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Monte-Carlo Simulation on Association Number of Titania Multicomponent Colloidal Clusters

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1. Introduction

Multicomponent colloidal clusters experiments were carried out in the Japanese Experiment Module (JEM) 'Kibo' in 2020. There are two kinds of microparticles, that is, positively charged ones and negatively charged ones. Thus, colloidal clusters are formed by Coulomb attraction force. The particles are made of titania (TiO_2). Since the density of particles is much larger than that of water, the particles are rapidly precipitated on the ground. Therefore, to make the sedimentation period long enough to form clusters, we used the microgravity environment. The colloidal samples gelled by ultraviolet light on orbit to fix the particles coordinates. After recovery of the samples, association number of clusters was investigated at the Yamanaka lab. To understand the distribution of association number, Monte-Carlo simulation was executed. The simulation results are compared with the experimental ones.

2. Experimental Results

The association number of titania clusters was investigated. All results are shown in another paper[1)]. In this paper, the population distribution of association number of $m \geq 1$ is shown in **Figure 1** as a summary of the titania cluster experiment. In this figure, the population of $m=0$ excluded since this position is not a cluster but a single particle.

The tendency of distribution monotonously decreases with increasing association number. However, the population of m=4 in the case of NaCl concentration of 50 μM does not decrease and is the same population as that of m=3. This is a good result because a titania tetrahedron is quite valuable from the engineering view of point due to a minimum element of a diamond structure. The titania cluster with diamond structure is theoretically predicted as a perfect photonic material. Thus, we execute Monte Carlo simulation to know why the population of m=4 and 50 μM is larger than the other cases

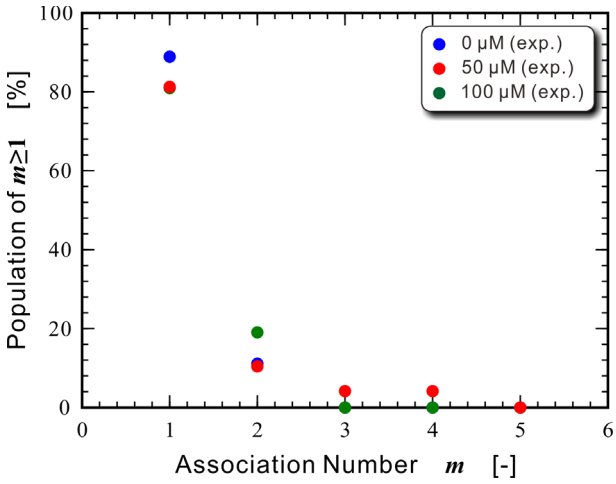

Figure 1. Distribution of association number of m≥1. Cluster population of m≥1 is calculated by excluding single particles.

3. Monte Carlo Simulation

We provided a Metropolis Monte Carlo simulation code. This is one of Markov chain Monte Carlo codes. The Metropolis method is expressed as shown in Eq. (1).

$$\langle A \rangle = \frac{1}{Z}\int d\boldsymbol{r}\, A(\mathbf{r},\mathbf{p}) \exp\left\{-\frac{H(\mathbf{r},\mathbf{p})}{k_B T}\right\} \simeq \frac{1}{Z}\sum_{i}^{N} A(\boldsymbol{r}_i,\boldsymbol{p}_i) \exp\left\{-\frac{H(\mathbf{r}_i,\mathbf{p}_i)}{k_B T}\right\}, \tag{1}$$

where A Z, H, $\mathbf{r}$, $\mathbf{p}$ are targeting physical quantity, a partition function, the Hamiltonian, coordinates, and momentum, respectively. The partition function is described as follows.

$$Z = \int d\mathbf{r} \exp\left\{-\frac{H(\mathbf{r},\mathbf{p})}{k_B T}\right\} \simeq \sum_{i}^{N} \exp\left\{-\frac{H(\mathbf{r}_i,\mathbf{p}_i)}{k_B T}\right\}. \tag{2}$$

To calculate the Hamiltonian, the DLVO potential is used as the potential energy and the virial theorem is used to calculate the kinetic energy due to this system is not ideal gas.

4. Comparison

The simulation results are compared with the experimental ones in three cases, that is, NaCl concentration of 0, 50, and 100 μM. The comparison is shown in **Figure 2**. It is found that the simulation results are consistent with the experimental results. The population of association number of m=4 is largest in the 50 μM case in both the simulation and experimental results.

To know whether the concentration of 50 μM indicates the largest population, the simulation is executed in several NaCl concentration cases. The result is shown in **Figure 3**. This figure shows that the largest population exists at the concentration of 50 μM. The result also shows that the population is low in the range of 20 to 40 μM. It is difficult to understand this behavior because the Coulomb interaction decreases with increasing the concentration due to the shorter Debye length. More investigation is needed in future.

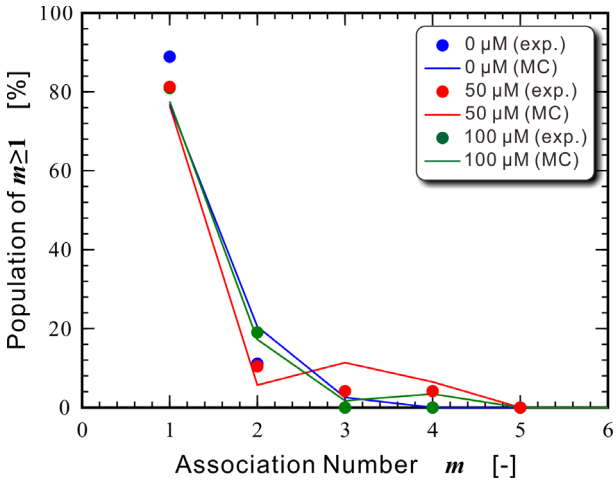

Figure 2. Comparison of simulation results with experimental results. The tendency of simulation result is consistent with the experimental one.

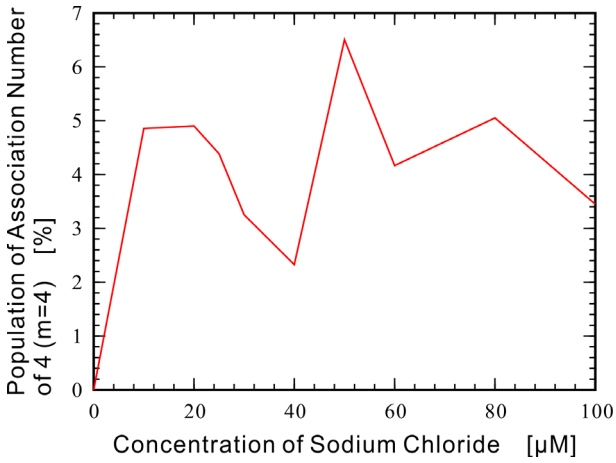

Figure 3. Population distribution of association number of m=4. The population of m=4 has the maximum at the NaCl concentration of 50 μM.

5. Conclusions

To explain the experimental results in space, we conducted the Monte Carlo simulation. The simulation results are compared with the experimental ones. The tendency of population distribution of simulation is consistent with the experiments. The simulation also indicates that the population is largest at the concentration of 50 μM.

References

1) H. Miki. T. Ishigami, J. Yamanaka, T. Okuzono, A. Toyotama, J. Mata, H. Komazawa, Y. Takeda, M. Minami, M. Fujita, M. Doi, T. Higuchi, H. Takase, S. Adachi, T. Sakashita, T. Shimaoka, M. Nagai, Y. Watanabe and S. Fukuyama: Clustering of charged colloidal particles in the microgravity environment of space, npj Microgravity, **9** (2023) 33, DOI: https://doi.org/10.1038/s41526-023-00280-5.