

# Supporting Information

## State-of-the-art quantum chemistry meets variable reaction coordinate transition state theory to solve the puzzling case of the $\text{H}_2\text{S} + \text{Cl}$ system

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## Input file for RRKM-ME calculations using the MESS package for the $\text{H}_2\text{S} + \text{Cl}$ reaction

In the following, the input file for the VRC-VTSTin calculation is reported. All the important molecular properties are enclosed. The nomenclature used is the same used in Section 3.4.

# VRC-VTSTin

```
!*****
!           GLOBAL SECTION
!*****
!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!
!
!
TemperatureList[K]          200 225 250 275 300 325 350 375 400  425 430 450 475 500 600 700 800 900 1000
PressureList[atm]          0.5 1
!
!
EnergyStepOverTemperature  .2      ! [Discretization energy step (global relax matrix)] / T
ExcessEnergyOverTemperature 30     ! [Highest barrier in the model (global relax matrix)] / T
ModelEnergyLimit[kcal/mol]  400    ! Highest reference energy used in the calculation ( or ReferenceEnergy[kcal/mol])
!
CalculationMethod          direct  ! direct or low-eigenvalue
!
WellCutoff                 20     ! well truncation parameter : Max { dissociation limit (min barrier rel. to bottom of the well) / T }
ChemicalEigenvalueMax      0.2    ! Max chemical eigenvalue / Lowest Collision relaxation eigenvalue
!
ReductionMethod            diagonalization ! [low eigenvalue method only] diagonalization or projection (default)
!
!!!!!!!!!!test!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!
!WellCutoff                10
!ChemicalEigenvalueMin     1.e-6    #only for direct diagonalization method
!!!!!!!!!!test!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!
AtomDistanceMin[bohr]     1.3
!!
RateOutput                 rate.out      ! output file name for rate coefficients
!
!
!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!
!*****
!           MODEL SECTION
!*****
!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!
!
!
Model
!
  EnergyRelaxation
  Exponential               ! Currently the only possible energy relaxation model
    Factor[1/cm]           260        ! (Delta_E_down)^(0) @ standard T (300 K)
    Power                  0.875     ! Power n in the expression (Delta_E_down) = (Delta_E_down)^(0) (T/T0)^(n)
    ExponentCutoff         10        ! if (Delta_E) / (Delta_E_down) > value transition probability is zero
  End
!
  CollisionFrequency        ! Collision frequency model
  LennardJones             ! Currently the only possible collisional frequency model based on LJ potential
    Epsilons[K]           90.58 617.0 ! Epsilon_1 and Epsilon_2
    Sigmas[angstrom]      3.54 5.62  ! Sigma_1 and Sigma_2
    Masses[amu]           39.948 69.0 ! Masses of the buffer gas molecule and of the complex (check order)
  End
!
!*****
```

```

!
!*****
!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!
!*****
! REACTANTS
!*****

Bimolecular REACS
Fragment REACT1
RRHO
Geometry[angstrom]      3
  S      0.00000000  0.00000000  0.10283700
  H      0.00000000  0.96718500 -0.82269200
  H      0.00000000 -0.96718500 -0.82269200
Core RigidRotor
SymmetryFactor  2.0000000000000000
End
  Frequencies[1/cm]      3
  1217.89 2735.01 2750.20
ZeroEnergy[kcal/mol]      0.
ElectronicLevels[1/cm]      1
  0      1
!*****
End
!*****
Fragment REACT2
Atom
Name Cl
ElectronicLevels[1/cm]      2
  0      4
  881.00 2
!*****
End
!*****
GroundEnergy[kcal/mol] 0.0
End
Bimolecular PRODS
Fragment PROD1
RRHO
Geometry[angstrom]      2
  H      0.00000000  0.00000000 -1.26323400
  S      0.00000000  0.00000000  0.07895200
Core RigidRotor
SymmetryFactor  1.0000000000000000
End
  Frequencies[1/cm]      1
  2719.96
ZeroEnergy[kcal/mol]      0.
ElectronicLevels[1/cm]      2
  0.0000000000000000  2.0000000000000000
  376.9599999999998  2.0000000000000000
!*****
End
!*****

```

```

Fragment PROD2
RRHO
Geometry[angstrom]      2
  H      0.00000000  0.00000000 -1.20561900
  Cl     0.00000000  0.00000000  0.07091900
Core RigidRotor
SymmetryFactor  1.0000000000000000
End
  Frequencies[1/cm]    1
  3006.51
ZeroEnergy[kcal/mol]   0.
ElectronicLevels[1/cm] 1
  0.0000000000000000  1.0000000000000000

```

!\*\*\*\*\*

```

End
GroundEnergy[kcal/mol] -12.00
End

```

!\*\*\*\*\*

```

Well WR
Species
RRHO ! well
Geometry[angstrom]    4
  H      0.86853200 -1.21794200  0.97069800
  S     -0.05263800 -1.26295000  0.00000000
  H      0.86853200 -1.21794200 -0.97069800
  Cl    -0.05263800  1.33194700  0.00000000
Core RigidRotor
SymmetryFactor  1.0000000000000000
End
  Frequencies[1/cm]    6
  235.50 361.30 394.60 1211.08 2735.16 2749.92
ZeroEnergy[kcal/mol]  -8.58
ElectronicLevels[1/cm] 1
  0.0000000000000000  2.0000000000000000

```

End

End

!\*\*\*\*\*

```

Well WP
Species
RRHO ! well
Geometry[angstrom]    4
  S     -1.89837200 -0.07864900 -0.00000300
  H     -1.96120000  1.26293700 -0.00000900
  H      0.58255000 -0.07505800  0.00001500
  Cl    1.86780000  0.00414800  0.00000200
Core RigidRotor
SymmetryFactor  1.0000000000000000
End
  Frequencies[1/cm]    6
  92.55 144.45 254.96 397.14 2715.09 2844.56
ZeroEnergy[kcal/mol]  -13.88
ElectronicLevels[1/cm] 1
  0.0000000000000000  2.0000000000000000

```

End

End

```

!*****
Barrier B1 REACS WR
RRHO
Stoichiometry H2S1Cl1
Core Rotd
File flux_lr_so9.dat
SymmetryFactor 2.2200000000
End
Frequencies[1/cm] 3
1217.89 2735.01 2750.20
ZeroEnergy[kcal/mol] 0.0
ElectronicLevels[1/cm] 1
0.0000000000000000 2.0000000000000000
End
!*****
Barrier B3 WP PRODS
RRHO
Stoichiometry H2S1Cl1
Core PhaseSpaceTheory
FragmentGeometry[angstrom] 2
H 0.00000000 0.00000000 -1.26323400
S 0.00000000 0.00000000 0.07895200
FragmentGeometry[angstrom] 2
H 0.00000000 0.00000000 -1.20561900
Cl 0.00000000 0.00000000 0.07091900
SymmetryFactor 1.0000000000000000
PotentialPrefactor[au] 230.
PotentialPowerExponent 6
End
Frequencies[1/cm] 2
2719.96 3006.51
ZeroEnergy[kcal/mol] -12.00
ElectronicLevels[1/cm] 2
0.0000000000000000 2.0000000000000000
376.95999999999998 2.0000000000000000
End
!*****
Barrier B2 WR WP
Variational
RRHO ! 90
Geometry[angstrom] 4
Cl 1.36787 0.01974 -0.00798
H -0.27379 -0.19797 0.71945
S -1.44332 -0.05571 -0.01648
H -1.39984 1.28048 0.08033
Core RigidRotor
SymmetryFactor 0.5000000000000000
End
Frequencies[1/cm] 5
2733.8 2093.0 1136.8 394.25 189.40
ZeroEnergy[kcal/mol] -1.3891033240988111
ElectronicLevels[1/cm] 1
0.0000000000000000 2.0000000000000000
End
!*****
RRHO ! 91

```

```

Geometry[angstrom]      4
Cl  1.36762  0.01969 -0.00777
H   -0.26174 -0.19606  0.70931
S   -1.44342 -0.05572 -0.01638
H   -1.39997  1.28056  0.07988
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End
      Frequencies[1/cm]      5
      2733.4   2039.3   1129.8   396.99   191.83
ZeroEnergy[kcal/mol]  -1.3246034340405763
ElectronicLevels[1/cm]      1
      0.0000000000000000   2.0000000000000000
End
!*****
RRHO      !      92
Geometry[angstrom]      4
Cl  1.36736  0.01964 -0.00756
H   -0.24947 -0.19415  0.69944
S   -1.44353 -0.05573 -0.01629
H   -1.40008  1.28065  0.07946
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End
      Frequencies[1/cm]      5
      2733.0   1982.1   1122.1   399.78   194.59
ZeroEnergy[kcal/mol]  -1.2720111512322105
ElectronicLevels[1/cm]      1
      0.0000000000000000   2.0000000000000000
End
!*****
RRHO      !      93
Geometry[angstrom]      4
Cl  1.36710  0.01959 -0.00735
H   -0.23696 -0.19225  0.68988
S   -1.44364 -0.05574 -0.01620
H   -1.40016  1.28073  0.07908
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End
      Frequencies[1/cm]      5
      2732.5   1921.6   1113.6   402.59   197.68
ZeroEnergy[kcal/mol]  -1.2310834484781283
ElectronicLevels[1/cm]      1
      0.0000000000000000   2.0000000000000000
End
!*****
RRHO      !      94
Geometry[angstrom]      4
Cl  1.36684  0.01954 -0.00714
H   -0.22418 -0.19036  0.68068
S   -1.44375 -0.05574 -0.01612
H   -1.40021  1.28081  0.07873
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End

```

```

Frequencies[1/cm]          5
2732.1   1858.4   1104.3   405.40   201.13
ZeroEnergy[kcal/mol]      -1.2010483570396526
ElectronicLevels[1/cm]    1
0.0000000000000000      2.0000000000000000

```

End

!\*\*\*\*\*

```

RRHO      !          95
Geometry[angstrom]      4
Cl  1.36658  0.01949 -0.00694
H   -0.21112 -0.18850  0.67187
S   -1.44388 -0.05575 -0.01606
H   -1.40024  1.28089  0.07841
Core RigidRotor
SymmetryFactor  0.5000000000000000

```

End

```

Frequencies[1/cm]          5
2731.6   1793.2   1094.0   408.20   204.92
ZeroEnergy[kcal/mol]      -1.1871230884349664
ElectronicLevels[1/cm]    1
0.0000000000000000      2.0000000000000000

```

End

!\*\*\*\*\*

```

RRHO      !          96
Geometry[angstrom]      4
Cl  1.36633  0.01944 -0.00674
H   -0.19777 -0.18666  0.66349
S   -1.44402 -0.05576 -0.01600
H   -1.40024  1.28097  0.07812
Core RigidRotor
SymmetryFactor  0.5000000000000000

```

End

```

Frequencies[1/cm]          5
2731.1   1727.3   1082.9   410.98   209.05
ZeroEnergy[kcal/mol]      -1.1814743148651377
ElectronicLevels[1/cm]    1
0.0000000000000000      2.0000000000000000

```

End

!\*\*\*\*\*

```

RRHO      !          97
Geometry[angstrom]      4
Cl  1.36608  0.01939 -0.00655
H   -0.18414 -0.18488  0.65558
S   -1.44418 -0.05577 -0.01596
H   -1.40022  1.28105  0.07786
Core RigidRotor
SymmetryFactor  0.5000000000000000

```

End

```

Frequencies[1/cm]          5
2730.6   1661.8   1070.8   413.69   213.48
ZeroEnergy[kcal/mol]      -1.1950082180404342
ElectronicLevels[1/cm]    1
0.0000000000000000      2.0000000000000000

```

End

!\*\*\*\*\*

```

RRHO      !          98

```

```

Geometry[angstrom]      4
Cl  1.36583  0.01935 -0.00636
H   -0.17022 -0.18315  0.64815
S   -1.44435 -0.05577 -0.01593
H   -1.40017  1.28112  0.07763
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End
      Frequencies[1/cm]      5
      2730.1  1598.0  1057.9  416.27  218.14
ZeroEnergy[kcal/mol]  -1.2196770344013361
ElectronicLevels[1/cm]      1
      0.0000000000000000  2.0000000000000000
End
!*****
RRHO      !      99
Geometry[angstrom]      4
Cl  1.36560  0.01930 -0.00617
H   -0.15606 -0.18147  0.64119
S   -1.44454 -0.05578 -0.01591
H   -1.40011  1.28119  0.07742
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End
      Frequencies[1/cm]      5
      2729.6  1537.3  1044.1  418.64  222.94
ZeroEnergy[kcal/mol]  -1.2664584242452357
ElectronicLevels[1/cm]      1
      0.0000000000000000  2.0000000000000000
End
!*****
RRHO      !      100
Geometry[angstrom]      4
Cl  1.36537  0.01926 -0.00599
H   -0.14168 -0.17985  0.63466
S   -1.44475 -0.05578 -0.01589
H   -1.40002  1.28126  0.07723
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End
      Frequencies[1/cm]      5
      2729.1  1482.5  1032.3  423.97  228.94
ZeroEnergy[kcal/mol]  -1.3085493681677438
ElectronicLevels[1/cm]      1
      0.0000000000000000  2.0000000000000000
End
!*****
RRHO      !      101
Geometry[angstrom]      4
S   0.00000  0.00000  0.00000
H   0.00000  0.00000  1.34174
H   1.48190  0.00000 -0.02582
Cl  2.55774 -1.20674  0.18741
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End

```



Frequencies[1/cm] 5  
2728.7 1427.7 1014.0 422.35 232.63

ZeroEnergy[kcal/mol] -1.3981992624439694

ElectronicLevels[1/cm] 1  
0.0000000000000000 2.0000000000000000

End

!\*\*\*\*\*

RRHO ! 102

Geometry[angstrom] 4

Cl 1.36495 0.01917 -0.00564

H -0.11227 -0.17684 0.62311

S -1.44523 -0.05579 -0.01590

H -1.39980 1.28137 0.07693

Core RigidRotor

SymmetryFactor 0.5000000000000000

End

Frequencies[1/cm] 5  
2728.3 1375.8 995.55 420.71 235.90

ZeroEnergy[kcal/mol] -1.5096615201384820

ElectronicLevels[1/cm] 1  
0.0000000000000000 2.0000000000000000

End

!\*\*\*\*\*

RRHO ! 103

Geometry[angstrom] 4

Cl 1.36477 0.01913 -0.00547

H -0.09731 -0.17545 0.61803

S -1.44550 -0.05579 -0.01592

H -1.39967 1.28141 0.07682

Core RigidRotor

SymmetryFactor 0.5000000000000000

End

Frequencies[1/cm] 5  
2727.8 1337.9 981.72 424.41 241.57

ZeroEnergy[kcal/mol] -1.6083262095437896

ElectronicLevels[1/cm] 1  
0.0000000000000000 2.0000000000000000

End

!\*\*\*\*\*

RRHO ! 104

Geometry[angstrom] 4

Cl 1.36459 0.01909 -0.00531

H -0.08224 -0.17411 0.61327

S -1.44579 -0.05579 -0.01595

H -1.39952 1.28145 0.07671

Core RigidRotor

SymmetryFactor 0.5000000000000000

End

Frequencies[1/cm] 5  
2727.5 1299.2 964.92 424.74 245.39

ZeroEnergy[kcal/mol] -1.7513611790346379

ElectronicLevels[1/cm] 1  
0.0000000000000000 2.0000000000000000

End

!\*\*\*\*\*

RRHO ! 105

```

Geometry[angstrom]      4
Cl  1.36443  0.01905 -0.00515
H   -0.06706 -0.17283  0.60886
S   -1.44609 -0.05579 -0.01598
H   -1.39937  1.28148  0.07662
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End
      Frequencies[1/cm]      5
      2727.2   1264.2   947.82   424.63   248.54
ZeroEnergy[kcal/mol]  -1.9223692408117470
ElectronicLevels[1/cm]      1
      0.0000000000000000   2.0000000000000000
End
!*****
RRHO      !      106
Geometry[angstrom]      4
Cl  1.36428  0.01902 -0.00499
H   -0.05180 -0.17160  0.60478
S   -1.44642 -0.05578 -0.01602
H   -1.39920  1.28150  0.07652
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End
      Frequencies[1/cm]      5
      2726.9   1232.5   930.41   424.04   250.93
ZeroEnergy[kcal/mol]  -2.1221223636140656
ElectronicLevels[1/cm]      1
      0.0000000000000000   2.0000000000000000
End
!*****
RRHO      !      107
Geometry[angstrom]      4
Cl  1.36415  0.01898 -0.00484
H   -0.03646 -0.17044  0.60103
S   -1.44676 -0.05578 -0.01606
H   -1.39902  1.28152  0.07644
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End
      Frequencies[1/cm]      5
      2726.6   1204.1   912.74   423.02   252.55
ZeroEnergy[kcal/mol]  -2.3381561325375895
ElectronicLevels[1/cm]      1
      0.0000000000000000   2.0000000000000000
End
!*****
RRHO      !      108
Geometry[angstrom]      4
Cl  1.36403  0.01895 -0.00469
H   -0.02104 -0.16934  0.59760
S   -1.44712 -0.05578 -0.01611
H   -1.39884  1.28153  0.07636
Core RigidRotor
      SymmetryFactor  0.5000000000000000
End

```

```

Frequencies[1/cm]          5
2726.4    1179.1    894.84    421.60    253.56
ZeroEnergy[kcal/mol]      -2.5886382283970186
ElectronicLevels[1/cm]          1
0.0000000000000000    2.0000000000000000

```

End

!\*\*\*\*\*

```

RRHO      !          109
Geometry[angstrom]      4
Cl  1.36393  0.01891 -0.00455
H   -0.00556 -0.16831  0.59449
S   -1.44751 -0.05578 -0.01617
H   -1.39864  1.28154  0.07628

Core RigidRotor
      SymmetryFactor  0.5000000000000000

```

End

```

Frequencies[1/cm]          5
2726.2    1158.1    876.80    419.86    254.22
ZeroEnergy[kcal/mol]      -2.8595888902462985
ElectronicLevels[1/cm]          1
0.0000000000000000    2.0000000000000000

```

End

!\*\*\*\*\*

```

RRHO      !          110
Geometry[angstrom]      4
Cl  1.36384  0.01888 -0.00441
H    0.00998 -0.16735  0.59173
S   -1.44791 -0.05577 -0.01623
H   -1.39844  1.28154  0.07621

Core RigidRotor
      SymmetryFactor  0.5000000000000000

```

End

```

Frequencies[1/cm]          5
2726.0    1142.0    858.69    417.86    254.79
ZeroEnergy[kcal/mol]      -3.1430175373966329
ElectronicLevels[1/cm]          1
0.0000000000000000    2.0000000000000000

```

End

!\*\*\*\*\*

```

! Tunneling Eckart
! ImaginaryFrequency[1/cm]  1076.56
! WellDepth[kcal/mol]      7.35
! WellDepth[kcal/mol]      12.65
! End

```

```

      Tunneling Read
CutoffEnergy[1/cm]        3500
ImaginaryFrequency[1/cm]  1076.5599999999999
File imactint.dat
End

```

!\*\*\*\*\*

End

End

## VRC-TST reactive flux: flux\_lr\_so9.dat

0	7388.88
13.9008	16396
28.4966	30980.7
43.8222	52764.1
59.9141	83468.9
76.8106	125276
94.552	180699
113.18	252106
132.74	342081
153.278	452744
174.843	580815
197.486	725672
221.261	889861
246.224	1.08211e+06
272.436	1.30653e+06
299.959	1.56775e+06
328.858	1.87055e+06
359.201	2.21851e+06
391.062	2.62067e+06
424.516	3.08577e+06
459.643	3.62356e+06
496.526	4.2451e+06
535.253	4.96392e+06
575.916	5.79245e+06
618.613	6.7453e+06
663.444	7.84349e+06
710.517	9.08811e+06
759.944	1.05128e+07
811.842	1.21388e+07
866.335	1.3987e+07
923.552	1.60929e+07
983.631	1.84841e+07
1046.71	2.12299e+07
1112.95	2.43237e+07
1182.5	2.78659e+07
1255.52	3.14448e+07
1332.2	3.48072e+07
1412.71	3.8266e+07
1497.25	4.19167e+07
1586.01	4.56694e+07
1679.21	4.96416e+07
1777.07	5.3989e+07
1879.83	5.86709e+07
1987.72	6.37684e+07
2101.01	6.9319e+07
2219.96	7.53693e+07
2344.86	8.20702e+07
2476	8.95202e+07
2613.7	9.78191e+07

2758.29	1.07099e+08
2910.1	1.17503e+08
3069.51	1.29187e+08
3236.88	1.42265e+08
3412.63	1.56895e+08
3597.16	1.73316e+08
3790.92	1.91763e+08
3994.37	2.12575e+08
4207.99	2.36086e+08
4432.29	2.62665e+08
4667.8	2.92709e+08
4915.09	3.26624e+08
5174.75	3.6492e+08
5447.39	4.08341e+08
5733.66	4.57709e+08
6034.24	5.13803e+08
6349.85	5.77476e+08
6681.25	6.49699e+08
7029.21	7.31552e+08
7394.57	8.24541e+08
7778.2	9.30513e+08
8181.01	1.05119e+09
8603.96	1.18833e+09
9048.06	1.34386e+09
9514.36	1.52014e+09
10004	1.72116e+09
10518.1	1.94994e+09
11057.9	2.20985e+09
11624.7	2.50463e+09
12219.8	2.83886e+09
12844.7	3.21886e+09
13500.8	3.65113e+09
14189.8	4.1418e+09
14913.2	4.69888e+09
15672.7	5.32929e+09
16470.3	6.04446e+09
17307.7	6.85799e+09
18187	7.78227e+09
19110.2	8.82912e+09
20079.6	1.00109e+10
21097.5	1.13532e+10
22166.3	1.28807e+10
23288.5	1.4614e+10
24466.8	1.6568e+10
25704.1	1.87764e+10
27003.2	2.1293e+10
28367.2	2.41533e+10
29799.5	2.73799e+10
31303.4	3.10112e+10
32882.5	3.51457e+10
34540.5	3.98525e+10
36281.4	4.51663e+10
38109.4	5.11387e+10
40028.7	5.79183e+10
42044.1	6.56466e+10
44160.2	7.43746e+10

46382.1	8.41946e+10
48715.1	9.5299e+10
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