Molecular simulation has become an essential tool for studying organoclay systems at atomic resolution. Significant progress in simulation methodology and computing power has enabled simulations of increasingly large systems and long time scales. However, the accuracy of the simulations in predicting physical observables relies on both accurate energy functions and effective conformational sampling after the system reaches equilibrium. It is essential to prove the simulation systems reach equilibrium and the key properties such as energy and basal spacing are converged.

The convergence tests were performed to show that the 3 ns simulation time is enough for our CTMA-Mt case. The square deviation of energy, basal spacing sampled from nearest 0.2 ns as a function of time in the last 1 ns of NPT ensemble simulation for 0.33, 1.0, 1.66CEC case were calculated to show the degree of convergence (Fig. S1-S3). The threshold was set to be 0.1 in all tests, which means that the properties are converged when the square deviation remains below 0.1. From the convergence tests, the square deviation of all cases remain below the chosen threshold. As a result, it can be concluded that the simulation systems reach equilibrated after 3 ns simulation.

Fig. S1 Convergence of total energy of the last 1 ns NPT ensemble simulations for 0.33, 1.0, 1.66 CTMA-Mt cases. The convergence threshold is denoted by a dashed black line.

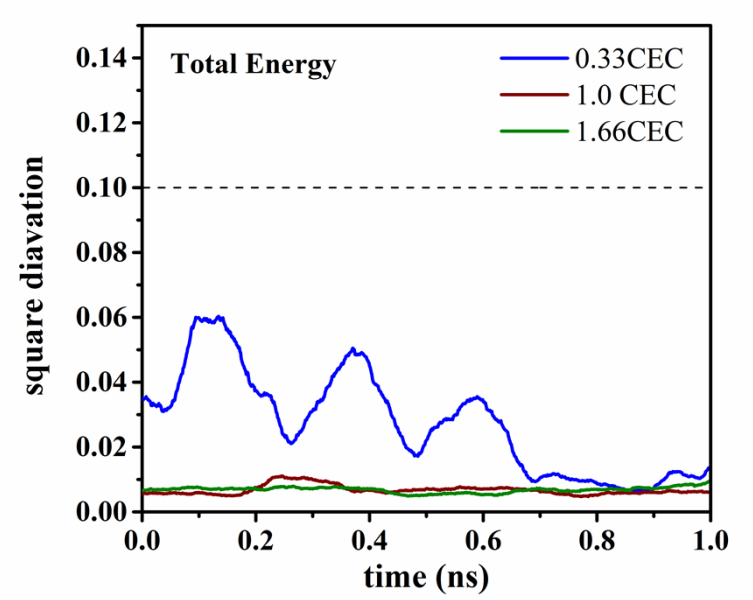

Fig. S2 Convergence of basal spacing of the last 1 ns NPT ensemble simulations for 0.33, 1.0, 1.66 CTMA-Mt cases. The convergence threshold is denoted by a dashed black line.