Supporting information: Development of The ReaxFF Reactive Force Field for Inherent Point Defects in the Si/Silica system

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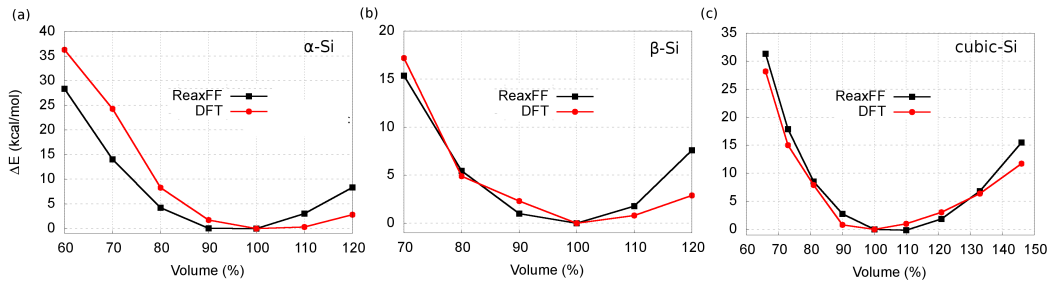

Figure S1: Equations of states of (a) alpha, (b) beta and (c) cubic phases of silicon predicted by QM and ReaxFF.

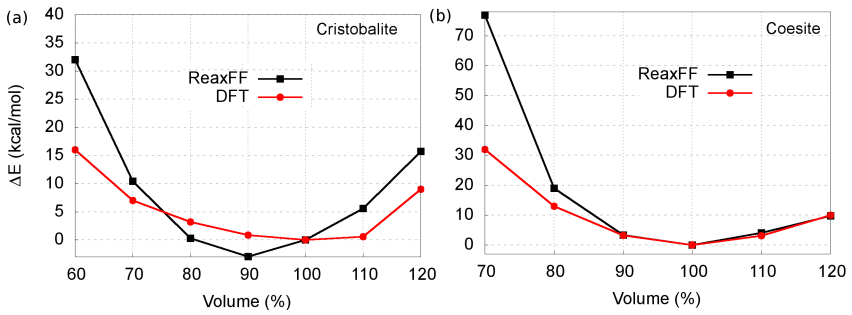

Figure S2: Equations of states of (a) cristobalite and (b) coesite phases of silica predicted by QM and ReaxFF.

Table S1: $ReaxFF_{present}$ **and QM[1] energies (in kcal/mol) of Si-O-H reactions.**

Reaction	$ReaxFF_{present}$	DFT
$Si(OH)_4 + Si(OH)_3O^- \rightarrow (OH)_3Si-O-Si(OH)_2O$ $+H_2O(dimeranion) + H_2O$	-24.4	-20.8
$(OH)_3Si-O-Si(OH)_2O + Si(OH)_4$ $\rightarrow (OH)_3Si-O-SiOH_2-O-SiOH_2O^- + H_2O$ $(trimeranion + H_2O)$	-18.6	-14.3
$(OH)_3Si-O-Si(OH)_2-O-Si(OH)_2O + Si(OH)_4$ $\rightarrow branchedquadrimeranion + H_2O$	-25.2	-28.9

ReaxFF Reactive Force Field Parameters for Si/O/H developed in the present work

The force field developed in this work can be used with the standalone ReaxFF program, as well as with the LAMMPS open-source MD-program and the ADF-program.

```
Reactive MD-force field:  Si/O/H force field 2019
 39        !  Number of general parameters
   50.0000 !Overcoordination parameter
    9.5469 !Overcoordination parameter
   26.5405 !Valency angle conjugation parameter
    1.7224 !Triple bond stabilisation parameter
    6.8702 !Triple bond stabilisation parameter
   12.1176 !Undercoordination parameter
   13.3056 !Undercoordination parameter
  -70.5044 !Triple bond stabilization energy
    0.0000 !Lower Taper-radius
   10.0000 !Upper Taper-radius
    2.8793 !Not used
   33.8667 !Valency undercoordination
    6.0891 !Valency angle/lone pair parameter
    1.0563 !Valency angle
    2.0384 !Valency angle parameter
    6.1431 !Not used
    6.9290 !Double bond/angle parameter
    0.3989 !Double bond/angle parameter:  overcoord
    3.9954 !Double bond/angle parameter:  overcoord
   -2.4837 !Not used
    5.7796 !Torsion/BO parameter
   10.0000 !Torsion overcoordination
    1.9487 !Torsion overcoordination
   -1.2327 !Conjugation 0 (not used)
    2.1645 !Conjugation 1.5591 !vdWaals shielding
    0.1000 !Cutoff for bond order (*100)
    2.1365 !Valency angle conjugation parameter
    0.6991 !Overcoordination parameter
   50.0000 !Overcoordination parameter
    1.8512 !Valency/lone pair parameter
    0.5000 !Not used 20.0000 !Not used
    5.0000 !Molecular energy (not used)
    0.0000 !Molecular energy (not used)
    2.6962 !Valency angle conjugation parameter
```

```
4        !  Nr of atoms; cov.r; valency;a.m;Rvdw;Evdw;gammaEEM;cov.r2;#
            alfa;gammavdW;valency;Eunder;Eover;chiEEM;etaEEM;n.u.
            cov r3;Elp;Heat inc.;n.u.;n.u.;n.u.;n.u.  ov/un;val1;n.u.;val3,vval4
H    0.8930   1.0000   1.0080   1.3550   0.0930   0.8203  -0.1000   1.0000
     8.2230  33.2894   1.0000   0.0000 121.1250   3.7248   9.6093   1.0000
    -0.1000   0.0000  61.6606   3.0408   2.4197   0.0003   1.0698   0.0000
   -19.4571   4.2733   1.0338   1.0000   2.8793   0.0000   0.0000   0.0000
O    1.2450   2.0000  15.9990   2.3890   0.1000   1.0898   1.0548   6.0000
     9.7300  13.8449   4.0000  37.5000 116.0768   8.5000   8.3122   2.0000
     0.9049   0.4056  59.0626   3.5027   0.7640   0.0021   0.9745   0.0000
    -3.5500   2.9000   1.0493   4.0000   2.9225   0.0000   0.0000   0.0000
Si   2.0175   4.0000  28.0600   2.0473   0.1835   0.8925   1.2962   4.0000
    12.3588   1.2523   4.0000  21.7115 139.9309   4.6988   6.0000   0.0000
    -1.0000   0.0000 128.2031   8.7895  23.9298   0.8381   0.8563   0.0000
    -4.7525   2.1607   1.0338   4.0000   2.5791   0.0000   0.0000   0.0000
X   -0.1000   2.0000   1.0080   2.0000   0.0000   0.0100  -0.1000   6.0000
    10.0000   2.5000   4.0000   0.0000   0.0000   5.00009999.9999   0.0000
    -0.1000   0.0000  -2.3700   8.7410  13.3640   0.6690   0.9745   0.0000
   -11.0000   2.7466   1.0338   2.0000   2.8793   0.0000   0.0000   0.0000
6    !  Nr of bonds; Edis1;LPpen;n.u.;pbe1;pbo5;13corr;pbo6
                     pbe2;pbo3;pbo4;n.u.;pbo1;pbo2;ovcorr
 1  1 153.3934   0.0000   0.0000  -0.4600   0.0000   1.0000   6.0000   0.7300
       6.2500   1.0000   0.0000   1.0000  -0.0790   6.0552   0.0000   0.0000
 1  2 160.0000   0.0000   0.0000  -0.5725   0.0000   1.0000   6.0000   0.5626
       1.1150   1.0000   0.0000   0.0000  -0.0920   4.2790   0.0000   0.0000
 2  2 142.2858 145.0000  50.8293   0.2506  -0.1000   1.0000  29.7503   0.6051
       0.3451  -0.1055   9.0000   1.0000  -0.1225   5.5000   1.0000   0.0000
 1  3 250.0000   0.0000   0.0000  -0.7128   0.0000   1.0000   6.0000   0.1186
      18.5790   1.0000   0.0000   1.0000  -0.0731   7.4983   0.0000   0.0000
 2  3 291.4687  16.2724   0.0000  -0.6232  -0.3000   1.0000  36.0000   0.6762
       9.6502  -0.4634  29.9983   1.0000  -0.1500   7.6885   1.0000   0.0000
 3  3  78.6948  12.3833  30.0000   0.2706  -0.3000   1.0000  16.0000   0.0100
       0.7257  -0.9788   7.8412   1.0000  -0.0824   8.5016   0.0000   0.0000
```

```
 3     !  Nr of off-diagonal terms; Ediss;Ro;gamma;rsigma;rpi;rpi2
  1  2   0.0283   1.2885  10.9190   0.9215  -1.0000  -1.0000
  1  3   0.1659   1.4000  11.7054   1.3437  -1.0000  -1.0000
  2  3   0.1556   2.1618  10.4851   1.7448   1.3513  -1.0000
 18     !  Nr of angles;at1;at2;at3;Thetao,o;ka;kb;pv1;pv2
  1  1  1   0.0000   27.9213   5.863   0.0000   0.0000   0.0000   1.0400
  1  1  2   0.0000   8.5744   3.0000   0.0000   0.0000   0.0000   1.0421
  2  1  2   0.0000  15.0000   2.8900   0.0000   0.0000   0.0000   2.8774
  1  2  1  85.8000   9.8453   2.2720   0.0000   2.8635   0.0000   1.5800
  1  2  2  75.6935  50.0000   2.0000   0.0000   1.0000   0.0000   1.1680
  2  2  2  80.7324  30.4554   0.9953   0.0000   1.6310  50.0000   1.0783
  3  3  3  71.0490  32.4076   1.2648   0.0000   0.0133   0.0000   1.2899
  1  3  3  79.0296   4.8472   8.0000   0.0000   4.0000   0.0000   1.0400
  1  3  1  76.7122  31.0021   2.4370   0.0000   0.1930   0.0000   1.0400
  2  3  3  97.1370  39.9525   1.0978   0.0000   0.5073   0.0000   1.3154
  1  3  2  77.4530  40.0000   1.1121   0.0000   4.0000   0.0000   1.1201
  2  3  2  95.2092  40.0000   1.1635   0.0000   0.3525   0.0000   3.0000
  3  2  3  47.4477   8.8967   0.5000   0.0000   4.0000   0.0000   1.0400
  1  2  3  80.0391   4.5313   8.0000   0.0000   3.3561   0.0000   1.8653
  2  2  3  20.0000   8.1399   2.7859   0.0000   3.0206   0.0000   1.6650
  1  1  3   0.0000  72.1101   7.6682   0.0000   2.7489   0.0000   1.0400
  3  1  3   0.0000  59.2363   2.3343   0.0000   1.3778   0.0000   2.3874
  2  1  3   0.0000  15.0000   2.8577   0.0000   4.0000   0.0000   1.1566
 8 !  Nr of torsions;at1;at2;at3;at4;;V1;V2;V3;V2(BO);vconj;n.u;n
  1  2  2  1   2.5000  -4.0000   0.9000  -2.5000  -1.0000   0.0000   0.0000
  1  2  2  2   0.8302  -4.0000  -0.7763  -2.5000  -1.0000   0.0000   0.0000
  2  2  2  2  -2.5000  -4.0000   1.0000  -2.5000  -1.0000   0.0000   0.0000
  0  1  2  0   0.0000   0.1000   0.0200  -2.5415   0.0000   0.0000   0.0000
  0  2  2  0   0.5511  25.4150   1.1330  -5.1903  -1.0000   0.0000   0.0000
  1  3  3  1   0.0000   0.0000   0.0640  -2.4426   0.0000   0.0000   0.0000
  1  3  3  3   0.0000   0.0000   0.1587  -2.4426   0.0000   0.0000   0.0000
  0  1  3  0   0.0000   0.0000   0.1200  -2.4847   0.0000   0.0000   0.0000
 1         !  Nr of hydrogen bonds;at1;at2;at3;Rhb;Dehb;vhb1
  2  1  2   2.1200  -3.5800   1.4500  19.5000
```

ReaxFF Reactive Force Field Parameters for Si/O/H developed by Fogarty et al.[2]

```
Reactive MD-force field:  Si/O/H force field 2010
 39        !  Number of general parameters
   50.0000 !Overcoordination parameter
    9.5469 !Overcoordination parameter
   26.5405 !Valency angle conjugation parameter
    1.7224 !Triple bond stabilisation parameter
    6.8702 !Triple bond stabilisation parameter
   12.1176 !Undercoordination parameter
   13.3056 !Undercoordination parameter
  -70.5044 !Triple bond stabilization energy
    0.0000 !Lower Taper-radius
   10.0000 !Upper Taper-radius
    2.8793 !Not used
   33.8667 !Valency undercoordination
    6.0891 !Valency angle/lone pair parameter
    1.0563 !Valency angle
    2.0384 !Valency angle parameter
    6.1431 !Not used
    6.9290 !Double bond/angle parameter
    0.3989 !Double bond/angle parameter:  overcoord
    3.9954 !Double bond/angle parameter:  overcoord
   -2.4837 !Not used
    5.7796 !Torsion/BO parameter
   10.0000 !Torsion overcoordination
    1.9487 !Torsion overcoordination
   -1.2327 !Conjugation 0 (not used)
    2.1645 !Conjugation 1.5591 !vdWaals shielding
    0.1000 !Cutoff for bond order (*100)
    2.1365 !Valency angle conjugation parameter
    0.6991 !Overcoordination parameter
   50.0000 !Overcoordination parameter
    1.8512 !Valency/lone pair parameter
    0.5000 !Not used 20.0000 !Not used
    5.0000 !Molecular energy (not used)
    0.0000 !Molecular energy (not used)
    2.6962 !Valency angle conjugation parameter
```

```
4        !  Nr of atoms; cov.r; valency;a.m;Rvdw;Evdw;gammaEEM;cov.r2;#
            alfa;gammavdW;valency;Eunder;Eover;chiEEM;etaEEM;n.u.
            cov r3;Elp;Heat inc.;n.u.;n.u.;n.u.;n.u.  ov/un;val1;n.u.;val3,vval4
H    0.8930   1.0000   1.0080   1.3550   0.0930   0.8203  -0.1000   1.0000
     8.2230  33.2894   1.0000   0.0000 121.1250   3.7248   9.6093   1.0000
    -0.1000   0.0000  61.6606   3.0408   2.4197   0.0003   1.0698   0.0000
   -19.4571   4.2733   1.0338   1.0000   2.8793   0.0000   0.0000   0.0000
O    1.2450   2.0000  15.9990   2.3890   0.1000   1.0898   1.0548   6.0000
     9.7300  13.8449   4.0000  37.5000 116.0768   8.5000   8.3122   2.0000
     0.9049   0.4056  59.0626   3.5027   0.7640   0.0021   0.9745   0.0000
    -3.5500   2.9000   1.0493   4.0000   2.9225   0.0000   0.0000   0.0000
Si   2.0175   4.0000  28.0600   2.0473   0.1835   0.8925   1.2962   4.0000
    12.3588   1.2523   4.0000  21.7115 139.9309   4.6988   6.0000   0.0000
    -1.0000   0.0000 128.2031   8.7895  23.9298   0.8381   0.8563   0.0000
    -4.7525   2.1607   1.0338   4.0000   2.5791   0.0000   0.0000   0.0000
X   -0.1000   2.0000   1.0080   2.0000   0.0000   0.0100  -0.1000   6.0000
    10.0000   2.5000   4.0000   0.0000   0.0000   5.00009999.9999   0.0000
    -0.1000   0.0000  -2.3700   8.7410  13.3640   0.6690   0.9745   0.0000
   -11.0000   2.7466   1.0338   2.0000   2.8793   0.0000   0.0000   0.0000
6    !  Nr of bonds; Edis1;LPpen;n.u.;pbe1;pbo5;13corr;pbo6
                     pbe2;pbo3;pbo4;n.u.;pbo1;pbo2;ovcorr
 1  1 153.3934   0.0000   0.0000  -0.4600   0.0000   1.0000   6.0000   0.7300
       6.2500   1.0000   0.0000   1.0000  -0.0790   6.0552   0.0000   0.0000
 1  2 160.0000   0.0000   0.0000  -0.5725   0.0000   1.0000   6.0000   0.5626
       1.1150   1.0000   0.0000   0.0000  -0.0920   4.2790   0.0000   0.0000
 2  2 142.2858 145.0000  50.8293   0.2506  -0.1000   1.0000  29.7503   0.6051
       0.3451  -0.1055   9.0000   1.0000  -0.1225   5.5000   1.0000   0.0000
 1  3 250.0000   0.0000   0.0000  -0.7128   0.0000   1.0000   6.0000   0.1186
      18.5790   1.0000   0.0000   1.0000  -0.0731   7.4983   0.0000   0.0000
 2  3 272.8709  18.4462   0.0000  -0.6107  -0.3000   1.0000  36.0000   0.8270
      10.2334  -0.5495  29.9954   1.0000  -0.1277   7.5863   1.0000   0.0000
 3  3  78.0276  54.0531  30.0000   0.5398  -0.3000   1.0000  16.0000   0.0476
       0.2865  -0.8055   7.1248   1.0000  -0.0681   8.6957   0.0000   0.0000
```

```
 3     !  Nr of off-diagonal terms; Ediss;Ro;gamma;rsigma;rpi;rpi2
  1  2   0.0283   1.2885  10.9190   0.9215  -1.0000  -1.0000
  1  3   0.1659   1.4000  11.7054   1.3437  -1.0000  -1.0000
  2  3   0.1330   2.0545  10.8315   1.7043   1.3773  -1.0000
 18     !  Nr of angles;at1;at2;at3;Thetao,o;ka;kb;pv1;pv2
  1  1  1   0.0000   27.9213   5.863   0.0000   0.0000   0.0000   1.0400
  1  1  2   0.0000   8.5744   3.0000   0.0000   0.0000   0.0000   1.0421
  2  1  2   0.0000  15.0000   2.8900   0.0000   0.0000   0.0000   2.8774
  1  2  1  85.8000   9.8453   2.2720   0.0000   2.8635   0.0000   1.5800
  1  2  2  75.6935  50.0000   2.0000   0.0000   1.0000   0.0000   1.1680
  2  2  2  80.7324  30.4554   0.9953   0.0000   1.6310  50.0000   1.0783
  3  3  3  71.0490  32.4076   1.2648   0.0000   0.0133   0.0000   1.2899
  1  3  3  77.2616   5.0190   7.8944   0.0000   4.0000   0.0000   1.0400
  1  3  1  75.7983  14.4132   2.8640   0.0000   4.0000   0.0000   1.0400
  2  3  3  99.8997  26.6610   2.1237   0.0000   0.0100   0.0000   1.4341
  1  3  2  73.6998  40.0000   1.8782   0.0000   4.0000   0.0000   1.1290
  2  3  2  98.2184  38.9429   0.7727   0.0000   1.1658   0.0000   2.2641
  3  2  3  39.2858   1.3068   5.6478   0.0000   3.8972   0.0000   3.0000
  1  2  3  79.2126   4.8973   8.0000   0.0000   1.0859   0.0000   2.1209
  2  2  3  82.7397  32.1198   1.8862   0.0000   0.1058   0.0000   1.5443
  1  1  3   0.0000  47.1300   6.0000   0.0000   1.6371   0.0000   1.0400
  3  1  3   0.0000  27.4206   6.0000   0.0000   1.6371   0.0000   1.0400
  2  1  3   0.0000   7.0550   3.9236   0.0000   1.6371   0.0000   1.0400
 8 !  Nr of torsions;at1;at2;at3;at4;;V1;V2;V3;V2(BO);vconj;n.u;n
  1  2  2  1   2.5000  -4.0000   0.9000  -2.5000  -1.0000   0.0000   0.0000
  1  2  2  2   0.8302  -4.0000  -0.7763  -2.5000  -1.0000   0.0000   0.0000
  2  2  2  2  -2.5000  -4.0000   1.0000  -2.5000  -1.0000   0.0000   0.0000
  0  1  2  0   0.0000   0.1000   0.0200  -2.5415   0.0000   0.0000   0.0000
  0  2  2  0   0.5511  25.4150   1.1330  -5.1903  -1.0000   0.0000   0.0000
  1  3  3  1   0.0000   0.0000   0.0640  -2.4426   0.0000   0.0000   0.0000
  1  3  3  3   0.0000   0.0000   0.1587  -2.4426   0.0000   0.0000   0.0000
  0  1  3  0   0.0000   0.0000   0.1200  -2.4847   0.0000   0.0000   0.0000
 1         !  Nr of hydrogen bonds;at1;at2;at3;Rhb;Dehb;vhb1
  2  1  2   2.1200  -3.5800   1.4500  19.5000
```

References

(1) Trinh, T. T.; Jansen, A. P. J.; van Santen, R. A. Mechanism of Oligomerization Reactions of Silica. *The Journal of Physical Chemistry B* **2006**, *110*, 23099–23106, PMID: 17107150.

(2) Fogarty, J. C.; Aktulga, H. M.; Grama, A. Y.; van Duin, A. C. T.; Pandit, S. A. A reactive molecular dynamics simulation of the silica-water interface. *The Journal of Chemical Physics* **2010**, *132*, 174704.