Supporting Information for
“Diverse Phases of Carbonaceous Materials from Stochastic Simulations”

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Computational Details

All reactive molecular dynamics runs were carried out with the ReaxFF code available in the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) package.\textsuperscript{1} Periodic boundary conditions were applied in all directions, and the time step was set to 0.2 fs. The temperature was controlled through the Berendsen’s thermostat\textsuperscript{2} with a relaxation constant of 0.1 ps.

Generation of initial configurations

To start the \textit{PES Transformation and Structure Evolution – point (i)} initial carbon configurations were created by randomly dispersing small carbon clusters inside a cubic simulation box where the size was determined based on the chosen density. The clusters were further randomized, to create unbiased arrangements, by disassembling their atoms at high-temperature (8000 K) in the NVT ensemble, in a period of about 100 ps. Then, the final configurations were used as starting structures of a series of short (around 100 ps each) MD simulations based on a variety of force fields, with modified parameters, at a temperature (2000 K) lower than the graphitization threshold (approximately 2550K). The total simulation time (1 ns, at most) was sufficient to obtain relatively stable arrangements.

Estimate of cpu time

We use a Linux cluster: 1 node with 96 cores (see below), cpu GHz = 2.3. We run 100 ps of MD, time step 0.2 fs, saving structures every 1000 steps (0.2 ps), total number of collected snapshots = 500, whence a total cpu time $\approx 13$ h.

\begin{verbatim}
Architecture: x86_64
CPU op-mode(s): 32-bit, 64-bit
Byte Order: Little Endian
CPU(s): 96
On-line CPU(s) list: 0-95
Thread(s) per core: 2
Core(s) per socket: 12
Socket(s): 4
NUMA node(s): 4
Vendor ID: GenuineIntel
CPU family: 6
Model: 85
Model name: Intel(R) Xeon(R) Gold 5118 CPU @ 2.30GHz
Stepping: 4
CPU GHz: 2.3
\end{verbatim}

RMSD and RMSF Quantities
The root mean square deviation (RMSD) measures the deviation of a structure from a reference structure (RMSD=0.0 indicates a perfect overlap). The RMSD is defined as:

$$RMSD = \sqrt{\frac{\sum_{i=0}^{N} [m_i \ast (X_i - Y_i)^2]}{M}}$$

where $N$ is the number of atoms, $m_i$ is the mass of atom $i$, $X_i$ is the coordinate vector for target atom $i$, $Y_i$ is the coordinate vector for reference atom $i$, and $M$ is the total mass. If the RMSD is not mass-weighted, all $m_i=1$ and $M=N$.

The root mean square fluctuation (RMSF) of a structure is the time average of the deviation of a structure from its average value. It is calculated according to the equation below, where $x_i$ is the coordinates of particle $i$, and $\langle x_i \rangle$ is the ensemble average position of $i$:

$$RMSF = \sqrt{\langle (x_i - \langle x_i \rangle)^2 \rangle}$$

The RMSD quantifies how much a structure diverges from a reference over time, the RMSF reveals which areas (atoms) of the system are the most mobile. While RMSD is frequently calculated with respect to the initial state, the RMSF is calculated with respect to an average structure of the simulation. An area of the structure with high RMSF values frequently diverges from the average, indicating high mobility.

Note that we compute RMSD and RMSF after a rigid-body optimal alignment of the atomic coordinates in each snapshot to a reference structure (starting or average configuration).

**Definition of massaging protocol**

The parameters we have chosen to explore in the ‘massaging’ procedure, reported in Table 1, belong to the atom, bond, valence angle and torsion sections of the ReaxFF force field, which is divided into 7 parts, containing the general, atom, bond, off-diagonal, valence angle, torsion angle, and hydrogen bond parameters – we refer to the ReaxFF manual[^3] for a detailed description. As explained in the ReaxFF manual, all parameters without physical meaning are named after the partial energy contribution (valp1, vapl2, *etc*). In contrast, the other ones, like the torsional rotational barriers (V1, V2, V3), have names connected to physical quantities or descriptors.

[^3]: [Reference](#)
**Table S1.** Force field parameters selected for the PES transformation (*massaging*) simulations (50% parameter reduction). Root mean square deviations between the initial and final sampled geometry are reported in the last column. Parameters' names and descriptions have been extracted from the ReaxFF manual.

| Force Field Section | Parameter Identifier | Parameter FF Name | Description                                | RMSD (Å) |
|---------------------|----------------------|-------------------|--------------------------------------------|----------|
| ATOM                | P1                   | eps               | vDW dissociation                           | 8.7      |
|                     | P2                   | valp1             | En. Undercoord. En.                        | 8.1      |
| BOND                | P3                   | de1               | $D_{e}^{\sigma}$                           | 15.9     |
|                     | P4                   | de2               | $D_{e}^{\pi}$                              | 11.3     |
|                     | P5                   | de3               | $D_{e}^{\pi\pi}$                           | 9.1      |
|                     | P6                   | psi               | pbe1, bond En.                             | 8.5      |
|                     | P7                   | vover             | overcoord. penalty                         | 8.4      |
|                     | P8                   | psp               | pbe2, bond En.                             | 9.0      |
| ANGLE               | P9                   | th0               | eq. angle                                  | 8.6      |
|                     | P10                  | vka               | force const.                               | 7.6      |
|                     | P11                  | vka3              | force const.                               | 9.8      |
|                     | P12                  | vval              | En./bond ord.                              | 9.3      |
| TORSION             | P13                  | V1                | barrier 1                                  | 9.0      |
|                     | P14                  | V2                | barrier 2                                  | 8.9      |
|                     | P15                  | V3                | barrier 3                                  | 9.3      |
Figure S1. Root mean square deviation of all the atoms of the sampled configurations relative to the initial structure. The chosen prototype system contains 4176 atoms (simulation box = 42 × 38 × 45 Å³). NVT MD at 2000 K for 20 ps, structures are collected every 2 fs.
Figure S2. Initial and final structures obtained after 20 ps massaging MD at T=2000 K in the NVT ensemble. The massaged parameters are indicated with $P_i$ in each case, and the molecular structures are represented with cyan lines connecting the carbon atoms and filled colored rings. Different colors are used to distinguish the number of ring members. The initial structure is the same prototype system, containing 4176 atoms (simulation box = $42 \times 38 \times 45 \text{ Å}^3$), cited in Figure S1.
Table S2. Force field parameters randomly selected for the four-step-\textit{massage} NVT MD simulations at T=2000 K (50\% parameter reduction), starting from the small prototype model of a carbonaceous material.

| MM1 | MM2 | MM3 | MM4 | MM5 | MM6 | MM7 | MM8 | MM9 | MM10 |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|------|
| P8  | P1  | P9  | P11 | P12 | P10 | P7  | P13 | P4  | P15  |
| P11 | P8  | P15 | P2  | P6  | P1  | P15 | P4  | P4  | P11  |
| P9  | P7  | P8  | P6  | P14 | P8  | P13 | P11 | P7  | P4   |
| P1  | P3  | P14 | P11 | P5  | P12 | P9  | P2  | P7  | P2   |
Figure S3. Root mean square deviation of all the atoms of the configurations sampled at the end of each massage (forty in all) relative to the initial structure. The initial structure is the same prototype system containing 4176 atoms (simulation box = 42 × 38 × 45 Å³) shown in Figure S2. Each NVT MD simulation was carried out at 2000 K for 50 ps. The modified parameters are reported in Table S2.
Table S3. Structural descriptors of the geometries of the prototype structure obtained at the end of the 4-step-massage and after the GO procedure.

| MM # | C1 | C2 | C3 | C4 | A_{opt} | A_{opt2} | R5 | R6 | R7 | CN_c | sasa | D_{ave} | FV |
|------|----|----|----|----|---------|---------|----|----|----|------|------|--------|----|
| MM1  | 0.001 | 0.36 | 0.63 | 0.008 | 10.8 | 2.3 | 0.060 | 0.052 | 0.032 | 34.2 | 10159 | 8.0 | 48.1 |
| opt  | 0.001 | 0.36 | 0.63 | 0.009 | 6.6 | 2.2 | 0.060 | 0.053 | 0.032 | 33.9 | 9744 | 8.7 | 47.1 |
| MM2  | 0.017 | 0.70 | 0.28 | 0.000 | 10.4 | 1.8 | 0.013 | 0.039 | 0.009 | 32.5 | 4483 | 26.6 | 42.1 |
| opt  | 0.014 | 0.71 | 0.28 | 0.000 | 5.1 | 1.2 | 0.014 | 0.039 | 0.009 | 32.8 | 4291 | 23.5 | 41.9 |
| MM3  | 0.001 | 0.32 | 0.67 | 0.004 | 11.2 | 2.1 | 0.054 | 0.061 | 0.039 | 34.0 | 8952 | 9.6 | 46.5 |
| opt  | 0.000 | 0.32 | 0.68 | 0.004 | 5.2 | 1.6 | 0.054 | 0.062 | 0.039 | 34.0 | 9186 | 10.0 | 46.7 |
| MM4  | 0.000 | 0.04 | 0.94 | 0.021 | 19.0 | 2.3 | 0.112 | 0.132 | 0.087 | 39.9 | 10329 | 10.1 | 51.2 |
| opt  | 0.000 | 0.04 | 0.94 | 0.018 | 14.0 | 1.4 | 0.112 | 0.133 | 0.089 | 40.0 | 10118 | 9.5 | 51.5 |
| MM5  | 0.001 | 0.14 | 0.85 | 0.010 | 14.2 | 2.0 | 0.090 | 0.120 | 0.076 | 38.2 | 10186 | 7.8 | 50.0 |
| opt  | 0.000 | 0.13 | 0.86 | 0.011 | 8.3 | 1.3 | 0.090 | 0.121 | 0.076 | 38.2 | 10461 | 7.6 | 50.1 |
| MM6  | 0.000 | 0.10 | 0.88 | 0.018 | 15.0 | 1.9 | 0.105 | 0.146 | 0.083 | 39.6 | 10125 | 11.2 | 50.9 |
| opt  | 0.000 | 0.10 | 0.88 | 0.020 | 10.8 | 1.1 | 0.106 | 0.148 | 0.084 | 39.7 | 10095 | 11.1 | 51.1 |
| MM7  | 0.003 | 0.96 | 0.04 | 0.000 | 10.8 | 1.4 | 0.000 | 0.000 | 0.000 | 31.1 | 1521 | 44.4 | 38.9 |
| opt  | 0.001 | 0.93 | 0.07 | 0.000 | 5.1 | 2.7 | 0.000 | 0.000 | 0.000 | 31.6 | 1434 | 44.7 | 39.1 |
| MM8  | 0.000 | 0.04 | 0.94 | 0.020 | 18.6 | 1.7 | 0.104 | 0.168 | 0.094 | 42.5 | 9812 | 11.8 | 53.0 |
| opt  | 0.000 | 0.04 | 0.94 | 0.021 | 16.4 | 1.1 | 0.105 | 0.169 | 0.094 | 42.4 | 9929 | 11.5 | 52.7 |
| MM9  | 0.000 | 0.07 | 0.89 | 0.043 | 17.8 | 1.6 | 0.094 | 0.187 | 0.103 | 43.5 | 10042 | 12.2 | 53.1 |
| opt  | 0.000 | 0.07 | 0.89 | 0.044 | 13.9 | 1.1 | 0.094 | 0.187 | 0.104 | 43.0 | 9804 | 10.8 | 52.7 |
| MM10 | 0.000 | 0.06 | 0.92 | 0.027 | 17.9 | 1.7 | 0.099 | 0.173 | 0.099 | 43.3 | 9680 | 10.7 | 52.9 |
| opt  | 0.000 | 0.06 | 0.92 | 0.028 | 15.4 | 1.2 | 0.100 | 0.173 | 0.099 | 43.1 | 9645 | 12.5 | 52.7 |

D=Box density (g/cm³)

C1=Fraction of C atoms with coordination 1.

C2=Fraction of C atoms with coordination 2.

C3=Fraction of C atoms with coordination 3.

C4=Fraction of C atoms with coordination 4.

A_{opt}=Average difference between ideal 180° angle for sp C atoms and effective angles formed by those C atoms characterized by coordination 2. Large values correspond to “bended wires” deviating from ideal linearity (degrees).

A_{opt2}=Average difference between the ideal 120° angle for sp² C atoms and effective angles formed by those C atoms characterized by coordination 3. Large values correspond to “bended sheets” deviating from perfect planarity of ideal graphitic foils (degrees).

R5=Ratio between the number 5-member rings and the total number of atoms

R6=Ratio between the number 6-member rings and the total number of atoms

R7=Ratio between the number 7-member rings and the total number of atoms

CN_c=Coordination number (second sphere)

sasa=Solvent-accessible surface area (Å²)

D_{ave}=Average Density of the System (g/cm³)

FV=Fraction of Free-Volume (%)
Figure S4. Final representative structures of the ten four-step-*massages* described in Tables S2-S3. The initial structure is the same prototype system containing 4176 atoms (simulation box = 42 × 38 × 45 Å³) shown in Figure S2.
Table S4. Structural descriptors of the final geometries generated by the full DynReaxMas protocol (massages, equilibration, plus a final GO) using nine different DynReaxMas massages, namely: MM1/MM4, MM1/MM6, MM1/MM8, MM2/MM4, MM2/MM6, MM2/MM8, MM3/MM4, MM3/MM6, MM3/MM8, conducted at 2000 K and 1.15 g/cm$^3$ mass density (MM1/MM4 is simplified to M1M4 in the table for the sake of notation, and analogously for the other combinations).

| property          | M1M4  | M1M6  | M1M8  | M2M4  | M2M6  | M2M8  | M3M4  | M3M6  | M3M8  |
|-------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| density [g/cm$^3$] | 1.146 | 1.146 | 1.146 | 1.146 | 1.146 | 1.146 | 1.146 | 1.146 | 1.146 |
| PLD [Å]           | 4.74  | 6.94  | 6.90  | 5.99  | 7.36  | 4.62  | 6.02  | 5.83  | 8.24  |
| LCD [Å]           | 17.89 | 16.36 | 16.93 | 18.81 | 17.46 | 17.94 | 18.89 | 16.88 | 16.48 |
| $S_{AC,T}$ [m$^2$/g] | 852.4 | 797.7 | 868.5 | 853.60| 812.7 | 868.9 | 861.3 | 803.2 | 855.7 |
| $S_{AC,A}$ [m$^2$/g] | 602.7 | 644.4 | 742.7 | 723.70| 729.9 | 646.9 | 713.3 | 572.6 | 643.0 |
| $V_{PO,T}$ [cm$^3$/g] | 0.387 | 0.365 | 0.386 | 0.382 | 0.364 | 0.381 | 0.385 | 0.368 | 0.381 |
| $V_{PO,A}$ [cm$^3$/g] | 0.276 | 0.282 | 0.326 | 0.327 | 0.316 | 0.285 | 0.315 | 0.254 | 0.289 |
| $F_{Hel,T}$     | 0.44  | 0.42  | 0.44  | 0.44  | 0.42  | 0.44  | 0.42  | 0.44  | 0.44  |

PLD = Pore limiting diameter, Å  
LCD = Largest cavity diameter, Å  
$S_{AC,T}$ = Total accessible surface area, m$^2$/g  
$S_{AC,A}$ = Network accessible surface area, m$^2$/g  
$V_{PO,T}$ = Total probe-occupiable volume, cm$^3$/g  
$V_{PO,A}$ = Network accessible probe-occupiable volume, cm$^3$/g  
$F_{Hel,T}$ = Helium pore volume fraction

C$_1$=Fraction of C atoms with coordination 1  
C$_2$=Fraction of C atoms with coordination 2  
C$_3$=Fraction of C atoms with coordination 3  
C$_4$=Fraction of C atoms with coordination 4  
$A_{180}$=Average difference between ideal 180° angle for $sp$ C atoms and effective angles formed by those C atoms characterized by coordination 2. Large values correspond to “bended wires” deviating from ideal linearity (degrees).  
$A_{120}$=Average difference between the ideal 120° angle for $sp^2$ C atoms and effective angles formed by those C atoms characterized by coordination 3. Large values correspond to “bended sheets” deviating from perfect planarity of ideal graphitic foils (degrees).  
R$_5$= 100 x Ratio between the number 5-member rings and the total number of atoms  
R$_6$= 100 x Ratio between the number 6-member rings and the total number of atoms  
R$_7$= 100 x Ratio between the number 7-member rings and the total number of atoms  
CN$_5$=coordination number (second sphere)  
sasa=Solvent-accessible surface area (Å$^2$)  
D$_{ave}$=Average Density of the System (g/cm$^3$)  
FV=Fraction of Free-Volume (%)
Table S5. Structural descriptors of the final geometries generated by the full DynReaxMas protocol (massages, equilibration, plus a final GO) using nine different DynReaxMas massages, namely: MM1/MM4, MM1/MM6, MM1/MM8, MM2/MM4, MM2/MM6, MM2/MM8, MM3/MM4, MM3/MM6, MM3/MM8, conducted at 2000 K and 0.50 g/cm³ mass density (MM1/MM4 is simplified to m1m4 in the inset for the sake of notation, and analogously for the other combinations).

| property                  | M1M4 | M1M6 | M1M8 | M2M4 | M2M6 | M2M8 | M3M4 | M3M6 | M3M8 |
|---------------------------|------|------|------|------|------|------|------|------|------|
| density [g/cm³]           | 0.499| 0.499| 0.499| 0.499| 0.499| 0.499| 0.499| 0.499| 0.499|
| PLD [Å]                   | 19.24| 19.65| 19.34| 18.17| 25.86| 19.53| 17.82| 19.48| 16.95|
| LCD [Å]                   | 40.61| 43.82| 36.16| 44.02| 35.99| 38.80| 45.96| 48.78| 41.07|
| $\delta_{AC,T}$ [m²/g]   | 2287.9| 2376.8| 2371.1| 2221.8| 2198.4| 2363.5| 2297.2| 2258.0| 2409.0|
| $\delta_{AC,A}$ [m²/g]   | 2278.9| 2368.5| 2357.6| 2213.1| 2198.4| 2369.9| 2299.2| 2245.5| 2405.2|
| $V_{PO,T}$ [cm³/g]       | 1.524| 1.511| 1.522| 1.506| 1.518| 1.528| 1.516| 1.516| 1.521|
| $V_{PO,A}$ [cm³/g]       | 1.523| 1.508| 1.517| 1.519| 1.499| 1.519| 1.521| 1.510| 1.515|
| $F_{He,T}$               | 0.76 | 0.75 | 0.76 | 0.75 | 0.76 | 0.76 | 0.76 | 0.76 | 0.76 |

PLD = Pore limiting diameter, Å
LCD = Largest cavity diameter, Å
$\delta_{AC,T}$ = Total accessible surface area, m²/g
$\delta_{AC,A}$ = Network accessible surface area, m²/g
$V_{PO,T}$ = Total probe-occupiable volume, cm³/g
$V_{PO,A}$ = Network accessible probe-occupiable volume, cm³/g
$F_{He,T}$ = Helium pore volume fraction

| C₁  | C₂  | C₃  | C₄  | A₁(0) | A₁(02) | A₂(0) | A₂(02) | R₅  | R₆  | R₇  | CN₅ | sasa | D₀D | FV  |
|-----|-----|-----|-----|-------|--------|-------|--------|-----|-----|-----|-----|-----|-----|-----|
| 0.0000 | 0.0960 | 0.8860 | 0.0170 | 20.8670 | 0.9530 | 10.5600 | 16.0480 | 9.3190 | 37.8590 | 125287.0 | 3.7870 | 78.6630 |
| 0.0000 | 0.1390 | 0.8540 | 0.0070 | 13.7530 | 0.9050 | 10.6400 | 14.4120 | 8.0700 | 36.1660 | 131840.0 | 4.0210 | 78.2970 |
| 0.0000 | 0.0970 | 0.8920 | 0.0110 | 19.2340 | 0.9760 | 11.1790 | 15.6290 | 8.7920 | 37.1110 | 129226.0 | 4.4250 | 78.5680 |
| 0.0000 | 0.0950 | 0.8880 | 0.0170 | 19.5800 | 0.9710 | 10.7720 | 16.1080 | 8.4890 | 38.8540 | 121405.0 | 3.2940 | 78.6830 |
| 0.0000 | 0.1240 | 0.8680 | 0.0090 | 13.7460 | 1.0620 | 10.6520 | 14.2640 | 8.2100 | 39.1720 | 119028.0 | 3.4480 | 78.4650 |
| 0.0000 | 0.1020 | 0.8860 | 0.0120 | 19.2990 | 0.9770 | 10.9750 | 15.8840 | 8.4970 | 37.6120 | 127328.0 | 3.9390 | 78.5920 |
| 0.0000 | 0.0960 | 0.8680 | 0.0180 | 19.8080 | 0.8880 | 10.2890 | 16.3190 | 8.7480 | 38.4100 | 123729.0 | 3.7030 | 78.7540 |
| 0.0000 | 0.1180 | 0.8720 | 0.0090 | 13.7950 | 0.8760 | 10.6760 | 15.2860 | 8.1460 | 37.7050 | 123767.0 | 3.6960 | 78.6500 |
| 0.0000 | 0.1040 | 0.8840 | 0.0120 | 18.8810 | 0.8770 | 10.8960 | 16.1040 | 8.6450 | 37.2830 | 130090.0 | 3.3690 | 78.6540 |

C₁=Fraction of C atoms with coordination 1.
C₂=Fraction of C atoms with coordination 2.
C₃=Fraction of C atoms with coordination 3.
C₄=Fraction of C atoms with coordination 4.
A₁(0)=Average difference between ideal 180° angle for sp C atoms and effective angles formed by those C atoms characterized by coordination 2. Large values correspond to “bended wires” deviating from ideal linearity (degrees).
A₁(02)=Average difference between the ideal 120° angle for sp² C atoms and effective angles formed by those C atoms characterized by coordination 3. Large values correspond to “bended sheets” deviating from perfect planarity of ideal graphitic foils (degrees).
R₅= 100 x Ratio between the number 5-member rings and the total number of atoms
R₆= 100 x Ratio between the number 6-member rings and the total number of atoms
R₇= 100 x Ratio between the number 7-member rings and the total number of atoms
CN₅=Coordination number (second sphere)
sasa=Solvent-accessible surface area (Å²)
D₀D=Average Density of the System (g/cm³)
FV=Fraction of Free-Volume (%)
Table S6. Structural descriptors of the final geometries generated by the full DynReaxMas protocol (massages, equilibration, plus a final GO) using nine different DynReaxMas massages, namely: MM1/MM4, MM1/MM6, MM1/MM8, MM2/MM4, MM2/MM6, MM2/MM8, MM3/MM4, MM3/MM6, MM3/MM8, conducted at 2000 K and 0.16 g/cm$^3$ mass density (MM1/MM4 is simplified to m1m4 in the inset for the sake of notation, and analogously for the other combinations).

| property          | M1M4  | M1M6  | M1M8  | M2M4  | M2M6  | M2M8  | M3M4  | M3M6  | M3M8  |
|-------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| density [g/cm$^3$]| 0.164 | 0.164 | 0.164 | 0.164 | 0.164 | 0.164 | 0.164 | 0.164 | 0.164 |
| PLD [Å]           | 111.82| 114.66| 109.94| 57.61 | 78.46 | 105.71| 54.60 | 46.01 | 48.50 |
| LCD [Å]           | 135.93| 128.27| 134.34| 106.07| 99.82 | 119.66| 97.51 | 92.09 | 91.48 |
| $S_{AC}[m^2/g]$   | 1554.3| 1718.9| 1620.8| 2193.1| 1282.2| 2023.2| 3511.9| 3604.2| 3635.0|
| $S_{ACA}[m^2/g]$  | 1530.3| 1705.4| 1607.6| 2178.9| 1259.4| 1991.0| 3513.3| 3590.2| 3633.8|
| $V_{PO,T}[cm^3/g]$| 5.64 | 5.62 | 5.62 | 5.62 | 5.61 | 5.63 | 5.62 | 5.62 | 5.62 |
| $V_{PO,A}[cm^3/g]$ | 5.62 | 5.61 | 5.62 | 5.62 | 5.59 | 5.62 | 5.62 | 5.62 | 5.62 |
| $F_{He,T}$        | 0.92  | 0.92  | 0.92  | 0.92  | 0.92  | 0.92  | 0.92  | 0.92  | 0.92  |

PLD = Pore limiting diameter, Å
LCD = Largest cavity diameter, Å
$S_{AC} = $ Total accessible surface area, m$^2$/g
$S_{ACA} = $ Network accessible surface area, m$^2$/g
$V_{PO,T} = $ Total probe-occupiable volume, cm$^3$/g
$V_{PO,A} = $ Network accessible probe-occupiable volume, cm$^3$/g
$F_{He,T} = $ Helium pore volume fraction

C$_1$=Fraction of C atoms with coordination 1.
C$_2$=Fraction of C atoms with coordination 2.
C$_3$=Fraction of C atoms with coordination 3.
C$_4$=Fraction of C atoms with coordination 4.
$A_{180} = $Average difference between ideal 180° angle for $sp$ C atoms and effective angles formed by those C atoms characterized by coordination 2. Large values correspond to “bended wires” deviating from ideal linearity (degrees).
$A_{120} = $Average difference between the ideal 120° angle for $sp^2$ C atoms and effective angles formed by those C atoms characterized by coordination 3. Large values correspond to “bended sheets” deviating from perfect planarity of ideal graphitic foils (degrees).
R$_5 = $ 100 x Ratio between the number 5-member rings and the total number of atoms
R$_6 = $ 100 x Ratio between the number 6-member rings and the total number of atoms
CN$_5$=coordination number (second sphere)
sasa=Solvent-accessible surface area (Å$^2$)
$D_{ave} = $Average Density of the System (g/cm$^3$)
FV=Fraction of Free-Volume (%)
Convergence Check
For the three examined densities (1.15, 0.50, and 0.16 g/cm$^3$) and for all combinations of massages \{MM1,MM2,MM3\} x \{MM4,MM6,MM8\}, the final structure of the last massage was equilibrated for about fifty picoseconds (ps) using the original (unbiased, not-massaged) ReaxFF (C.ff) force field at T=2000 K. The average configuration obtained from the final ten ps of this equilibration was used for the subsequent GO steps whence the final descriptor analysis. To check convergence, the root mean squared deviations (RMSD) of all the atoms from the average structure were evaluated in the last ten ps of the equilibration dynamics: these are illustrated in Figure S5(left panel) focusing on the intermediate density (0.50 g/cm$^3$). A superimposition of the average structure with another structure having a RMSD of approximately 7.0 Å obtained during the last ten ps of the dynamics is also shown in Figure S5(right panel). Inspection of Figure S5 indicates that the RMSD are small and oscillate in a range of 6.2-8.5 Å. Visual inspection of the superimposition further confirms the validity of convergence.

Figure S5. On the left, distribution of the RMSD of all the structures sampled during the last ten ps of the equilibration dynamics with the C.ff force field at T=2000K relative to the average configuration. Results refer to mass density of 0.50 g/cm$^3$ and are reported for all combinations of massages: \{MM1,MM2,MM3\} x \{MM4,MM6,MM8\} (MM1/MM4 is simplified to m1m4 in the inset for the sake of notation, and analogously for the other combinations). On the right, a typical superimposition between all the atoms of the average structure and those of a configuration sampled during the last ten picoseconds of the equilibration dynamics having a RMSD=7 Å.
Figure S6. Final configurations obtained after the destruction massages. (a) MM1 (step 3 = P9), (b) MM2 (step 4 = P3) and (c) MM3 (step 1 = P9). Gray lines indicate carbon chains, whereas ring present in the configurations are identified by multicolored regions (3-member rings are red).
Effects of the temperature selected for the DynReaxMas massage procedure

Figure S7. Final configurations obtained after four different DynReaxMas massages, namely: MM1/MM4, MM1/MM8, MM3/MM4, MM3/MM8 (from top to bottom), at four different temperatures, namely: 1500, 2000, 2500, and 3000K (from left to right).
Figure S8. Final configurations (at LOW density – 0.16 g/cm³) obtained after nine different combinations of massages, namely: MM1/MM4, MM1/MM6, MM1/MM8 (first row), MM2/MM4, MM2/MM6, MM2/MM8 (second row), MM3/MM4, MM3/MM6, MM3/MM8 (third row).
Figure S9. Final configurations (at MEDIUM density – 0.50 g/cm$^3$) obtained after nine different combinations of massages, namely: MM1/MM4, MM1/MM6, MM1/MM8 (first row), MM2/MM4, MM2/MM6, MM2/MM8 (second row), MM3/MM4, MM3/MM6, MM3/MM8 (third row).
Figure S10. Final configurations (at HIGH density – 1.15 g/cm$^3$) obtained after nine different combinations of massages, namely: MM1/MM4, MM1/MM6, MM1/MM8 (first row), MM2/MM4, MM2/MM6, MM2/MM8 (second row), MM3/MM4, MM3/MM6, MM3/MM8 (third row).
Figure S11. Evolution of the given descriptors \([sp = C_2, sp2 = C_3, sp3 = C_4, 5\text{-}member \text{rings} = R_5, 6\text{-}member \text{rings} = R_6, 7\text{-}member \text{rings} = R_7, \Delta A_{(sp)}, \Delta A_{(sp2)}, \text{SASA, Local Density, Average Density} = D_{\text{ave}}, \text{Free Volume} = FV]\), as defined in Tables S3 and S4) during the various massages after destruction for MM1/MM8, MM2/MM6, and MM3/MM4 DynReaXMas runs at medium density \((0.50 \text{ g/cm}^3)\) and 2000K (MM1/MM4 is simplified to m1m4 in the inset, and analogously for the other combinations). Twenty points are reported from each parameter perturbation.
Figure S12. Contour plots (derived from the parameters evolution) highlighting the correlation among sp2 (or C3 in Tables S3 and S4), 6-member ring content (R6 in the Tables), and local density (multicolored regions) for MM1/MM8 (top), MM2/MM6 (medium), and MM3/MM4 (bottom) DynReaxMas runs, at medium-density (0.50 g/cm³) and 2000K.
Figure S13. Pore size distributions of the final structures deriving from four different DynReaxMas massages, namely: MM1/MM4, MM1/MM8, MM3/MM4, MM3/MM8, (MM1/MM4 is simplified to m1m4 in the inset for the sake of notation, and analogously for the other combinations) conducted at four different temperatures, namely: 1500, 2000, 2500, and 3000 K and at medium-density (0.50 g/cm$^3$).
Figure S14. Pore size distributions (PSD) of the final structures deriving from nine different DynReaxMas massages, namely: MM1/MM4, MM1/MM6, MM1/MM8 (top), MM2/MM4, MM2/MM6, MM2/MM8 (middle), MM3/MM4, MM3/MM6, MM3/MM8 (bottom), conducted at 2000 K and 1.15 g/cm$^3$ mass density (MM1/MM4 is simplified to m1m4 in the inset for the sake of notation, and analogously for the other combinations).
Figure S15. Pore size distributions (PSD) of the final structures deriving from nine different DynReaxMas massages, namely: MM1/MM4, MM1/MM6, MM1/MM8 (top), MM2/MM4, MM2/MM6, MM2/MM8 (middle), MM3/MM4, MM3/MM6, MM3/MM8 (bottom), conducted at 2000 K and 0.50 g/cm$^3$ mass density (MM1/MM4 is simplified to m1m4 in the inset for the sake of notation, and analogously for the other combinations).
Figure S16. Pore size distributions (PSD) of the final structures deriving from nine different DynReaxMas massages, namely: MM1/MM4, MM1/MM6, MM1/MM8 (top), MM2/MM4, MM2/MM6, MM2/MM8 (middle), MM3/MM4, MM3/MM6, MM3/MM8 (bottom), conducted at 2000 K and 0.16 g/cm$^3$ mass density (MM1/MM4 is simplified to m1m4 in the inset for the sake of notation, and analogously for the other combinations).
Figure S17. HRTEM images of the final configurations obtained at the end of MM1/MM4 (top left), MM1/MM8 (top right), MM3/MM4 (bottom left), MM3/MM8 (bottom right) at medium-density (0.50 g/cm$^3$) and T=3000K.
Figure S18. HRTEM images of the final configurations (at LOW density – 0.16 g/cm$^3$, T = 2000 K) obtained after nine different massages, namely: MM1/MM4, MM1/MM6, MM1/MM8 (first row), MM2/MM4, MM2/MM6, MM2/MM8 (second row), MM3/MM4, MM3/MM6, MM3/MM8 (third row).
Figure S19. HRTEM images of the final configurations (at MEDIUM density – 0.50 g/cm$^3$, T = 2000 K) obtained after nine different massages, namely: MM1/MM4, MM1/MM6, MM1/MM8 (first row), MM2/MM4, MM2/MM6, MM2/MM8 (second row), MM3/MM4, MM3/MM6, MM3/MM8 (third row).
Figure S20. HRTEM images of the final configurations (at HIGH density – 1.15 g/cm$^3$, T = 2000 K) obtained after nine different massages, namely: MM1/MM4, MM1/MM6, MM1/MM8 (first row), MM2/MM4, MM2/MM6, MM2/MM8 (second row), MM3/MM4, MM3/MM6, MM3/MM8 (third row).
Figure S21. Plots of Pair Distribution Functions (PDF) or $g(r)$ of the final configurations (at MEDIUM density – 0.50 g/cm$^3$) obtained after four different massages, namely: MM1/MM4, MM1/MM8, MM2/MM4, MM2/MM8 at different temperatures: $T = 1500, 2000, 2500, 3000$ K.
Figure S22. A narrow tunnel identified by the CAVER software in one of the sampled structures exhibiting a bimodal distribution of pore sizes. The tunnel corresponds to the continuous green line, and for the sake of clarity, we have highlighted with a pink aura its intersection with the wall of the periodic unit cell.
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