JOURNAL OF APPLIED CRYSTALLOGRAPHY

Volume 52 (2019)

Supporting information for article:

Molecular dynamics simulation of organic crystals: introducing the CLP-*dyncry* environment

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Summary

S1. Parametrization of bonded interactions in CLP-dyn

Table S1.

See Section 2.1.1 in the main text. Equilibrium bond lengths (expt, from averaged crystal structure data, calc from ab initio calculations) and bond stretching force constants in kJ mol^{-1} $angstrom^{-2}$ from MP2/6-31G** (New J. Chem. **2016**, *40*, 6848-6853) unless otherwise stated.

Bond	R_{expt}	$R°_{calc}$	k	system for R° and k calculation
-C≡C-	1.183	1.223	9620	but-1-yne
>C=C<	1.346-1.360	1.353	5600	butadiene
≡C - C≡	1.378	1.383	4540	buta-1,3-diyne
Car---Car	1.382	1.397	4640	benzene
Csp^2 - Csp^2	1.439	1.457-1.463	3400	butadiene
≡C - Csp^3	1.467	1.472	3340	but-1-yne
Csp^2 -Csp^3	1.503	1.513-1.515	3120	toluenes
Csp^3 - Csp^3	1.523	1.517-1.536	2800	butane
Csp^3- H	1.085	1.093	3630	ethane
Csp^2 - H	1.077	1.087	3630	benzene
C≡N	1.139	1.180	11500	acetonitrile
Csp^3 - N<	1.461	1.460	3540	trimethylamine
Csp^2 - O	1.369	1.381	4320	methoxybenzene
Csp^3 - O	1.435	1.432	3630	dimethylether
C=O	1.214	1.227	8200	acetone
Csp^2 - F	1.346	1.358	4200	fluorobenzene
Csp^3 - F	1.367	1.397	3950	fluoroethane
Csp^2 - Cl	1.735	1.742	2580	chlorobenzene
Csp^3 - Cl	1.771	1.784	2410	chloroethane
Csp^2 - Br	1.892	-		
Csp^2 - I	2.095	-		
N = O nitro	1.218	-		
N-H	-	1.018	5300	urea (this work)
O-H	-	0.987	4250	acetic acid (this work)

Table S2.

See Section 2.1.1 in the main text. Bond bending force field parameters from MP2/6-31G** ab initio calculations

	α°	k, kJ mol^{-1}	sample system
C-C-C	112.4	880	propane, bend of CCC and CCH angles in the CH_2 group
C=C-C	124.5	1030	propene, bend of CCC and CCH angles
C-O-C	112.4	972	dimethyl ether
C-C=O	123	894	acetone simultaneous bending of two angles
(O)=C-O-H	104	475	-COOH acid
(O)=C-N-H	120	940	-$CONH_2$ amide, bend of 2 CNH
(Ar)C-O-H	107	550	alcohol (phenol)
CCH	120	890	benzene simultaneous bending
HCH	106.5	530	propane, scissor mode at the methylene group
CCH	110	680	methyl
CCH	110	980	methylene

S2. Map of torsional potential energies from ab initio MP2/6-31G** calculation using Gaussian

See Section 2.1.1 in the main text. For each torsion, the schemes mark the involved atoms in red. The numbering in the schemes corresponds to the numbering in the pictures with the torsional profiles. Each profile must be fitted with the appropriate constants for the MD torsional energy function. For complex profiles, the fitting may not be obvious, or more than one torsion energy term must be employed.

$$E_{tors} = k[1 + fcos(m\tau)]$$

Where τ is the torsion angle in degrees, k = 1/2 barrier height, f is a phase factor, equal to +1 or -1, and m = 1,2 or 3.
The first derivative with respect to τ is:

$$E' = -kf[m \cdot sin(m\tau)]$$

With this function, the zero energy torsion can only be =, 60, 90, 120 or 180°. In the Monte Carlo module of the CLP package, a more flexible form of the torsional function is implemented, either $E_{tors} = k\{1 + fcos[m(\tau - \tau^0)]\}$, or in the form of a polynomial fitting the curve.

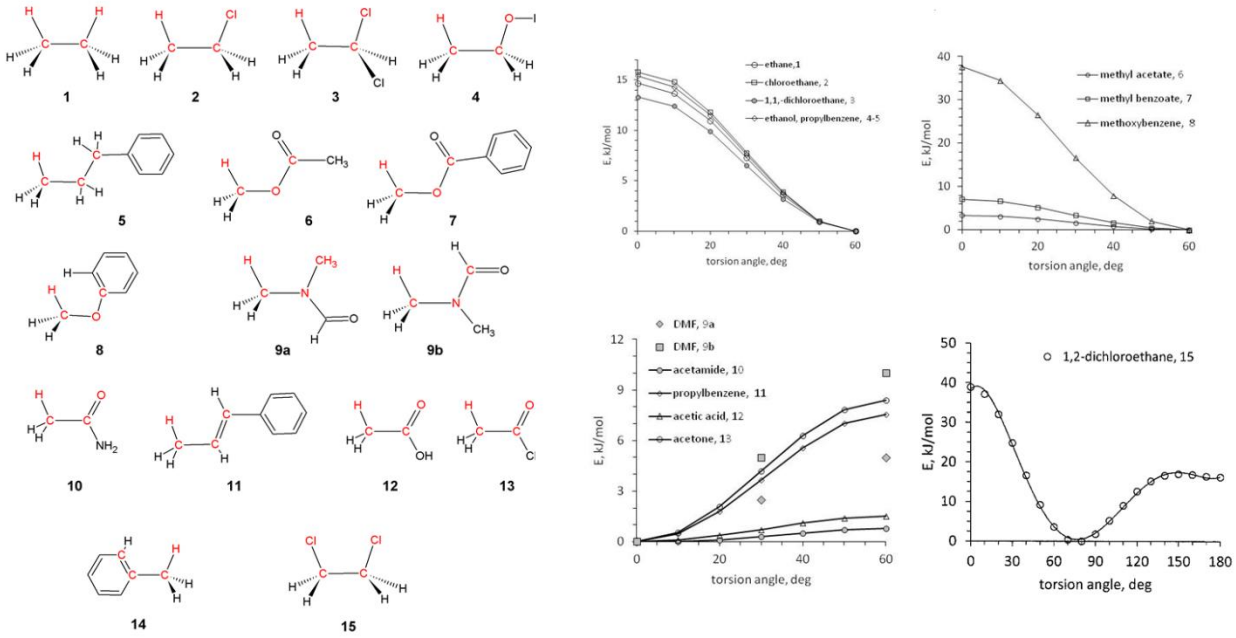

Figure S1.

Ab-initio torsional potentials for compounds 1-15.

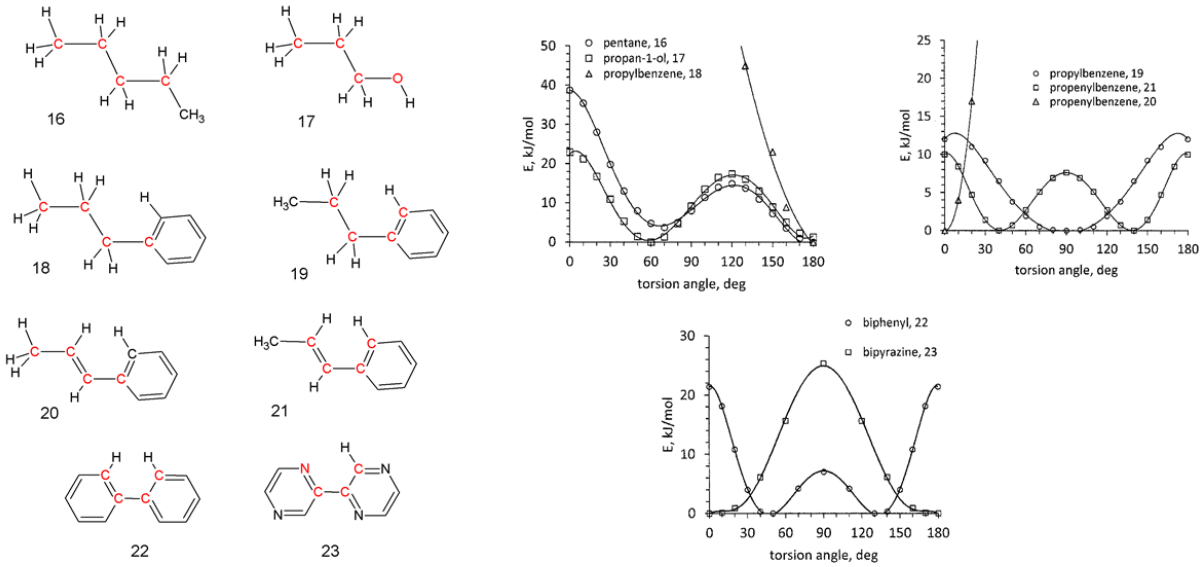

Figure S2.

Ab-initio torsional potentials for compounds 16-23.

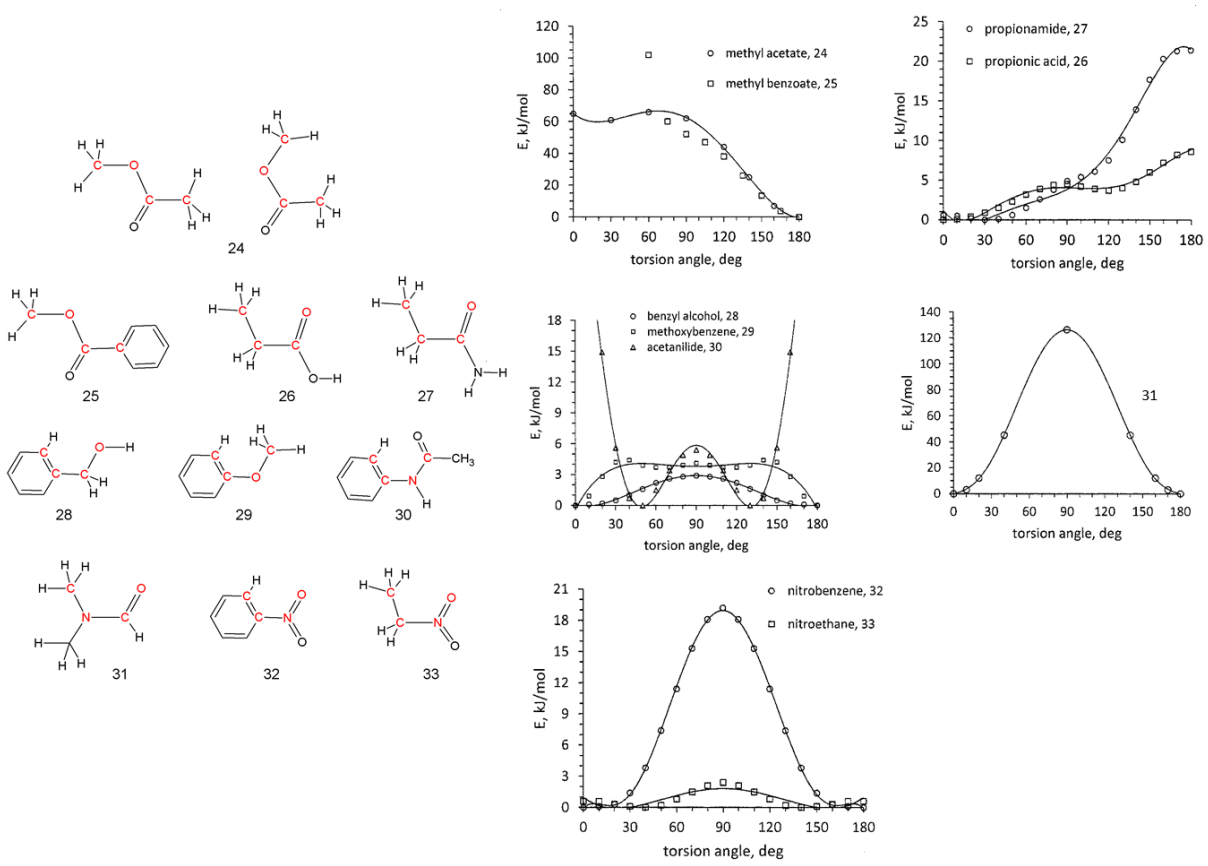

Figure S3.

Ab-initio torsional potentials for compounds 24-33.

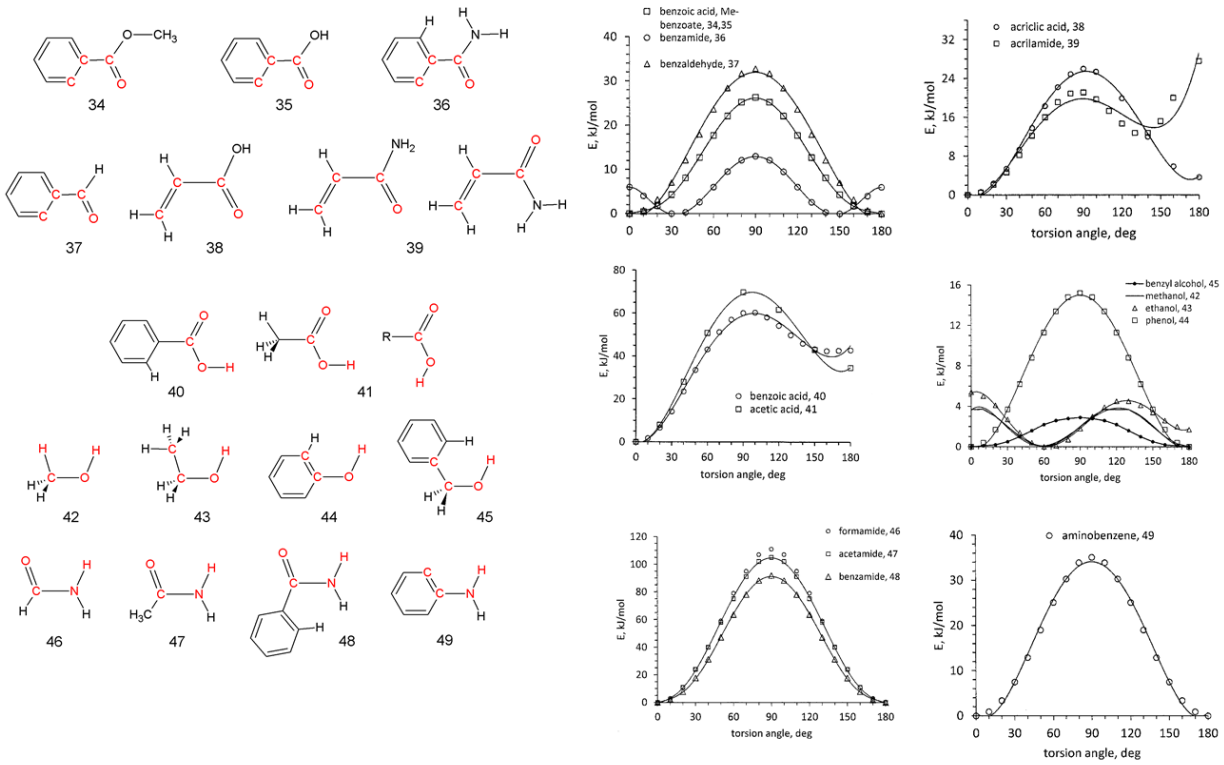

Figure S4.

Ab-initio torsional potentials for compounds 34-49.

S3. Automatic assignment of atom types in the ATB approach

See Section 2.1.2 in the main text. The Automated Topology Builder procedure assigns atom types based on a combination of geometry and point charge criteria, which attribute to the various sub-species different 6-12 potential energy parameters depending on their chemical environment (Table S1 SI). In general, this is a continuing challenge in the GROMOS environment, because no unique guidelines are available to define some kind of "universal" set of empirical parameters. Due to overlap between successive versions of the force fields, with superimposing re-parameterizations, unraveling back the philosophy under the different sub-species parameters is an awkward task.

For carbon atoms, the distinction between C "normal" and CAro "aromatic" is made on the basis of connectivity and the number of surrounding double bonds, but the difference is anyway very small. CPos is assigned when the point charge on the carbon atom is positive, usually when bound to oxygen but also to the methylene carbon in cyclohexadiene (q = +0.19). This atom type has a much more stabilizing potential and a minimum-energy distance much shorter than twice the accepted atomic radius of carbon. The parameters for the oxygen sub-species are strikingly different. Oxygen OM is the carboxylate oxygen (alanine), OA is the C-O-C oxygen in the ester group of the hydroxybenzoate derivative and in succinic and maleic anhydrides, OEOpt is the carbonyl oxygen, with a minimum energy separation some 30% longer than twice the atomic radius, and OAlc is the alcohol hydrogen. Nitrogen NL is the quaternary nitrogen in alanine, and NPri is the amino nitrogen in NH_2, but the same atom in the $HNCH_3$ group of methylurea is labelled as N, with a much shorter minimum distance. Finally, HC is hydrogen bound to carbon while HS14 is for hydrogen bonding donors with zero LJ potential since in these force fields the hydrogen bond is described only by the Coulombic interaction between point charges.

Table S3.

See Section 2.1.2 in the main text. Parameters for the 6-12 Lennard-Jones (L-J) potentials for atom types in the 54A7 GROMOS force field. The quoted distances (R_{min}, Å) and energies (E_{min}, kJ/mol) refer to the minimum of the X···X interaction curve. X···Y parameters are assigned by the geometric mean rule. R_{at} (Å) is the commonly accepted van der Waals radius.

Label	Chemical specie	C_6	C_{12}	R_{min}	E_{min}	R_{at}
Carbon atoms						
C	C aliphatic	2340.62	4937284	4.000	-0.28	1.77
Caro	C aromatic	2340.62	4456320	3.950	-0.31	
CPos	$C^{\delta+}$ (as in C=O)	2025.00	1000000	3.150	-1.03	
Oxygen atoms						
OM	$-COO^-$	2261.95	741493.3	2.950	-1.72	1.58
OA	C-O-C (esters)	2261.95	1505529	3.300	-0.85	
OEOpt	-C=O	3089.14	4774220	3.800	-0.50	
OAlc	-OH	1774.94	1210000	3.350	-0.65	
Nitrogen atoms						
NOpt	N aromatic	8262.81	29160000	4.400	-0.58	1.64
NPri	$-NH_2$	10816.00	49000000	4.550	-0.60	
N	-NH-R	2436.41	2319529	3.500	-0.64	
NL	$-NH_3^+$	2436.41	2319529	3.500	-0.64	
Hydrogen atoms						
HC	-CH, -OH	84.64	15129	2.650	-0.12	1.10
HS14[a]	$-NH_3^+$	0.00	0			

[a] To avoid collapse of the positively charged hydrogen onto negatively charged atoms when 1-4 interactions are computed, ATB uses a special polar hydrogen type (HS14) that exploits Lennard Jones (L-J) terms only for the 1-4 interactions. According to the original GROMOS formulation, such L-J terms are switched off when more distant pairs come into play.

S4. Topologies for Molecular Dynamics

S4.1 CLP Topology files

See Section 2.2.2 in the main text. Details on the structure of the files can be found at www.angelogavezzotti.it, link to CLP-*dyn*, documentation directory. Atomic charges are evaluated from the Mulliken partitioning of an unexpensive Extended Hückel wavefunction, but they are rescaled by an empirical 0.41 factor before any energy and force calculation (see also the user manual).

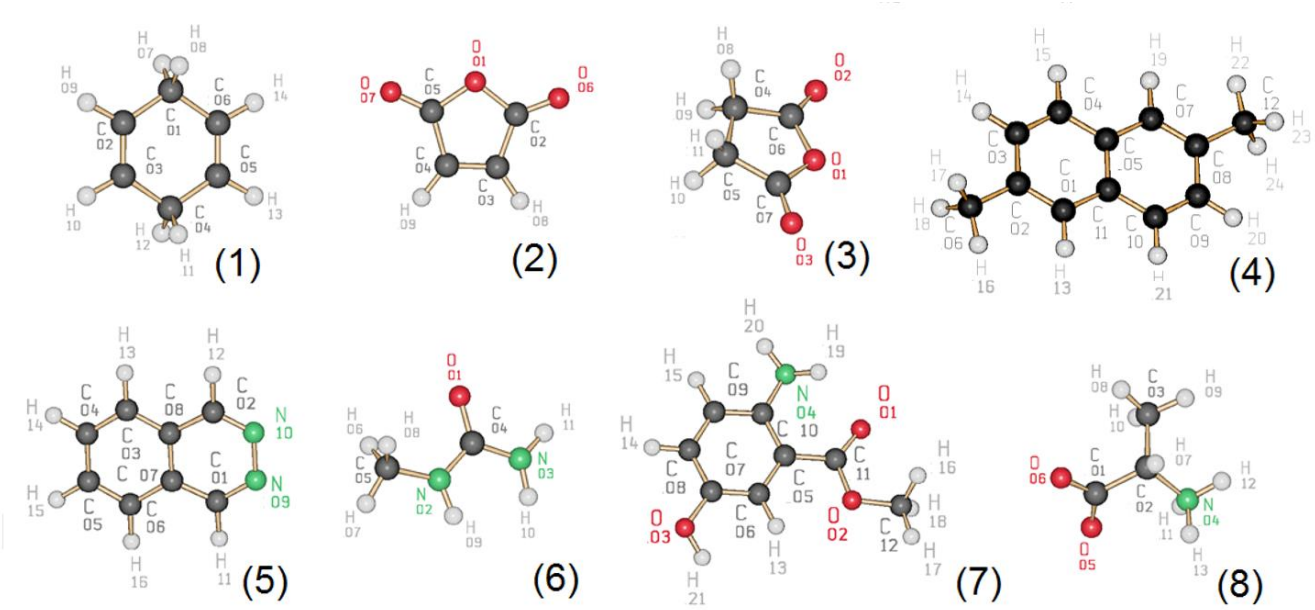

Figure S5.

Molecules investigated by MD, with the atom numbering scheme. See Figure 1 in the main text for further information.

CLP topologies: for each molecule, x,y,z, atom type, atomic point charge to be rescaled by the standard CLP factor, 0.41; list of stretching, bending and torsional functions; CLP standard rescaling parameters. Refer to Figure S5 above for the atom numbering.

1,4-cyclohexadiene (1)

Atomic point charges to be multiplied by the rescaling factor 0.41

```
14
 1   0.00205  -0.00575  -1.47764  13   -0.5014
 2  -0.00195   1.23767  -0.66357  12   -0.3534
 3   0.00195   1.24225   0.65373  12   -0.3531
 4  -0.00205   0.00575   1.47764  13   -0.5014
 5   0.00195  -1.23767   0.66357  12   -0.3534
 6  -0.00195  -1.24225  -0.65373  12   -0.3531
 7  -0.86860  -0.00838  -2.11668   3    0.3148
 8   0.87886  -0.00833  -2.10821   3    0.3156
 9  -0.00840   2.18681  -1.17878   2    0.2888
10   0.00838   2.19528   1.16181   2    0.2887
```

```
 11   0.86860   0.00838   2.11668   3    0.3148
 12  -0.87886   0.00833   2.10821   3    0.3156
 13   0.00840  -2.18681   1.17878   2    0.2888
 14  -0.00838  -2.19528  -1.16181   2    0.2887
  0  nslav-u
  0  ncore-v
  0  nslav-v
 87.0    0.0 volu-u,volu-v
 14  nstr-u
  1    2   1.486  3100.0
  1    6   1.486  3100.0
  3    4   1.486  3100.0
  4    5   1.486  3100.0
  2    3   1.317  3400.0
  5    6   1.317  3400.0
  1    7   1.080  3500.0
  1    8   1.080  3500.0
  2    9   1.080  3500.0
  3   10   1.080  3500.0
  4   11   1.080  3500.0
  4   12   1.080  3500.0
  5   13   1.080  3500.0
  6   14   1.080  3500.0
  0  nstr-v
 24  nbend-u
  1    2    3  123.0   1000.0
  1    6    5  123.0   1000.0
  2    3    4  123.0   1000.0
  4    5    6  123.0   1000.0
  2    1    6  113.0   900.0
  3    4    5  113.0   900.0
  1    2    9  118.0   900.0
  1    6   14  118.0   900.0
  2    3   10  118.0   900.0
  3    2    9  118.0   900.0
  4    3   10  118.0   900.0
  4    5   13  118.0   900.0
  5    6   14  118.0   900.0
  6    5   13  118.0   900.0
  2    1    7  109.5   900.0
  2    1    8  109.5   900.0
  3    4   11  109.5   900.0
  3    4   12  109.5   900.0
  5    4   11  109.5   900.0
  5    4   12  109.5   900.0
  6    1    7  109.5   900.0
  6    1    8  109.5   900.0
  7    1    8  109.5   500.0   H-C-H
 11    4   12  109.5   500.0
  0  nbend-v
 10  ntors-u
1  2  3  4  50.0 -1.0   1.0
2  3  4  5  50.0 -1.0   1.0
3  4  5  6  50.0 -1.0   1.0
4  5  6  1  50.0 -1.0   1.0
5  6  1  2  50.0 -1.0   1.0
6  1  2  3  50.0 -1.0   1.0
2  1  3  9  100.0  -1.0  1.0
3  2  4 10  100.0  -1.0  1.0
5  4  6 13  100.0  -1.0  1.0
6  5  1 14  100.0  -1.0  1.0
  0  ntors-v
  0  nlist-u
  0  nlist-v
 0.410   235.0   650.0   77000.0
  0  nintra
```

Maleic anhydride (2)

Atomic point charges to be multiplied by the rescaling factor 0.41

```
    9
    1   0.02552   0.92681   0.00431  23   -0.7536
    2   0.00300   0.10952  -1.11818  10    1.3867
    3   0.00238  -1.29766  -0.67111  12   -0.2214
    4   0.00257  -1.30367   0.66051  12   -0.2185
    5   0.00028   0.10436   1.12150  10    1.3867
    6  -0.01648   0.57192  -2.22305  27   -1.0952
    7  -0.01560   0.56736   2.22536  27   -1.0936
    8   0.00186  -2.16790  -1.31070   2    0.3046
    9   0.00409  -2.17954   1.29236   2    0.3044
    0  nslav-u
    0  ncore-v
    0  nslav-v
 79.4    0.0 volu-u,volu-v
    9  nstr-u    new
    1    2   1.388  4300.0   C-O
    1    5   1.388  4300.0
    2    6   1.198  8000.0   C=O
    5    7   1.198  8000.0
    3    4   1.332  3400.0   C=C
    2    3   1.479  3100.0   C-C
    4    5   1.479  3100.0
    4    9   1.080  3600.0   C-H
    3    8   1.080  3600.0
    0  nstr-v
   13  nbend-u
    1    2    3  108.33   900.0     O-C-C
    1    5    4  108.33   900.0
    1    2    6  121.06   900.0     O-C=O
    1    5    7  121.06   900.0
    2    1    5  107.56   900.0     C-O-C
    4    5    7  130.60   900.0     C-C=O
    3    2    6  130.60   900.0
    2    3    4  107.88  1000.0     C-C=C
    3    4    5  107.88  1000.0
    3    4    9  126.06   850.0     C-C-H
    4    3    8  126.06   850.0
    5    4    9  126.06   850.0
    2    3    8  126.06   850.0
    0  nbend-v
    9  ntors-u
  7  5  1  2  50.0   1.0   1.0    stiff 1-fold
  5  1  2  6  50.0   1.0   1.0
  7  5  4  3  50.0   1.0   1.0
  6  2  3  4  50.0   1.0   1.0
  5  4  3  2  100.0 -1.0   1.0
  5  1  4  7   100.0  -1.0  1.0   improper
  2  1  3  6   100.0  -1.0  1.0
  3  2  4  8   100.0  -1.0  1.0
  4  5  3  9   100.0  -1.0  1.0
    0  ntors-v
    0  nlist-u
    0  nlist-v
 0.410   235.0   650.0   77000.0
    0  nintra
```

Succinic anhydride (3)

Atomic point charges to be multiplied by the rescaling factor 0.41

```
   11
```

```
  1  -0.00753   0.91932  -0.00501  23   -0.7159
  2   0.00967   0.64674   2.20948  27   -1.1190
  3   0.00205   0.64863  -2.20881  27   -1.1225
  4  -0.01244  -1.30070   0.75705  13   -0.5393
  5   0.01235  -1.31114  -0.75073  13   -0.5375
  6  -0.00211   0.13995   1.13656  10    1.3282
  7  -0.00368   0.12896  -1.13900  10    1.3297
  8   0.84889  -1.81218   1.16067   3    0.3426
  9  -0.89819  -1.79204   1.13171   3    0.3453
 10  -0.84704  -1.82988  -1.14919   3    0.3428
 11   0.90000  -1.80272  -1.12064   3    0.3454
  0  nslav-u
  0  ncore-v
  0  nslav-v
 82.6    0.0 volu-u,volu-v
 11  nstr-u
  2    6   1.200  8000.0
  3    7   1.200  8000.0
  1    6   1.380  4300.0
  1    7   1.380  4300.0
  4    5   1.510  2800.0
  4    6   1.490  3100.0
  5    7   1.490  3100.0
  4    8   1.080  3600.0
  4    9   1.080  3600.0
  5   10   1.080  3600.0
  5   11   1.080  3600.0
  0  nstr-v
 19  nbend-u
  6    1    7  110.0   950.0
  1    6    2  120.0   880.0
  1    7    3  120.0   880.0
  2    6    4  130.0   880.0
  3    7    5  130.0   880.0
  1    6    4  110.0   880.0
  1    7    5  110.0   880.0
  4    5    7  105.0   880.0
  5    4    6  105.0   880.0
  5    4    8  110.0   750.0
  5    4    9  110.0   750.0
  6    4    8  110.0   750.0
  6    4    9  110.0   750.0
  4    5   10  110.0   750.0
  4    5   11  110.0   750.0
  7    5   10  110.0   750.0
  7    5   11  110-0   750.0
  8    4    9  108.0   530.0
 10    5   11  108.0   530.0
  0  nbend-v
  7  ntors-u
2  6  1  7  50.0  1.0   1.0
6  1  7  3  50.0  1.0   1.0
2  6  4  5  50.0  1.0   1.0
3  7  5  4  50.0  1.0   1.0
6  4  5  7  50.0 -1.0   1.0
6  1  4  2  100.0  -1.0  1.0
7  1  5  3  100.0  -1.0  1.0
  0  ntors-v
  0  nlist-u
  0  nlist-v
 0.410   235.0   650.0   77000.0
  0  nintra
```

2,6-dimetylnaphthalene (4)

Atomic point charges to be multiplied by the rescaling factor 0.41

```
 24
  1   0.02120  -1.04392   1.55154  12   -0.3686
  2   0.01077  -0.09688   2.54132  12    0.0605
  3   0.01089   1.27149   2.16792  12   -0.3218
  4   0.00298   1.65329   0.85918  12   -0.3127
  5  -0.00490   0.68603  -0.17957  14    0.0341
  6  -0.01826  -0.47918   3.99632  13   -0.8854
  7  -0.02120   1.04391  -1.55153  12   -0.3686
  8  -0.01076   0.09687  -2.54132  12    0.0605
  9  -0.01089  -1.27150  -2.16792  12   -0.3218
 10  -0.00298  -1.65330  -0.85918  12   -0.3127
 11   0.00491  -0.68604   0.17957  14    0.0341
 12   0.01826   0.47917  -3.99632  13   -0.8854
 13   0.04216  -2.08873   1.82300   2    0.2876
 14   0.01717   2.02877   2.93856   2    0.2869
 15   0.00235   2.70393   0.60964   2    0.2880
 16  -0.01829  -1.55554   4.08565   3    0.2963
 17   0.85291  -0.07684   4.49066   3    0.3173
 18  -0.91024  -0.07934   4.45594   3    0.3178
 19  -0.04216   2.08872  -1.82300   2    0.2876
 20  -0.01716  -2.02878  -2.93856   2    0.2869
 21  -0.00234  -2.70394  -0.60964   2    0.2880
 22   0.03763   1.55552  -4.08492   3    0.2963
 23   0.90013   0.06355  -4.46055   3    0.3173
 24  -0.86252   0.09345  -4.48701   3    0.3178
  0  nslav-u
  0  ncore-v
  0  nslav-v
159.0    0.0 volu-u,volu-v
 25  nstr-u
  1    2   1.370  4500.0  C-C ring
  1   11   1.418  4500.0
  2    3   1.418  4500.0
  3    4   1.363  4500.0
  4    5   1.419  4500.0
  5    7   1.418  4500.0
  5   11   1.418  4500.0
  7    8   1.370  4500.0
  8    9   1.418  4500.0
  9   10   1.363  4500.0
 10   11   1.419  4500.0
  2    6   1.505  3500.0 C-CMe
  8   12   1.505  3500.0
  1   13   1.080  3500.0  C-H ring
  3   14   1.080  3500.0
  4   15   1.080  3500.0
  7   19   1.080  3500.0
  9   20   1.080  3500.0
 10   21   1.080  3500.0
  6   16   1.080  3500.0  C-HMe
  6   17   1.080  3500.0
  6   18   1.080  3500.0
 12   22   1.080  3500.0
 12   23   1.080  3500.0
 12   24   1.080  3500.0
  0  nstr-v
 42  nbend-u
  2    1   11  120.0   1000.0 CCC ring
  3    4    5  120.0   1000.0
  5    7    8  120.0   1000.0
  9   10   11  120.0   1000.0
  1    2    3  120.0   1000.0
  2    3    4  120.0   1000.0
  7    8    9  120.0   1000.0
  8    9   10  120.0   1000.0
  4    5    7  122.0   1000.0 central C's
  1   11   10  122.0   1000.0
  4    5   11  119.0   1000.0
  7    5   11  119.0   1000.0
```

```
  1   11    5  119.0   1000.0
  5   11   10  119 0   1000.0
  1    2    6  120.0    900.0 C-C-Me
  3    2    6  120.0    900.0
  7    8   12  120.0    900.0
  9    8   12  120.0    900.0
  2    1   13  120.0    850.0 CCH ring
 11    1   13  120.0    850.0
  3    4   15  120.0    850.0
  5    4   15  120.0    850.0
  5    7   19  120.0    850.0
  8    7   19  120.0    850.0
  9   10   21  120.0    850.0
 11   10   21  120.0    850.0
  2    3   14  120.0    850.0
  4    3   14  120.0    850.0
  8    9   20  120.0    850.0
 10    9   20  120.0    850.0
  2    6   16  109.47   700.0  C-C-HMe
  2    6   17  109.47   700.0
  2    6   18  109.47   700.0
  8   12   22  109.47   700.0
  8   12   23  109.47   700.0
  8   12   24  109.47   700.0
 16    6   17  109.47   500.0  HMe-C-HMe
 16    6   18  109.47   500.0
 17    6   18  109.47   500.0
 22   12   23  109.47   500.0
 22   12   24  109.47   500.0
 23   12   24  109.47   500.0
  0  nbend-v
 23  ntors-u
 1  2  3  4  50.0 -1.0   1.0 CCCC ring T°=0
 2  3  4  5  50.0 -1.0   1.0
 3  4  5 11  50.0 -1.0   1.0
 4  5 11  1  50.0 -1.0   1.0
 5 11  1  2  50.0 -1.0   1.0
11  1  2  3  50.0 -1.0   1.0
11  5  7  8  50.0 -1.0   1.0
 5  7  8  9  50.0 -1.0   1.0
 7  8  9 10  50.0 -1.0   1.0
 8  9 10 11  50.0 -1.0   1.0
 9 10 11  5  50.0 -1.0   1.0
 1  2 11 13  100.0  -1.0  1.0
 2  3  1  6  100.0  -1.0  1.0
 3  4  2 14  100.0  -1.0  1.0
 4  5  3 15  100.0  -1.0  1.0
 5  7 11  4  100.0  -1.0  1.0
 7  8  5 19  100.0  -1.0  1.0
 8  9  7 12  100.0  -1.0  1.0
 9 10  8 20  100.0  -1.0  1.0
10 11  9 21  100.0  -1.0  1.0
11  1  5 10  100.0  -1.0  1.0
 1  2  6 16   5.0  1.0   3.0 C-C-C-H methyl
 7  8 12 22   5.0  1.0   3.0 C-C-C-H methyl
   0  ntors-v
   0  nlist-u
   0  nlist-v
  0.410   235.0   650.0   77000.0
   0  nintra
```

2,3-diazanaphthalene (phthalazine) (5)

Atomic point charges to be multiplied by the rescaling factor 0.41

```
  16
```

```
  1  -0.00902  -1.30207   1.23362  12    0.1735
  2   0.01014   1.29890   1.23086  12    0.1722
  3  -0.00968   1.38042  -1.24990  12   -0.2705
  4  -0.00401   0.69371  -2.41787  12   -0.2449
  5   0.00490  -0.69180  -2.42892  12   -0.2492
  6   0.00999  -1.37834  -1.26084  12   -0.2701
  7  -0.00299  -0.69066  -0.03346  14    0.0865
  8  -0.00077   0.68866  -0.04076  14    0.0909
  9  -0.00576  -0.68619   2.37542  18   -0.5900
 10   0.00626   0.68662   2.36872  18   -0.5839
 11  -0.01713  -2.38175   1.25801   2    0.2740
 12   0.02297   2.37851   1.25580   2    0.2732
 13  -0.02104   2.46037  -1.25627   2    0.2852
 14  -0.00650   1.23370  -3.35318   2    0.2832
 15   0.00780  -1.22434  -3.36843   2    0.2847
 16   0.02407  -2.45818  -1.27194   2    0.2851
  0  nslav-u
  0  ncore-v
  0  nslav-v
119.5    0.0 volu-u,volu-v
 17  nstr-u
  1    7   1.409  4500.0  ring C-C
  2    8   1.409  4500.0
  7    8   1.379  4500.0
  3    8   1.400  4500.0
  6    7   1.400  4500.0
  3    4   1.355  4500.0
  5    6   1.355  4500.0
  4    5   1.386  4500.0
  1    9   1.295  5500.0  C-N
  2   10   1.295  5500.0
  9   10   1.373  5500.0  N-N
  1   11   1.080  3500.0
  2   12   1.080  3500.0
  3   13   1.080  3500.0
  4   14   1.080  3500.0
  5   15   1.080  3500.0
  6   16   1.080  3500.0
  0  nstr-v
 26  nbend-u
  1    9   10  118.0  1000.0 CNN
  2   10    9  118.0  1000.0
  8    2   10  126.0  1000.0 NCC
  7    1    9  126.0  1000.0 CCC rings
  4    3    8  120.0  1000.0
  3    4    5  120.0  1000.0
  4    5    6  120.0  1000.0
  5    6    7  120.0  1000.0
  6    7    8  120.0  1000.0
  3    8    7  120.0  1000.0
  1    7    6  125.0  1000.0
  2    8    3  125.0  1000.0
  1    7    8  116.0  1000.0
  2    8    7  116.0  1000.0
  7    1   11  117.0   850.0  CCH
  9    1   11  117.0   850.0
  8    2   12  117.0   850.0
 10    2   12  117.0   850.0
  4    3   13  120.0   850.0
  8    3   13  120.0   850.0
  3    4   14  120.0   850.0
  5    4   14  120.0   850.0
  4    5   15  120.0   850.0
  6    5   15  120.0   850.0
  5    6   16  120.0   850.0
  7    6   16  120.0   850.0
  0  nbend-v
 19  ntors-u
1  9 10  2  50.0 -1.0   1.0
9 10  2  8  50.0 -1.0   1.0
```

```
10  2  8  7  50.0 -1.0   1.0
 2  8  7  1  50.0 -1.0   1.0
 8  7  1  9  50.0 -1.0   1.0
 7  1  9 10  50.0 -1.0   1.0
 7  8  3  4  50.0 -1.0   1.0
 8  3  4  5  50.0 -1.0   1.0
 3  4  5  6  50.0 -1.0   1.0
 4  5  6  7  50.0 -1.0   1.0
 5  6  7  8  50.0 -1.0   1.0
 1  7 11  9  100.0  -1.0  1.0
 2  8 12 10  100.0  -1.0  1.0
 3  4 13  8  100.0  -1.0  1.0
 4  5 14  3  100.0  -1.0  1.0
 5  6 15  4  100.0  -1.0  1.0
 6  7 16  5  100.0  -1.0  1.0
 7  6  8  1  100.0  -1.0  1.0
 8  3  2  7  100.0  -1.0  1.0
   0  ntors-v
   0  nlist-u
   0  nlist-v
  0.410   235.0   650.0   77000.0
   0  nintra
```

Methylurea (6)

Atomic point charges to be multiplied by the rescaling factor 0.41

```
 11
   1   0.00121  -1.30400  -0.47945  27   -1.2843
   2   0.01035   0.64993   0.65834  21   -0.6511
   3  -0.01806   0.64533  -1.61806  21   -0.9755
   4   0.00434  -0.05564  -0.47598  10    1.2133
   5  -0.00657   0.02736   1.95453  13   -0.6380
   6   0.78152  -0.70899   2.01059   3    0.2871
   7   0.14679   0.77753   2.71621   3    0.2881
   8  -0.96075  -0.45405   2.11023   3    0.2857
   9   0.02701   1.64259   0.53844   7    0.4870
  10   0.02762   1.64337  -1.57484   7    0.4930
  11   0.09215   0.13648  -2.47186   7    0.4947
   0  nslav-u
   0  ncore-v
   0  nslav-v
  70.3    0.0 volu-u,volu-v
  10  nstr-u
   1    4   1.250  8000.0
   2    4   1.340  4000.0
   2    5   1.440  3500.0
   3    4   1.340  4000.0
   2    9   1.000  5000.0
   3   10   1.000  5000.0
   3   11   1.000  5000.0
   5    6   1.080  3500.0
   5    7   1.080  3500.0
   5    8   1.080  3500.0
   0  nstr-v
  15  nbend-u
   1    4    2  122.0   900.0
   1    4    3  122.0   900.0
   2    4    3  116.0   900.0
   4    2    5  122.0   900.0
   4    2    9  115.0   940.0
   4    3   10  118.0   940.0
   4    3   11  118.0   940.0
   5    2    9  123.0   940.0
   6    5    7  109.47   530.0
   6    5    8  109.47   530.0
```

```
  7    5    8  109.47   530.0
  2    5    6  109.47   680.0
  2    5    7  109.47   680.0
  2    5    8  109.47   680.0
 10    3   11  124.0    600.0
  0  nbend-v
  6  ntors-u
8  5  2  4  3.0     1.0   3.0
9  2  4  1  50.0   -1.0   2.0
1  4  3 11  50.0   -1.0   2.0
3 10 11  4  100.0  -1.0  1.0
2  4  5  9  100.0  -1.0  1.0
4  2  1  3  100.0  -1.0  1.0
  0  ntors-v
  0  nlist-u
  0  nlist-v
 0.410   235.0   650.0   77000.0
  0  nintra
```

Methyl-2-amino-5-hydroxybenzoate (7)

Atomic point charges to be multiplied by the rescaling factor 0.41

```
 21
  1   0.09284   0.93302   2.37369  27   -1.1595
  2  -0.06234  -1.25649   2.01002  23   -0.7439
  3  -0.00023  -1.99477  -2.87718  29   -1.3795
  4   0.01809   2.61089   0.25757  17   -1.0967
  5   0.02218   0.14615   0.12364  12   -0.0781
  6   0.02032  -1.00167  -0.68631  12   -0.3206
  7   0.00971  -0.90283  -2.05591  12    0.6561
  8  -0.01866   0.35324  -2.65180  12   -0.2969
  9  -0.02716   1.48833  -1.87392  12   -0.3633
 10  -0.01666   1.41746  -0.47808  12    0.3495
 11   0.02817   0.01518   1.59164  10    1.3067
 12  -0.04059  -1.43618   3.42943  13   -0.4167
 13   0.02740  -1.97783  -0.22430   2    0.2964
 14  -0.03405   0.43814  -3.72840   2    0.2955
 15  -0.04230   2.45617  -2.35293   2    0.2903
 16  -0.76435  -0.77783   3.88675   3    0.2854
 17  -0.28580  -2.46097   3.66638   3    0.2843
 18   0.94482  -1.20531   3.80644   3    0.2859
 19  -0.38696   2.52326   1.16760   8    0.5024
 20  -0.26317   3.36189  -0.33983   8    0.4989
 21   0.34288  -2.79005  -2.37739   5    0.8040
  0  nslav-u
  0  ncore-v
  0  nslav-v
149.5    0.0 volu-u,volu-v
 21  nstr-u standards CLPdyn
  1   11   1.208  8000. C=O
  2   11   1.342  3500. C-O
  2   12   1.431  3500. C-O
  3    7   1.366  3500. C-O alc
  3   21   1.000  4200. O-H
  4   10   1.402  3200. C-N?
  4   19   1.000  5000. N-H
  4   20   1.000  5000.
  5    6   1.405  4500. C-Carom
  6    7   1.373  4500.
  7    8   1.391  4500.
  8    9   1.376  4500.
  9   10   1.398  4500.
  5   10   1.407  4500.
  5   11   1.474  3100. C-COOH
  6   13   1.080  3500. C-H arom
```

```
   8   14   1.080  3500.
   9   15   1.080  3500.
  12   16   1.080  3500. C-H methyl
  12   17   1.080  3500.
  12   18   1.080  3500.
   0  nstr-v
  32  nbend-u  standards CLPdyn
  11    2   12  115.30   900.0 C-O-C
   7    3   21  109.43   550.0 C-O-H alc
  10    4   20  108.59   900.0 CNH
  10    4   19  113.13   900.0 CNH
  19    4   20  119.71   500.0 HNH
   6    5   10  119.45  1000.0 ring
   6    5   11  120.11  1000.0
  10    5   11  120.42  1000.0
   5    6    7  121.08  1000.0
   5    6   13  119.46   850.0 CCH ring
   7    6   13  119.46   850.0
   3    7    6  122.82  1000.0
   3    7    8  117.66  1000.0
   6    7    8  119.51  1000.0
   7    8    9  120.20  1000.0
   7    8   14  119.90   850.0
   9    8   14  119.90   850.0
   8    9   10  121.51  1000.0
   8    9   15  119.25   850.0
  10    9   15  119.24   850.0
   4   10    5  122.95  1000.0 exocyclic NH2
   4   10    9  118.75  1000.0
   5   10    9  118.24  1000.0
   1   11    2  121.47   900.0 O-C-O
   1   11    5  125.29   900.0 O-C-C
   2   11    5  113.24   900.0
   2   12   16  109.47   700.0 C-C-H methyl
   2   12   17  109.47   700.0
   2   12   18  109.47   700.0
  16   12   17  109.47   500.0 H-C-H methyl
  16   12   18  109.47   500.0
  17   12   18  109.47   500.0
   0  nbend-v
  19  ntors-u
 1 11  5  6  50.0  1.0   1.0 O=C-C-C stiff
 1 11  2 12  50.0 -1.0   1.0 O=C-O-C stiff
11  2 12 16   5.0  1.0   3.0 C-O-C-H methyl **new
 5 10  9  8  50.0 -1.0   1.0 benzene ring stiff
10  5  6  7  50.0 -1.0   1.0
 8  7  6  5  50.0 -1.0   1.0
 9  8  7  6  50.0 -1.0   1.0
10  9  8  7  50.0 -1.0   1.0
 6  5 10  9  50.0 -1.0   1.0
 5 10  4 19  15.0 -1.0   1.0 C-C-N-H  180,check **new
 9 10  4 20  15.0 -1.0   1.0 C-C-N-H   0,check **new
 6  7  3 21  7.5  -1.0   1.0 C-C-O-H alc **new
 5 11 10  6 100.0 -1.0   1.0  improper
 6  5  7 13 100.0 -1.0   1.0
 7  8  6  3 100.0 -1.0   1.0
 8  9 14  7 100.0 -1.0   1.0
 9 10 15  8 100.0 -1.0   1.0
10  5  9  4 100.0 -1.0   1.0
11  1  5  2 100.0 -1.0   1.0
   0  ntors-v
   0  nlist-u
   0  nlist-v
  0.410   235.0   650.0   77000.0
   0  nintra
```

L-alanine (8)

Atomic point charges to be multiplied by the rescaling factor 0.41

```
   13
    1   0.03517   0.06833  -0.80562  10    1.3833
    2   0.38540  -0.24919   0.64996  13   -0.0133
    3  -0.55383  -1.30007   1.22261  13   -0.8666
    4   0.31851   0.98670   1.47235  16   -0.7034
    5  -0.57441   1.12119  -1.04878  27   -1.2121
    6   0.39526  -0.78183  -1.65833  27   -1.2392
    7   1.39721  -0.62411   0.69512   3    0.2969
    8  -0.41935  -2.23085   0.69160   3    0.3016
    9  -0.33346  -1.44773   2.26952   3    0.2989
   10  -1.57557  -0.96762   1.11321   3    0.2965
   11  -0.61775   1.41381   1.42932   8    0.4855
   12   0.51932   0.75554   2.45578   8    0.4849
   13   1.03522   1.64934   1.14347   8    0.4870
    0  nslav-u
    0  ncore-v
    0  nslav-v
 84.1    0.0 volu-u,volu-v
   12  nstr-u
    1    2   1.530  3100.0 O2C-C
    1    5   1.241  3500.0  C=O
    1    6   1.257  3500.0
    2    3   1.521  2800.0  C-C
    2    4   1.486  3500.0  C-N
    2    7   1.080  3600.0  C-H
    3    8   1.080  3600.0
    3    9   1.080  3600.0
    3   10   1.080  3600.0
    4   11   1.030  5300.0  N-H
    4   12   1.030  5300.0
    4   13   1.030  5300.0
    0  nstr-v
   21  nbend-u
    2    1    5  118.35   900.0
    2    1    6  116.06   900.0
    5    1    6  125.58   900.0
    3    2    4  109.78   900.0
    3    2    7  108.84   800.0
    4    2    7  107.92   800.0
    1    2    3  111.10   900.0
    1    2    4  110.09   900.0
    1    2    7  109.04   800.0
    2    3    8  109.47   800.0
    2    3    9  109.47   800.0
    2    3   10  109.47   800.0
    8    3    9  109.47   500.0
    8    3   10  109.47   500.0
    9    3   10  109.47   500.0
    2    4   11  111.26   900.0
    2    4   12  109.45   900.0
    2    4   13  109.09   900.0
   11    4   12  108.07   600.0
   11    4   13  110.63   600.0
   12    4   13  108.28   600.0
    0  nbend-v
    4  ntors-u
  6  1  2  3   2.5 -1.0   1.0 OCCC
 11  4  2  3   7.5  1.0   3.0 HNCC
  8  3  2  4   7.5  1.0   3.0 HCCN
  1  5  2  6  100.0 -1.0   1.0 improper
    0  ntors-v
    0  nlist-u
    0  nlist-v
  0.410   235.0   650.0   77000.0
    0  nintra
```

S4.2 Gromacs Topology files

See Section 2.2.2 in the main text. The atom numbering in the Gromacs input corresponds to that employed in CLP-*dyn*. For example, the 14th atom in the topology file of 1,4-cyclohexadiene corresponds to H14 in the CLP input, the 7th atom of maleic anhydride to O7, and so on. Refer to Figure S5 above for the atom numbering. Labels in the "atom" column are irrelevant. For each molecule, atom type, atomic point charges and masses; list of stretching, bending and torsional functions. The arbitrary name "SOLU" has been given to all residues.

1,4-cyclohexadiene (1)

```
; Include forcefield parameters
#include "./gromos54a7_atb.ff/forcefield.itp"

[ moleculetype ]
; Name   nrexcl
SOLU     6
[ atoms ]
;  nr  type  resnr  resid  atom  cgnr  charge    mass
    1  CPos    1    SOLU     C1    1    0.190  12.0110
    2     C    1    SOLU     C2    2   -0.215  12.0110
    3     C    1    SOLU     C3    3   -0.215  12.0110
    4  CPos    1    SOLU     C4    4    0.190  12.0110
    5     C    1    SOLU     C5    5   -0.215  12.0110
    6     C    1    SOLU     C6    6   -0.215  12.0110
    7    HC    1    SOLU     H7    7    0.010   1.0080
    8    HC    1    SOLU     H8    8    0.010   1.0080
    9    HC    1    SOLU     H9    9    0.110   1.0080
   10    HC    1    SOLU    H10   10    0.110   1.0080
   11    HC    1    SOLU    H11   11    0.010   1.0080
   12    HC    1    SOLU    H12   12    0.010   1.0080
   13    HC    1    SOLU    H13   13    0.110   1.0080
   14    HC    1    SOLU    H14   14    0.110   1.0080
; total charge of the molecule:  -0.000
[ bonds ]
;  ai   aj  funct   c0         c1
   10    3    2   0.1090   1.2300e+07
    3    4    2   0.1510   3.7279e+06
    3    2    2   0.1330   1.1800e+07
    4    5    2   0.1510   3.7279e+06
    4   12    2   0.1100   1.2100e+07
    4   11    2   0.1100   1.2100e+07
    5   13    2   0.1090   1.2300e+07
    5    6    2   0.1330   1.1800e+07
    6   14    2   0.1090   1.2300e+07
    6    1    2   0.1510   3.7279e+06
    1    8    2   0.1100   1.2100e+07
    1    7    2   0.1100   1.2100e+07
    1    2    2   0.1510   3.7279e+06
    2    9    2   0.1090   1.2300e+07
[ pairs ]
;  ai   aj  funct  ;  all 1-4 pairs but the ones excluded in GROMOS it
[ angles ]
;  ai   aj   ak  funct   angle     fc
   10    3    4    2    120.00   505.00
   10    3    2    2    120.00   505.00
    4    3    2    2    126.00   640.00
    3    4    5    2    111.00   530.00
    3    4   12    2    109.00  1680.51
    3    4   11    2    109.00  1680.51
    5    4   12    2    109.00  1680.51
    5    4   11    2    109.00  1680.51
   12    4   11    2    104.00   490.00
    4    5   13    2    120.00   505.00
    4    5    6    2    126.00   640.00
   13    5    6    2    120.00   505.00
    5    6   14    2    120.00   505.00
    5    6    1    2    126.00   640.00
   14    6    1    2    120.00   505.00
```

```
    6    1    8    2    109.00  1680.51
    6    1    7    2    109.00  1680.51
    6    1    2    2    111.00   530.00
    8    1    7    2    104.00   490.00
    8    1    2    2    109.00  1680.51
    7    1    2    2    109.00  1680.51
    3    2    1    2    126.00   640.00
    3    2    9    2    120.00   505.00
    1    2    9    2    120.00   505.00
[ dihedrals ]
; GROMOS improper dihedrals
;  ai   aj   ak   al  funct   angle     fc
    3   10    4    2    2      0.00   167.36
    2    3    1    9    2      0.00   167.36
    5    4   13    6    2      0.00   167.36
    6    5   14    1    2      0.00   167.36
[ dihedrals ]
;  ai   aj   ak   al  funct    ph0      cp     mult
    3    4    5    6    1    180.00     1.00    6
    4    3    2    1    1    180.00    41.80    2
    4    5    6    1    1    180.00    41.80    2
    5    6    1    2    1    180.00     1.00    6
    6    1    2    3    1    180.00     1.00    6
    2    3    4    5    1    180.00     1.00    6
[ exclusions ]
;  ai   aj  funct  ;  GROMOS 1-4 exclusions

; Include Position restraint file
#ifdef POSRES
#include "posre.itp"
#endif

[ system ]
; Name
1,4-cyclohexadiene crystal

[ molecules ]
; Compound        #mols
SOLU              300
```

Maleic anhydride (2)

```
; Include forcefield parameters
#include "./gromos54a7_atb.ff/forcefield.itp"

[ moleculetype ]
; Name   nrexcl
SOLU     6
[ atoms ]
;  nr  type  resnr  resid  atom  cgnr  charge    mass
    1    OA    1    SOLU     O1    1   -0.414  15.9994
    2  CAro    1    SOLU     C2    2    0.701  12.0110
    3  CAro    1    SOLU     C3    3   -0.217  12.0110
    4  CAro    1    SOLU     C4    4   -0.217  12.0110
    5  CAro    1    SOLU     C5    5    0.701  12.0110
    6 OEOpt    1    SOLU     O6    6   -0.477  15.9994
    7 OEOpt    1    SOLU     O7    7   -0.477  15.9994
    8    HC    1    SOLU     H8    8    0.200   1.0080
    9    HC    1    SOLU     H9    9    0.200   1.0080
; total charge of the molecule:   0.000
[ bonds ]
;  ai   aj  funct   c0         c1
    6    2    2   0.1210   2.7321e+07
    2    3    2   0.1480   5.7300e+06
    2    1    2   0.1390   8.6600e+06
    3    4    2   0.1330   1.1800e+07
    3    8    2   0.1090   1.2300e+07
    4    9    2   0.1090   1.2300e+07
    4    5    2   0.1480   5.7300e+06
    5    1    2   0.1390   8.6600e+06
    5    7    2   0.1210   2.7321e+07
[ pairs ]
;  ai   aj  funct  ;  all 1-4 pairs but the ones excluded in GROMOS it
[ angles ]
;  ai   aj   ak  funct   angle     fc
    6    2    3    2    126.00   640.00
```

```
    6    2    1    2    124.00   730.00
    3    2    1    2    109.50   520.00
    2    3    4    2    109.50   520.00
    2    3    8    2    120.00   505.00
    4    3    8    2    126.00   575.00
    3    4    9    2    126.00   575.00
    3    4    5    2    109.50   520.00
    9    4    5    2    120.00   505.00
    4    5    1    2    109.50   520.00
    4    5    7    2    126.00   640.00
    1    5    7    2    124.00   730.00
    2    1    5    2    109.50   450.00
[ dihedrals ]
; GROMOS improper dihedrals
;  ai   aj   ak   al  funct   angle     fc
    5    4    1    7    2      0.00   167.36
    2    6    3    1    2      0.00   167.36
    3    2    4    8    2      0.00   167.36
    4    3    9    5    2      0.00   167.36
[ dihedrals ]
;  ai   aj   ak   al  funct    ph0      cp     mult
    2    3    4    5    1    180.00    41.80    2
    3    2    1    5    1    180.00    41.80    2
    3    4    5    1    1    180.00    41.80    2
    4    5    1    2    1    180.00    41.80    2
    1    2    3    4    1    180.00    41.80    2
[ exclusions ]
;  ai   aj  funct  ;  GROMOS 1-4 exclusions

; Include Position restraint file
#ifdef POSRES
#include "posre.itp"
#endif

[ system ]
; Name
maleic anhydride crystal

[ molecules ]
; Compound        #mols
SOLU              360
```

Succinic anhydride (3)

```
; Include forcefield parameters
#include "./gromos54a7_atb.ff/forcefield.itp"

[ moleculetype ]
; Name   nrexcl
SOLU     6
[ atoms ]
;  nr  type  resnr  resid  atom  cgnr  charge    mass
    1    OA    1    SOLU     O7    1   -0.382  15.9994
    2 OEOpt    1    SOLU     O6    2   -0.510  15.9994
    3 OEOpt    1    SOLU     O1    3   -0.510  15.9994
    4     C    1    SOLU     C4    4   -0.151  12.0110
    5     C    1    SOLU     C3    5   -0.151  12.0110
    6  CPos    1    SOLU     C5    6    0.652  12.0110
    7  CPos    1    SOLU     C2    7    0.652  12.0110
    8    HC    1    SOLU    H11    8    0.100   1.0080
    9    HC    1    SOLU     H9    9    0.100   1.0080
   10    HC    1    SOLU     H8   10    0.100   1.0080
   11    HC    1    SOLU    H10   11    0.100   1.0080
; total charge of the molecule:   0.000
[ bonds ]
;  ai   aj  funct   c0         c1
    2    6    2   0.1210   2.7321e+07
    6    1    2   0.1390   8.6600e+06
    6    4    2   0.1520   5.4300e+06
    1    7    2   0.1390   8.6600e+06
    7    5    2   0.1520   5.4300e+06
    7    3    2   0.1210   2.7321e+07
    5   11    2   0.1090   1.2300e+07
    5   10    2   0.1090   1.2300e+07
    5    4    2   0.1530   7.1500e+06
    4    8    2   0.1090   1.2300e+07
```

```
    4    9    2   0.1090   1.2300e+07
[ pairs ]
;  ai   aj  funct  ;  all 1-4 pairs but the ones excluded in GROMOS it
[ angles ]
;  ai   aj   ak  funct   angle     fc
    2    6    1    2    124.00   730.00
    2    6    4    2    126.00   640.00
    1    6    4    2    109.50   520.00
    6    1    7    2    109.50   450.00
    1    7    5    2    109.50   520.00
    1    7    3    2    124.00   730.00
    5    7    3    2    126.00   640.00
    7    5   11    2    109.00  1680.51
    7    5   10    2    109.00  1680.51
    7    5    4    2    109.50   520.00
   11    5   10    2    107.57   484.00
   11    5    4    2    113.00   545.00
   10    5    4    2    113.00   545.00
    6    4    5    2    109.50   520.00
    6    4    8    2    109.00  1680.51
    6    4    9    2    109.00  1680.51
    5    4    8    2    113.00   545.00
    5    4    9    2    113.00   545.00
    8    4    9    2    107.57   484.00
[ dihedrals ]
; GROMOS improper dihedrals
;  ai   aj   ak   al  funct   angle     fc
    7    1    5    3    2      0.00   167.36
    6    2    1    4    2      0.00   167.36
[ dihedrals ]
;  ai   aj   ak   al  funct    ph0      cp     mult
    2    6    4    5    1    180.00     1.00    6
    6    1    7    3    1    180.00    24.00    2
    7    5    4    6    1    180.00     1.00    3
    4    6    1    7    1    180.00    24.00    2
    3    7    5    4    1    180.00     1.00    6
[ exclusions ]
;  ai   aj  funct  ;  GROMOS 1-4 exclusions

; Include Position restraint file
#ifdef POSRES
#include "posre.itp"
#endif

[ system ]
; Name
succinic anhydride crystal

[ molecules ]
; Compound        #mols
SOLU              360
```

2,6-dimetylnaphthalene (4)

```
Include forcefield parameters
#include "./gromos54a7_atb.ff/forcefield.itp"

[ moleculetype ]
; Name   nrexcl
SOLU    12
[ atoms ]
;  nr  type  resnr  resid  atom  cgnr  charge    mass
    1  CAro    1    SOLU     C1    1   -0.367  12.0110
    2  CAro    1    SOLU     C2    2    0.308  12.0110
    3  CAro    1    SOLU     C3    3   -0.289  12.0110
    4  CAro    1    SOLU     C4    4   -0.176  12.0110
    5  CAro    1    SOLU     C5    5    0.118  12.0110
    6     C    1    SOLU     C6    6   -0.385  12.0110
    7  CAro    1    SOLU     C7    7   -0.367  12.0110
    8  CAro    1    SOLU     C8    8    0.308  12.0110
    9  CAro    1    SOLU     C9    9   -0.289  12.0110
   10  CAro    1    SOLU    C10   10   -0.176  12.0110
   11  CAro    1    SOLU    C11   11    0.118  12.0110
   12     C    1    SOLU    C12   12   -0.385  12.0110
   13    HC    1    SOLU    H13   13    0.165   1.0080
   14    HC    1    SOLU    H14   14    0.161   1.0080
```

```
   15    HC    1    SOLU    H15   15    0.138   1.0080
   16    HC    1    SOLU    H16   16    0.109   1.0080
   17    HC    1    SOLU    H17   17    0.109   1.0080
   18    HC    1    SOLU    H18   18    0.109   1.0080
   19    HC    1    SOLU    H19   19    0.165   1.0080
   20    HC    1    SOLU    H20   20    0.161   1.0080
   21    HC    1    SOLU    H21   21    0.138   1.0080
   22    HC    1    SOLU    H22   22    0.109   1.0080
   23    HC    1    SOLU    H23   23    0.109   1.0080
   24    HC    1    SOLU    H24   24    0.109   1.0080
; total charge of the molecule:   0.000
[ bonds ]
;  ai   aj  funct   c0         c1
   17    6    2   0.1090   1.2300e+07
    6   16    2   0.1090   1.2300e+07
    6   18    2   0.1090   1.2300e+07
    6    2    2   0.1520   5.4300e+06
    2    3    2   0.1430   8.1800e+06
    2    1    2   0.1390   8.6600e+06
    3   14    2   0.1090   1.2300e+07
    3    4    2   0.1380   1.1000e+07
    4   15    2   0.1090   1.2300e+07
    4    5    2   0.1430   8.1800e+06
    5    7    2   0.1430   8.1800e+06
    5   11    2   0.1430   8.1800e+06
    7   19    2   0.1090   1.2300e+07
    7    8    2   0.1390   8.6600e+06
    8   12    2   0.1520   5.4300e+06
    8    9    2   0.1430   8.1800e+06
   12   22    2   0.1090   1.2300e+07
   12   24    2   0.1090   1.2300e+07
   12   23    2   0.1090   1.2300e+07
    9   20    2   0.1090   1.2300e+07
    9   10    2   0.1380   1.1000e+07
   10   21    2   0.1090   1.2300e+07
   10   11    2   0.1430   8.1800e+06
   11    1    2   0.1430   8.1800e+06
    1   13    2   0.1090   1.2300e+07
[ pairs ]
;  ai   aj  funct  ;  all 1-4 pairs but the ones excluded in GROMOS it
[ angles ]
;  ai   aj   ak  funct   angle     fc
   17    6   16    2    108.00   465.00
   17    6   18    2    108.00   465.00
   17    6    2    2    111.30   632.00
   16    6   18    2    108.00   465.00
   16    6    2    2    111.30   632.00
   18    6    2    2    111.30   632.00
    6    2    3    2    120.00   560.00
    6    2    1    2    120.00   560.00
    3    2    1    2    120.00   560.00
    2    3   14    2    120.00   505.00
    2    3    4    2    120.00   560.00
   14    3    4    2    120.00   505.00
    3    4   15    2    120.00   505.00
    3    4    5    2    120.00   560.00
   15    4    5    2    120.00   505.00
    4    5    7    2    120.00   560.00
    4    5   11    2    120.00   560.00
    7    5   11    2    120.00   560.00
    5    7   19    2    120.00   505.00
    5    7    8    2    120.00   560.00
   19    7    8    2    120.00   505.00
    7    8   12    2    120.00   560.00
    7    8    9    2    120.00   560.00
   12    8    9    2    120.00   560.00
    8   12   22    2    111.30   632.00
    8   12   24    2    111.30   632.00
    8   12   23    2    111.30   632.00
   22   12   24    2    108.00   465.00
   22   12   23    2    108.00   465.00
   24   12   23    2    108.00   465.00
    8    9   20    2    120.00   505.00
    8    9   10    2    120.00   560.00
   20    9   10    2    120.00   505.00
    9   10   21    2    120.00   505.00
    9   10   11    2    120.00   560.00
   21   10   11    2    120.00   505.00
    5   11   10    2    120.00   560.00
```

```
    5   11    1    2    120.00   560.00
   10   11    1    2    120.00   560.00
    2    1   11    2    120.00   560.00
    2    1   13    2    120.00   505.00
   11    1   13    2    120.00   505.00
[ dihedrals ]
; GROMOS improper dihedrals
;  ai   aj   ak   al  funct   angle     fc
    2    6    3    1    2      0.00   167.36
    1    2   11   13    2      0.00   167.36
   11    5   10    1    2      0.00   167.36
    5    4    7   11    2      0.00   167.36
    4    3   15    5    2      0.00   167.36
    3    2   14    4    2      0.00   167.36
    7    5   19    8    2      0.00   167.36
    8    7   12    9    2      0.00   167.36
    9    8   20   10    2      0.00   167.36
   10    9   21   11    2      0.00   167.36
[ dihedrals ]
;  ai   aj   ak   al  funct    ph0      cp     mult
   17    6    2    1    1    180.00     1.00    6
    2    3    4    5    1    180.00    41.80    2
    3    2    1   11    1    180.00    41.80    2
    3    4    5   11    1    180.00    41.80    2
    5    7    8    9    1    180.00    41.80    2
    5   11    1    2    1    180.00    41.80    2
    7    5   11   10    1    180.00    41.80    2
    7    8   12   23    1    180.00     1.00    6
    7    8    9   10    1    180.00    41.80    2
    8    9   10   11    1    180.00    41.80    2
    9   10   11    5    1    180.00    41.80    2
   11    5    7    8    1    180.00    41.80    2
    1    2    3    4    1    180.00    41.80    2
[ exclusions ]
;  ai   aj  funct  ;  GROMOS 1-4 exclusions

; Include Position restraint file
#ifdef POSRES
#include "posre.itp"
#endif

[ system ]
; Name
2,6-Dimethylnaphthalene

[ molecules ]
; Compound        #mols
SOLU              240
```

2,3-diazanaphthalene (phthalazine) (5)

```
; Include forcefield parameters
#include "./gromos54a7_atb.ff/forcefield.itp"

[ moleculetype ]
; Name   nrexcl
SOLU     12
[ atoms ]
;  nr  type  resnr  resid  atom  cgnr  charge    mass
    1  CAro    1    SOLU     C6    1    0.313  12.0110
    2  CAro    1    SOLU     C5    2    0.313  12.0110
    3  CAro    1    SOLU     C3    3   -0.344  12.0110
    4  CAro    1    SOLU     C2    4   -0.016  12.0110
    5  CAro    1    SOLU     C1    5   -0.016  12.0110
    6  CAro    1    SOLU     C8    6   -0.344  12.0110
    7  CAro    1    SOLU     C7    7    0.106  12.0110
    8  CAro    1    SOLU     C4    8    0.106  12.0110
    9  NOpt    1    SOLU     N2    9   -0.393  14.0067
   10  NOpt    1    SOLU     N1   10   -0.393  14.0067
   11    HC    1    SOLU     H5   11    0.016   1.0080
   12    HC    1    SOLU     H4   12    0.016   1.0080
   13    HC    1    SOLU     H3   13    0.198   1.0080
   14    HC    1    SOLU     H2   14    0.120   1.0080
   15    HC    1    SOLU     H1   15    0.120   1.0080
   16    HC    1    SOLU     H6   16    0.198   1.0080
; total charge of the molecule:   0.000
```

```
[ bonds ]
;  ai    aj  funct   c0         c1
   15     5     2   0.1090   1.2300e+07
    5     6     2   0.1390   8.6600e+06
    5     4     2   0.1420   3.2236e+06
    6    16     2   0.1090   1.2300e+07
    6     7     2   0.1410   6.5389e+06
    7     1     2   0.1430   8.1800e+06
    7     8     2   0.1420   3.2236e+06
    1    11     2   0.1090   1.2300e+07
    1     9     2   0.1320   1.2000e+07
    9    10     2   0.1380   4.4633e+06
   10     2     2   0.1320   1.2000e+07
    2    12     2   0.1090   1.2300e+07
    2     8     2   0.1430   8.1800e+06
    8     3     2   0.1410   6.5389e+06
    3    13     2   0.1090   1.2300e+07
    3     4     2   0.1390   8.6600e+06
    4    14     2   0.1090   1.2300e+07
[ pairs ]
;  ai    aj  funct  ;  all 1-4 pairs but the ones excluded in GROMOS it
[ angles ]
;  ai    aj    ak  funct   angle     fc
   15     5     6     2    120.00   505.00
   15     5     4     2    120.00   505.00
    6     5     4     2    120.00   560.00
    5     6    16     2    120.00   505.00
    5     6     7     2    120.00   560.00
   16     6     7     2    120.00   505.00
    6     7     1     2    126.00   640.00
    6     7     8     2    120.00   560.00
    1     7     8     2    120.00   560.00
    7     1    11     2    120.00   505.00
    7     1     9     2    126.00   640.00
   11     1     9     2    120.00   505.00
    1     9    10     2    119.00  2211.40
    9    10     2     2    119.00  2211.40
   10     2    12     2    120.00   505.00
   10     2     8     2    126.00   640.00
   12     2     8     2    120.00   505.00
    7     8     2     2    120.00   560.00
    7     8     3     2    120.00   560.00
    2     8     3     2    126.00   640.00
    8     3    13     2    120.00   505.00
    8     3     4     2    120.00   560.00
   13     3     4     2    120.00   505.00
    5     4     3     2    120.00   560.00
    5     4    14     2    120.00   505.00
    3     4    14     2    120.00   505.00
[ dihedrals ]
; GROMOS improper dihedrals
;  ai    aj    ak    al  funct   angle     fc
    5    15     6     4     2      0.00   167.36
    4     5     3    14     2      0.00   167.36
    3     8    13     4     2      0.00   167.36
    8     7     2     3     2      0.00   167.36
    2    10    12     8     2      0.00   167.36
    1     7    11     9     2      0.00   167.36
    7     6     1     8     2      0.00   167.36
    6     5    16     7     2      0.00   167.36
[ dihedrals ]
;  ai    aj    ak    al  funct    ph0       cp     mult
    5     6     7     8     1    180.00    41.80    2
    6     5     4     3     1    180.00    41.80    2
    7     1     9    10     1    180.00    41.80    2
    7     8     3     4     1    180.00    41.80    2
    1     7     8     2     1    180.00    41.80    2
    1     9    10     2     1    180.00    41.80    2
    9    10     2     8     1    180.00    41.80    2
   10     2     8     7     1    180.00    41.80    2
    8     7     1     9     1    180.00    41.80    2
    8     3     4     5     1    180.00    41.80    2
    4     5     6     7     1    180.00    41.80    2
[ exclusions ]
;  ai    aj  funct  ;  GROMOS 1-4 exclusions

; Include Position restraint file
#ifdef POSRES
#include "posre.itp"
```

```
#endif

[ system ]
; Name
2,3-diazanaphthalene

[ molecules ]
; Compound        #mols
SOLU              384
```

Methylurea (6)

```
; Include forcefield parameters
#include "./gromos54a7_atb.ff/forcefield.itp"

[ moleculetype ]
; Name   nrexcl
SOLU     6
[ atoms ]
;  nr  type  resnr  resid  atom  cgnr  charge    mass
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    2     N    1    SOLU     N2    2   -0.594  14.0067
    3  NPri    1    SOLU     N1    3   -0.923  14.0067
    4  CPos    1    SOLU     C1    4    0.769  12.0110
    5  CPos    1    SOLU     C2    5    0.029  12.0110
    6    HC    1    SOLU     H6    6    0.055   1.0080
    7    HC    1    SOLU     H4    7    0.055   1.0080
    8    HC    1    SOLU     H5    8    0.055   1.0080
    9  HS14    1    SOLU     H3    9    0.382   1.0080
   10  HS14    1    SOLU     H1   10    0.394   1.0080
   11  HS14    1    SOLU     H2   11    0.394   1.0080
; total charge of the molecule:   0.000
[ bonds ]
;  ai   aj  funct   c0         c1
    1    4    2   0.1230   1.6600e+07
    4    2    2   0.1360   1.0200e+07
    4    3    2   0.1390   8.6600e+06
    2    9    2   0.1010   2.1076e+07
    2    5    2   0.1450   5.2319e+06
    5    6    2   0.1090   1.2300e+07
    5    8    2   0.1090   1.2300e+07
    5    7    2   0.1090   1.2300e+07
    3   11    2   0.1010   2.1076e+07
    3   10    2   0.1010   2.1076e+07
[ pairs ]
;  ai   aj  funct  ;  all 1-4 pairs but the ones excluded in GROMOS it
[ angles ]
;  ai   aj   ak  funct   angle     fc
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    1    4    3    2    124.00   730.00
    2    4    3    2    114.00  1559.41
    4    2    9    2    116.00   465.00
    4    2    5    2    122.00   700.00
    9    2    5    2    116.00   465.00
    2    5    6    2    108.00   465.00
    2    5    8    2    108.00   465.00
    2    5    7    2    108.00   465.00
    6    5    8    2    108.53   443.00
    6    5    7    2    108.53   443.00
    8    5    7    2    108.53   443.00
    4    3   11    2    116.00   465.00
    4    3   10    2    116.00   465.00
   11    3   10    2    113.00   545.00
[ dihedrals ]
; GROMOS improper dihedrals
;  ai   aj   ak   al  funct   angle     fc
    4    1    2    3    2      0.00   167.36
[ dihedrals ]
;  ai   aj   ak   al  funct    ph0      cp     mult
    1    4    2    5    1    180.00    33.50    2
    1    4    3   10    1    180.00    33.50    2
    4    2    5    7    1      0.00     3.77    6
[ exclusions ]
;  ai   aj  funct  ;  GROMOS 1-4 exclusions
```

```
; Include Position restraint file
#ifdef POSRES
#include "posre.itp"
#endif

[ system ]
; Name
Methylurea crystal

[ molecules ]
; Compound        #mols
SOLU              400
```

Methyl-2-amino-5-hydroxybenzoate (7)

```
; Include forcefield parameters
#include "./gromos54a7_atb.ff/forcefield.itp"

[ moleculetype ]
; Name   nrexcl
SOLU     12
[ atoms ]
;  nr  type  resnr  resid  atom  cgnr  charge    mass
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    2    OA    1    SOLU     O2    2   -0.277  15.9994
    3  OAlc    1    SOLU     O3    3   -0.634  15.9994
    4  NPri    1    SOLU     N1    4   -0.882  14.0067
    5  CAro    1    SOLU     C1    5   -0.319  12.0110
    6  CAro    1    SOLU     C2    6   -0.309  12.0110
    7  CAro    1    SOLU     C3    7    0.393  12.0110
    8  CAro    1    SOLU     C4    8   -0.210  12.0110
    9  CAro    1    SOLU     C5    9   -0.302  12.0110
   10  CAro    1    SOLU     C6   10    0.439  12.0110
   11  CPos    1    SOLU     C7   11    0.762  12.0110
   12  CPos    1    SOLU     C8   12   -0.189  12.0110
   13    HC    1    SOLU     H4   13    0.136   1.0080
   14    HC    1    SOLU     H5   14    0.179   1.0080
   15    HC    1    SOLU     H6   15    0.177   1.0080
   16    HC    1    SOLU     H9   16    0.123   1.0080
   17    HC    1    SOLU     H8   17    0.123   1.0080
   18    HC    1    SOLU     H7   18    0.123   1.0080
   19  HS14    1    SOLU     H2   19    0.390   1.0080
   20  HS14    1    SOLU     H3   20    0.390   1.0080
   21  HS14    1    SOLU     H1   21    0.463   1.0080
; total charge of the molecule:   0.000
[ bonds ]
;  ai   aj  funct   c0         c1
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   11    5    2   0.1480   5.7300e+06
   11    2    2   0.1360   1.0200e+07
    5   10    2   0.1430   8.1800e+06
    5    6    2   0.1410   6.5389e+06
   10    9    2   0.1410   6.5389e+06
   10    4    2   0.1380   1.1000e+07
    9   15    2   0.1090   1.2300e+07
    9    8    2   0.1390   8.6600e+06
    8   14    2   0.1090   1.2300e+07
    8    7    2   0.1400   8.5400e+06
    7    6    2   0.1390   8.6600e+06
    7    3    2   0.1380   1.1000e+07
    6   13    2   0.1090   1.2300e+07
    3   21    2   0.0972   1.9581e+07
    4   20    2   0.1010   2.1076e+07
    4   19    2   0.1010   2.1076e+07
    2   12    2   0.1430   8.1800e+06
   12   16    2   0.1090   1.2300e+07
   12   17    2   0.1090   1.2300e+07
   12   18    2   0.1090   1.2300e+07
[ pairs ]
;  ai   aj  funct  ;  all 1-4 pairs but the ones excluded in GROMOS it
[ angles ]
;  ai   aj   ak  funct   angle     fc
    1   11    5    2    121.00   685.00
    1   11    2    2    124.00   730.00
```

```
    5   11    2    2    115.00   610.00
   11    5   10    2    120.00   560.00
   11    5    6    2    120.00   560.00
   10    5    6    2    120.00   560.00
    5   10    9    2    120.00   560.00
    5   10    4    2    126.00   640.00
    9   10    4    2    120.00   560.00
   10    9   15    2    120.00   505.00
   10    9    8    2    120.00   560.00
   15    9    8    2    120.00   505.00
    9    8   14    2    120.00   505.00
    9    8    7    2    120.00   560.00
   14    8    7    2    120.00   505.00
    8    7    6    2    120.00   560.00
    8    7    3    2    117.00   635.00
    6    7    3    2    121.00   685.00
    5    6    7    2    120.00   560.00
    5    6   13    2    120.00   505.00
    7    6   13    2    120.00   505.00
    7    3   21    2    109.50   450.00
   10    4   20    2    115.00   460.00
   10    4   19    2    115.00   460.00
   20    4   19    2    120.00   445.00
   11    2   12    2    116.00   620.00
    2   12   16    2    110.30   524.00
    2   12   17    2    110.30   524.00
    2   12   18    2    110.30   524.00
   16   12   17    2    109.50   448.00
   16   12   18    2    109.50   448.00
   17   12   18    2    109.50   448.00
[ dihedrals ]
; GROMOS improper dihedrals
;  ai   aj   ak   al  funct   angle     fc
    5   11   10    6    2      0.00   167.36
    6    5    7   13    2      0.00   167.36
    7    8    6    3    2      0.00   167.36
    8    9   14    7    2      0.00   167.36
    9   10   15    8    2      0.00   167.36
   10    5    9    4    2      0.00   167.36
   11    1    5    2    2      0.00   167.36
[ dihedrals ]
;  ai   aj   ak   al  funct    ph0      cp     mult
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    1   11    2   12    1    180.00    24.00    2
   11    2   12   18    1      0.00     1.26    3
    5   10    9    8    1    180.00    41.80    2
    5   10    4   19    1    180.00     5.86    2
   10    5    6    7    1    180.00    41.80    2
   10    9    8    7    1    180.00    41.80    2
    9   10    4   20    1    180.00     5.86    2
    9    8    7    6    1    180.00    41.80    2
    8    7    6    5    1    180.00    41.80    2
    6    5   10    9    1    180.00    41.80    2
    6    7    3   21    1    180.00    16.70    2
[ exclusions ]
;  ai   aj  funct  ;  GROMOS 1-4 exclusions

; Include Position restraint file
#ifdef POSRES
#include "posre.itp"
#endif

[ system ]
; Name
Methyl-2-amino-5-hydroxybenzoate crystal

[ molecules ]
; Compound        #mols
SOLU              384
```

L-alanine (8)

```
; Include forcefield parameters
#include "./gromos54a7_atb.ff/forcefield.itp"

[ moleculetype ]
```

```
; Name   nrexcl
SOLU     6
[ atoms ]
;  nr  type  resnr  resid  atom  cgnr  charge    mass
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    2  CPos    1    SOLU     C2    2    0.280  12.0110
    3     C    1    SOLU     C3    3   -0.390  12.0110
    4    NL    1    SOLU     N1    4   -0.513  14.0067
    5    OM    1    SOLU     O1    5   -0.680  15.9994
    6    OM    1    SOLU     O2    6   -0.680  15.9994
    7    HC    1    SOLU     H4    7    0.040   1.0080
    8    HC    1    SOLU     H6    8    0.113   1.0080
    9    HC    1    SOLU     H5    9    0.113   1.0080
   10    HC    1    SOLU     H7   10    0.113   1.0080
   11  HS14    1    SOLU     H1   11    0.339   1.0080
   12  HS14    1    SOLU     H2   12    0.339   1.0080
   13  HS14    1    SOLU     H3   13    0.339   1.0080
; total charge of the molecule:  -0.000
[ bonds ]
;  ai   aj  funct   c0         c1
    6    1    2   0.1250   1.3400e+07
    1    5    2   0.1250   1.3400e+07
    1    2    2   0.1570   2.4342e+06
    2    7    2   0.1090   1.2300e+07
    2    3    2   0.1520   5.4300e+06
    2    4    2   0.1520   5.4300e+06
    3    9    2   0.1090   1.2300e+07
    3    8    2   0.1090   1.2300e+07
    3   10    2   0.1090   1.2300e+07
    4   11    2   0.1020   1.7782e+07
    4   12    2   0.1020   1.7782e+07
    4   13    2   0.1020   1.7782e+07
[ pairs ]
;  ai   aj  funct  ;  all 1-4 pairs but the ones excluded in GROMOS it
[ angles ]
;  ai   aj   ak  funct   angle     fc
    6    1    5    2    126.00   770.00
    6    1    2    2    115.00   610.00
    5    1    2    2    115.00   610.00
    1    2    7    2    108.00   465.00
    1    2    3    2    111.00   530.00
    1    2    4    2    108.00   465.00
    7    2    3    2    110.30   524.00
    7    2    4    2    106.75   503.00
    3    2    4    2    111.00   530.00
    2    3    9    2    111.30   632.00
    2    3    8    2    111.30   632.00
    2    3   10    2    111.30   632.00
    9    3    8    2    108.53   443.00
    9    3   10    2    108.53   443.00
    8    3   10    2    108.53   443.00
    2    4   11    2    100.00   475.00
    2    4   12    2    115.00   460.00
    2    4   13    2    115.00   460.00
   11    4   12    2    111.00   530.00
   11    4   13    2    111.00   530.00
   12    4   13    2    111.00   530.00
[ dihedrals ]
; GROMOS improper dihedrals
;  ai   aj   ak   al  funct   angle     fc
    1    6    5    2    2      0.00   167.36
[ dihedrals ]
;  ai   aj   ak   al  funct    ph0      cp     mult
    1    2    3    9    1      0.00     5.92    3
    1    2    4   11    1    180.00     1.00    3
    5    1    2    3    1    180.00     1.00    6
[ exclusions ]
;  ai   aj  funct  ;  GROMOS 1-4 exclusions

; Include Position restraint file
#ifdef POSRES
#include "posre.itp"
#endif

[ system ]
; Name
L-alanine crystal

[ molecules ]
```

```
; Compound        #mols
SOLU              432
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S5. Equilibration

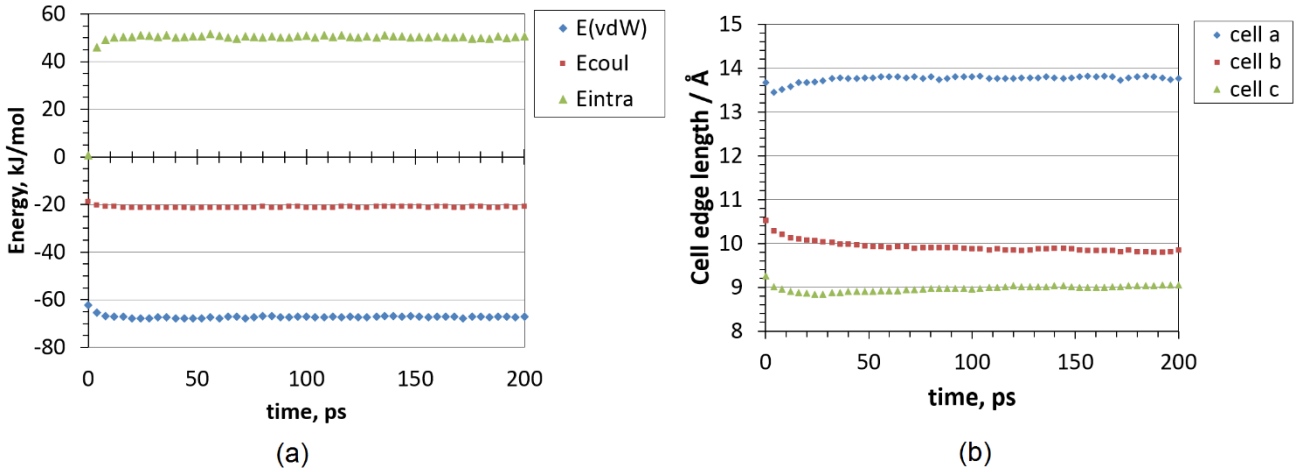

Figure S6.

See Section 2.2.2 in the main text. Change in energy components (a, left) and lattice parameters (b, right) of 2,3-diazanaphthalene in CLP-*dyncry* as a function of the simulation time. After ~50 ps the system is fully equilibrated. The other substances here studied show an essentially identical behaviour.

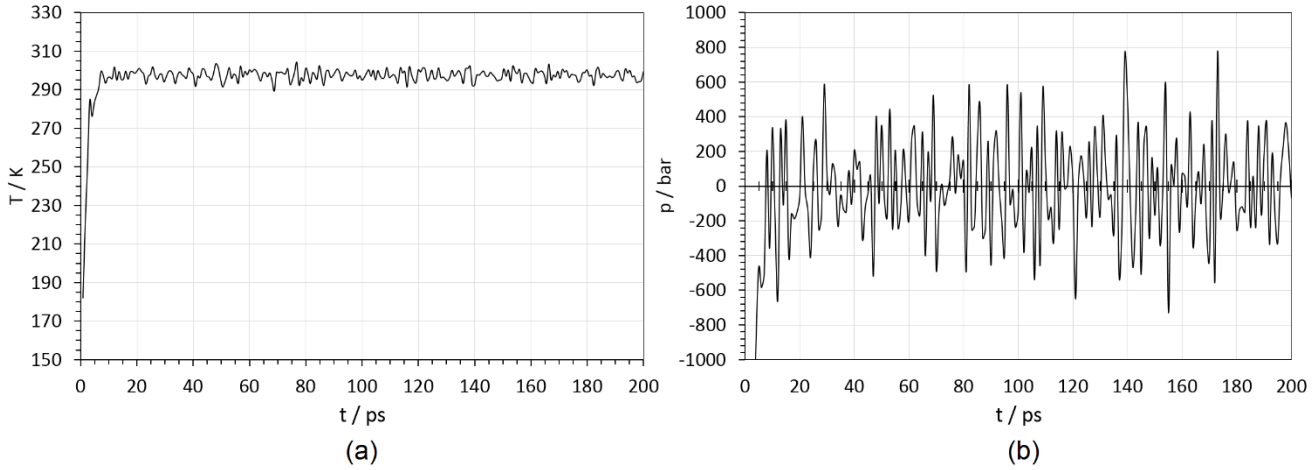

Figure S7.

See Section 2.2.2 in the main text. Temperature (a, left) and pressure (b, right) fluctuations of 2,3-diazanaphthalene in CLP-*dyncry* as a function of the simulation time. The other substances here studied show an essentially analogous behaviour.

S6. Equilibrium properties

See Table 1 (Section 3.1) and Figure 2 (Section 3.2) in the main text.

Table S4.

Crystal cell parameters (Å) and densities ($g \cdot cm^{-3}$), with percent deviation from experiment. First row, CLP-dyn, second row, G-dyn, third row, experimental. When available, experimental estimated standard deviations are reported.

Compound	ID[a]	*T* / K	*a* / Å	*b*/ Å	*c*/ Å	density / g·cm–3
Cyclohexadiene	(1)	153	7.168 +1.4 6.879 -2.7 7.070(3)	6.677 -6.9 6.975 -2.7 7.169(2)	9.775 -1.3 9.641 -2.7 9.909(2)	1.138 +7.4 1.151 +8.5 1.060
Maleic anhydride	(2)	298	6.872 -4.3 7.409 +3.2 7.180	11.794 +5.0 11.713 +4.3 11.231	5.261 -2.4 5.595 +3.8 5.390	1.528 +1.9 1.341 -10.5 1.499
Succinic anhydride	(3)	298	5.526 +1.8 5.501 +1.4 5.426(<1)	7.062 +1.2 7.022 +0.7 6.975(<1)	11.653 -0.5 11.893 +1.5 11.717(<1)	1.462 -2.5 1.447 -3.5 1.499
Succinic anhydride	(3)	225	5.477 +1.5 5.446 +1.7 5.395(<1)	6.973 +1.0 7.001 +1.4 6.907(<1)	11.590 -0.5 11.857 +1.8 11.649(<1)	1.502 -1.9 1.460 -4.6 1.531
Succinic anhydride	(3)	100	5.413 +1.1 5.446 +1.7 5.355(<1)	6.868 +0.8 6.905 +2.0 6.816(<1)	11.511 -0.4 11.775 +1.8 11.563(<1)	1.553 -1.4 1.491 -5.3 1.575
2,6-dimethylnaphthalene	(4)	298	7.095 -4.8 7.244 +2.8 7.454(2)	6.055 -0.4 6.160 -1.3 6.083(2)	20.229 +1.8 20.550 -2.3 20.095(5)	1.194 +3.9 1.132 +0.7
2,6-dimethylnaphthalene	(4)	143	6.883 -6.0 7.290 -0.5 7.323(1)	5.987 -0.6 5.990 -0.5 6.022(1)	20.077 +0.5 19.926 -0.3 19.985(5)	1.255 +6.6 1.193 +1.3 1.177
2,3-diazanaphthalene	(5)	298	13.780 +0.6 14.056 +2.6 13.695(5)	9.849 -6.7 10.837 +2.7 10.557(5)	9.015 -2.9 9.525 +5.7 9.285(5)	1.412 +8.9 1.192 -6.8 1.279
Methylurea	(6)	298	8.319 -1.9 8.960 +5.6 8.477(1)	7.100 +1.7 7.048 +1.0 6.981(1)	6.895 -0.4 7.244 +4.6 6.923(1)	1.208 +0.6 1.076 -10.3 1.200
Benzoate derivative	(7)	130	4.947 -0.5 5.150 +3.6 4.973(2)	11.377 +4.2 11.330 +3.7 10.923(2)	13.097 -6.9 14.588 +3.7 14.074(6)	1.506 +3.6 1.304 -10.3 1.453
L-alanine	(8)	298	6.002 -0.4 6.082 +1.0 6.025(7)	12.251 -0.7 12.590 +2.2 12.324(14)	5.824 +0.7 5.956 +3.0 5.783(6)	1.383 +0.9 1.279 -5.8 1.370
L-alanine	(8)	23	5.827 -1.7 6.006 +1.3 5.928(1)	12.060 -1,6 12.431 +1.4 12.260(1)	5.832 +0.7 5.881 +1.5 5.794(<1)	1.444 +2.8 1.348 -4.0 1.405

[a] See Scheme 1 and Figure 1 in the main text

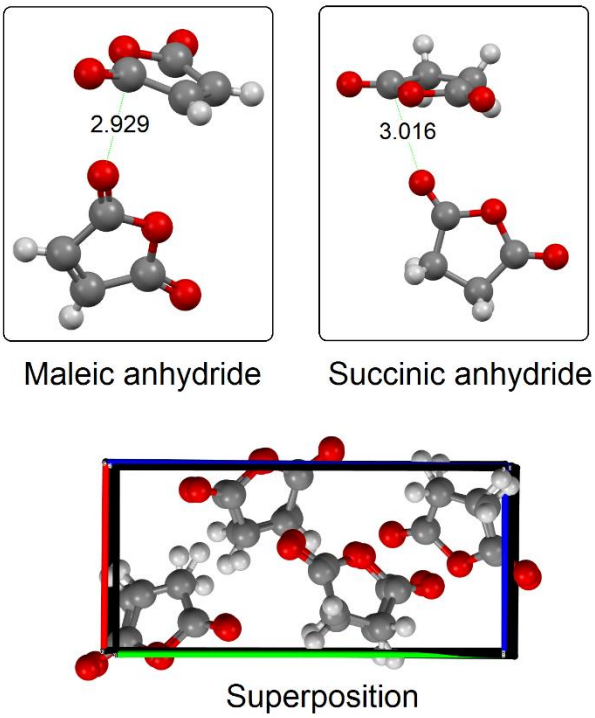

Figure S8.

See Figure 2 in the main text and Table S4 above. Comparison of maleic and succinic anhydride short contacts and crystal packings (distances in Å). When drawing the structure superposition, the maleic anhydride crystal was reflected into the (*a*,*b*) plane to get the same handedness as the succinic anhydride structure.

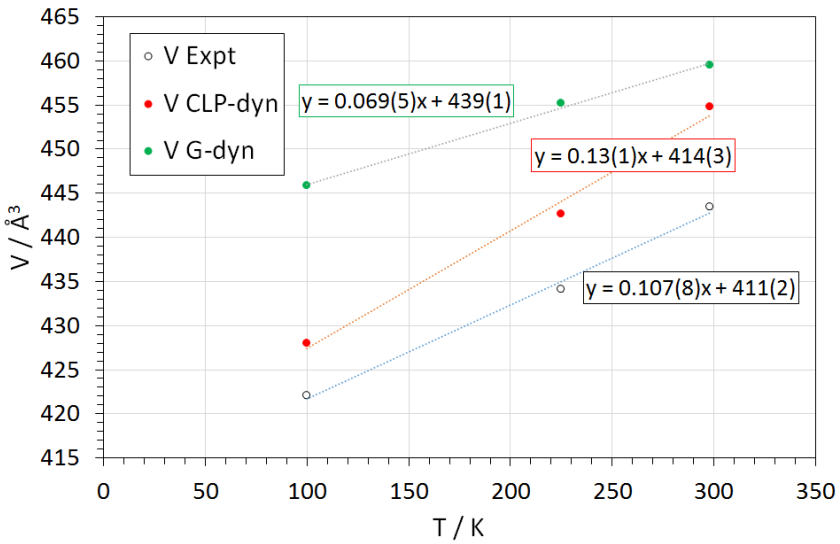

Figure S9.

See Table S4 above and Figure 2 in the main text. Cell volume of succinic anhydride as a function of *T*. Experimental values (white points) are compared with MD estimates from CLP-*dyncry* (red points, $1/V(dV/dT) \sim 2.8\cdot10^{-4}$ K^{-1} and G-*dyn* (green points, $1/V(dV/dT) \sim 1.7\cdot10^{-4}$ K^{-1}). The experimental estimate of $1/V(dV/dT)$ is $2.6\cdot10^{-4}$ K^{-1}.

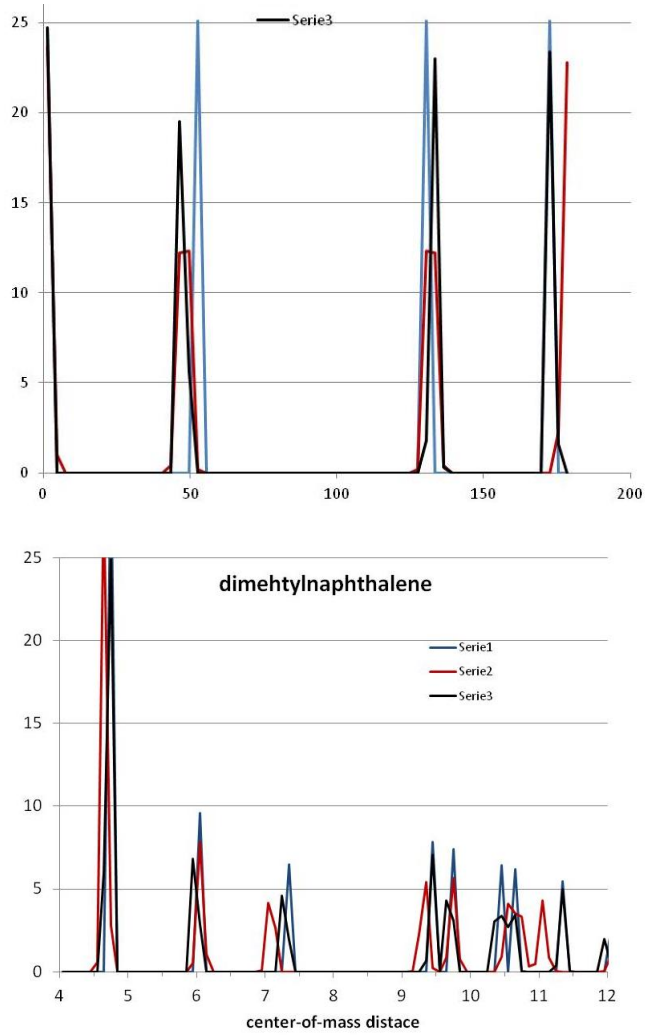

Figure S10. See Section 3.3.3 in the main text. 2,6-Dimethylnaphthalene crystal. Top: distribution of angles between central C-C vectors, bottom: c.o.m. RDF. Blue, crystal; red, CLP-*dyncry* simulation, black: G-*dyn* simulation.

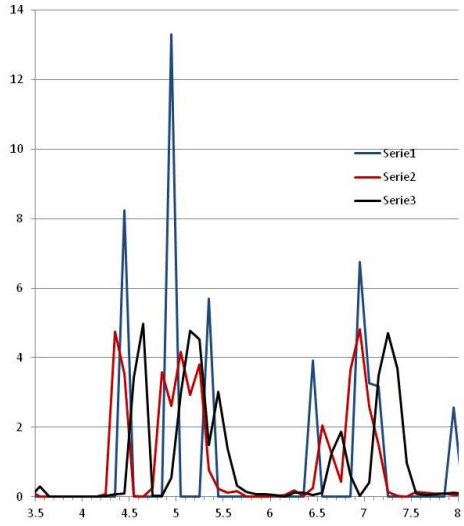

Figure S11. See Section 3.3.4 in the main text. Methylurea crystal, c.o.m. RDF. Blue, crystal; red, CLP-*dyncry* simulation, black: G-*dyn* simulation.

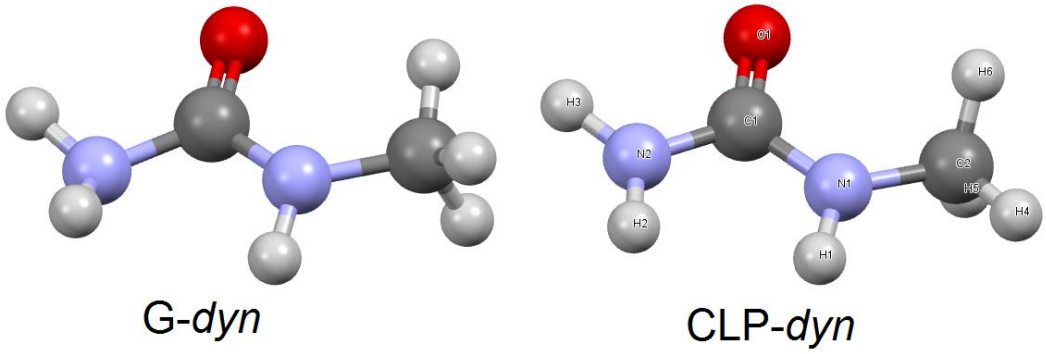

Figure S12.

See Section 3.3.4 in the main text. Methylurea conformers used as reference structures in G-*dyn* (left) and CLP-*dyncry* (right) simulations. The different pyramidalization of the $–NH_2$ group and the different torsion at the terminal $–CH_3$ group are evident.

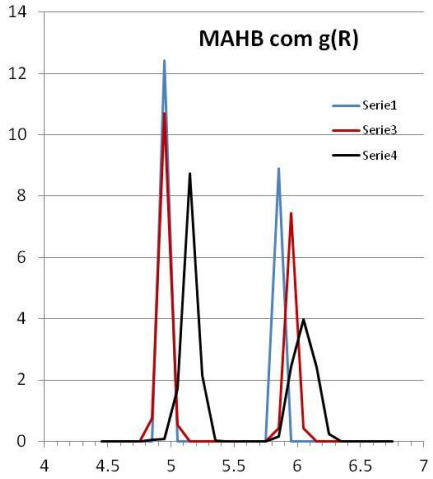

Figure S13.

See Section 3.3.4 and Figure 10 in the main text. Comparison of centre of mass RDF's from G-*dyn* and CLP-*dyncry* calculations to the experimental one for the benzoate derivative (7). Blue, crystal; red, CLP-*dyncry*, black: G-*dyn* simulation.

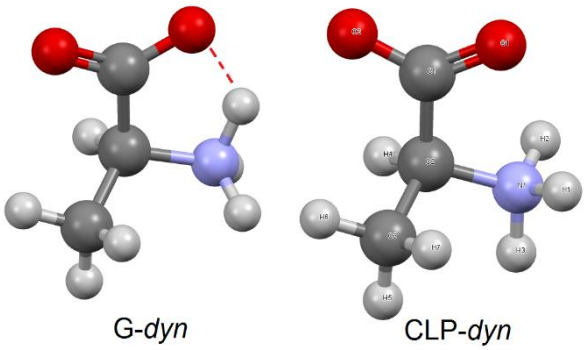

Figure S14.

See Section 3.3.4 and Figure 11 in the main text. L-alanine conformers used as reference structures in G-*dyn* (left) and CLP-*dyncry* (right) simulations. The intramolecular hydrogen bond between oppositely charged carboxy and ammonium groups (dashed red line) is present only in the gas-phase optimized geometry.