

Supporting Information

Ergodic-to-nonergodic phase inversion and reentrant ergodicity transition in DNA-nanoclay dispersions

Najmul Arfin¹ and H. B. Bohidar^{1,2*}

¹Polymer and Biophysics Laboratory, School of Physical Sciences

²Special Centre for Nanosciences

Jawaharlal Nehru University, New Delhi-110067, India

*Corresponding author email : bohi0700@mail.jnu.ac.in

Tel: +91 11 2670 4699, Fax: +91 11 2674 1837

Theoretical Model

We propose a simple and crude lattice model to capture the main experimental results. Here profile of electrostatic interaction energy between DNA and laponite on a lattice for different concentrations were studied. Role of hydration in this model is totally ignored. Further extension of this model using more computational and rigorous mathematics will better explain the experimental finding in better and quantitative manner.

Laponite particle which has negatively charged surface and positively charged edge was assumed to be a dipole. Laponite charge and partial charge on DNA is represented as q_D and q_{DNA} respectively. Here laponite (dipole) is assumed to have high charge (q_D) as compared to DNA partial charge (q_{DNA}). This is because on close approach of laponite towards DNA, laponite will see only a fraction of charge on DNA as compared to total charge present on long DNA strand.

Initially, the system was modeled on a 50×50 square lattice where laponite platelets were allowed to occupy all the sites with random orientations. For this configuration total electrostatic energy were calculated keeping in mind only nearest neighbor interaction between dipoles for the sake of simplicity. In next step, volume fraction of DNA on the square lattice was increased by randomly selecting a site and replacing dipole site with a DNA. Total electrostatic energy for this DNA-L configuration were calculated by assuming only nearest neighbor interactions which may consist of DNA-L and L-L interactions. Further increase in DNA concentration was done by again randomly replacing a Laponite site with that of DNA using random number generator. Total electrostatic energy was again calculated for this DNA-L configuration on lattice by assuming only nearest neighbor interactions which may consist of DNA-L, L-L and DNA-DNA interactions. These energies were then plotted against different DNA-L concentrations to get energy profile.

Thus, replacing laponite sites with DNA on lattice sites eventually gives experimental realization having DNA-L system having different volume fraction. Ensemble averaging of energy for each volume fraction was done over 100,000 times by using different random seed for random replacement of laponite with DNA. Energy profile was obtained by calculating total electrostatic energy for different volume fraction of the system.

Figure S1 shows plot of electrostatic interaction energy as a function of DNA-L concentration for three different q_D . It is to be noticed that for acquiring minimum interaction energy a specific charge ratio between q_D and q_{DNA} was required. FigureS1 ($q_D = 100$) depicts that system is stable and maximum interaction between DNA and laponite occurs at intermediate volume fraction of DNA and laponite which is analogous to region (ii) of the experimental results. However charge ratio between DNA and laponite is the crucial factor in achieving region (ii) as was seen in experiments.

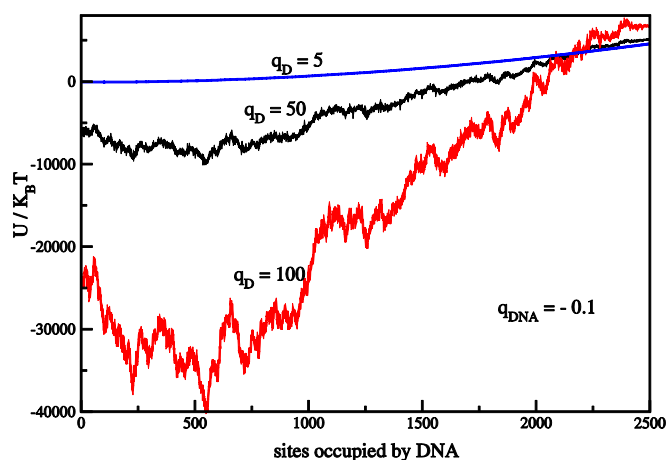


Figure S1. Variation of potential energy as a function of volume fraction of DNA for different $q_D:q_{DNA}$. Note that stable energy configuration is achieved as was seen in experiments (region (ii)) for $q_D = 100$.

Thus, a simple lattice model revealed that for some intermediate DNA-L concentration stable energy configuration exists where maximum interaction occurred (onset of non-ergodic phase) between DNA and laponite.