Using Molecular Dynamics to simulate realistic structures of nitrocellulose of different nitration levels

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1 Supplementary information

1.1 Additional Parameters for OPLS-aa

Additional parameters were needed to describe the nitrate groups of nitrocellulose. The values illustrated in figure 1 were found using LigParGen to obtained parameters for HNO_3.
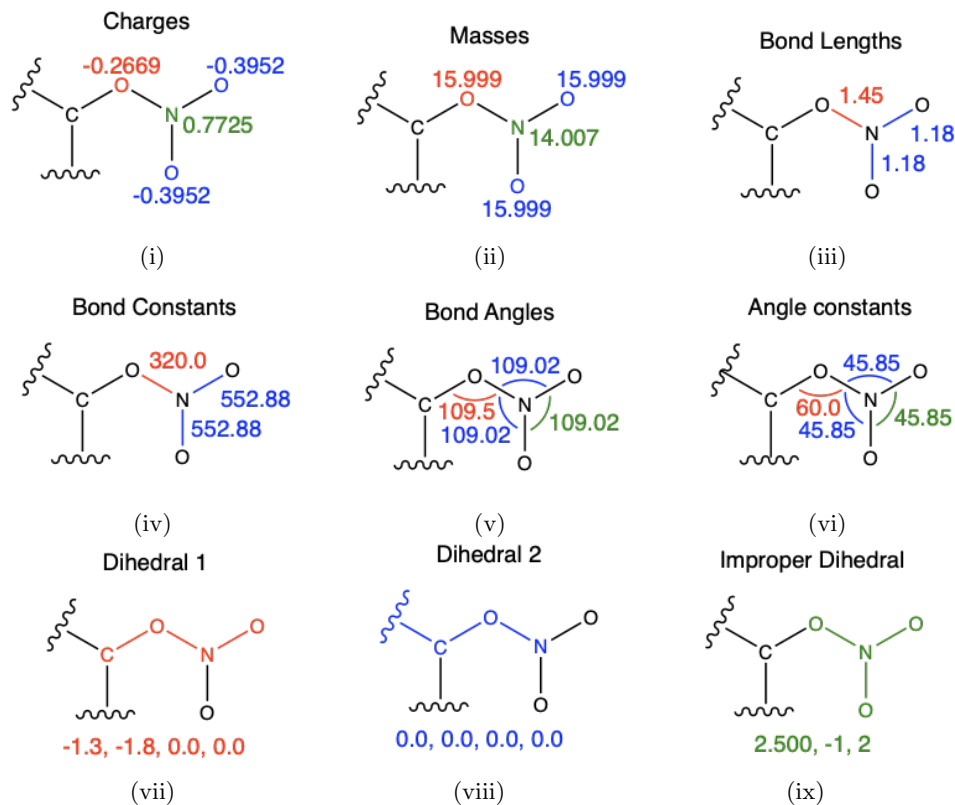
Figure 1: Parameters for ONO_2 groups

To add these parameters to the forcefield, the following lines were added to the oplsaa.lt file provided on the MolTemplate GitHub.

```
OPLSAA {
    write_once("In Init") {
    ...
    improper_style cvff
    ...
    } #(end of init parameters
    write_once("In Charges") {
        ...
        set type @atom:907 charge   0.7725    # N
        set type @atom:908 charge  -0.3952    # -O
        set type @atom:909 charge  -0.2669    # -O-
        ...
    } #(end of atom partial charges)

    write_once("Data Masses") {
        ...
        @atom:907 14.007
```

```
18        @atom:908 15.999
19        @atom:909 15.999
20        ...
21    } #(end of atom masses)
22
23    ...
24    replace{ @atom:907 @atom:907_b111_a111_d111_i111 }
25    replace{ @atom:908 @atom:908_b112_a112_d112_i112 }
26    replace{ @atom:909 @atom:909_b020_a020_d020_i020 }
27    ...
28
29    write_once("In Settings") {
30        ...
31        pair_coeff @atom:907_b111_a111_d111_i111 @atom:907
    _b111_a111_d111_i111 0.17 3.3
32        pair_coeff @atom:908_b112_a112_d112_i112 @atom:908
    _b112_a112_d112_i112 0.17 2.96
33        pair_coeff @atom:909_b020_a020_d020_i020 @atom:909
    _b020_a020_d020_i020 0.14 2.9
34        ...
35    } #(end of pair_coeffs)
36
37    write_once("In Settings") {
38        ...
39        bond_coeff @bond:020_111 320.00 1.45
40        bond_coeff @bond:111_112 552.88 1.18
41        ...
42    } #(end of bond_coeffs)
43
44    write_once("Data Bonds By Type") {
45        ...
46        @bond:020_111 @atom:*_b020_a*_d*_i* @atom:*_b111_a*_d*_i*
47        @bond:111_112 @atom:*_b111_a*_d*_i* @atom:*_b112_a*_d*_i*
48        ...
49    } #(end of bonds by type)
50
51    write_once("In Settings") {
52        ...
53        angle_coeff @angle_013_020_111  60.00 190.50
54        angle_coeff @angle_020_111_112  45.85 109.02
55        angle_coeff @angle_112_111_112. 45.85 109.02
56        ...
57    } #(end of angle_coeffs)
58
59    write_once("Data Angles By Type") {
60        ...
61        @angle:013_020_111 @atom:*_b*_a013_d*_i* @atom:*_b*_a020_d*_i*
    @atom:*_b*_a111_d*_i*
62        @angle:020_111_112 @atom:*_b*_a020_d*_i* @atom:*_b*_a111_d*_i*
    @atom:*_b*_a112_d*_i*
63        @angle:112_111_112 @atom:*_b*_a112_d*_i* @atom:*_b*_a111_d*_i*
    @atom:*_b*_a112_d*_i*
64        ...
65    } #(end of angles by type)
66
67    write_once("In Settings") {
```

```
68        ...
69        dihedral_coeff @dihedral:112_111_020_013 -1.3 -1.8  0.0   0.0
70        dihedral_coeff @dihedral:111_020_013_046  0.0  0.0  0.0   0.0
71        ...
72    } #(end of dihedral_coeffs)
73
74    write_once("Data Dihedrals By Type") {
75        ...
76        @dihedral:112_111_020_013 @atom:*_b*_a*_d112_i* @atom:*_b*_a*
      _d111_i* @atom:*_b*_a*_d020_i* @atom:*_b*_a*_d013_i*
77        @dihedral:111_020_013_046 @atom:*_b*_a*_d111_i* @atom:*_b*_a*
      _d020_i* @atom:*_b*_a*_d013_i* @atom:*_b*_a*_d046_i*
78        ...
79    } #(end of dihedrals by type)
80
81    write_once("In Settings") {
82        ...
83        improper_coeff @improper:020_112_111_112 2.500 -1 2 # (
      moltemplate)
84        ...
85    } #(end of improper_coeffs)
86
87    write_once("Data Impropers By Type (opls_imp.py)") {
88        ...
89        @improper:020_112_111_112 @atom:*_b*_a*_d*_i020* @atom:*_b*_a*_d*
      _i112* @atom:*_b*_a*_d*_i111* @atom:*_b*_a*_d*_i112*
90        ...
91    } #(end of impropers by type)
92 }
```

Listing 1: Lines added to OPLSAA file provided on the Moltemplete GitHub

1.2   Moltemplate input file

The structure was built up using Moltemplate, building units into chains, then planes and then the whole structures. The unit shown here is a "master" unit. This means that it has both nitrated and non-nitrated functional groups for positions a through to f. These are denoted by comments including *Cell* or *NCell*. Once it has been decided whether a particular position is nitrated or not, the relevant lines are kept and unnecessary ones deleted - eg. if position **a** is to be nitrated, all lines containing

```
1     # aCell
```

should be deleted, and

```
1     # aNCell
```

be kept, and vice versa for the

The force-field file "my_oplsaa.lt" is the force-field file provided for LAMMPS with the added lines described in section 1.1. The bonds within this module are also listed. When chains are being created there also needs to be a bond between 07C and 04O of neighbouring units.

1.2 Moltemplate input file

```
1  import "my_oplsaa.lt"
2
3  cUnit_xyz inherits OPLSAA {
4
5    write('Data Atoms') {
6      $atom:01C   $mol @atom:135  0.00    0.148  -0.342   0.449
7      $atom:02C   $mol @atom:100  0.00   -0.032  -1.499  -0.536   # aCell
8      $atom:02C   $mol @atom:125  0.00   -0.032  -1.499  -0.536   # aNCell
9      $atom:03C   $mol @atom:100  0.00    0.439  -1.116  -1.918   # bCell
10     $atom:03C   $mol @atom:125  0.00    0.439  -1.116  -1.918   # bNCell
11     $atom:04C   $mol @atom:911  0.00   -0.082   0.244  -2.336
12     $atom:05C   $mol @atom:125  0.00    0.055   1.296  -1.296
13     $atom:06C   $mol @atom:99   0.00   -0.662   2.599  -1.588   # cCell
14     $atom:01O   $mol @atom:122  0.00   -0.535  -0.701   1.636
15     $atom:02O   $mol @atom:96   0.00    0.766  -2.559  -0.003   # aCell
16     $atom:02O   $mol @atom:909  0.00    0.766  -2.559  -0.003   # aNCell
17     $atom:03O   $mol @atom:96   0.00    0.032  -2.127  -2.864   # bCell
18     $atom:03O   $mol @atom:909  0.00    0.032  -2.127  -2.864   # bNCell
19     $atom:04O   $mol @atom:122  0.00    0.535   0.701  -3.554
20     $atom:05O   $mol @atom:122  0.00   -0.528   0.807  -0.055
21     $atom:06O   $mol @atom:96   0.00   -0.001   3.284  -2.637   # cCell
22     $atom:06O   $mol @atom:909  0.00   -2.049   2.357  -1.743   # cNCell
23     $atom:01H   $mol @atom:136  0.00    1.093  -0.153   0.621
24     $atom:02H   $mol @atom:118  0.00   -0.972  -1.774  -0.567   # aCell
25     $atom:02H   $mol @atom:127  0.00   -0.972  -1.774  -0.567   # aNCell
26     $atom:03H   $mol @atom:118  0.00    1.775  -1.075  -1.898   # bCell
27     $atom:03H   $mol @atom:127  0.00    1.775  -1.075  -1.898   # bNCell
28     $atom:04H   $mol @atom:127  0.00   -1.041   0.143  -2.517
29     $atom:05H   $mol @atom:127  0.00    1.004   1.484  -1.131
30     $atom:06H   $mol @atom:118  0.00   -1.576   2.411  -1.846   # cCell
31     $atom:06H   $mol @atom:127  0.00   -0.518   3.215  -0.855   # cNCell
32     $atom:07H   $mol @atom:118  0.00   -0.676   3.155  -0.794   # cCell
33     $atom:07H   $mol @atom:118  0.00   -0.308   2.998  -2.398   # cNCell
34     $atom:08H   $mol @atom:97   0.00    0.581  -2.603   0.955   # aCell
35     $atom:01N   $mol @atom:907  0.00    2.168  -2.028   0.042   # aNCell
36     $atom:12O   $mol @atom:908  0.00    3.221  -2.839   0.608   # aNCell
37     $atom:13O   $mol @atom:908  0.00    2.467  -0.713  -0.474   # aNCell
38     $atom:09H   $mol @atom:97   0.00    0.593  -1.956  -3.651   # bCell
39     $atom:02N   $mol @atom:907  0.00   -1.464  -2.232  -2.870   # bNCell
40     $atom:14O   $mol @atom:908  0.00   -2.196  -1.555  -1.825   # bNCell
41     $atom:15O   $mol @atom:908  0.00   -2.172  -3.016  -3.856   # bNCell
42     $atom:11H   $mol @atom:97   0.00   -0.569   4.012  -2.958   # cCell
43     $atom:03N   $mol @atom:907  0.00   -2.822   3.298  -0.867   # cNCell
44     $atom:16O   $mol @atom:908  0.00   -4.266   3.269  -0.875   # cNCell
45     $atom:17O   $mol @atom:908  0.00   -2.123   4.234  -0.017   # cNCell
46     $atom:07C   $mol @atom:135  0.00   -0.148   0.342   5.639
47     $atom:08C   $mol @atom:100  0.00    0.032   1.499   4.654   # dCell
48     $atom:08C   $mol @atom:125  0.00    0.032   1.499   4.654   # dNCell
49     $atom:09C   $mol @atom:100  0.00   -0.439   1.116   3.272   # eCell
50     $atom:09C   $mol @atom:125  0.00   -0.439   1.116   3.272   # eNCell
51     $atom:10C   $mol @atom:911  0.00    0.082  -0.244   2.855
52     $atom:11C   $mol @atom:125  0.00   -0.055  -1.296   3.911
53     $atom:12C   $mol @atom:99   0.00    0.662  -2.599   3.602   # fCell
54     $atom:12C   $mol @atom:124  0.00    0.662  -2.599   3.602   # fNCell
55     $atom:08O   $mol @atom:96   0.00   -0.766   2.559   5.187   # dCell
56     $atom:08O   $mol @atom:909  0.00   -0.766   2.559   5.187   # dNCell
```

```
57    $atom:09O    $mol @atom:96    0.00    -0.032    2.127    2.326    # eCell
58    $atom:09O    $mol @atom:909   0.00    -0.032    2.127    2.326    # eNCell
59    $atom:10O    $mol @atom:122   0.00     0.528   -0.807    5.135
60    $atom:11O    $mol @atom:96    0.00     0.001   -3.284    2.553    # fCell
61    $atom:11O    $mol @atom:909   0.00     2.064   -2.394    3.586    # fNCell
62    $atom:12H    $mol @atom:136   0.00    -1.093    0.153    5.811
63    $atom:13H    $mol @atom:118   0.00     0.972    1.774    4.623    # dCell
64    $atom:13H    $mol @atom:127   0.00     0.972    1.774    4.623    # dNCell
65    $atom:14H    $mol @atom:118   0.00    -1.420    1.078    3.280    # eCell
66    $atom:14H    $mol @atom:127   0.00    -1.420    1.078    3.280    # eNCell
67    $atom:15H    $mol @atom:127   0.00     1.041   -0.143    2.673
68    $atom:16H    $mol @atom:127   0.00    -1.004   -1.484    4.059
69    $atom:17H    $mol @atom:118   0.00     1.576   -2.411    3.334    # fCell
70    $atom:17H    $mol @atom:127   0.00     0.444   -3.258    4.280    # fNCell
71    $atom:18H    $mol @atom:118   0.00     0.676   -3.155    4.396    # fCell
72    $atom:18H    $mol @atom:127   0.00     0.371   -2.929    2.738    # fNCell
73    $atom:20H    $mol @atom:97    0.00    -0.581    2.603    6.145    # dCell
74    $atom:04N    $mol @atom:907   0.00     0.122    3.697    5.596    # dNCell
75    $atom:18O    $mol @atom:908   0.00     1.550    3.647    5.386    # dNCell
76    $atom:19O    $mol @atom:908   0.00    -0.478    4.857    6.212    # dNCell
77    $atom:21H    $mol @atom:97    0.00    -0.593    1.956    1.539    # eCell
78    $atom:05N    $mol @atom:907   0.00     1.460    2.096    2.172    # eNCell
79    $atom:20O    $mol @atom:908   0.00     2.288    2.961    1.184    # eNCell
80    $atom:21O    $mol @atom:908   0.00     2.288    1.214    2.961    # eNCell
81    $atom:22H    $mol @atom:97    0.00     0.569   -4.012    2.232    # fCell
82    $atom:06N    $mol @atom:907   0.00     2.518   -1.944    4.943    # fNCell
83    $atom:22O    $mol @atom:908   0.00     1.528   -1.818    5.986    # fNCell
84    $atom:23O    $mol @atom:908   0.00     3.906   -1.660    5.226    # fNCell
85   }
86
87   write('Data Bond List') {
88     $bond:01C02C        $atom:01C        $atom:02C
89     $bond:01C01O        $atom:01C        $atom:01O
90     $bond:01C05O        $atom:01C        $atom:05O
91     $bond:01C01H        $atom:01C        $atom:01H
92     $bond:02C03C        $atom:02C        $atom:03C
93     $bond:02C02O        $atom:02C        $atom:02O
94     $bond:02C02H        $atom:02C        $atom:02H
95     $bond:03C04C        $atom:03C        $atom:04C
96     $bond:03C03O        $atom:03C        $atom:03O
97     $bond:03C03H        $atom:03C        $atom:03H
98     $bond:04C05C        $atom:04C        $atom:05C
99     $bond:04C04O        $atom:04C        $atom:04O
100    $bond:04C04H        $atom:04C        $atom:04H
101    $bond:05C06C        $atom:05C        $atom:06C
102    $bond:05C05O        $atom:05C        $atom:05O
103    $bond:05C05H        $atom:05C        $atom:05H
104    $bond:06C06O        $atom:06C        $atom:06O
105    $bond:06C06H        $atom:06C        $atom:06H
106    $bond:06C07H        $atom:06C        $atom:07H
107    $bond:02O08H        $atom:02O        $atom:08H        # aCell
108    $bond:02O01N        $atom:02O        $atom:01N        # aNCell
109    $bond:01N12O        $atom:01N        $atom:12O        # aNCell
110    $bond:01N13O        $atom:01N        $atom:13O        # aNCell
111    $bond:03O09H        $atom:03O        $atom:09H        # bCell
112    $bond:03O02N        $atom:03O        $atom:02N        # bNCell
```

```
113        $bond:02N14O           $atom:02N           $atom:14O           # bNCell
114        $bond:02N15O           $atom:02N           $atom:15O           # bNCell
115        $bond:06011H           $atom:06O           $atom:11H           # cCell
116        $bond:06003N           $atom:06O           $atom:03N           # cNCell
117        $bond:03N16O           $atom:03N           $atom:16O           # cNCell
118        $bond:03N17O           $atom:03N           $atom:17O           # cNCell
119        $bond:07C08C           $atom:07C           $atom:08C
120        $bond:07C10O           $atom:07C           $atom:10O
121        $bond:07C12H           $atom:07C           $atom:12H
122        $bond:08C09C           $atom:08C           $atom:09C
123        $bond:08C08O           $atom:08C           $atom:08O
124        $bond:08C13H           $atom:08C           $atom:13H
125        $bond:09C10C           $atom:09C           $atom:10C
126        $bond:09C09O           $atom:09C           $atom:09O
127        $bond:09C14H           $atom:09C           $atom:14H
128        $bond:10C11C           $atom:10C           $atom:11C
129        $bond:10C01O           $atom:10C           $atom:01O
130        $bond:10C15H           $atom:10C           $atom:15H
131        $bond:11C12C           $atom:11C           $atom:12C
132        $bond:11C10O           $atom:11C           $atom:10O
133        $bond:11C16H           $atom:11C           $atom:16H
134        $bond:12C11O           $atom:12C           $atom:11O
135        $bond:12C17H           $atom:12C           $atom:17H
136        $bond:12C18H           $atom:12C           $atom:18H
137        $bond:08020H           $atom:08O           $atom:20H           # dCell
138        $bond:08004N           $atom:08O           $atom:04N           # dNCell
139        $bond:04N18O           $atom:04N           $atom:18O           # dNCell
140        $bond:04N19O           $atom:04N           $atom:19O           # dNCell
141        $bond:09021H           $atom:09O           $atom:21H           # eCell
142        $bond:09005N           $atom:09O           $atom:05N           # eNCell
143        $bond:05N20O           $atom:05N           $atom:20O           # eNCell
144        $bond:05N21O           $atom:05N           $atom:21O           # eNCell
145        $bond:11022H           $atom:11O           $atom:22H           # fCell
146        $bond:11006N           $atom:11O           $atom:06N           # fNCell
147        $bond:06N22O           $atom:06N           $atom:22O           # fNCell
148        $bond:06N23O           $atom:06N           $atom:23O           # fNCell
149    }
150
151 } # cUnit_xyz
```

Listing 2: moltemplate input file to create single unit of nitrocellulose.

1.3 Sample LAMMPS input files

```
1 ## -- Init section --
2 units real
3 include "system.in.init"        # <-- contains text written to "In Init"
4
5 ## -- Settings Section --
6 include "system.in.settings"
7 include "system.in.charges"
8
9 ## -- Run section --
10 timestep         0.5
11 variable d internal 0.0
```

```
12  variable dsq equal v_d*v_d
13
14  neigh_modify delay 0 every 1 check yes
15  kspace_style pppm 1.0e-4
16  thermo_style custom step temp press etotal pe cella cellb cellc cellalpha
        cellbeta cellgamma
17  #thermo_style custom cella cellb cellc cellalpha cellbeta cellgamma ##
        check the unit cell dimensions
18
19  dump 1 all custom 100000 traj_0N_d.lammpstrj id mol type x y z ix iy iz #
        every 50 ps, 20 frames per ns
20  dump 2 all dcd 10000 traj_0N_d.dcd # every 5 ps, 200 frames per ns (
        required for MDAnalysis of RDF, RMSD and hydrogen bonds)
21  dump 3 all xyz 100000 traj_0N_d.xyz # every 50 ps, 20 frames per ns
22
23  thermo 200                        # 10 per ps
```

Listing 3: Parameters set for all input files

```
1   ## -- Atom definition section --
2
3   boundary p p p
4   read_data "system.data"
5   change_box all triclinic
6
7   change_box all xy final  -7.43
8   change_box all xz final   0.00
9   change_box all yz final   0.00
10  change_box all x final 0 62.27
11  change_box all y final 0 65.19
12  change_box all z final 0 83.04
13
14  ## parameters as above ##
15
16  min_style cg
17
18  ## Minimisation
19  minimize 1.0e-4 1.0e-6 100000 400000
20
21  write_data  system_after_min.data
```

Listing 4: minimisation script

```
1   read_data        "system_after_min.data"
2
3   ## parameters as above ##
4
5   fix press01 all npt temp 1.0 1.0 $(100.0*dt) tri 100 1 $(1000.0*dt)
6   run             2380            # 1.19 ps
7   write_data system_after_00001ps_press.data
8   unfix press01
9
10  fix temp300 all npt temp 1.0 300.0 $(100.0*dt) tri 1 1 $(1000.0*dt)
11  run             6000000         # 3 ns
12  write_data system_after_03002ps_temp.data
```

```
13  unfix temp300
```

Listing 5: decompression/heating script (example for 0N; decompression time 1.19 ps)

```
1  read_data       "system_after_03002ps_temp.data"
2
3  ## parameters as above ##
4
5  fix prod01 all npt temp 300.0 300.0 $(100.0*dt) tri 1 1 $(1000.0*dt)
6  run             20000000        # 10 ns
7  write_data system_after_13002ps_prod.data
```

Listing 6: production phase script (example for 0N; decompression time 1.19 ps)
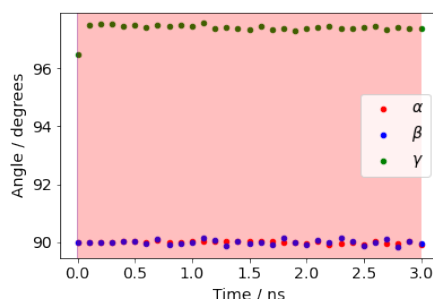
1.4   Average density values for structures created

Table 1: Average Density for structures simulated, used in the plot in figure **??**

Structure name	Density / $\mathrm{g\,cm^{-3}}$
0N	1.585
1Na	1.540
1Nb	1.568
1Nc	1.490
1Nd	1.529
1Ne	1.571
1Nf	1.502
3Ncfa	1.629
3Ncfb	1.630
3Ncfd	1.626
3Ncfe	1.673
45N	1.637
6N	1.673

1.5   Investigation of lattice parameters
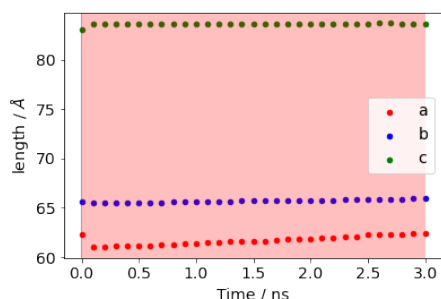
(i)   Cellulose lattice lengths

(ii)   Cellulose lattice angles
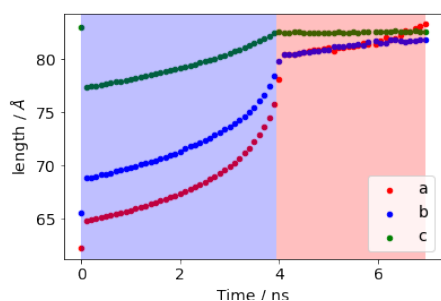
(iii)   12 wt% nitrocellulose lattice lengths
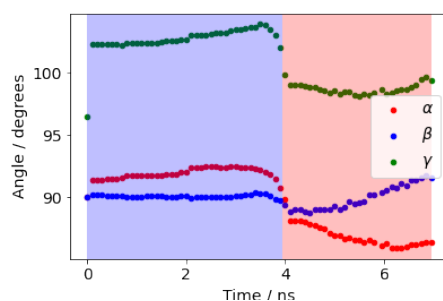
(iv)   12 wt% nitrocellulose lattice angles

Figure 2: Lattice parameters for both cellulose and 12 wt% nitrocellulose during the decompression (blue background, duration 0.001 ns for cellulose, and 3.9 ns for 12 wt%) and heating (red background, duration 3 ns for both structures). In both cases these stages were followed by 10 ns of production phase at 300 K and 1 atm, not shown here, during which equilibration was achieved. It is clear that the cellulose is not changing phase, and the 12 wt% does go through a large change.
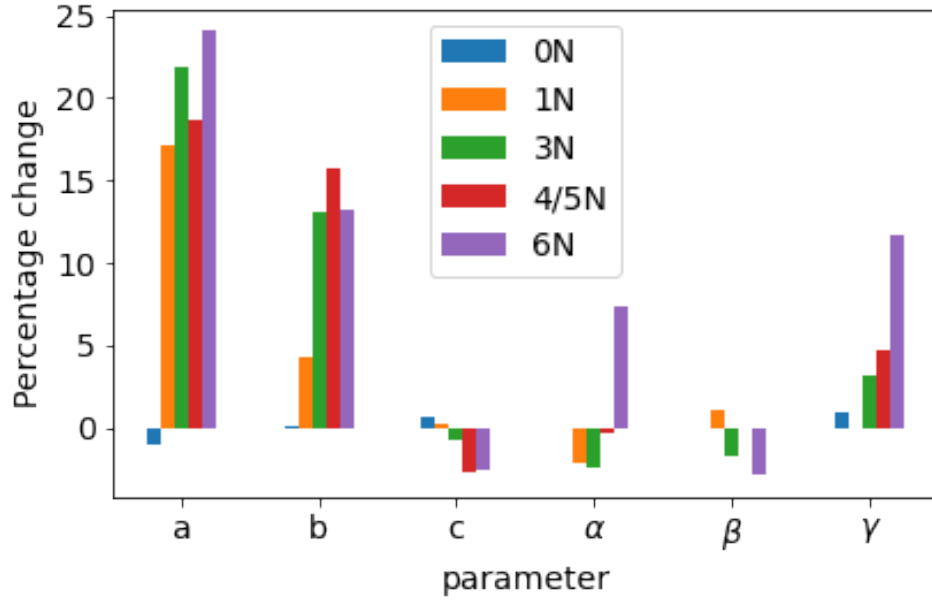
Figure 3:   Percentage changes in each lattice parameter in each of the structures, between the initial crystal structure generated, and the average values during the 10 ns production phase. For the structures where multiple structures were simulated, the percentage shown is the average of all the simulations.

For figure 3 the percentage changes were calculated using

$$\text{Percentage change} = \frac{P_{calc} - P_{original}}{P_{original}} * 100 \tag{1}$$

where P is the specific parameter being calculated.

The variation for cellulose is all within 1 %. This is as expected, as the structure for the cellulose is based on crystallographic coordinates, without any added atoms, so it would not be expected to change much. For all of the other structures, a and b are increasing for all of the structures. Length c has the least variation in all of the dimensions, this is because it is the same as the axis of the chain. For the structures where c is decreasing from the initial length, this means that the chains are bending themselves a little, implying a loss of crystallinity.

1.6   Investigation of 3Ncfe phase change
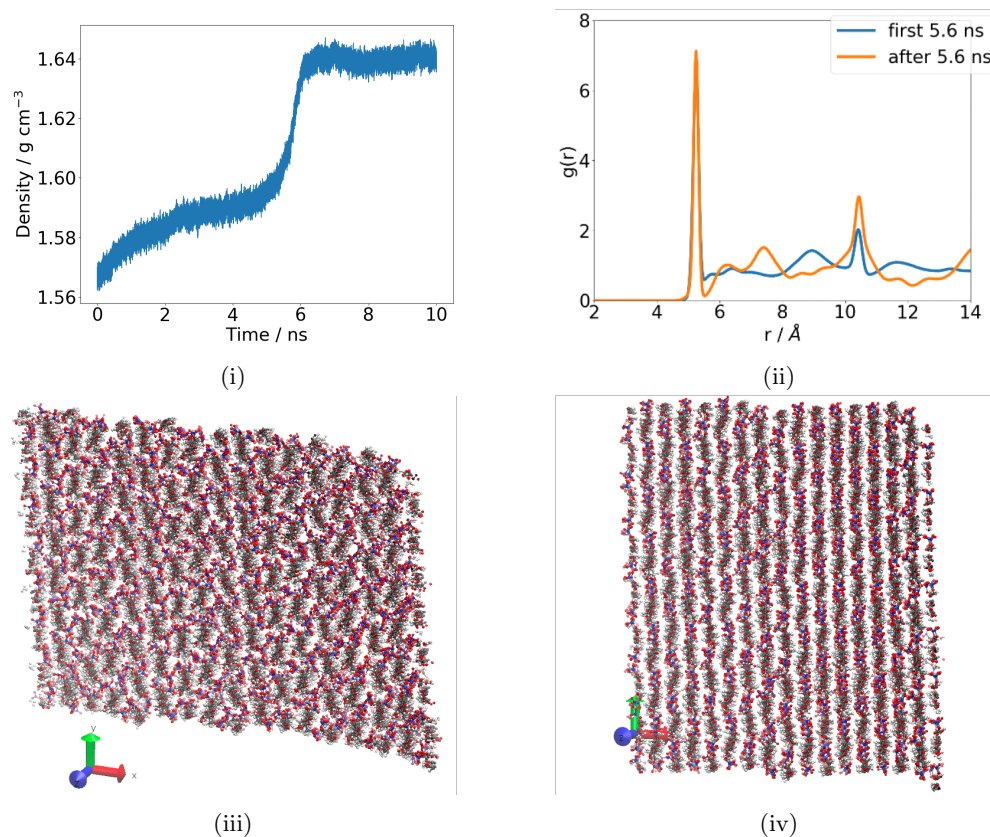

(i)

(ii)

(iii)

(iv)

Figure 4: When the simulation of 3Ncfe structure was run, the density during the 10 ns production phase was found to take a major shift at around 5.6 ns (4i). When the RDF of the structure before and after this shift are plotted (4ii), it can be seen that there is a significant difference in the crystallinity of the structure. This can also be seen visually (4iii), (4iv). Further tests were run on this structure, discussed in table 2

Table 2: Results of tests run on 3Ncfe. After the original simulation showed a density change, and a corresponding change visually and in the RDF, 5 new geometries were tested, with very small changes to the initial geometry. Of the 5 tests, two of the simulations failed. Of the remaining 3, all of them showed a slight change of density, but this did not cause a significant change to the crystallinity, according to the RDF and visualisations

Test number	Structure	Completed	Density change	RDF change	Visual Change
Original	Original geometry	Yes	Yes	Yes	Yes
Test 1	Shift 1st H of each unit by (0.001, 0.0, 0.0)	No	X	X	X
Test 2	Shift 2nd H of each unit by (0.001, 0.0, 0.0)	Yes	Yes	No	No
Test 3	Shift 3rd H of each unit by (0.001, 0.0, 0.0)	No	X	X	X
Test 4	Shift 4th H of each unit by (0.001, 0.0, 0.0)	Yes	Yes	No	No
Test 5	Shift 5th H of each unit by (0.001, 0.0, 0.0)	Yes	Yes	No	No

1.7 Analysing annealing cycles on 0N

As previously mentioned the cellulose structure at the end of the 10 ns production phase was still very crystalline.

Therefore, following the protocol in figure **??**, annealing cycles were run. Although the RDFs during the 700 K steps seem to be fairly paracrystalline, all of the structures at 300 K are crystalline, and running multiple annealing cycles on the structure does not improve the situation - the RDF for the first annealing cycle is the same as for the tenth cycle, as illustrated in figure 5.

Annealing cycles were also run on the 4/5N structures, to see if the paracrystallinity could be increased. The structures stayed at a similar level of crystallinity.
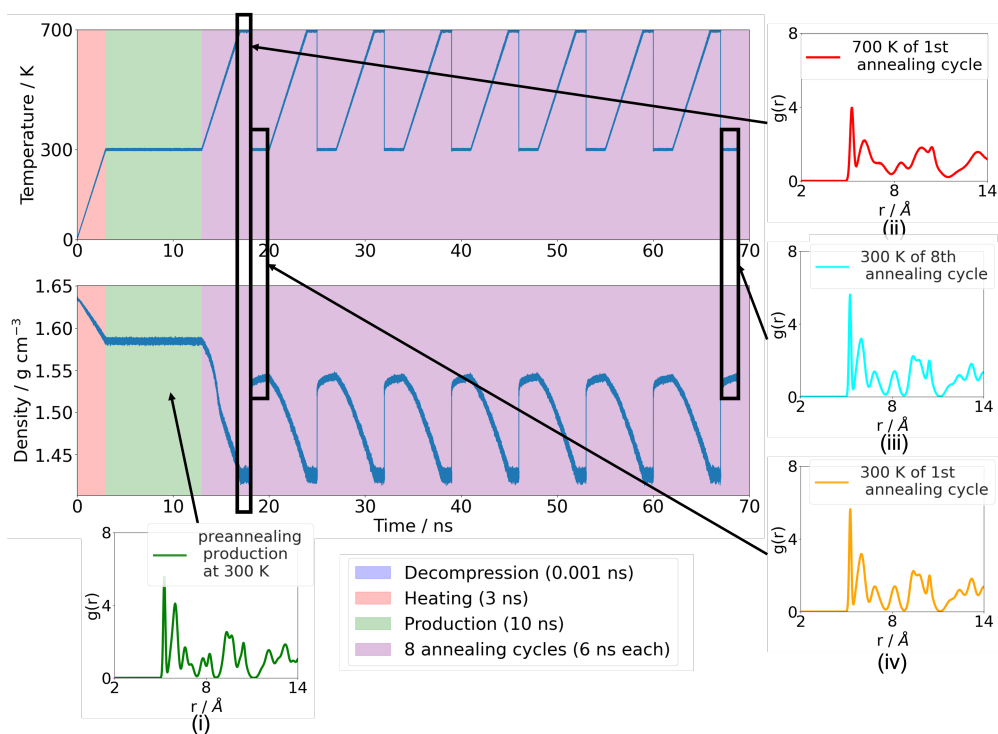
Figure 5: During the entire simulation, the pressure is kept constant and the temperature is varied as shown on the upper line of the plot. The change in density is inversely proportional to the change in temperature. RDF plot (i) represents the structure shown in the 0N final structure in figure 7b of the main paper, which is very crystalline. RDF plot (ii) is for the structure during the first 1 ns at 700 K, where the paracrystallinity has definitely increased. The remaining two RDF graphs are for the 2 ns at 300 K of the first annealing cycle (iv), and the final annealing cycle (iii). These two overlay exactly, showing that even after running 8 cycles the paracrystallinity has not been improved.