Mechanical Size Effects of Amorphous Polymer Derived Ceramics at Nanoscale: Experiments and ReaxFF Simulations

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ReaxFF reactive force field

Force Field Parameter Development

ReaxFF is a bond-order dependent potential [1, 2], where the connectivity between all atom pairs is modified depending on the local atomic environment during the molecular dynamics (MD) simulation. ReaxFF has shown applicability in various research areas ranging from chemistry to mechanics of materials [3-5]. This feature allows ReaxFF to reliably describe the chemical reaction process as well as the energetics involved. In this perspective, ReaxFF is a prominent tool to simulate large systems that are not often studied by quantum mechanical (QM) methods at a comparable computational cost of nonreactive potentials. The total energy of the system in ReaxFF generally consists of contributions from bond-order dependent terms and non-bonded interaction terms. Energy terms such as a bond, valence angle, and torsion angle contribute to the bond-order dependent terms, whereas van der Waals and Coulomb energies are the partial contributions of the non-bonded interactions. Bond order is calculated and modified based on the interatomic distances of all atom pairs at every iteration. Hence, the energy contributions from bond-order dependent terms vanish upon bond dissociation, while leaving only the non-bonded interactions to be considered afterward. For both types of non-bonded interactions, ReaxFF employs a shielded potential to avoid excessive repulsions and unphysical charge distributions at short distances. The electronegativity equalization method

(EEM) [6] is used to derive the atomic charges. Further details of the ReaxFF potential forms can be found in publications by van Duin and Chenoweth.[1, 7]

In general, ReaxFF reactive force field parameters undergo an optimization process prior to performing ReaxFF MD simulations for accurate investigation of the target system. Typically, QM calculations and experimental values comprise a training set and force field parameter optimizations are carried out against these dataset to minimize the total error using a successive one-parameter search technique based on parabolic extrapolation method.[8] For the current investigation, we developed a new Si/O/C/H/N ReaxFF potential by strategically re-optimizing the CHON-2017_weak force field [9] with two existing trainsets, Pitman et al. [10] for Si/O/H interactions and Newsome et al.[11] for C/O/Si interactions, to adequately address the stress analysis of silicon oxycarbide (SiOC) ceramic fibers and assess the temperature effects. As previously mentioned, the basis of our new force field is from parameters of CHON-2017_weak. While this force field has successfully demonstrated its capability in reproducing the properties of the functionalized hydrocarbon system and the hydrocarbon-water mixtures, it does not include Si parameters and consequently, lack the applicability when it comes to describing any system with Si component. For this reason, we first incorporated the Si related parameters from literatures and then reoptimized them, since the significant parameter correlation in ReaxFF makes it necessary as the C, H and O element parameter changes made in the new generation C/H/O/N ReaxFF force field (CHON_2017_weak) affect the bond order of the atom pairs. Here, Si/O/H interactions such as equations of state of bulk-Si and different SiO_2 polymorphs, binding and dissociation of a single water molecule from a $Si(OH)_4$ molecule were taken into account according to the Pitman et al. training set. Meanwhile, C/O/Si interactions such as a Si-C bond, combinations of all C/O/Si angles, and the equation of state of the SiC crystals were also included for retraining the force field parameters to ensure proper description of the dissociation and bending energies. Table S1 provides a comprehensive

outlook of the list of parameters that had been optimized in this study. Energy profile comparisons of QM calculations and ReaxFF are shown in the Supporting Information (Figure S1), and the good agreement of these results suggest that we obtained highly transferrable force field that could provide reasonable simulation results for our system of interest as well as SiO_2/water or SiC/O_2/water systems which were initially the target system for the trainset used in this work.

Table S1. List of reoptimized parameters for the new ReaxFF Si/O/C/H/N potential

Element parameters

Atom	Reoptimized from
Si	Pitman

Off-diagonal parameters

Terms	Reoptimized from
C-Si	Newsome
H-Si	Pitman
O-Si	Pitman

Bond parameters

Bond	Reoptimized from
C-Si	Newsome
H-Si	Pitman
O-Si	Pitman
Si-Si	Pitman

Angle parameters

Angle	Reoptimized from	Angle	Reoptimized from
O-Si-Si	Pitman	C-C-Si	Newsome
O-Si-O	Pitman	C-Si-C	Newsome
Si-O-Si	Pitman	Si-C-Si	Newsome
H-O-Si	Pitman	C-Si-Si	Newsome
O-O-Si	Pitman	H-C-Si	Newsome
		C-Si-H	Newsome
		C-O-Si	Newsome
		C-Si-O	Newsome
		O-C-Si	Newsome

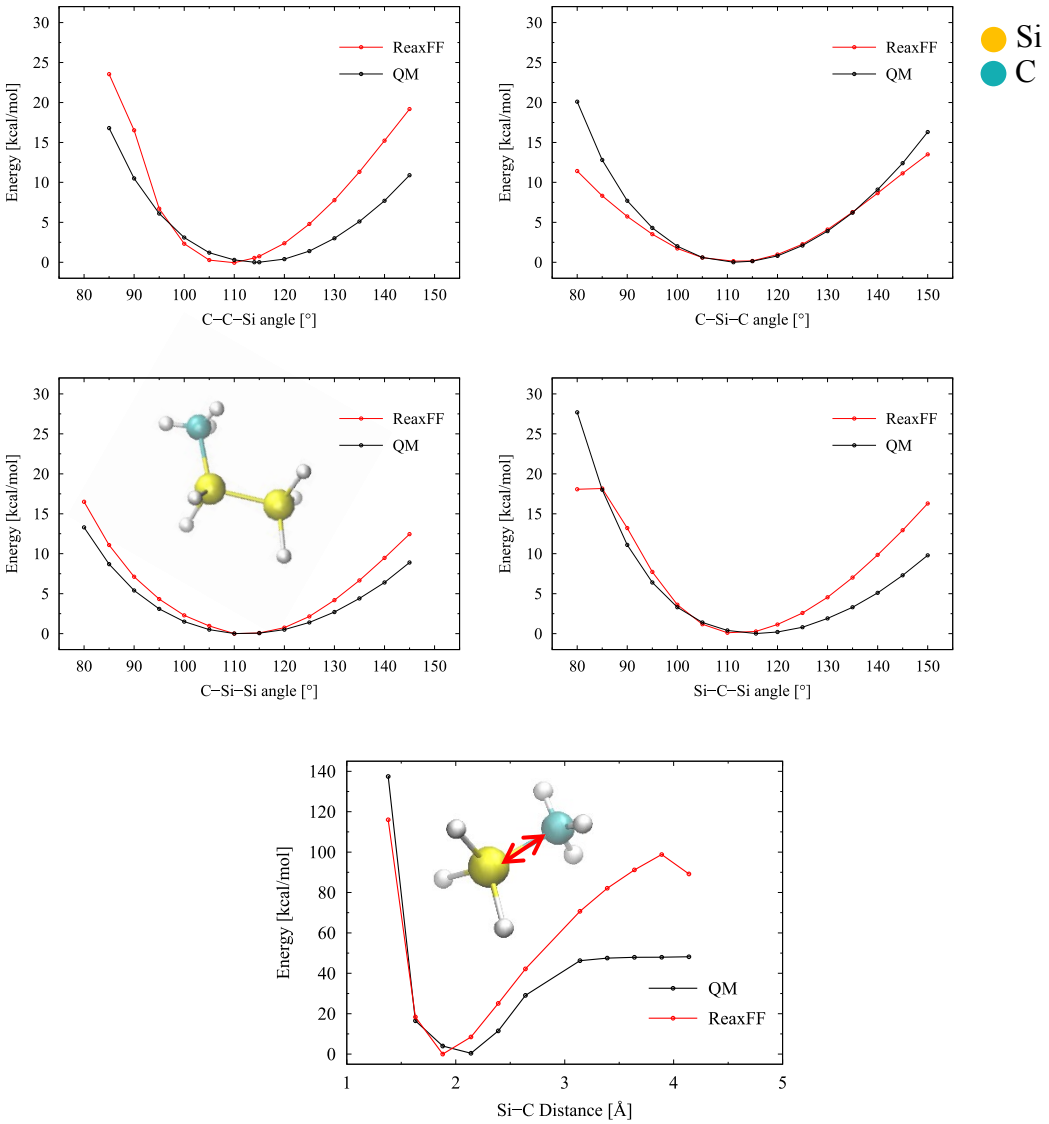

Figure S1: Quantum mechanics and ReaxFF energy comparisons of bond dissociation and bending energy

ReaxFF reactive force field parameters for Si/O/C/H/N

```
Reactive MD-force field: Si/O/C/H/N_2018
 39       ! Number of general parameters
   50.0000 !Overcoordination parameter
    9.5469 !Overcoordination parameter
    1.6725 !Valency angle conjugation parameter
    1.7224 !Triple bond stabilisation parameter
    6.8702 !Triple bond stabilisation parameter
   60.4850 !C2-correction
    1.0588 !Undercoordination parameter
    4.6000 !Triple bond stabilisation parameter
   12.1176 !Undercoordination parameter
   13.3056 !Undercoordination parameter
  -20.0000 !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)
    1.7602 !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)
    0.7903 !Valency angle conjugation parameter
 12    ! 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
 C     1.3727    4.0000   12.0000    2.0270    0.1113    0.5516    1.1706    4.0000
```

```
      9.2293    4.5389    4.0000   30.0000   79.5548    4.4087    7.0601    0.0000
      1.1168    0.0000  181.0000   14.5210   24.9431    6.7313    0.8563    0.0000
     -6.7437    5.6329    1.0564    4.0000    2.9663    0.0000    0.0000    0.0000
H     0.8924    1.0000    1.0080    1.6791    0.0709    0.7390   -0.1000    1.0000
      8.3519   39.1732    1.0000    0.0000  121.1250    3.5442    9.3848    1.0000
     -0.1000    0.0000   61.6606    2.8222    2.1441    0.0003    1.0698    0.0000
    -18.1423    5.3143    1.0338    1.0000    2.8793    0.0000    0.0000    0.0000
O     1.2450    2.0000   15.9990    2.3396    0.1000    1.1000    1.0548    6.0000
      9.3187   12.5083    4.0000   37.5000  116.0768    8.5000    8.4783    2.0000
      0.9049    0.1000   59.0626    3.4340    0.7722    0.0021    0.9745    0.0000
     -3.5352    3.2703    1.0493    4.0000    2.9225    0.0000    0.0000    0.0000
N     1.2333    3.0000   14.0000    1.9690    0.1513    1.0000    1.1748    5.0000
      9.6326    9.1048    4.0000   32.4838  100.0000    6.6335    7.1473    2.0000
      1.0433    2.3175  119.9837    0.4648    9.7074    2.2521    0.9745    0.0000
     -7.0000    4.0000    1.0183    4.0000    2.8793    0.0000    0.0000    0.0000
S     1.9673    2.0000   32.0600    2.1729    0.3000    1.0336    1.5359    6.0000
     10.3008    4.9055    4.0000   52.9998  112.1416    6.5000    8.2545    2.0000
      1.4601    9.7177   71.1843    5.7487   23.2859   12.7147    0.9745    0.0000
    -11.0000    2.7466    1.0338    6.2998    2.8793    0.0000    0.0000    0.0000
Mg    1.8315    2.0000   24.3050    2.2464    0.1806    0.5020    1.0000    2.0000
     10.9186   27.1205    3.0000   38.0000    0.0000    0.9499    5.6130    0.0000
     -1.3000    0.0000  220.0000   49.9248    0.3370    0.0000    0.0000    0.0000
     -1.0823    2.3663    1.0564    6.0000    2.9663    0.0000    0.0000    0.0000
P     1.5994    3.0000   30.9738    1.7000    0.1743    1.0000    1.3000    5.0000
      9.1909   12.0377    5.0000    0.0000    0.0000    1.7681    6.9748    0.0000
     -1.0000   13.1397  125.6300    0.2526   16.9194   24.7770    0.0000    0.0000
     -9.7896    3.3929    1.0338    5.0000    2.8793    0.0000    0.0000    0.0000
Na    2.0300    1.0000   22.9898    2.3334    0.1481    0.8765   -1.0000    1.0000
     11.0000    9.8000    1.0000    0.0000    0.0000   -3.8501    5.9459    0.0000
     -1.0000    0.0000   67.5458  100.0000   10.0000    0.2500    0.8563    0.0000
     -2.5766    2.5000    1.0338    6.0000    2.5791    0.0000    0.0000    0.0000
Cu    1.9202    2.0000   63.5460    1.9221    0.2826    1.0000    0.1000    1.0000
     10.9889  100.0000    1.0000    0.0000    0.0000    2.7875    6.0000    0.0000
     -1.0000    0.0000   80.7000   34.9555    0.4988    0.0000    0.8563    0.0000
     -5.1872    3.1491    1.0000    4.0000    2.5791    0.0000    0.0000    0.0000
Cl    1.7140    1.0000   35.4500    1.9139    0.2000    0.3837   -1.0000    7.0000
     11.5345   10.1330    1.0000    0.0000    0.0000    9.9614    6.5316    0.0000
     -1.0000    3.5750  143.1770    6.2293    5.2294    0.1542    0.8563    0.0000
    -10.2080    2.9867    1.0338    6.2998    2.5791    0.0000    0.0000    0.0000
X    -0.0998    2.0000    1.0080    2.0000    0.0000    1.0000   -0.1000    6.0000
     10.0000    2.5000    4.0000    0.0000    0.0000    8.5000    1.5000    0.0000
     -0.1000    0.0000   -2.3700    8.7410   13.3640    0.6690    0.9745    0.0000
    -11.0000    2.7466    1.0338    4.0000    2.8793    0.0000    0.0000    0.0000
Si    2.1932    4.0000   28.0600    1.8951    0.1737    0.8112    1.2962    4.0000
     11.3429    5.2054    4.0000   21.7115  139.9309    4.0081    5.7104    0.0000
     -1.0000    0.0000  128.2031    9.0751   23.8188    0.8381    0.8563    0.0000
     -4.1684    2.0754    1.0338    4.0000    2.5791    1.4000    0.2000   13.0000
48       ! Nr of bonds; Edis1;LPpen;n.u.;pbe1;pbo5;13corr;pbo6
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pbe2;pbo3;pbo4;Etrip;pbo1;pbo2;ovcorr

1	1	91.8588	120.2415	50.6788	0.3723	-0.1000	1.0000	34.6637	0.7015
		6.8558	-0.1605	7.9139	1.0000	-0.0719	6.9788	1.0000	0.0000
1	2	185.0817	0.0000	0.0000	-0.4949	0.0000	1.0000	6.0000	0.6864
		8.2839	1.0000	0.0000	1.0000	-0.0847	6.4150	0.0000	0.0000
2	2	165.6186	0.0000	0.0000	-0.4194	0.0000	1.0000	6.0000	0.6197
		6.3245	1.0000	0.0000	1.0000	-0.0879	6.0634	0.0000	0.0000
1	3	148.9918	115.2723	93.5449	-0.6834	-0.3639	1.0000	18.9178	0.6790
		0.4084	-0.4385	7.7418	1.0000	-0.1798	4.9342	0.0000	0.0000
2	3	159.4722	0.0000	0.0000	-0.5725	0.0000	1.0000	6.0000	0.5505
		1.1150	1.0000	0.0000	0.0000	-0.0920	4.2790	0.0000	0.0000
3	3	142.2858	85.0000	0.0000	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	4	186.9647	90.5539	69.0633	-1.7565	-0.1960	1.0000	28.5338	0.1967
		0.0446	-0.3644	6.5414	1.0000	-0.2803	4.6293	1.0000	0.0000
3	4	128.8596	167.8643	40.0000	0.3819	-0.1539	1.0000	34.9972	0.1900
		1.0110	-0.3716	7.0805	1.0000	-0.1265	6.8843	1.0000	0.0000
4	4	160.1592	82.5526	153.9884	0.4110	-0.0934	1.0000	12.4304	0.5899
		0.1538	-0.1473	11.9187	1.0000	-0.0753	5.4371	1.0000	0.0000
2	4	173.4757	0.0000	0.0000	-0.2904	0.0000	1.0000	6.0000	0.6709
		0.5347	1.0000	0.0000	1.0000	-0.1855	5.2649	0.0000	0.0000
1	5	150.8132	59.3363	55.2528	-0.0628	-0.5211	1.0000	18.9617	0.3219
		0.3317	-0.2289	7.5946	1.0000	-0.1946	5.9455	1.0000	0.0000
2	5	143.4377	0.0000	0.0000	-0.2944	0.0000	1.0000	6.0000	0.6034
		9.5627	1.0000	0.0000	1.0000	-0.0516	7.0960	1.0000	0.0000
3	5	0.0000	0.0000	0.0000	0.5563	-0.4038	1.0000	49.5611	0.6000
		0.4259	-0.4577	12.7569	1.0000	-0.1100	7.1145	1.0000	0.0000
4	5	0.0000	0.0000	0.0000	0.4438	-0.2034	1.0000	40.3399	0.6000
		0.3296	-0.3153	9.1227	1.0000	-0.1805	5.6864	1.0000	0.0000
5	5	140.8887	84.9350	68.6860	-0.4111	-0.4781	1.0000	17.8574	-0.1336
		0.2881	-0.2494	9.8436	1.0000	-0.1806	7.4732	1.0000	0.0000
2	6	58.6896	0.0000	0.0000	-0.0203	-0.1418	1.0000	13.1260	0.0230
		8.2136	-0.1310	0.0000	1.0000	-0.2692	6.4254	0.0000	24.4461
3	6	87.0227	0.0000	43.3991	0.0030	-0.3000	1.0000	36.0000	0.0250
		0.0087	-0.2500	12.0000	1.0000	-0.0439	6.6073	1.0000	24.4461
6	6	32.3808	0.0000	0.0000	-0.0076	-0.2000	0.0000	16.0000	0.2641
		4.8726	-0.2000	10.0000	1.0000	-0.0729	4.6319	0.0000	0.0000
1	7	0.0000	0.0000	0.0000	0.2171	-0.1418	1.0000	13.1260	0.6000
		0.3601	-0.2500	20.0000	1.0000	-0.2000	10.0000	1.0000	0.0000
2	7	0.0000	0.0000	0.0000	0.2250	-0.1418	1.0000	13.1260	0.6000
		0.3912	-0.1310	0.0000	1.0000	-0.2000	10.0000	0.0000	0.0000
3	7	250.0000	182.8454	0.0000	0.0607	-0.5000	1.0000	25.0000	0.4547
		0.7012	-0.2197	15.9047	1.0000	-0.2026	6.4270	1.0000	0.0000
4	7	130.0000	0.0000	0.0000	0.2171	-0.1418	1.0000	13.1260	0.6000
		0.3601	-0.1310	10.7257	1.0000	-0.0869	5.3302	1.0000	0.0000
6	7	0.1000	0.0000	0.0000	0.2500	-0.5000	1.0000	35.0000	0.6000
		0.5000	-0.5000	20.0000	1.0000	-0.2000	10.0000	1.0000	0.0000
7	7	0.0000	0.0000	0.0000	0.2171	-0.5000	1.0000	35.0000	0.6000

		0.5000	-0.5000	20.0000	1.0000	-0.2000	10.0000	1.0000	0.0000
2	8	0.0000	0.0000	0.0000	-1.0000	-0.3000	1.0000	36.0000	0.7000
		10.1151	-0.3500	25.0000	1.0000	-0.1053	8.2003	1.0000	0.0000
3	8	76.0753	0.0000	0.0000	-0.4452	-0.3000	1.0000	36.0000	0.6433
		5.6834	-0.3500	25.0000	1.0000	-0.0539	8.0273	1.0000	0.0000
4	8	0.0000	0.0000	0.0000	-1.0000	-0.3000	1.0000	36.0000	0.7000
		10.1151	-0.3500	25.0000	1.0000	-0.1053	8.2003	1.0000	0.0000
6	8	0.1000	0.0000	0.0000	0.2500	-0.5000	1.0000	35.0000	0.6000
		0.5000	-0.5000	20.0000	1.0000	-0.2000	10.0000	1.0000	0.0000
7	8	0.1000	0.0000	0.0000	0.2500	-0.5000	1.0000	35.0000	0.6000
		0.5000	-0.5000	20.0000	1.0000	-0.2000	10.0000	1.0000	0.0000
8	8	27.8052	0.0000	0.0000	0.4022	0.3000	0.0000	25.0000	0.4894
		0.6222	-0.4000	12.0000	1.0000	-0.0500	5.3362	0.0000	0.0000
8	12	0.0000	0.0000	0.0000	1.0000	0.3000	0.0000	26.0000	1.0000
		0.5000	0.0000	12.0000	1.0000	-0.2000	10.0000	0.0000	0.0000
4	6	0.0000	0.0000	0.0000	-1.0000	-0.3000	1.0000	36.0000	0.7000
		10.1151	-0.3500	25.0000	1.0000	-0.1053	8.2003	1.0000	0.0000
1	9	0.0000	0.0000	0.0000	0.2000	-0.1418	1.0000	13.1260	0.5000
		0.5000	-0.2000	20.0000	1.0000	-0.1000	9.0000	0.0000	0.0000
2	9	0.0000	0.0000	0.0000	0.2000	-0.1418	1.0000	13.1260	0.5000
		0.5000	-0.2000	20.0000	1.0000	-0.1000	9.0000	0.0000	0.0000
3	9	81.4346	0.0000	0.0000	-0.1594	-0.3000	1.0000	36.0000	0.0025
		0.2904	-0.2500	12.0000	1.0000	-0.0742	9.3638	0.0000	0.0000
4	9	96.5322	0.0000	0.0000	0.9970	-0.3000	1.0000	36.0000	0.5095
		0.7247	-0.2500	12.0000	1.0000	-0.1175	9.9985	0.0000	0.0000
9	9	73.6263	0.0000	0.0000	0.0209	-0.2000	0.0000	16.0000	0.3414
		0.4703	-0.2000	15.0000	1.0000	-0.1319	5.9254	0.0000	0.0000
2	10	109.1686	0.0000	0.0000	-0.1657	-0.2000	0.0000	16.0000	1.2500
		2.8463	-0.2000	15.0000	1.0000	-0.1111	5.2687	0.0000	0.0000
3	10	0.0000	0.0000	0.0000	0.5000	-0.2000	0.0000	16.0000	0.5000
		1.0001	-0.2000	15.0000	1.0000	-0.1000	10.0000	0.0000	0.0000
9	10	118.3052	0.0000	0.0000	-0.1168	-0.2000	0.0000	16.0000	0.0697
		2.9176	-0.2000	15.0000	1.0000	-0.1316	5.3624	0.0000	0.0000
10	10	0.2500	0.0000	0.0000	0.1803	-0.2000	0.0000	16.0000	0.3356
		0.9228	-0.2000	15.0000	1.0000	-0.1178	5.6715	0.0000	0.0000
1	8	0.0000	0.0000	0.0000	-1.0000	-0.3000	1.0000	36.0000	0.7000
		10.1151	-0.3500	25.0000	1.0000	-0.1053	8.2003	1.0000	0.0000
1	10	0.0000	0.0000	0.0000	0.5000	-0.2000	0.0000	16.0000	0.5000
		1.0001	-0.2000	15.0000	1.0000	-0.1000	10.0000	0.0000	0.0000
4	10	0.0000	0.0000	0.0000	0.5000	-0.2000	0.0000	16.0000	0.5000
		1.0001	-0.2000	15.0000	1.0000	-0.1000	10.0000	0.0000	0.0000
1	12	99.0142	100.8152	0.0000	0.0973	-0.5558	1.0000	17.2117	0.4287
		0.2384	-0.2529	10.1181	1.0000	-0.1172	5.7351	1.0000	0.0000
2	12	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
3	12	310.7157	5.0116	0.0000	-0.7381	-0.3000	1.0000	36.0000	0.7580
		10.8085	-0.2343	29.6431	1.0000	-0.1164	8.8837	6.0658	0.0000
12	12	70.9120	54.0531	30.0000	0.4931	-0.3000	1.0000	16.0000	0.0392

```
         0.2476   -0.8055    7.1248    1.0000   -0.1009    8.7229    0.0000    0.0000
 28     ! Nr of off-diagonal terms; Ediss;Ro;gamma;rsigma;rpi;rpi2
  1  2    0.1163    1.3831    9.9402    1.1471   -1.0000   -1.0000
  1  3    0.0729    2.0641    9.6888    1.4359    1.1638    1.0702
  2  3    0.0283    1.2853   10.9173    0.9259   -1.0000   -1.0000
  1  4    0.1268    1.9685    9.7237    1.3409    1.2578    1.1715
  2  4    0.1140    1.4837    9.0464    1.0812   -1.0000   -1.0000
  3  4    0.2000    1.8111    9.3860    1.4308    1.0787    1.2599
  1  5    0.1618    1.7943   10.1042    1.7489    1.3150    1.4031
  2  5    0.0764    1.5838   10.1462    1.4206   -1.0000   -1.0000
  3  5    0.1022    1.9887   10.0605    1.5799    1.4000   -1.0000
  4  5    0.1505    1.9000   10.5104    1.8000    1.4000   -1.0000
  2  6    0.0100    1.6000   13.2979    1.8670   -1.0000   -1.0000
  3  6    0.0809    1.7000   11.4606    1.5177   -1.0000   -1.0000
  1  7    0.2291    1.7500   10.0393   -1.0000   -1.0000   -1.0000
  2  7    0.3000    1.9262   10.0302   -1.0000   -1.0000   -1.0000
  3  7    0.1573    1.8644   10.1395    1.7435    1.4412   -1.0000
  6  7    0.1801    1.8566    9.8498    0.1000   -1.0000   -1.0000
  3  8    0.1592    1.8283   11.7256    1.6655   -1.0000   -1.0000
  1  9    0.0500    1.7500   12.3500    0.1000   -1.0000   -1.0000
  2  9    0.0300    1.5200   12.5000    0.1000   -1.0000   -1.0000
  3  9    0.0348    1.7637   12.3562    1.7228   -1.0000   -1.0000
  4  9    0.0478    1.7704   12.8051    1.6100   -1.0000   -1.0000
  2 10    0.0568    1.6740    9.6297    1.2200   -1.0000   -1.0000
  3 10    0.1927    2.2551   11.2308   -1.0000   -1.0000   -1.0000
  9 10    0.1402    2.1604   10.9786    1.7505   -1.0000   -1.0000
  1 12    0.0322    2.0505   13.4936    1.6976    1.4577   -1.0000
  2 12    0.2000    1.5207   12.9535    1.2125   -1.0000   -1.0000
  3 12    0.2000    1.8970   11.2796    1.7248    1.2186   -1.0000
  8 12    0.0216    1.5025   11.8792   -1.0000   -1.0000   -1.0000
127     ! Nr of angles;at1;at2;at3;Thetao,o;ka;kb;pv1;pv2
  1  1  1  66.0347   45.0000    1.1248    0.0000    3.0000   70.0000    2.4142
  1  1  2  58.9292   15.1099    4.4631    0.0000    0.0287    0.0000    1.8837
  2  1  2  64.8992   17.4932    1.9470    0.0000    3.0000    0.0000    1.0135
  1  2  2   0.0000    0.0000    6.0000    0.0000    0.0000    0.0000    1.0400
  1  2  1   0.0000    7.5000    5.0000    0.0000    0.0000    0.0000    1.0400
  2  2  2   0.0000   27.9213    5.8635    0.0000    0.0000    0.0000    1.0400
  1  1  3  51.7494   42.2658    0.7914    0.0000    4.0000   58.6562    1.7461
  3  1  3  72.7766    9.2311    7.5000  -20.0000    1.1419    0.0000    1.4603
  1  1  4  58.4756   41.7584    0.6211    0.0000    1.4838    0.0000    1.6722
  3  1  4  79.3051   13.4668    1.9350    0.0000    0.4036   32.5000    1.3503
  4  1  4  65.2722    4.5000    0.4741    0.0000    0.9683    0.0000    2.1819
  2  1  3  59.1567   27.4483    1.1736    0.0000    0.1000    0.0000    1.6111
  2  1  4  61.4983   11.5971    2.1170    0.0000    0.1000    0.0000    1.2079
  1  2  4   0.0000    0.0019    6.3000    0.0000    0.0000    0.0000    1.0400
  1  3  1  68.7170    6.5547    0.5383    0.0000    2.2753    0.0000    1.8466
  1  3  3  82.6613   45.0000    1.3380    0.0000    1.1034   68.1072    1.6232
  1  3  4  90.0000   18.7269    1.4975    0.0000    3.0479    0.0000    1.0965
```

3	3	3	90.0000	25.0984	1.6910	0.0000	1.6310	50.0000	1.0000
3	3	4	82.4170	34.0230	1.0276	0.0000	2.6572	0.0000	1.4288
4	3	4	72.6356	45.0000	1.1114	0.0000	3.0072	0.0000	1.0000
1	3	2	90.0000	8.9828	2.3727	0.0000	1.2146	0.0000	3.0000
2	3	3	79.5453	50.0000	2.1630	0.0000	3.0000	0.0000	1.2391
2	3	4	69.4868	16.3078	4.4440	0.0000	0.2502	0.0000	1.8860
2	3	2	85.7876	9.3298	2.0747	0.0000	2.8632	0.0000	1.6905
1	4	1	81.6738	11.0200	1.6084	0.0000	2.7249	0.0000	1.4096
1	4	3	88.8926	9.1088	4.8453	0.0000	2.1900	0.0000	3.0000
1	4	4	71.4079	13.7291	2.6469	0.0000	2.9290	0.0000	3.0000
3	4	3	74.7164	43.0000	1.1816	-18.0069	3.0584	0.0000	1.1322
3	4	4	81.1388	40.7000	0.9549	-0.9193	3.0760	0.0000	1.0000
4	4	4	72.1841	29.8125	1.9658	0.0000	2.9239	0.0000	2.9000
1	4	2	88.7155	6.5761	1.8952	0.0000	0.2894	0.0000	1.1321
2	4	3	76.0014	10.1041	7.3108	0.0000	0.4346	0.0000	3.0000
2	4	4	90.0000	42.7711	1.4427	0.0000	0.1000	0.0000	1.1775
2	4	2	83.5503	12.3251	4.8243	0.0000	0.1777	0.0000	1.0020
1	2	3	0.0000	17.0624	0.9927	0.0000	0.6225	0.0000	1.0000
1	2	4	0.0000	19.3874	0.2427	0.0000	0.2078	0.0000	1.1303
1	2	5	0.0000	15.0000	3.0000	0.0000	0.0000	0.0000	1.0400
3	2	3	0.0000	1.0000	2.5054	0.0000	0.0000	0.0000	4.0000
3	2	4	0.0000	12.4279	2.9774	0.0000	0.1224	0.0000	1.8030
4	2	4	0.0000	20.0000	3.6106	0.0000	0.1460	0.0000	1.0000
2	2	3	0.0000	5.8938	3.0000	0.0000	0.0000	0.0000	1.0629
2	2	4	0.0000	0.0019	6.0000	0.0000	0.0000	0.0000	1.0400
1	1	5	74.4180	33.4273	1.7018	0.1463	0.5000	0.0000	1.6178
1	5	1	79.7037	28.2036	1.7073	0.1463	0.5000	0.0000	1.6453
2	1	5	63.3289	29.4225	2.1326	0.0000	0.5000	0.0000	3.0000
1	5	2	85.9449	38.3109	1.2492	0.0000	0.5000	0.0000	1.1000
1	5	5	80.0000	25.0000	2.0000	0.0000	0.5000	0.0000	1.3830
2	5	2	85.0000	15.1317	2.0000	0.0000	0.5000	0.0000	2.0000
2	5	5	97.0064	32.1121	2.0242	0.0000	0.5000	0.0000	2.8568
2	2	5	0.0000	0.0019	6.0000	0.0000	0.0000	0.0000	1.0400
5	4	5	62.0000	33.4273	1.7018	0.1463	0.5000	0.0000	1.0500
3	5	3	77.0699	39.4349	2.1313	-30.0000	0.9567	0.0000	1.1483
1	5	3	70.0000	35.0000	3.4223	0.0000	1.3550	0.0000	1.2002
1	5	4	70.0000	35.0000	3.4223	0.0000	1.3550	0.0000	1.2002
4	1	5	60.0000	35.0000	3.0223	0.0000	2.3550	0.0000	1.2002
3	5	4	70.0000	35.0000	3.4223	0.0000	1.3550	0.0000	1.2002
5	1	7	70.0000	35.0000	3.4223	0.0000	1.3550	0.0000	1.2002
1	3	5	73.0990	33.8942	1.2098	0.0000	0.8161	0.0000	1.1776
3	3	5	83.9753	31.0715	3.5590	0.0000	0.8161	0.0000	1.1776
2	3	5	76.9521	20.0000	2.0903	0.0000	1.0000	0.0000	1.0400
2	6	2	0.0000	49.8261	0.2093	0.0000	2.0870	0.0000	2.2895
2	2	6	0.0000	39.7818	3.1505	0.0000	1.1296	0.0000	1.1110
6	2	6	0.0000	0.5047	0.8000	0.0000	0.8933	0.0000	4.6650
2	6	6	0.0000	8.7037	0.0827	0.0000	3.5597	0.0000	1.1198
3	6	3	0.0000	9.2317	0.1000	0.0000	1.0000	0.0000	1.0920

6	3	6	0.0008	25.0000	8.0000	0.0000	1.0000	0.0000	3.0000
2	3	6	66.0423	5.0000	1.0000	0.0000	1.0000	0.0000	1.2500
2	6	3	0.0000	0.5000	0.1000	0.0000	1.0000	0.0000	3.0000
3	3	6	70.0000	20.0000	1.0000	0.0000	1.0000	0.0000	1.2500
3	7	3	90.0000	6.2396	1.3578	-24.9000	1.5156	0.0000	1.6302
2	3	7	100.0000	11.5040	1.5163	0.0000	3.6982	0.0000	1.0400
3	3	7	60.0000	40.0000	4.0000	0.0000	1.0000	0.0000	1.0400
3	2	7	0.0000	10.0000	1.0000	0.0000	1.0000	0.0000	1.0400
6	3	7	41.7961	3.4184	7.4578	0.0000	-0.2958	0.0000	1.0542
7	3	7	90.0000	16.5250	7.0313	0.0000	-0.0396	0.0000	3.0000
1	3	7	56.0610	4.2889	4.5235	0.0000	2.6593	0.0000	2.4280
2	7	3	75.0000	25.0000	2.0000	0.0000	1.0000	0.0000	1.2500
3	7	7	70.0000	25.0000	2.0000	0.0000	1.0000	0.0000	1.2500
3	9	3	96.2265	4.5610	12.0000	0.0000	0.3211	0.0000	1.5204
3	9	3	0.0000	9.1552	7.9919	0.0000	0.1660	0.0000	1.5386
9	3	9	100.0000	10.1065	6.0000	0.0000	1.0000	0.0000	3.6601
2	3	9	55.0417	3.5032	3.9979	0.0000	1.5171	0.0000	1.0400
3	3	9	70.0000	30.0000	2.0000	0.0000	1.0000	0.0000	1.2500
3	9	9	66.7783	14.3146	0.7911	0.0000	1.0000	0.0000	1.2333
3	9	10	96.6924	9.4823	5.7883	0.0000	0.2248	0.0000	2.2640
3	9	10	0.0000	3.8549	3.7230	0.0000	0.1482	0.0000	1.0400
9	10	9	0.0000	11.2336	6.8851	0.0000	1.0000	0.0000	1.0893
9	9	10	90.0000	5.0811	5.2147	0.0000	1.0000	0.0000	1.8538
10	9	10	0.0100	21.1482	0.3506	0.0000	1.0000	0.0000	1.4361
3	2	10	0.0000	0.0100	0.5211	0.0000	0.0000	0.0000	1.3859
3	9	4	100.0000	28.1532	12.0000	0.0000	0.2932	0.0000	1.6489
3	9	4	0.0000	22.7457	2.9039	0.0000	0.5593	0.0000	1.9764
4	9	4	87.0081	27.6432	3.9735	0.0000	4.0000	0.0000	1.4578
4	9	4	0.0000	22.8998	3.1077	0.0000	3.0000	0.0000	1.0696
9	4	9	100.0000	10.1065	6.0000	0.0000	1.0000	0.0000	3.6601
2	4	9	80.0000	3.5601	3.3645	0.0000	1.5171	0.0000	1.0400
3	4	9	70.0000	30.0000	2.0000	0.0000	1.0000	0.0000	1.2500
4	3	9	70.0000	30.0000	2.0000	0.0000	1.0000	0.0000	1.2500
4	4	9	70.0000	30.0000	2.0000	0.0000	1.0000	0.0000	1.2500
4	9	9	66.7783	14.3146	0.7911	0.0000	1.0000	0.0000	1.2333
4	9	10	95.2122	5.7090	12.0000	0.0000	0.2248	0.0000	2.8936
4	9	10	0.0000	9.0054	7.9511	0.0000	0.1482	0.0000	1.6245
4	2	10	0.0000	15.0000	2.8900	0.0000	0.0000	0.0000	2.8774
1	3	9	55.0000	15.0000	1.0000	0.0000	1.0000	0.0000	1.5000
1	4	9	55.0000	15.0000	1.0000	0.0000	1.0000	0.0000	1.5000
12	12	12	78.5339	36.4328	1.0067	0.0000	0.1694	0.0000	1.6608
2	12	12	77.2616	5.0190	7.8944	0.0000	4.0000	0.0000	1.0400
2	12	2	75.7983	14.4132	2.8640	0.0000	4.0000	0.0000	1.0400
3	12	12	90.0000	26.2447	8.0000	0.0000	0.8005	0.0000	2.2967
2	12	3	73.6998	40.0000	1.8782	0.0000	4.0000	0.0000	1.1290
3	12	3	75.7337	31.2440	1.3341	0.0000	0.1772	0.0000	2.7107
12	3	12	83.0371	4.8857	1.4424	0.0000	3.1590	0.0000	1.0400
2	3	12	90.0000	6.8271	1.5367	0.0000	1.5618	0.0000	1.0585

```
 3   3  12 101.5854   29.0118    0.9642    0.0000    1.7259    0.0000    2.4039
 2   2  12   0.0000   47.1300    6.0000    0.0000    1.6371    0.0000    1.0400
12   2  12   0.0000   27.4206    6.0000    0.0000    1.6371    0.0000    1.0400
 3   2  12   0.0000    5.0000    1.0000    0.0000    1.0000    0.0000    1.2500
 1   1  12  72.9199   19.5505    1.8653    0.0000    0.2042    0.0000    1.0633
 1  12   1  69.5338   17.8814    2.1791    0.0000    0.7456    0.0000    1.3913
12   1  12  68.2950   16.5938    1.7725    0.0000    0.2200    0.0000    1.3891
 1  12  12  69.2676   17.7813    1.8831    0.0000    1.1316    0.0000    1.0400
 2   1  12  74.1618   12.0055    2.4299    0.0000    0.3085    0.0000    1.2939
 1  12   2  71.7254   14.6023    2.3629    0.0000    0.7282    0.0000    1.2763
 1   2  12   0.0000    2.5000    1.0000    0.0000    1.0000    0.0000    1.2500
 1   3  12  84.5450   10.7378    0.9500    0.0000    0.3898    0.0000    1.1285
 1  12   3  70.4005   38.2218    1.7533    0.0000    1.7600    0.0000    1.0400
 3   1  12  69.4907    6.8475    1.1906    0.0000    1.0563    0.0000    1.1300
71     ! Nr of torsions;at1;at2;at3;at4;;V1;V2;V3;V2(BO);vconj;n.u;n
 1   1   1   1    0.3619    5.0000    0.2257   -9.0000   -2.4400    0.0000    0.0000
 1   1   1   2    0.2330   56.0330   -0.0369   -6.3749   -1.6179    0.0000    0.0000
 2   1   1   2   -0.0662   44.7480    0.3945   -6.5879   -3.0000    0.0000    0.0000
 1   1   1   3    2.0835   80.0000    0.2861   -5.8354   -1.1000    0.0000    0.0000
 2   1   1   3    2.5000   15.1318    1.0000   -6.3682   -1.0978    0.0000    0.0000
 3   1   1   3   -1.4902   25.2456   -1.0000   -2.5000   -0.8614    0.0000    0.0000
 1   1   3   1   -1.0000   -5.0000   -0.5000   -2.5000   -0.9000    0.0000    0.0000
 1   1   3   2   -2.1885    5.0000   -0.9943   -9.0000   -0.9000    0.0000    0.0000
 2   1   3   1    0.8086   63.4490   -0.1261   -9.0000   -0.9000    0.0000    0.0000
 2   1   3   2   -2.5000   73.5049    0.8863   -4.0386   -1.1000    0.0000    0.0000
 2   1   3   3   -0.9280  100.0000    1.0000   -3.9313   -2.8274    0.0000    0.0000
 3   1   3   1   -2.5000    9.3090   -1.0000   -8.2407   -3.0437    0.0000    0.0000
 3   1   3   2    2.5000   21.4615   -0.0315   -2.5000   -3.0476    0.0000    0.0000
 1   3   3   2   -2.5000    8.2305   -0.2363   -2.7513   -2.9498    0.0000    0.0000
 2   3   3   2   -2.5000  -25.0000   -1.0000   -2.5000    0.0000    0.0000    0.0000
 1   3   3   3    1.5042  -24.6836    1.0000   -2.5000   -0.9972    0.0000    0.0000
 2   3   3   3    0.2942   70.1282   -0.7126   -6.1638    0.0000    0.0000    0.0000
 3   3   3   3   -2.5000  -25.0000    1.0000   -2.5000   -0.9000    0.0000    0.0000
 1   1   4   2    0.7954   67.9069    0.3504   -8.0000   -1.9825    0.0000    0.0000
 2   1   4   2    0.4566   46.1133    0.4807   -5.7278   -2.1051    0.0000    0.0000
 3   1   4   2    1.0000   26.3717    1.0000   -3.5214   -2.5261    0.0000    0.0000
 3   1   1   4   -0.5802    7.2544    1.0000   -2.5000   -0.9511    0.0000    0.0000
 4   1   1   4   -0.2416   15.8213   -0.5000   -3.2327   -1.7241    0.0000    0.0000
 1   1   4   1   -1.0000   51.5166    0.1228   -6.5755   -1.6589    0.0000    0.0000
 3   1   4   1   -1.0000   14.9858    1.0000   -8.0000   -1.8038    0.0000    0.0000
 2   1   1   4    1.0000    0.0100    0.8120   -2.8086   -1.9000    0.0000    0.0000
 4   1   4   2    1.0000   51.6090   -0.5000   -3.9390   -2.0202    0.0000    0.0000
 2   1   4   1   -0.2342   68.6969    0.1904   -5.3454   -0.0100    0.0000    0.0000
 4   1   3   1   -0.8484   15.6632   -0.3865   -2.5000   -3.0437    0.0000    0.0000
 4   1   3   2    0.0107    3.7120   -0.5000   -7.9280   -3.0476    0.0000    0.0000
 1   1   1   4    0.9187   -5.0000    1.0000   -6.3298   -2.0000    0.0000    0.0000
 4   1   4   1   -0.8232   16.3430    0.5477   -7.0692   -2.0051    0.0000    0.0000
 0   1   2   0    0.0000    0.0000    0.0000   -4.0000    0.0000    0.0000    0.0000
```

```
0  2  2  0    0.0000    0.0000    0.0000   -4.0000    0.0000    0.0000    0.0000
0  2  3  0    0.0000    0.1000    0.0200   -4.0000    0.0000    0.0000    0.0000
0  1  1  0    0.0000   50.0000    0.3000   -4.0000   -2.0000    0.0000    0.0000
0  3  3  0    0.5511   25.4150    1.1330   -5.1903   -1.0000    0.0000    0.0000
0  1  4  0    0.2176   40.4126    0.3535   -3.9875   -2.0051    0.0000    0.0000
0  2  4  0    0.0000    0.1000    0.0100   -5.0000    0.0000    0.0000    0.0000
0  3  4  0    1.1397   61.3225    0.5139   -3.8507   -2.7831    0.0000    0.0000
0  4  4  0    0.7265   44.3155    1.0000   -4.4046   -2.0000    0.0000    0.0000
4  1  4  4   -0.0949    8.7582    0.3310   -7.9430   -2.0000    0.0000    0.0000
0  1  5  0    0.8251   92.1468    0.7176   -4.2341    0.0000    0.0000    0.0000
0  5  5  0    0.1291   -5.0000    0.9649   -5.0903    0.0000    0.0000    0.0000
0  2  5  0    0.0000    0.0000    0.0000    0.0000    0.0000    0.0000    0.0000
0  6  6  0    0.0000    0.0000    0.1200   -2.4426    0.0000    0.0000    0.0000
0  2  6  0    0.0000    0.0000    0.1200   -2.4847    0.0000    0.0000    0.0000
0  3  6  0    0.0000    0.0000    0.1200   -2.4703    0.0000    0.0000    0.0000
1  1  3  3   -0.0002   20.1851    0.1601   -9.0000   -2.0000    0.0000    0.0000
1  3  3  1    0.0002   80.0000   -1.5000   -4.4848   -2.0000    0.0000    0.0000
3  1  3  3   -0.1583   20.0000    1.5000   -9.0000   -2.0000    0.0000    0.0000
2  3  9  3   -1.5000    6.8333   -0.1978   -1.4683    0.0000    0.0000    0.0000
2  3  9  4   -0.6181    7.1542   -0.0047   -1.6577    0.0000    0.0000    0.0000
2  4  9  3   -1.5000    1.7820   -1.0000   -5.4916    0.0000    0.0000    0.0000
2  4  9  4   -0.1959    2.3626   -1.0000   -3.0702    0.0000    0.0000    0.0000
2  1  4  9    0.0000   10.0000    0.3000   -6.0000   -1.0000    0.0000    0.0000
2  3  9 10    0.1589   12.5000    0.4388   -1.5000    0.0000    0.0000    0.0000
7  1  1  7    1.0657   78.7067    0.6978   -8.4833   -1.7255    0.0000    0.0000
0  1  7  0    1.4891   35.1275    0.8254   -2.8518    0.0000    0.0000    0.0000
0  7  7  0   -0.3570   38.0764    0.1676   -6.7257    0.0000    0.0000    0.0000
1  1  1  7   -0.1661   19.8637    0.2351  -10.8000   -1.7255    0.0000    0.0000
2  1  3  7   -1.5000   99.6206    0.0516   -8.1129    0.0000    0.0000    0.0000
2  3  7  3   -1.5000   -1.0000    0.2523   -2.7183    0.0000    0.0000    0.0000
1  3  7  3   -1.5000   50.2166    0.9871   -3.6395    0.0000    0.0000    0.0000
7  3  7  3    1.5000   -0.9128    0.0100   -6.2667    0.0000    0.0000    0.0000
1  1  3  7    0.0000    0.1000    0.0100   -4.0000    0.0000    0.0000    0.0000
2 12 12  2    0.0000    0.0000    0.0640   -2.4426    0.0000    0.0000    0.0000
2 12 12 12    0.0000    0.0000    0.1587   -2.4426    0.0000    0.0000    0.0000
0  2 12  0    0.0000    0.0000    0.1200   -2.4847    0.0000    0.0000    0.0000
3  1  1 12   -0.5740   22.4215    0.8787   -2.7603   -1.1000    0.0000    0.0000
1  1 12  3   -0.5740   22.4215    0.8787   -2.7603   -1.1000    0.0000    0.0000
9    ! Nr of hydrogen bonds;at1;at2;at3;Rhb;Dehb;vhb1
3  2  3    2.1493   -3.8387    1.5230   28.0017
3  2  4    2.0543   -6.3062    1.4500   19.5000
4  2  3    1.7500   -1.8134    1.4500   19.5000
4  2  4    1.8886   -1.8024    1.4500   19.5000
3  2  5    1.5000   -2.0000    1.4500   19.5000
4  2  5    1.5000   -2.0000    1.4500   19.5000
5  2  3    1.5000   -2.0000    1.4500   19.5000
5  2  4    1.5000   -2.0000    1.4500   19.5000
5  2  5    1.5000   -2.0000    1.4500   19.5000
```

Deconvoluted XPS Data

Table S2: XPS peak fitting [12, 13] results for SiOC polymer derived ceramic.

Element	Element Atomic %	Component Name	Binding Energy (eV)	Component Atomic %
C 1s	34.5%	C = C	284.0	35.47
		C - C	284.4	20.65
		C – Si	283.5	7.28
		C – O – Si	284.8	29.00
		C – O	288.0	7.62
O 1s	37.3%	O – Si	532.2	97.12
		O – C	529.5	2.88
Si 2p	28.2%	Si – O – Si	103.0	57.46
		Si – C	102.4	42.54

Elastic Modulus of SiOC Ceramics

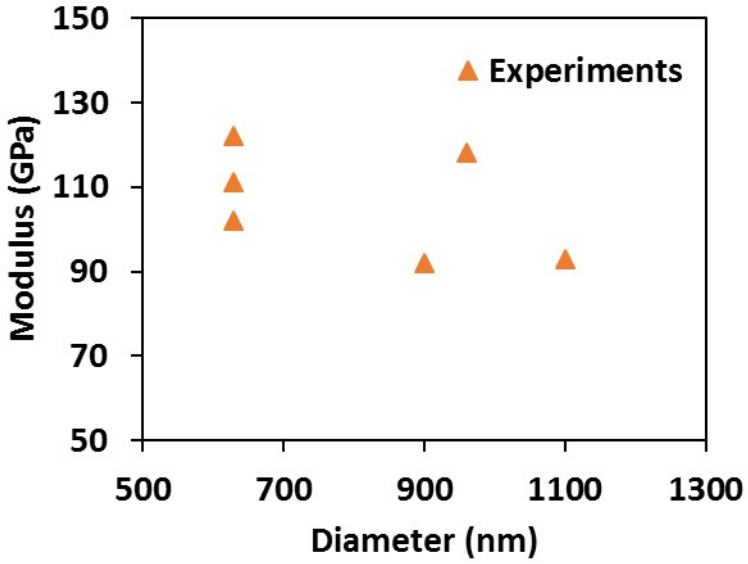

Figure S2: Dependence of experimentally measured tensile modulus on fiber diameter.

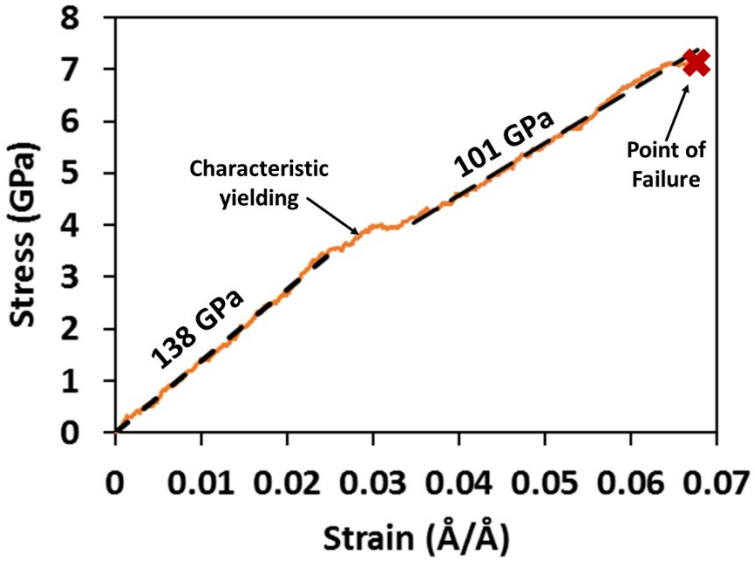

Figure S3: Characteristic failure during virtual tensile testing observed at ~3% at 27°C.

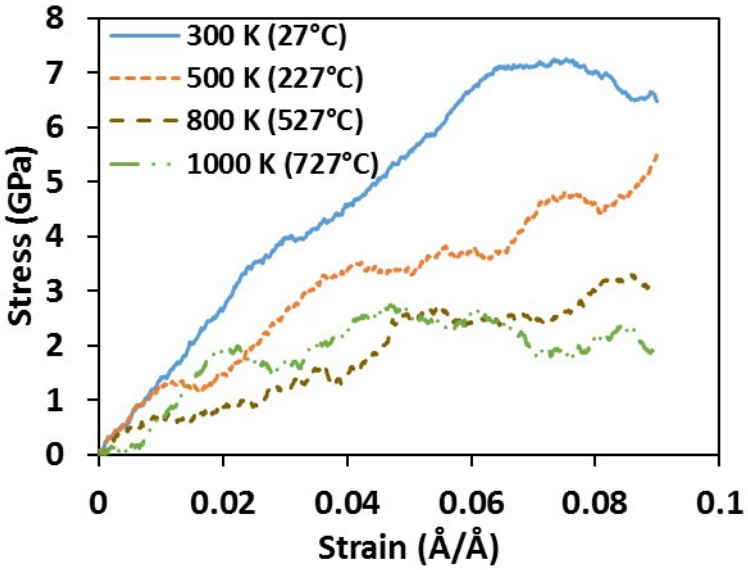

Figure S4: Stress vs. strain behavior of a flawless SiOC molecular structure at variable temperatures.

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