use these rigorous formulations for optimizing

conical intersections at the QM/MM level. To circumvent this bottleneck, several approximate treatments have been proposed, e.g. the sequential QM conical intersection optimization and MM minimization based on the electrostatic potential (ESP) fitting approximation. [18]

In this work, we adopted the penalty function method of Ciminelli et al. [19] implemented in the DL-FIND module of the ChemShell-3.5 package. [20] Its objective function is defined as

$$f(R) = \frac{E_I + E_J}{2} + c_1 c_2^2 \ln[1 + (\frac{E_J - E_I}{c_2})^2] \quad (3)$$

in which E_I and E_J are the QM/MM energies of electronic states I and J. c_1 and c_2 are two parameters, for which we used the recommended values of $5.0 \text{ (kcal/mol)}^{-1}$ and 5.0 kcal/mol . [19] Because this method only needs the gradients of the electronic states involved, the expensive computation of derivative coupling vectors is avoided. Full QM/MM conical intersection optimization based on the penalty function method was found to be a practical approach for the large condensed-phase systems studied presently.

The QM/MM-based penalty function method was also employed to optimize the crossing points between electronic states with different spin.

4 Convergence Criteria

In minimizations, the following convergence thresholds had to be satisfied: (1) energy converged to within 5.0×10^{-5} hartree; (2) maximum force component less than 4.5×10^{-3} hartree/bohr; (3) root-mean-square force less than 3.0×10^{-3} hartree/bohr; (4) maximum step size component less than 1.8×10^{-3} bohr; (5) root-mean-square step size less than 1.2×10^{-3} bohr.

For the optimization of conical intersections and crossing points, we adopted a more rigorous energy threshold and loosened the other thresholds by a factor of 2: (1) energy converged to within 1.0×10^{-5} hartree; (2) maximum force component less than 9.0×10^{-3} hartree/bohr; (3) root-mean-square force less than 6.0×10^{-3} hartree/bohr; (4) maximum step size component less than 3.6×10^{-3} bohr; (5) root-mean-square step size less than 2.4×10^{-3} bohr.

5 Energy Gaps at Intersection Points

Table 1 lists the computed energy gaps at the conical intersections and crossing points optimized with the QM/MM penalty function method (see above). It is evident that the two involved electronic states are essentially degenerate at the QM(CASSCF)/MM level (used for geometry optimization), and they remain close in energy in the single-point QM(CASPT2//CASSCF)/MM calculations.

Table 1: Energy Gaps (kcal/mol) at the Conical Intersections and Crossing Points Computed by the QM(CASSCF)/MM and QM(CASPT2//CASSCF)/MM (in parentheses) Methods.

| | S2S1-MIN | S2T2-MIN | T2T1-MIN | S1T1-MIN | S1S0-MIN | T1S0-MIN |
|------------|------------|------------|------------|------------|------------|------------|
| ΔE | 0.03 (1.8) | 0.08 (1.3) | 0.10 (0.3) | 0.58 (0.3) | 0.19 (2.9) | 0.03 (1.2) |

6 Geometric Parameters of S1S0-MIN

As discussed in the main text, the differences in the electronic and geometric structures of the S_1 state of 4-thiothymine and thymine cause their qualitatively different photophysics. Fig. 2 shows that the S_1/S_0 conical intersection (S1S0-MIN) of 4-thiothymine is structurally different from that of thymine. The $X4'-C4-C5-C5'$ dihedral angle is 1.2° ($X=S$) in 4-thiothymine and ca. 95° ($X=O$) in thymine (see Table 2) indicating that the methyl substituent is coplanar with the ring in the former and essentially perpendicular in the latter. The H6 atom is distorted out of the ring plane more strongly in 4-thiothymine than in thymine, as reflected in the $C4-C5-C6-H6'$ dihedral angles of 95.7° and ca. 130° , respectively. Finally, S1S0-MIN has a very elongated $C4-S4'$ bond in 4-thiothymine (1.98 Å, larger than the typical C-S single bond length of 1.82 Å) while the $C4-O4'$ bond remains short (ca. 1.2 Å, in the range of a typical C=O double bond).

Table 2: Selected Key Geometric Parameters of S1S0-MIN from Present and Previous Electronic Structure Computations.

| 4-thiothymine | C4-C5-C6-H6' | C4-S4' | S4'-C4-C5-C5' |
|--|--------------|--------|---------------|
| QM(SA-3-CAS(10,8)/6-31G*)/MM | 95.7 | 1.98 | 1.2 |
| thymine | C4-C5-C6-H6' | C4-O4' | O4'-C4-C5-C5' |
| OM2/MRCI ^a | 126.5 | 1.224 | 92.8 |
| CASSCF(14,10)/ANO-S ^b | 126.6 | 1.200 | 87.7 |
| MR-CISD(4,4)/SA-3-CAS(12,9)/3-21G ^c | 125.5 | 1.204 | 105.0 |
| SA-2-CAS(6,6)/3-21G ^c | 130.5 | 1.235 | 101.1 |
| SA-2-CAS(6,6)/6-31G* ^c | 129.3 | 1.213 | 94.5 |
| SA-2-CAS(6,6)/cc-pVDZ ^d | 133.6 | 1.183 | 87.9 |

^aFrom Lan, Z.G.; Fabiano, E; Thiel, W. [21]

^bFrom Merchán, M.; González-Luque, R.; Climent, T.; Serrano-Andrés, L.; Rodríguez, E.; Reguero, M.; Peláez D. [22]

^cFrom Zechmann, G.; Barbatti, M. [23]

^dFrom Perun, S.; Sobolewski, A. L.; Domcke, W. [24]

7 Single-point QM Energies

To evaluate the effects of the MM environment (ribose and solvent water molecules) on the energetics, we have performed single-point CASPT2 calculations for the QM part of each QM(CASSCF)/MM optimized structure. The single-point energies are compiled in Table 3. The adiabatic energies of T1-MIN, S1-MIN, and T2-MIN do not differ much at the QM and QM/MM levels (by less than 5 kcal/mol), and they remain close to each other in both cases. The adiabatic energy of S2-MIN is affected more strongly by the MM environment (lowered by

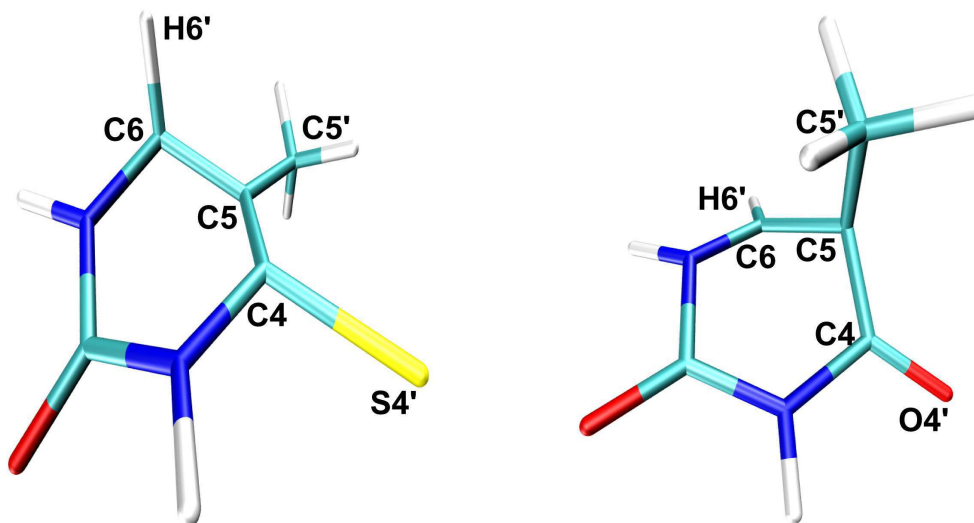


Figure 2: Optimized structures of the S_1/S_0 conical intersection of 4-thiothymine (left) and of thymine (right, SA-2-CAS(6,6)/6-31G* geometry from the work of Zechmann and Barbatti [23])

12.1 kcal/mol). The influence of the MM environment is more pronounced in the case of the conical intersections and crossing points: in the single-point QM calculations, the two relevant states remain close to each other only for S1T1-MIN and T2T1-MIN, but they considerably differ in energy in the other cases. For instance, the S_2-T_2 energy gap of S2T2-MIN increases from 1.3 kcal/mol at the QM(CASPT2//CASSCF)/MM level to 23.3 kcal/mol in the single-point CASPT2 calculation of the QM region. This demonstrates the importance of environmental effects on the geometric structures and energies of crossing points.

Table 3: Relative Energies (in kcal/mol) from QM(CASPT2//CASSCF)/MM Optimizations and from CASPT2 Single-Point Calculations of the QM Regions of QM(CASSCF)/MM Optimized Structures (see Section 12 for Full Cartesian Coordinates)

| | relaxed QM/MM Energy | unrelaxed QM Energy |
|------------------|-----------------------------|-----------------------------|
| S0-MIN | 0.00 | 0.00 |
| S_1 (vertical) | 69.2 | 62.8 |
| S_2 (vertical) | 94.6 | 92.4 |
| T1-MIN | 52.8 | 57.5 |
| S_1 -MIN | 58.6 | 56.9 |
| T2-MIN | 55.8 | 55.2 |
| S_2 -MIN | 71.8 | 83.9 |
| S1S0-MIN | 91.7(S_0)/94.6(S_1) | 74.5(S_0)/87.6(S_1) |
| T1S0-MIN | 91.4(S_0)/92.6(T_1) | 86.8(S_0)/92.9(T_1) |
| S1T1-MIN | 60.0(T_1)/59.7(S_1) | 57.7(T_1)/59.9(S_1) |
| S2S1-MIN | 72.5(S_1)/74.3(S_2) | 61.4(S_1)/83.8(S_2) |
| S2T2-MIN | 71.7(T_2)/73.0(S_2) | 60.4(T_2)/83.7(S_2) |
| T2T1-MIN | 60.2(T_1)/60.5(T_2) | 54.1(T_1)/56.3(T_2) |

8 Spin-Orbit Couplings

Spin-orbit couplings (SOCs) were calculated at the CASSCF(10,8)/6-31G* level with and without MM point charges. The atomic mean-field approximation (AMFI) method [25, 26] implemented in MOLPRO2010 package [27] was employed. Table 4 compiles the computed SOC values.

The SOC values were found to be quite sensitive to the absence or presence of MM point charges. They are influenced by polarization effects from the surrounding water molecules and the nearby ribose group, which thus affect the electronic properties and the mechanistic photophysics of 4-thiothymidine. The effects of the water molecules and ribose group on the spin-orbit couplings are state-specific and non-additive (see Table 4).

Table 4: Absolute Values of QM/MM Calculated Cartesian Components of the Spin-Orbit Coupling Matrix Elements (\hat{H}' : SOC Operator, Unit: cm^{-1}) at Selected Stationary Points. Values Given in A / B / C / D Notation: A, QM Gas-Phase Value; B, QM/MM with MM Charges of Ribose Group Included; C, QM/MM with MM Charges of Water Molecules Included; D, QM/MM with All MM Charges Included.

| structure | | X, Y | Z |
|-----------|--|-------------------------|---------------------------|
| S2T2-MIN | $ \langle S_2 \hat{H}' T_2 \rangle $ | 13.2 / 1.2 / 5.1 / 3.0 | 81.0 / 2.2 / 76.2 / 3.1 |
| S1T1-MIN | $ \langle S_1 \hat{H}' T_1 \rangle $ | 13.7 / 12.9 / 2.0 / 1.8 | 82.7 / 71.1 / 11.8 / 10.2 |
| S2-MIN | $ \langle S_2 \hat{H}' T_1 \rangle $ | 6.6 / 5.3 / 12.4 / 0.8 | 22.5 / 2.3 / 45.2 / 93.2 |
| S2-MIN | $ \langle S_2 \hat{H}' T_2 \rangle $ | 13.1 / 1.2 / 5.3 / 2.8 | 80.6 / 2.1 / 77.1 / 3.0 |
| S1-MIN | $ \langle S_1 \hat{H}' T_1 \rangle $ | 13.7 / 0.9 / 0.6 / 0.7 | 82.7 / 40.1 / 63.2 / 60.6 |

mediate the coupling between a singlet with $M_S=0$ and triplet substates with $M_S=+1$ and $M_S=-1$. Their absolute values are identical in the AMFI approximation. They are smaller than the Z components at the QM level and are generally further reduced by inclusion of the MM point charges.

The Z components of the SOC matrix elements mediate the coupling between a singlet with $M_S=0$ and the triplet substate with $M_S=0$. Their absolute values (A) are usually quite large (around 80 cm^{-1} in all cases except for $\langle S_2 | \hat{H}' | T_1 \rangle$ in S2-MIN). Including the ribose MM charges (B) lowers $\langle S_2 | \hat{H}' | T_2 \rangle$ dramatically in S2T2-MIN and S2-MIN, and both values remain low at the full QM/MM level (D). By contrast, $\langle S_1 | \hat{H}' | T_1 \rangle$ in S1T1-MIN is mostly reduced when incorporating the water MM charges (C), whereas $\langle S_1 | \hat{H}' | T_1 \rangle$ in S1-MIN remains sizable at all QM/MM levels, e.g. 60.6 cm^{-1} at (D). Finally, $\langle S_2 | \hat{H}' | T_1 \rangle$ in S2-MIN increases dramatically when including the MM point charges, up to the full QM/MM value of 93.2 cm^{-1} at (D).

9 Local Hydrogen-Bonding Networks

We have examined the local hydrogen-bonding (HB) networks of all optimized structures near the $S4'$ and $O2'$ acceptor atoms and the $N3H3'$ donor moiety. The corresponding parts of the QM/MM optimized structures are shown in Figures 3–13, and the relevant HB distances are listed in Table 5. The HB networks near $O2'$ and $N3H3'$ are rather stable: there are always three HBs and one HB, respectively, which are strong having HB distances

Table 5: Hydrogen-Bond Distances (in Å) around Acceptor Atoms ($O2'$ and $S4'$) and Donor Moiety ($N3-H3'$)

| | HB ($O2'$) | | | HB ($H3'$) | HB ($S4'$) | | |
|----------|--------------|------|------|--------------|--------------|------|------|
| S0-MIN | 1.57 | 1.62 | 1.67 | 1.52 | 2.15 | 2.20 | 2.40 |
| S1-MIN | 1.55 | 1.62 | 1.65 | 1.52 | 2.36 | 2.45 | 2.55 |
| S2-MIN | 1.56 | 1.62 | 1.67 | 1.52 | 2.21 | 2.31 | 2.80 |
| T1-MIN | 1.55 | 1.61 | 1.66 | 1.52 | 2.18 | 2.22 | 2.50 |
| T2-MIN | 1.56 | 1.61 | 1.61 | 1.53 | 2.43 | 2.82 | - |
| S1S0-MIN | 1.53 | 1.61 | 1.66 | 1.52 | 2.23 | 2.28 | 2.40 |
| S1T1-MIN | 1.56 | 1.62 | 1.65 | 1.52 | 2.25 | 2.43 | - |
| S2S1-MIN | 1.56 | 1.62 | 1.67 | 1.52 | 2.21 | 2.30 | 2.80 |
| S2T2-MIN | 1.56 | 1.62 | 1.68 | 1.52 | 2.20 | 2.31 | 2.80 |
| T2T1-MIN | 1.56 | 1.62 | 1.63 | 1.52 | 2.44 | 2.80 | 2.88 |
| S0T1-MIN | 1.56 | 1.63 | 1.64 | 1.50 | 2.17 | 2.18 | 2.61 |

of typically 1.5–1.6 Å that vary only slightly for a given HB (maximum fluctuation of 0.07 Å). By contrast, the number of HBs varies near the $S4'$ acceptor atom: for example, there are only two (rather than three) HBs in T2-MIN and S1T1-MIN. Moreover, the distances and orientations of the HBs around $S4'$ differ significantly between the different structures (e.g. within a range of 0.6 Å for the second HB). Thus, the HB network near $S4'$ appears to be quite variable.

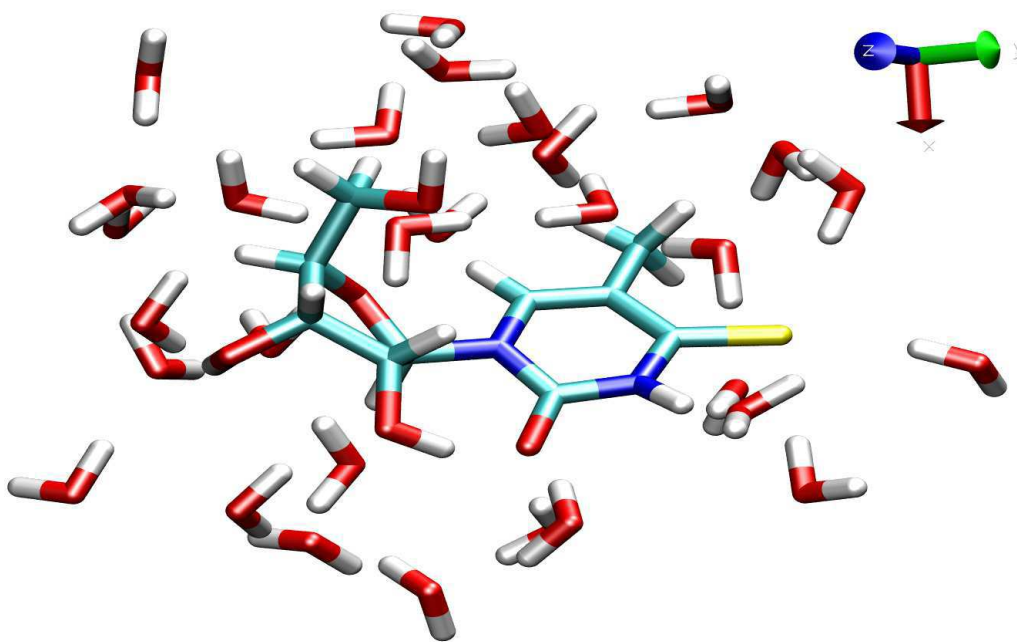


Figure 3: S0-MIN

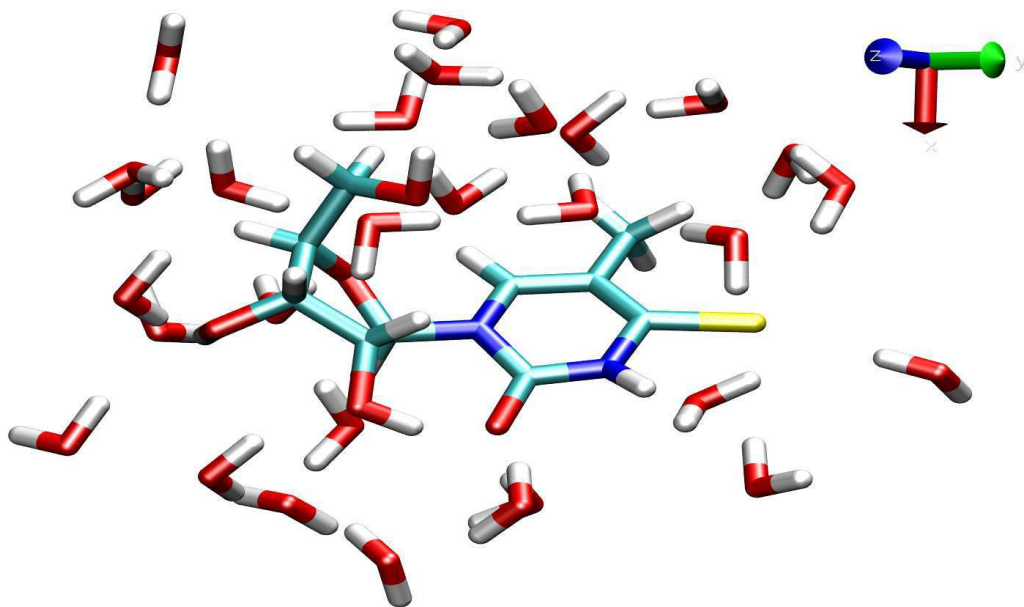


Figure 4: S1-MIN

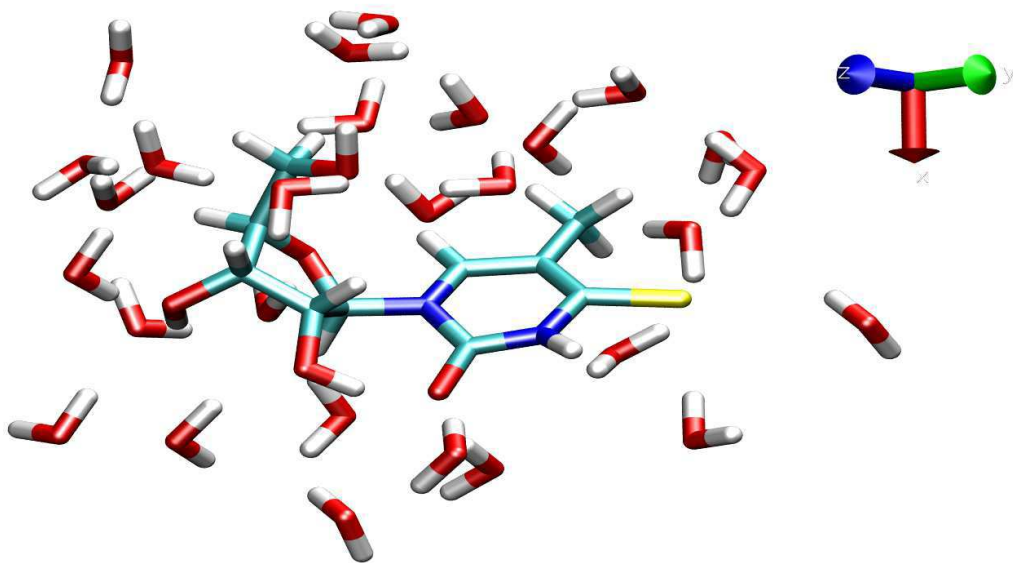


Figure 5: S2-MIN

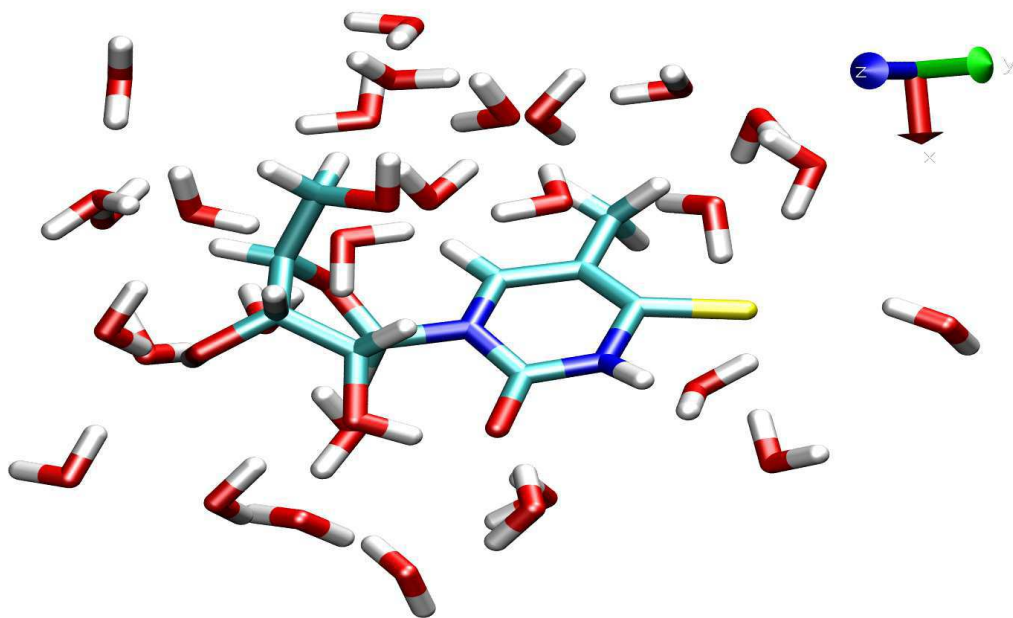


Figure 6: T1-MIN

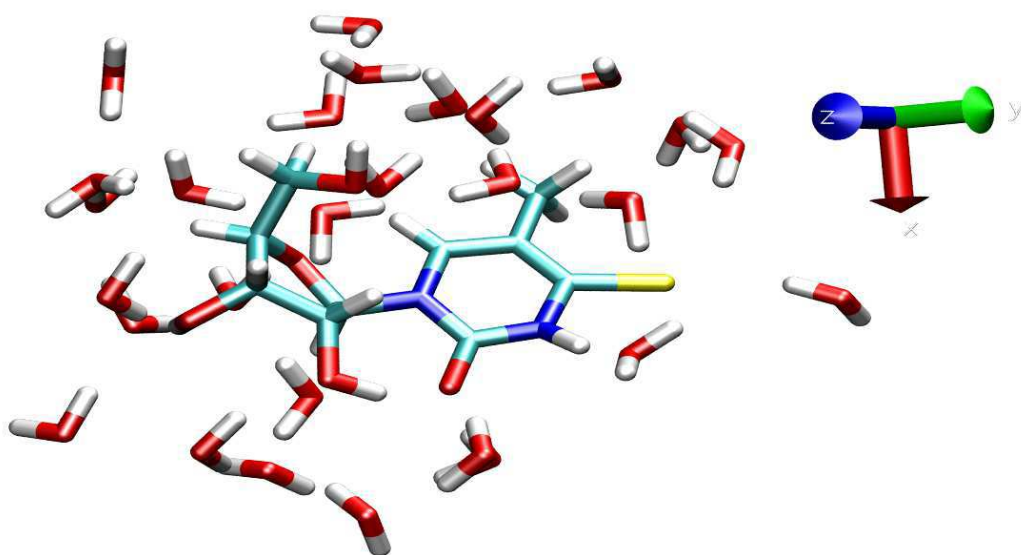


Figure 7: T2-MIN

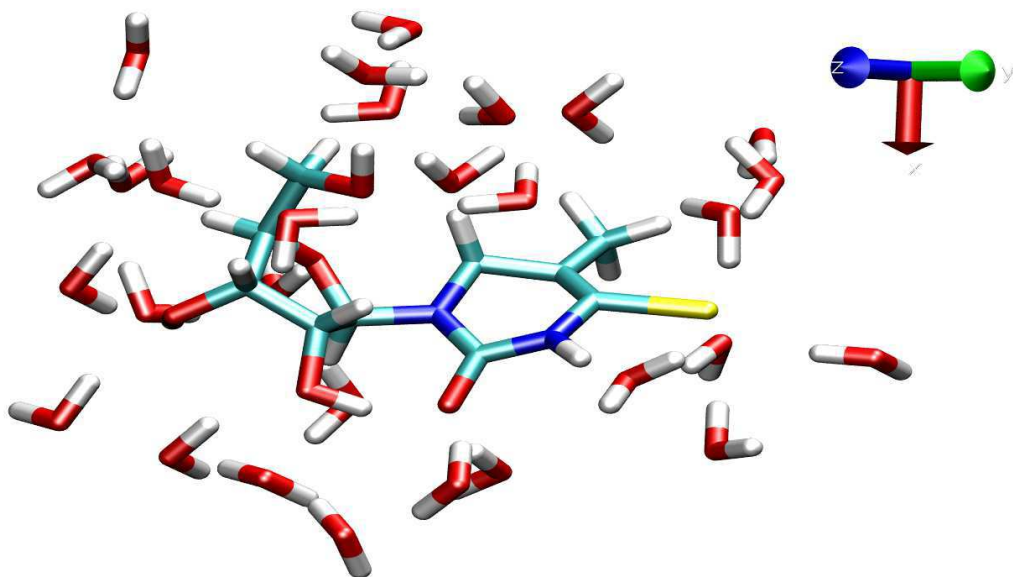


Figure 8: S1S0-MIN

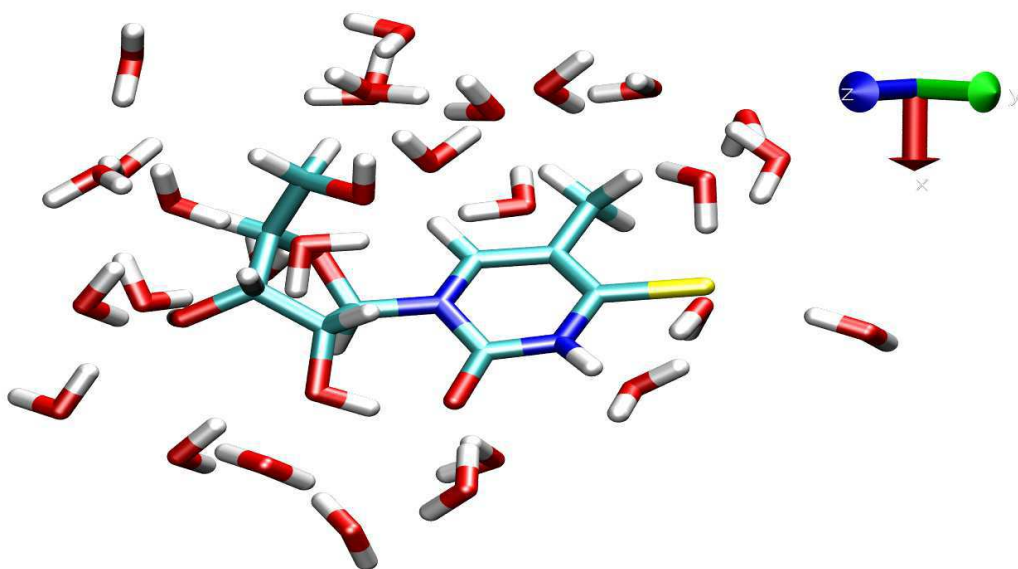


Figure 9: S1T1-MIN

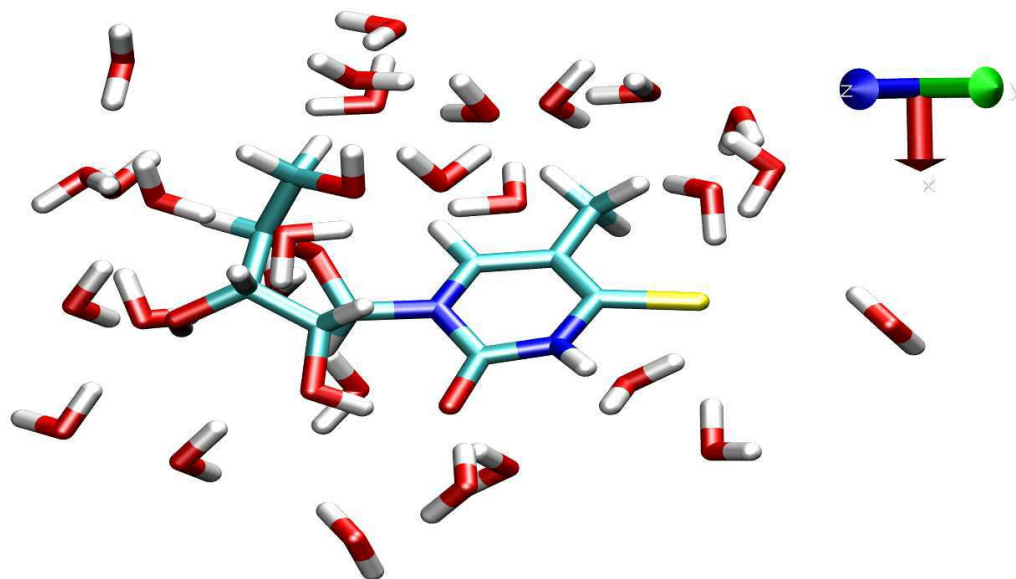


Figure 10: S2S1-MIN

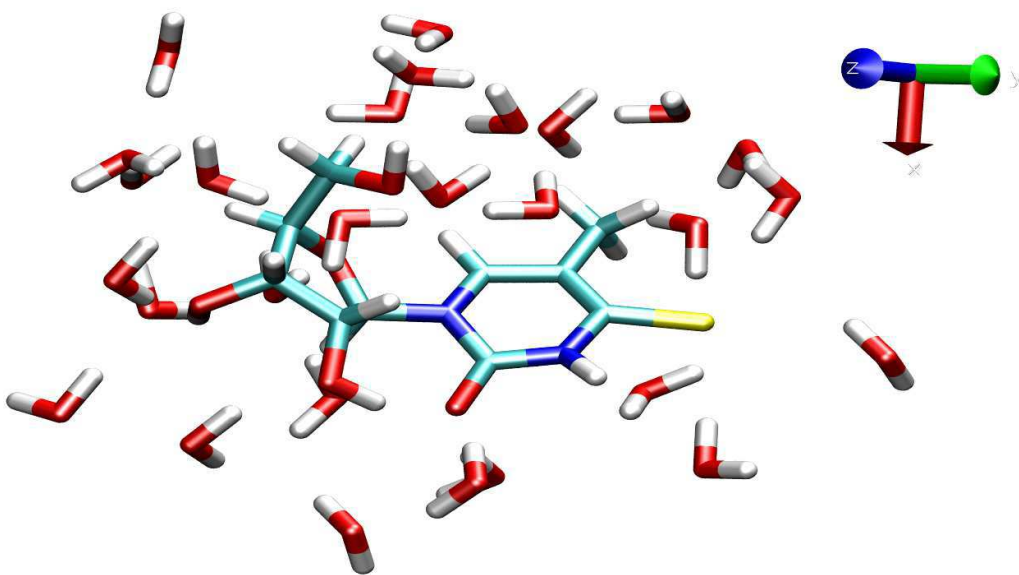


Figure 11: S2T2-MIN

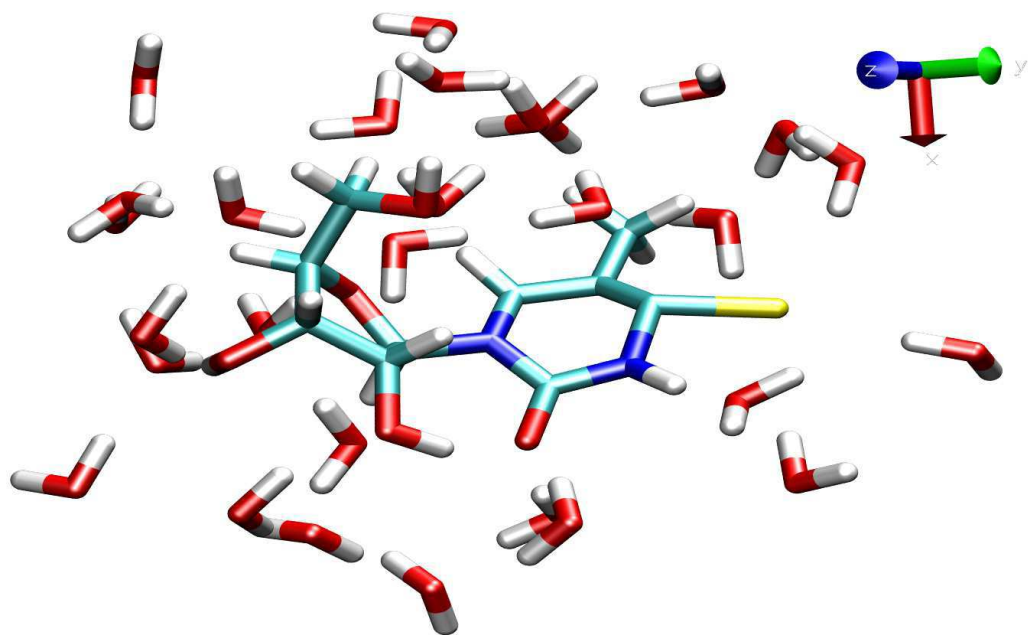


Figure 12: T1S0-MIN

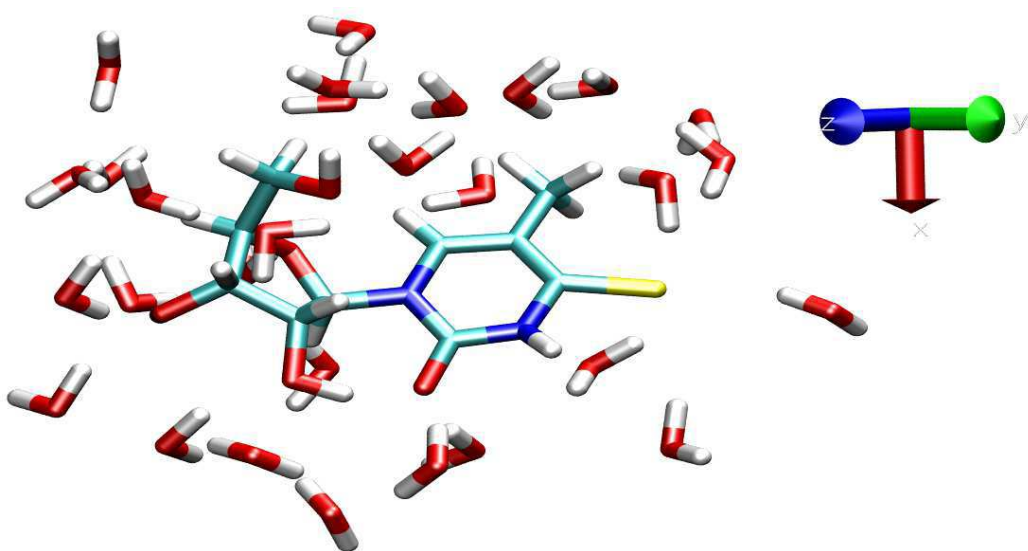


Figure 13: T2T1-MIN

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11 . Force Fields

Topological file

```
MASS  4 HT      1.00800 H ! TIPS3P WATER HYDROGEN
MASS  75 OT     15.99940 O ! TIPS3P WATER OXYGEN
MASS  201 NC=O  14.006700
MASS  202 C=C   12.011000
MASS  203 C=O   12.011000
MASS  204 CR    12.011000
MASS  205 HCMM  1.007940
MASS  206 OR    15.999400
MASS  207 HNCO  1.007940
MASS  208 O=C   15.999400
MASS  209 HOR   1.007940
MASS  210 S=C   32.066000
```

```
AUTOGENERATE ANGLES DIHE
DEFA FIRS NONE LAST NONE
```

```
RESI LIG  0.000
GROUP
ATOM N1  NC=O -0.4691
ATOM C4  C=C  -0.0410
ATOM C5  C=C  -0.1238
ATOM C7  C=O  0.4256
ATOM N2  NC=O -0.4900
ATOM C8  C=O  0.6900
ATOM C1  CR   0.2800
ATOM H1  HCMM 0.0000
ATOM H2  HCMM 0.0000
ATOM C2  CR   0.2800
ATOM H3  HCMM 0.0000
ATOM O1  OR  -0.5600
ATOM C3  CR   0.5801
ATOM H4  HCMM 0.0000
ATOM H5  HCMM 0.1500
ATOM C6  CR   0.1382
ATOM H6  HCMM 0.0000
ATOM H7  HCMM 0.0000
ATOM H8  HCMM 0.0000
ATOM H9  HNCO 0.3700
ATOM O2  O=C  -0.5700
ATOM C9  CR   0.2800
ATOM H10 HCMM 0.0000
ATOM C10 CR   0.2800
ATOM H11 HCMM 0.0000
ATOM O3  OR  -0.6800
ATOM O4  OR  -0.6800
ATOM H12 HOR  0.4000
ATOM H13 HOR  0.4000
ATOM O5  OR  -0.6800
ATOM H14 HOR  0.4000
ATOM S1  S=C  -0.3800
BOND H7  C6
BOND H8  C6
BOND C6  H6
BOND C6  C5
```

BOND S1 C7
 BOND C5 C7
 BOND C5 C4
 BOND C7 N2
 BOND H5 C4
 BOND C4 N1
 BOND N2 H9
 BOND N2 C8
 BOND N1 C8
 BOND N1 C3
 BOND C8 O2
 BOND H4 C3
 BOND C3 O1
 BOND C3 C10
 BOND O1 C2
 BOND H12 O4
 BOND H1 C1
 BOND O4 C10
 BOND C10 H11
 BOND C10 C9
 BOND C2 C1
 BOND C2 H3
 BOND C2 C9
 BOND C1 O5
 BOND C1 H2
 BOND O5 H14
 BOND H13 O3
 BOND C9 O3
 BOND C9 H10
 IMPH N1 C8 C3 C4
 IMPH C4 C5 N1 H5
 IMPH C5 C7 C4 C6
 IMPH C7 S1 C5 N2
 IMPH C3 O1 N1 C10
 IMPH C3 O1 N1 H4
 IMPH C2 C1 O1 C9
 IMPH C2 C1 O1 H3
 IMPH C1 O5 C2 H1
 IMPH C1 H1 C2 H2
 IMPH C6 H7 C5 H8
 IMPH C6 H7 C5 H6
 IMPH N2 C8 C7 H9
 IMPH C8 N2 N1 O2
 IMPH C9 C10 C2 O3
 IMPH C9 C10 C2 H10
 IMPH C10 C9 C3 O4
 IMPH C10 O4 C3 H11
 IC N1 C4 C5 C7 1.37 121.23 0.04 120.79 1.44
 IC N1 C4 C5 C6 1.37 121.23 -179.96 121.64 1.50
 IC N1 C8 N2 C7 1.37 115.23 -0.04 126.50 1.38
 IC N1 C8 N2 H9 1.37 115.23 179.99 116.76 1.09
 IC N1 C3 O1 C2 1.53 109.58 -127.70 109.99 1.46
 IC N1 C3 C10 C9 1.53 113.29 146.12 101.86 1.53
 IC N1 C3 C10 H11 1.53 113.29 4.80 139.98 1.09
 IC N1 C3 C10 O4 1.53 113.29 -105.61 81.62 1.43
 IC C4 N1 C8 N2 1.37 122.21 0.08 115.23 1.38
 IC C4 N1 C8 O2 1.37 122.21 -179.96 123.05 1.22
 IC C4 N1 C3 O1 1.37 123.05 -30.94 109.58 1.42
 IC C4 N1 C3 H4 1.37 123.05 85.57 103.02 1.09

| | | | | | | | | |
|-------|-----|-----|-----|------|--------|---------|--------|------|
| IC C4 | N1 | C3 | C10 | 1.37 | 123.05 | -151.41 | 113.29 | 1.53 |
| IC C4 | C5 | C7 | N2 | 1.34 | 120.79 | -0.00 | 114.04 | 1.38 |
| IC C4 | C5 | C7 | S1 | 1.34 | 120.79 | 179.97 | 125.36 | 1.57 |
| IC C4 | C5 | C6 | H6 | 1.34 | 121.64 | 60.02 | 109.49 | 1.09 |
| IC C4 | C5 | C6 | H7 | 1.34 | 121.64 | -179.98 | 109.50 | 1.09 |
| IC C4 | C5 | C6 | H8 | 1.34 | 121.64 | -59.97 | 109.51 | 1.09 |
| IC C5 | C4 | N1 | C8 | 1.34 | 121.23 | -0.08 | 122.21 | 1.37 |
| IC C5 | C4 | N1 | C3 | 1.34 | 121.23 | 177.30 | 123.05 | 1.53 |
| IC C5 | C7 | N2 | C8 | 1.44 | 114.04 | -0.00 | 126.50 | 1.38 |
| IC C5 | C7 | N2 | H9 | 1.44 | 114.04 | 179.97 | 116.74 | 1.09 |
| IC C7 | C5 | C4 | H5 | 1.44 | 120.79 | 177.31 | 118.69 | 1.08 |
| IC C7 | C5 | C6 | H6 | 1.44 | 117.57 | -119.99 | 109.49 | 1.09 |
| IC C7 | C5 | C6 | H7 | 1.44 | 117.57 | 0.01 | 109.50 | 1.09 |
| IC C7 | C5 | C6 | H8 | 1.44 | 117.57 | 120.03 | 109.51 | 1.09 |
| IC C7 | N2 | C8 | O2 | 1.38 | 126.50 | -180.00 | 121.72 | 1.22 |
| IC N2 | C7 | C5 | C6 | 1.38 | 114.04 | -180.00 | 117.57 | 1.50 |
| IC N2 | C8 | N1 | C3 | 1.38 | 115.23 | -177.51 | 114.69 | 1.53 |
| IC C8 | N1 | C4 | H5 | 1.37 | 122.21 | -177.31 | 120.02 | 1.08 |
| IC C8 | N1 | C3 | O1 | 1.37 | 114.69 | 146.62 | 109.58 | 1.42 |
| IC C8 | N1 | C3 | H4 | 1.37 | 114.69 | -96.87 | 103.02 | 1.09 |
| IC C8 | N1 | C3 | C10 | 1.37 | 114.69 | 26.15 | 113.29 | 1.53 |
| IC C8 | N2 | C7 | S1 | 1.38 | 126.50 | -179.97 | 120.60 | 1.57 |
| IC C1 | C2 | O1 | C3 | 1.52 | 108.84 | 105.59 | 109.99 | 1.42 |
| IC C1 | C2 | C9 | H10 | 1.52 | 115.77 | 30.02 | 109.51 | 1.09 |
| IC C1 | C2 | C9 | C10 | 1.52 | 115.77 | -86.27 | 102.80 | 1.53 |
| IC C1 | C2 | C9 | O3 | 1.52 | 115.77 | 156.55 | 116.52 | 1.42 |
| IC H1 | C1 | C2 | H3 | 1.07 | 109.46 | -79.97 | 109.46 | 1.09 |
| IC H1 | C1 | C2 | O1 | 1.07 | 109.46 | 33.69 | 108.84 | 1.46 |
| IC H1 | C1 | C2 | C9 | 1.07 | 109.46 | 150.91 | 115.77 | 1.53 |
| IC H1 | C1 | O5 | H14 | 1.07 | 109.47 | 92.50 | 109.54 | 0.96 |
| IC H2 | C1 | C2 | H3 | 1.07 | 109.43 | 40.00 | 109.46 | 1.09 |
| IC H2 | C1 | C2 | O1 | 1.07 | 109.43 | 153.66 | 108.84 | 1.46 |
| IC H2 | C1 | C2 | C9 | 1.07 | 109.43 | -89.12 | 115.77 | 1.53 |
| IC H2 | C1 | O5 | H14 | 1.07 | 109.49 | -27.51 | 109.54 | 0.96 |
| IC C2 | C1 | O5 | H14 | 1.52 | 109.50 | -147.50 | 109.54 | 0.96 |
| IC C2 | O1 | C3 | H4 | 1.46 | 109.99 | 119.98 | 109.52 | 1.09 |
| IC C2 | O1 | C3 | C10 | 1.46 | 109.99 | -3.97 | 107.84 | 1.53 |
| IC C2 | C9 | C10 | C3 | 1.53 | 102.80 | -34.79 | 101.86 | 1.53 |
| IC C2 | C9 | C10 | H11 | 1.53 | 102.80 | 119.98 | 109.50 | 1.09 |
| IC C2 | C9 | C10 | O4 | 1.53 | 102.80 | -119.99 | 109.48 | 1.43 |
| IC C2 | C9 | O3 | H13 | 1.53 | 116.52 | 33.38 | 109.43 | 0.96 |
| IC H3 | C2 | C1 | O5 | 1.09 | 109.46 | 160.02 | 109.50 | 1.43 |
| IC H3 | C2 | O1 | C3 | 1.09 | 104.51 | -137.55 | 109.99 | 1.42 |
| IC H3 | C2 | C9 | H10 | 1.09 | 113.01 | -97.35 | 109.51 | 1.09 |
| IC H3 | C2 | C9 | C10 | 1.09 | 113.01 | 146.36 | 102.80 | 1.53 |
| IC H3 | C2 | C9 | O3 | 1.09 | 113.01 | 29.18 | 116.52 | 1.42 |
| IC O1 | C2 | C1 | O5 | 1.46 | 108.84 | -86.32 | 109.50 | 1.43 |
| IC O1 | C2 | C9 | H10 | 1.46 | 104.44 | 149.67 | 109.51 | 1.09 |
| IC O1 | C2 | C9 | C10 | 1.46 | 104.44 | 33.37 | 102.80 | 1.53 |
| IC O1 | C2 | C9 | O3 | 1.46 | 104.44 | -83.80 | 116.52 | 1.42 |
| IC O1 | C3 | C10 | C9 | 1.42 | 107.84 | 24.66 | 101.86 | 1.53 |
| IC O1 | C3 | C10 | H11 | 1.42 | 107.84 | -116.66 | 139.98 | 1.09 |
| IC O1 | C3 | C10 | O4 | 1.42 | 107.84 | 132.93 | 81.62 | 1.43 |
| IC C3 | N1 | C4 | H5 | 1.53 | 123.05 | 0.07 | 120.02 | 1.08 |
| IC C3 | N1 | C8 | O2 | 1.53 | 114.69 | 2.45 | 123.05 | 1.22 |
| IC C3 | O1 | C2 | C9 | 1.42 | 109.99 | -18.62 | 104.44 | 1.53 |
| IC C3 | C10 | C9 | H10 | 1.53 | 101.86 | -151.12 | 109.46 | 1.09 |
| IC C3 | C10 | C9 | O3 | 1.53 | 101.86 | 88.67 | 107.43 | 1.42 |
| IC C3 | C10 | O4 | H12 | 1.53 | 81.62 | 80.33 | 109.50 | 0.96 |

| | | | | | | | | |
|--------|-----|------|-----|------|--------|---------|--------|------|
| IC H4 | C3 | C10 | C9 | 1.09 | 113.50 | -96.85 | 101.86 | 1.53 |
| IC H4 | C3 | C10 | H11 | 1.09 | 113.50 | 121.83 | 139.98 | 1.09 |
| IC H4 | C3 | C10 | O4 | 1.09 | 113.50 | 11.42 | 81.62 | 1.43 |
| IC H5 | C4 | C5 | C6 | 1.08 | 118.69 | -2.70 | 121.64 | 1.50 |
| IC C6 | C5 | C7 | S1 | 1.50 | 117.57 | -0.03 | 125.36 | 1.57 |
| IC H9 | N2 | C7 | S1 | 1.09 | 116.74 | 0.00 | 120.60 | 1.57 |
| IC H9 | N2 | C8 | O2 | 1.09 | 116.76 | 0.03 | 121.72 | 1.22 |
| IC C9 | C2 | C1 | O5 | 1.53 | 115.77 | 30.90 | 109.50 | 1.43 |
| IC C9 | C10 | O4 | H12 | 1.53 | 109.48 | -179.99 | 109.50 | 0.96 |
| IC H10 | C9 | C10 | H11 | 1.09 | 109.46 | 3.64 | 109.50 | 1.09 |
| IC H10 | C9 | C10 | O4 | 1.09 | 109.46 | 123.68 | 109.48 | 1.43 |
| IC H10 | C9 | O3 | H13 | 1.09 | 110.66 | 159.33 | 109.43 | 0.96 |
| IC C10 | C9 | O3 | H13 | 1.53 | 107.43 | -81.22 | 109.43 | 0.96 |
| IC H11 | C10 | C9 | O3 | 1.09 | 109.50 | -116.57 | 107.43 | 1.42 |
| IC H11 | C10 | O4 | H12 | 1.09 | 109.48 | -59.94 | 109.50 | 0.96 |
| IC O3 | C9 | C10 | O4 | 1.42 | 107.43 | 3.47 | 109.48 | 1.43 |
| IC C8 | C3 | *N1 | C4 | 0.00 | 0.00 | 180.00 | 0.00 | 0.00 |
| IC C5 | N1 | *C4 | H5 | 0.00 | 0.00 | 180.00 | 0.00 | 0.00 |
| IC C7 | C4 | *C5 | C6 | 0.00 | 0.00 | 180.00 | 0.00 | 0.00 |
| IC S1 | C5 | *C7 | N2 | 0.00 | 0.00 | 180.00 | 0.00 | 0.00 |
| IC O1 | N1 | *C3 | C10 | 0.00 | 0.00 | 120.00 | 0.00 | 0.00 |
| IC O1 | N1 | *C3 | H4 | 0.00 | 0.00 | -120.00 | 0.00 | 0.00 |
| IC C1 | O1 | *C2 | C9 | 0.00 | 0.00 | 120.00 | 0.00 | 0.00 |
| IC C1 | O1 | *C2 | H3 | 0.00 | 0.00 | -120.00 | 0.00 | 0.00 |
| IC O5 | C2 | *C1 | H1 | 0.00 | 0.00 | 120.00 | 0.00 | 0.00 |
| IC H1 | C2 | *C1 | H2 | 0.00 | 0.00 | -120.00 | 0.00 | 0.00 |
| IC H7 | C5 | *C6 | H8 | 0.00 | 0.00 | 120.00 | 0.00 | 0.00 |
| IC H7 | C5 | *C6 | H6 | 0.00 | 0.00 | -120.00 | 0.00 | 0.00 |
| IC C8 | C7 | *N2 | H9 | 0.00 | 0.00 | 180.00 | 0.00 | 0.00 |
| IC N2 | N1 | *C8 | O2 | 0.00 | 0.00 | 180.00 | 0.00 | 0.00 |
| IC C10 | C2 | *C9 | O3 | 0.00 | 0.00 | 120.00 | 0.00 | 0.00 |
| IC C10 | C2 | *C9 | H10 | 0.00 | 0.00 | -120.00 | 0.00 | 0.00 |
| IC C9 | C3 | *C10 | O4 | 0.00 | 0.00 | 120.00 | 0.00 | 0.00 |
| IC O4 | C3 | *C10 | H11 | 0.00 | 0.00 | -120.00 | 0.00 | 0.00 |

RESI TIP3 0.000 ! tip3p water model, generate using noangle nodihedral
GROUP
ATOM OH2 OT -0.834
ATOM H1 HT 0.417
ATOM H2 HT 0.417
BOND OH2 H1 OH2 H2 H1 H2 ! the last bond is needed for shake
ANGLE H1 OH2 H2 ! required
ACCEPTOR OH2
PATCHING FIRS NONE LAST NONE

Parameter file

BONDS
HCMM CR 342.991 1.0930
CR C=C 326.655 1.4820
S=C C=O 340.760 1.6650
C=C C=O 328.526 1.4680
C=C C=C 684.039 1.3330
C=O NC=O 419.491 1.3690
HCMM C=C 372.066 1.0830
C=C NC=O 455.474 1.3620
NC=O HNCO 479.511 1.0150
NC=O CR 335.651 1.4360
C=O O=C 931.963 1.2220

CR OR 363.214 1.4180
CR CR 306.432 1.5080
HOR OR 560.905 0.9720
OT HT 450.000 0.9572 ! ALLOW TIP3P WAT
HT HT 0.000 1.5139 ! ALLOW WAT

ANGLES

C=C NC=O C=O 71.966 120.7030
C=C NC=O CR 72.254 118.9160
C=O NC=O CR 59.084 119.6000
NC=O C=C C=C 72.182 120.8280
NC=O C=C HCMM 48.001 114.8590
C=C C=C HCMM 38.502 121.0040
C=C C=C C=O 39.221 111.2970
C=C C=C CR 48.361 122.1410
C=O C=C CR 50.232 116.1040
C=C C=O NC=O 74.988 111.7210
C=C C=O S=C 63.402 124.8500
NC=O C=O S=C 72.326 123.1500
C=O NC=O C=O 51.024 120.2740
C=O NC=O HNCO 41.380 120.2770
NC=O C=O NC=O 116.009 114.9230
NC=O C=O O=C 65.273 127.1520
HCMM CR HCMM 37.134 108.8360
HCMM CR CR 45.770 110.5490
HCMM CR OR 56.205 108.5770
CR CR OR 71.390 108.1330
CR CR CR 61.243 109.6080
CR OR CR 86.143 106.9260
NC=O CR OR 103.055 108.5680
NC=O CR HCMM 53.255 107.6460
NC=O CR CR 75.564 109.9600
C=C CR HCMM 45.482 110.2920
CR OR HOR 57.069 106.5030
HT OT HT 55.000 104.5200 ! ALLOW TIP3P WAT

DIHEDRALS

NC=O C=C C=C C=O 6.000 2 180.00
NC=O C=C C=C CR 6.000 2 180.00
NC=O C=O NC=O C=O 3.000 2 180.00
NC=O C=O NC=O HNCO 1.748 2 180.00
NC=O C=O NC=O HNCO 0.645 3 0.00
NC=O CR OR CR 0.100 3 0.00
NC=O CR CR CR 0.150 3 0.00
NC=O CR CR HCMM 0.213 3 0.00
NC=O CR CR OR 0.150 3 0.00
C=C NC=O C=O NC=O 3.000 2 180.00
C=C NC=O C=O O=C 3.000 2 180.00
C=C NC=O CR OR 0.150 3 0.00
C=C NC=O CR HCMM 0.150 3 0.00
C=C NC=O CR CR 0.150 3 0.00
C=C C=C C=O NC=O 0.048 1 0.00
C=C C=C C=O NC=O 0.791 2 180.00
C=C C=C C=O NC=O 0.190 3 0.00
C=C C=C C=O S=C 1.250 2 180.00
C=C C=C CR HCMM 0.251 1 0.00
C=C C=C CR HCMM -0.205 2 180.00
C=C C=C CR HCMM -0.268 3 0.00
C=C C=C NC=O C=O 3.000 2 180.00

| | | | |
|-------------------|--------|---|--------|
| C=C C=C NC=O CR | 3.000 | 2 | 180.00 |
| C=C C=O NC=O C=O | 3.000 | 2 | 180.00 |
| C=C C=O NC=O HNCO | -0.143 | 1 | 0.00 |
| C=C C=O NC=O HNCO | 3.571 | 2 | 180.00 |
| C=C C=O NC=O HNCO | 0.060 | 3 | 0.00 |
| C=O C=C C=C HCMM | 0.900 | 2 | 180.00 |
| C=O C=C CR HCMM | -0.054 | 3 | 0.00 |
| C=O NC=O C=O O=C | 0.388 | 1 | 0.00 |
| C=O NC=O C=O O=C | -0.292 | 2 | 180.00 |
| C=O NC=O C=O O=C | -0.072 | 3 | 0.00 |
| NC=O C=O C=C CR | 1.250 | 2 | 180.00 |
| NC=O C=O NC=O CR | 3.000 | 2 | 180.00 |
| C=O NC=O C=C HCMM | 3.000 | 2 | 180.00 |
| C=O NC=O CR OR | 0.500 | 3 | 0.00 |
| C=O NC=O CR HCMM | -1.050 | 1 | 0.00 |
| C=O NC=O CR HCMM | 0.681 | 2 | 180.00 |
| C=O NC=O CR HCMM | 0.011 | 3 | 0.00 |
| C=O NC=O CR CR | -0.513 | 1 | 0.00 |
| C=O NC=O CR CR | 0.347 | 2 | 180.00 |
| C=O NC=O CR CR | 0.474 | 3 | 0.00 |
| C=O NC=O C=O S=C | 3.000 | 2 | 180.00 |
| CR CR OR CR | -0.341 | 1 | 0.00 |
| CR CR OR CR | 0.378 | 2 | 180.00 |
| CR CR OR CR | 0.378 | 3 | 0.00 |
| CR CR CR HCMM | 0.320 | 1 | 0.00 |
| CR CR CR HCMM | -0.315 | 2 | 180.00 |
| CR CR CR HCMM | 0.132 | 3 | 0.00 |
| CR CR CR CR | 0.051 | 1 | 0.00 |
| CR CR CR CR | 0.341 | 2 | 180.00 |
| CR CR CR CR | 0.166 | 3 | 0.00 |
| CR CR CR OR | -0.344 | 1 | 0.00 |
| CR CR CR OR | 0.878 | 2 | 180.00 |
| CR CR CR OR | 0.238 | 3 | 0.00 |
| HCMM CR CR HCMM | 0.142 | 1 | 0.00 |
| HCMM CR CR HCMM | -0.693 | 2 | 180.00 |
| HCMM CR CR HCMM | 0.157 | 3 | 0.00 |
| HCMM CR CR OR | -0.327 | 1 | 0.00 |
| HCMM CR CR OR | 0.536 | 2 | 180.00 |
| HCMM CR CR OR | 0.140 | 3 | 0.00 |
| HCMM CR OR HOR | 0.298 | 1 | 0.00 |
| HCMM CR OR HOR | -0.138 | 2 | 180.00 |
| HCMM CR OR HOR | 0.173 | 3 | 0.00 |
| CR CR OR HOR | 0.135 | 2 | 180.00 |
| CR CR OR HOR | 0.118 | 3 | 0.00 |
| CR OR CR HCMM | 0.285 | 1 | 0.00 |
| CR OR CR HCMM | 0.160 | 2 | 180.00 |
| CR OR CR HCMM | 0.285 | 3 | 0.00 |
| OR CR CR OR | 0.204 | 1 | 0.00 |
| OR CR CR OR | 0.699 | 2 | 180.00 |
| OR CR CR OR | 0.480 | 3 | 0.00 |
| CR NC=O C=C HCMM | 3.000 | 2 | 180.00 |
| CR NC=O C=O O=C | -0.160 | 1 | 0.00 |
| CR NC=O C=O O=C | 3.147 | 2 | 180.00 |
| CR NC=O C=O O=C | -0.073 | 3 | 0.00 |
| HCMM C=C C=C CR | 6.000 | 2 | 180.00 |
| CR C=C C=O S=C | 1.250 | 2 | 180.00 |
| HNCO NC=O C=O S=C | 3.000 | 2 | 180.00 |
| HNCO NC=O C=O O=C | 0.718 | 1 | 0.00 |
| HNCO NC=O C=O O=C | 2.487 | 2 | 180.00 |

HNCO NC=O C=O O=C -0.227 3 0.00

IMPROPER

NC=O C=O CR C=C -1.439 0 0.00
C=C C=C NC=O HCMM 1.439 0 0.00
C=C C=O C=C CR 1.871 0 0.00
C=O S=C C=C NC=O 9.356 0 0.00
CR OR NC=O CR 0.000 0 0.00
CR OR NC=O HCMM 0.000 0 0.00
CR CR OR CR 0.000 0 0.00
CR CR OR HCMM 0.000 0 0.00
CR OR CR HCMM 0.000 0 0.00
CR HCMM CR HCMM 0.000 0 0.00
CR HCMM C=C HCMM 0.000 0 0.00
NC=O C=O C=O HNCO -2.159 0 0.00
C=O NC=O NC=O O=C 8.132 0 0.00
CR CR CR OR 0.000 0 0.00
CR CR CR HCMM 0.000 0 0.00

NONBONDED nbxmod 5 atom cdiel shift vatom vdistance vswitch -
cutnbn 14.0 ctofnb 12.0 ctonnb 10.0 eps 1.0 e14fac 1.0 wmin 1.5

NC=O 0.000000 -0.200000 1.850000
C=C 0.000000 -0.068000 2.090000
C=O 0.000000 -0.110000 2.000000
CR 0.000000 -0.055000 2.175000 0.000000 -0.010000 1.900000
HCMM 0.000000 -0.022000 1.320000
OR 0.000000 -0.152100 1.770000
HNCO 0.000000 -0.046000 0.224500
O=C 0.000000 -0.120000 1.700000 0.000000 -0.120000 1.400000
HOR 0.000000 -0.046000 0.224500
S=C 0.000000 -0.450000 2.000000
OT 0.000000 -0.152100 1.768200 ! ALLOW WAT
HT 0.000000 -0.046000 0.224500 ! ALLOW WAT

12. Cartesian Coordinates of All Optimized Structures

XYZ format (Angstrom)

Full system (3329 atoms, the first 32 atoms are for 4-thiothymidine)

1. S0-MIN
2. S1-MIN
3. S2-MIN
4. T1-MIN
5. T2-MIN
6. S2S1-MIN
7. S2T2-MIN
8. T2T1-MIN
9. S1T1-MIN
10. S1S0-MIN
11. T1S0-MIN

1. S0-MIN

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C -0.3728471 0.6880117 -2.8481644
C 0.5810925 1.7827077 -2.7623278
N 1.5658123 1.6527412 -1.8129100

C 1.7407284 0.6026622 -0.9968964
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H -2.6486953 -1.5015403 2.6189204
C -0.8979719 -2.1229844 1.5246234
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8. T2T1-MIN

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