**Supplemental Data**

In the following supplementary data, we present the energy and temperature *versus* time control graphics and energetic characteristics, for the system with MMT-O (Figure 1 and Table 1) and with MMT-O (Figure 2 and Table 2).

It is important to note that, in the subsequent simulation times after around 20 ps for the first system (Figure 1) and around 30 ps (Figure 2) the molecules are still being slightly reorganized due to the electrostatic interactions optimization procedure.

Here, one of the main points we would like to convey to the readers is precisely to show that long dynamic trajectories are not necessarily mandatory to obtain all the essential information about fundamental motions and intra and intermolecular interactions. Careful observation of the systems during the entire time of the trajectories would be essential.

P_acetyl_Figure1_a_b_c_dyn363K_100ps_see_stearic.tif

P_acetyl_3rd_calc_100ps__graph_Ext_Txt_Fig1d.tif

**Figure 1.** (Top, left) Initial system structure submitted for dynamics trajectory. Organic compounds are highlight in CPK: amylose, in violet; oleic acid, in medium blue; palmitic, in light green; stearic, in light blue. Water molecules are represented in stick. (Top, center and right; down, left) Snapshots of intermediate fluctuations of the dynamics trajectory corresponding to 40 ps and 100 ps, both frames already energetically optimized. Through those images, we are able to follow the adhesion of the majority of fatty acids over the amylose structure, progressively coiled. The non polar chains of some fatty acids seem to keep its mobility, essentially the palmitic acids (green arrows). Water molecules, rejected by fatty acids non polar termini, flow to the free amylose surface and stabilize through hydrogen bonds. (Down, right) Snapshots of control graphics for Energy (kcal mol-1) vs. time (ps) and temperature (K) vs. time (ps).

**Table 1.** Characteristics of the first molecular system modeled “Acetylated Pequi” in aqueous medium and Forcite energetic data.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Chemical species | Number of compounds | Number  of  atoms | Significant contributions to total energy | Energetic values (kcal.mol-1)  (0 ps) | Energetic values (kcal.mol-1)  (100 ps) |
| 5% Acetyl amylose | 1 | 2127 | **Valence energy (diagonal terms)** | -2133.04 | -2265.40 |
| Oleic acid | 18 | 972 | Bond | 484.14 | 481.06 |
| Palmitic acid | 14 | 700 | Angle | 1505.69 | 1432.25 |
| Stearic acid | 1 | 56 | Torsion | -4123.24 | -4179.23 |
| Water | 1166 | 3498 | **Valence energy (cross terms)** | -712.79 | -697.56 |
|  |  |  | Stretch-Bend-Stretch | -99.31 | -97.99 |
|  |  |  | Torsion-Bend-Bend | -182.22 | -157.91 |
|  |  |  | Bend-Torsion-Bend | -443.09 | -452.38 |
|  |  |  | **Non-bond energy** | -13252.96 | -13550.69 |
|  |  |  | van der Waals | 251.88 | 26.52 |
|  |  |  | Electrostatic | -13504.84 | -13577.21 |
| Total | 1200 | 7353 |  | -16098.78 | -16513.65 |

P_MMT_acetyl_Fig2_ops_40ps_100ps.tif

Acethyl_MMT_100ps_graphs_Ext_Txt.tif

**Figure 2.** Clockwise. Top view of the starting system (0 ps). Cetrimonium ions are dispersed into the unit cell (in medium blue). Snapshots of system fluctuations over MMT-O surface at 40 and 100 ps. (Down) Control graphics cards (E x 104 (kcal.mol-1) x t (ps)) and (T (K) x t (ps)) of the 100 ps dynamic trajectory, at 363 K. The oligomer acetyl amylose is colored violet; the atoms of oleic acids, gray; palmitic acids, green; light blue for stearic; cetrimonium ions, in medium blue and, all of them are shown in CPK. MMT-Na+ is mostly highlighted in thick traces.

**Table 2.** Characteristics of the second molecular system modeled “Acetylated Pequi” – MMT-O, in aqueous medium and Forcite energetic data.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Chemical species | Number of compounds | Number  of  atoms/ions | Significant contributions to total energy | Energetic values (kcal.mol-1)  (0 ps) | Energetic values (kcal.mol-1)  (100 ps) |
| 5% Acetyl amylose, fatty acids, H2O | 1200 | 7353 | **Valence energy (diagonal terms)** | 38707.23 | 38223.00 |
| MMT | 1 | 10466 | Bond | 34248.87 | 34089.29 |
| Na+ | 250 | 250 | Angle | 9039.24 | 8769.44 |
| C19H42N+ | 9 | 620 | Torsion | -4581.38 | -4636.27 |
|  |  |  | **Valence energy (cross terms)** | -755.24 | -733.84 |
|  |  |  | Stretch-Bend-Stretch | -106.01 | -90.73 |
|  |  |  | Torsion-Bend-Bend | -188.59 | -157.53 |
|  |  |  | Bend-Torsion-Bend | -471.24 | -490.71 |
|  |  |  | **Non-bond energy** | -1672453.60 | -1673698.83 |
|  |  |  | van der Waals | 778.90 | -352.33 |
|  |  |  | Electrostatic | -13504.84 | -1673346.51 |
| Total | 1461 | 18069 |  | -1673232.50 | -1636209.67 |

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