

Multiscale Molecular Modelling of ATPFueled Supramolecular Polymerisation and Depolymerisation

Original

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(Article begins on next page)

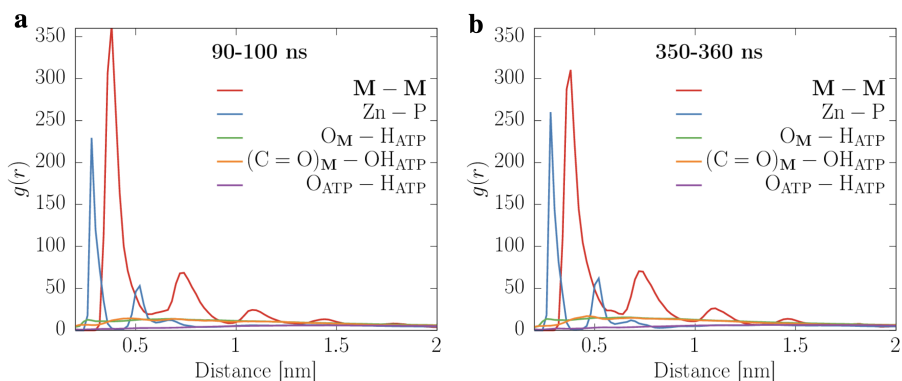


Figure S1: a) Radial distribution functions $g(r)$ between different groups of atoms, for the AA-MD simulation of the pre-stacked \mathbf{M} fiber in water with ATP. $g(r)$ is computed: among the \mathbf{M} centres of mass (red: core-core stacking), between the Zn and the P atoms (blue: electrostatic metal-phosphate interactions), between the \mathbf{M} oxygens and the ATP hydrogens (green: \mathbf{M} -ATP hydrogen-bonding), between the oxygens of the carbonyl groups in \mathbf{M} and the hydrogens in the ribose hydroxyl groups (orange: specific hydrogen bonding option), and between the ATP oxygens and ATP hydrogens (purple: inter-ATP hydrogen-bonding). The $g(r)$ is measured averaging the data calculated from 90 to 100 ns of MD. b) same as a) but averaged from 350 to 360 ns of MD.

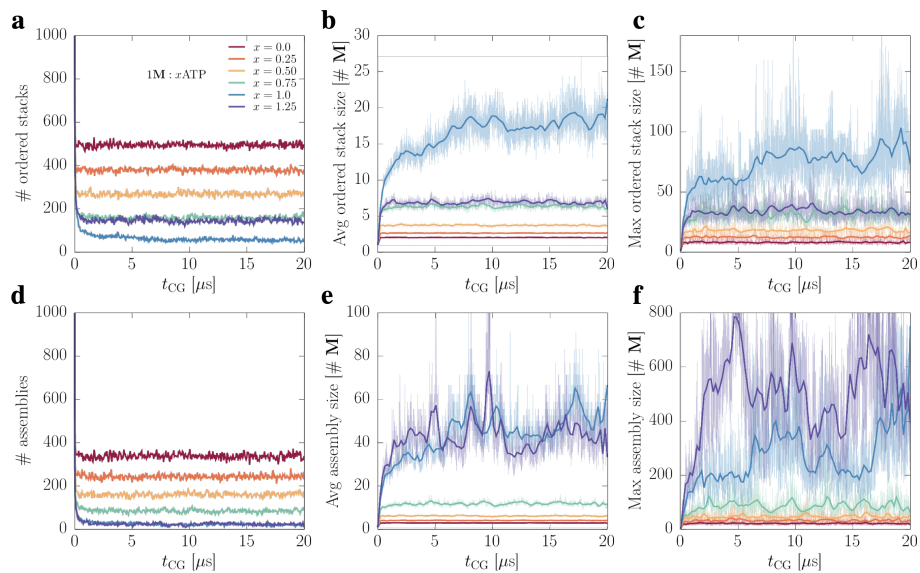


Figure S2: Self-assembly with different ATP concentrations. a) Number of ordered stacks detected versus the CG-MD time. b) Evolution of the average size of ordered stacks. The thick solid line is obtained with Bezier smoothing of the raw data (colored with 0.65 transparency). d) Evolution of the maximum size of ordered stacks. e) Number of assemblies (including disordered) detected versus the CG-MD time. d) Evolution of the average size of assemblies. d) Evolution of the maximum size of assemblies. All panels use the color coding indicated in a)

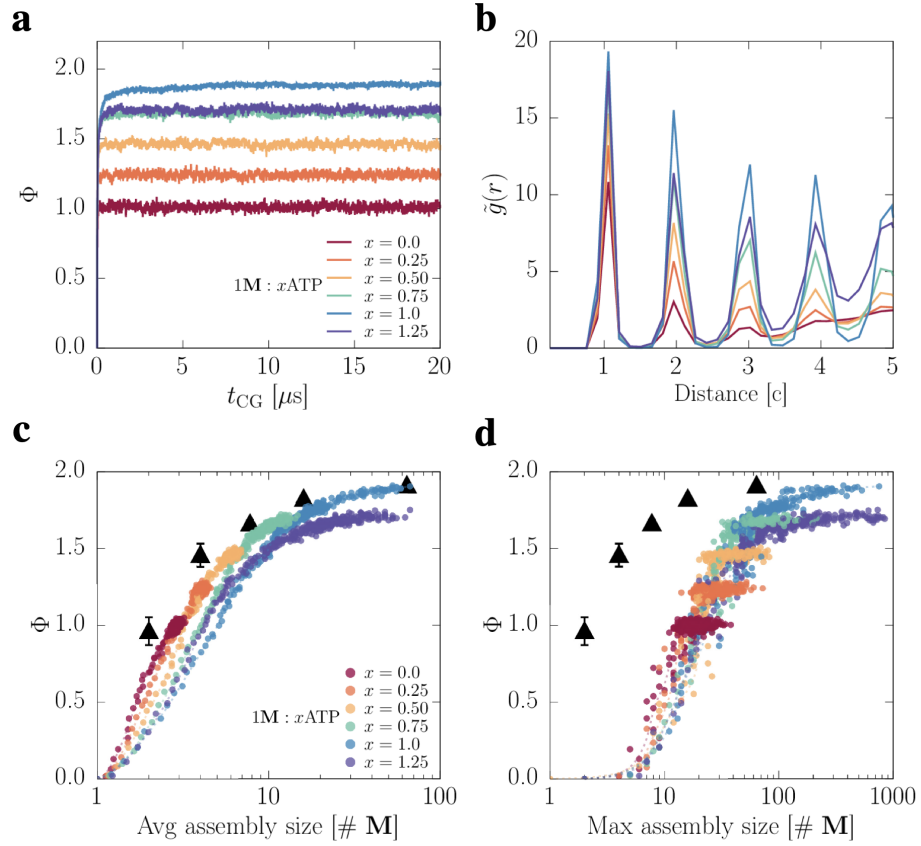


Figure S3: Ordering evolution at different ATP concentrations. a) Evolution of the order parameter Φ . b) Radial distribution function $\tilde{g}(r)$. $\tilde{g}(r)$ is measured averaging the data from $t = 19.9 \mu s$ to $t = 20.0 \mu s$, and it is displayed as a function of the stacking distance $c = 0.53 \text{ nm}$. c) Φ as a function of average assembly size. The black triangles indicate the Φ of pre-stacked assemblies of $\mathbf{M} - \text{ATP}$. g) Φ as a function of maximum assembly size. The black triangles indicate the Φ of pre-stacked assemblies of $\mathbf{M} - \text{ATP}$. All panels use the color coding indicated in a)

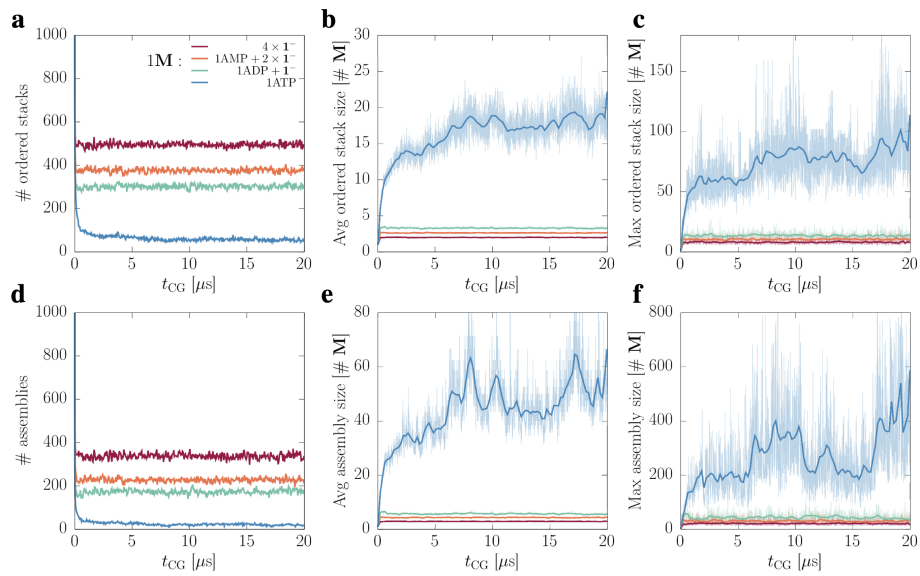


Figure S4: Self-assembly with different fuels. a) Number of ordered stacks detected versus the CG-MD time. b) Evolution of the average size of ordered stacks. The thick solid line is obtained with Bezier smoothing of the raw data (colored with 0.65 transparency). d) Evolution of the maximum size of ordered stacks. e) Number of assemblies (including disordered) detected versus the CG-MD time. d) Evolution of the average size of assemblies. d) Evolution of the maximum size of assemblies. All panels use the color coding indicated in a)

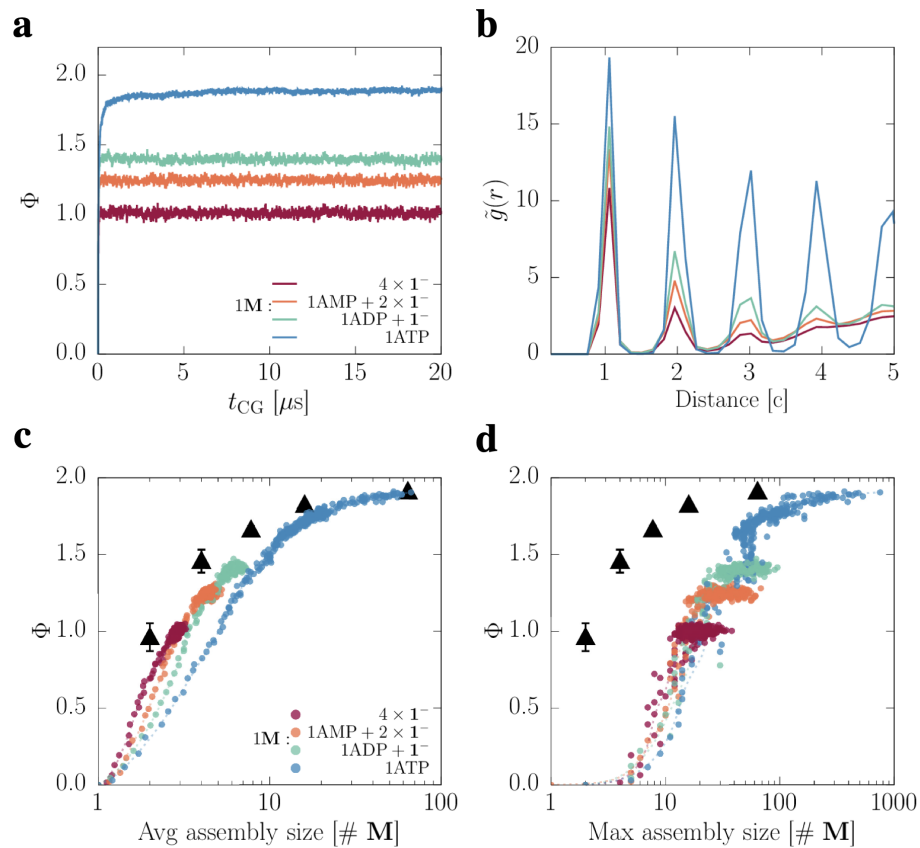


Figure S5: Ordering evolution with different fuels. a) Evolution of the order parameter Φ . b) Radial distribution function $\tilde{g}(r)$. $\tilde{g}(r)$ is measured averaging the data from $t = 19.9 \mu s$ to $t = 20.0 \mu s$, and it is displayed as a function of the stacking distance $c = 0.53$ nm. c) Φ as a function of average assembly size. The black triangles indicate the Φ of pre-stacked assemblies of $\mathbf{M} - \text{ATP}$. g) Φ as a function of maximum assembly size. The black triangles indicate the Φ of pre-stacked assemblies of $\mathbf{M} - \text{ATP}$. All panels use the color coding indicated in a)

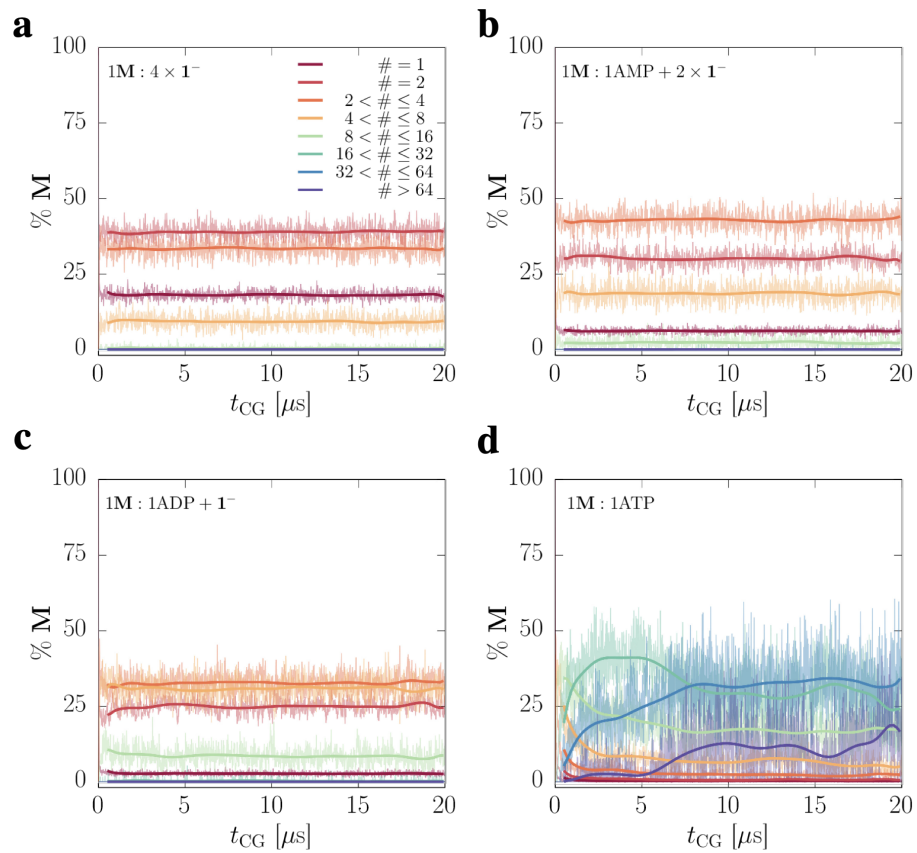


Figure S6: Sizes of ordered stacks with different fuels. Each panel shows the fraction of monomers belonging to ordered stacks of a certain size (or range of sizes) plotted vs the simulation time. The fuel composition is indicated at the upper left corner of each panel. All panels use the color coding indicated in a)

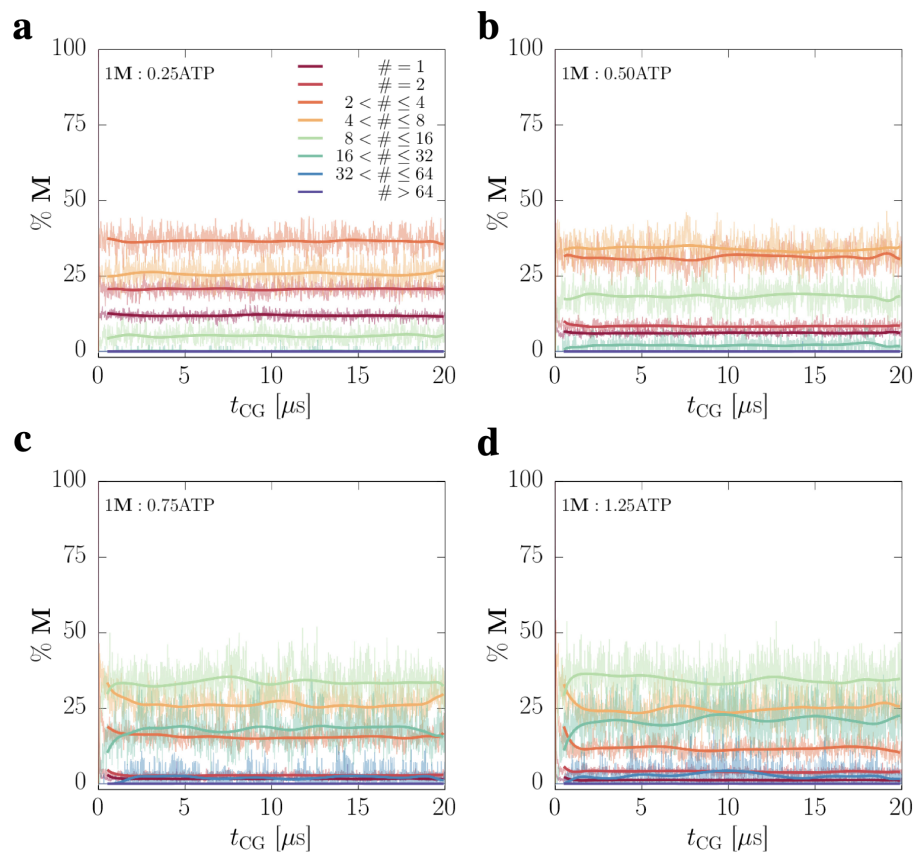


Figure S7: Sizes of ordered stacks at different ATP concentrations. Each panel shows the fraction of monomers belonging to ordered stacks of a certain size (or range of sizes) plotted vs the simulation time. The ATP concentration is indicated at the upper left corner of each panel. All panels use the color coding indicated in a)

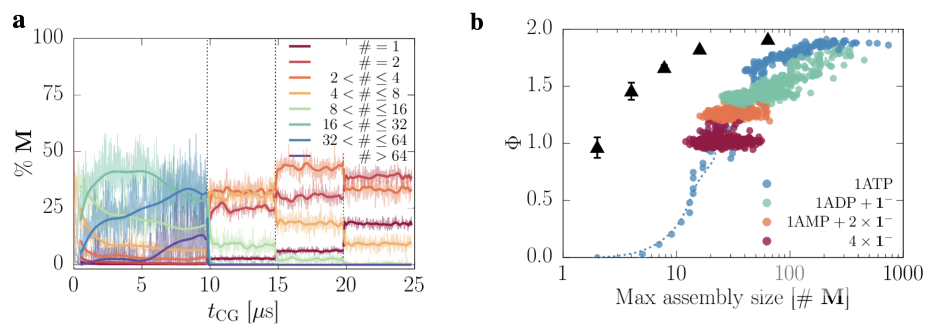


Figure S8: Disassembly triggered by successive cleavage of fuel bonds. a) fraction of monomers belonging to stacks of a certain size (or range of sizes) plotted vs the CG-MD time. The thick solid lines represent the exponential moving average (smoothed with Bezier algorithm) of the raw data (colored with 0.65 transparency). b) Φ as a function of maximum assembly size. The black triangles indicate the Φ of pre-stacked assemblies of $\mathbf{M} - \text{ATP}$.

Minimalistic CG Model Parameters

The force field parameters of the minimalistic CG model are reported in the following, in GROMACS .itp file format. The non-bonded interaction parameters for the different bead types are reported first, and then the CG bead composition and bonded interaction parameters of each molecular specie are indicated.

```
; Minimalistic CG model for fueled self-assembly

; Bead types and non-bonded interactions
[ defaults ]
1 2

[ atomtypes ]

F  0    47.000    0.000    A  0.0    0.0
M  0    72.000    0.000    A  0.0    0.0
P  0    72.000    0.000    A  0.0    0.0
Q  0    47.000    0.000    A  0.0    0.0

[ nonbond_params ]

; i j  funda c6 c12
; self terms
M  M  1  4.700000e-01 30.000000e+00 ;
F  F  1  4.700000e-01 0.200000e+00 ;
P  P  1  4.700000e-01 0.200000e+00 ;
Q  Q  1  4.700000e-01 0.200000e+00 ;

; cross terms
M  F  1  4.700000e-01 0.200000e+00 ;
P  F  1  4.700000e-01 0.200000e+00 ;
P  M  1  4.700000e-01 0.200000e+00 ;
Q  M  1  4.700000e-01 0.200000e+00 ;
Q  F  1  4.700000e-01 0.200000e+00 ;
Q  P  1  4.700000e-01 0.200000e+00 ;

; Monomer M

[ moleculetype ]
; molname      nrexcl
MON             1

[ atoms ]
;id type resnr residu atom cgnr  charge
1  M  1      MONM    M1  1  0
```

2	P	1	MON	P1	2	0
3	P	1	MON	P2	3	0
4	P	1	MON	P3	4	0
5	P	1	MON	P4	5	0
6	P	1	MON	P5	6	0
7	P	1	MON	P6	7	0
8	Q	1	MON	Q1	8	2
9	P	1	MON	P7	4	0
10	P	1	MON	P8	5	0
11	P	1	MON	P9	6	0
12	P	1	MON	P10	7	0
13	Q	1	MON	Q2	9	2
14	P	1	MON	P11	4	0
15	P	1	MON	P12	5	0
16	P	1	MON	P13	6	0
17	P	1	MON	P14	7	0

[bonds]

;	i	j	funct	length	force.c.
1	2	1	0.47	15000	
1	3	1	0.47	15000	
1	4	1	0.47	15000	
1	5	1	0.47	15000	
1	6	1	0.47	15000	
1	7	1	0.47	15000	
2	3	1	0.47	15000	
3	4	1	0.47	15000	
4	5	1	0.47	15000	
5	6	1	0.47	15000	
6	7	1	0.47	15000	
7	2	1	0.47	15000	
2	5	1	0.94	10000	
3	6	1	0.94	10000	
4	7	1	0.94	10000	
8	2	1	0.47	15000	
8	9	1	0.47	15000	
8	10	1	0.47	15000	
8	11	1	0.47	15000	
8	12	1	0.47	15000	
11	2	1	0.47	15000	
11	10	1	0.47	15000	
10	9	1	0.47	15000	
9	12	1	0.47	15000	
11	12	1	0.94	10000	
2	9	1	0.94	10000	
2	10	1	0.814	10000	

```

13  5  1  0.47  15000
13 14  1  0.47  15000
13 15  1  0.47  15000
13 16  1  0.47  15000
13 17  1  0.47  15000
 5 16  1  0.47  15000
16 15  1  0.47  15000
15 14  1  0.47  15000
14 17  1  0.47  15000
16 17  1  0.94  10000
 5 14  1  0.94  10000
 5 15  1  0.814 10000
 1  8  1  0.94  10000
 1 13  1  0.94  10000

; ATP

[ moleculetype ]
; molname      nrexcl
ATP            1

[ atoms ]
;id type resnr residu atom cgnr  charge

 1  F  1      ATP   F1  1  -1
 2  F  1      ATP   F2  2  -1
 3  F  1      ATP   F3  3  -1
 4  F  1      ATP   F4  4  -1

[bonds]
;  i  j  funct  length  force.c.
 1  2  1  0.3  15000
 2  3  1  0.3  15000
 3  4  1  0.3  15000

[angles]
 1  2  3  2  180.0  50
 2  3  4  2  180.0  50

; ADP

[ moleculetype ]
; molname      nrexcl
ADP            1

[ atoms ]

```

```
;id type resnr residu atom cgnr charge
```

```
1 F 1 ADP F1 1 -1
2 F 1 ADP F2 2 -1
3 F 1 ADP F3 3 -1
```

```
[bonds]
```

```
; i j funct length force.c.
1 2 1 0.3 15000
2 3 1 0.3 15000
```

```
[angles]
```

```
1 2 3 2 180.0 50
```

```
;AMP
```

```
[ moleculetype ]
```

```
; molname nrexcl
AMP 1
```

```
[ atoms ]
```

```
;id type resnr residu atom cgnr charge
```

```
1 F 1 AMP F1 1 -1
2 F 1 AMP F2 2 -1
```

```
[bonds]
```

```
; i j funct length force.c.
1 2 1 0.3 15000
```

```
;Negatively charged ion
```

```
[ moleculetype ]
```

```
; molname nrexcl
NEG 1
```

```
[ atoms ]
```

```
;id type resnr residu atom cgnr charge
```

```
1 F 1 NEG F1 1 -1
```

```
;Positively charged ion
```

```
[ moleculetype ]
```

```
; molname nrexcl
POS 1
```

```
[ atoms ]  
;id type resnr residu atom cgnr charge  
1 F 1 POS F1 1 +1
```