p-ISSN 2089 – 0133 e-ISSN 2477-6416
URL : https://jurnal.uns.ac.id/ijap/article/view/87643
Indonesian Journal of Applied Physics (IJAP) Vol. 16 No. 1 page 30
DOI : https://doi.org/10.13057/ijap.v16i1.87643

MODELING DISTRIBUTION PATTERNS OF COLLOIDAL PARTICLES ON ELECTROPHORETIC DEPOSITION DEMONSTRATED USING KINETIC MONTE CARLO SIMULATION

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Received 2024-06-05, Revised 2026-10-16, Accepted 2026-04-01,
Available Online, Published Regularly April 2026

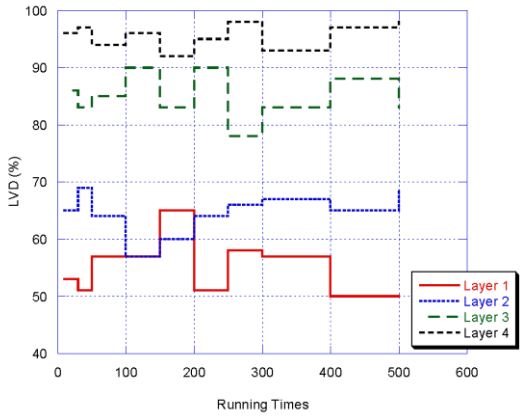
ABSTRACT

Electrophoretic deposition (EPD) patterns of colloidal SiO2 particles (400 nm) in solution were analysed and demonstrated using the kinetic Monte Carlo (KMC) simulation method. The behaviour of the colloidal particles which formed electrical double layers in solution was described by the DLVO theory. The dynamic particle migrations of the EPD process were estimated by the KMC simulation to statistically determine the probable particle distribution based on thermodynamic theories. Results show that the particle distribution and deposition in solution were significantly influenced by the presence of the zeta potential value (η) in the solution, and applied voltage in the EPD system. The particles aggregated together in the solution with a small η value due to the strong Van der Waals attractive forces among the particles. If a large η value was adopted, all the particles moved away from central and scattered around the edges the system space because of the strong electrostatic (repulsive) forces among them. The external voltage applied to the EPD system in different η value can affect the formation of particle deposition pattern. The EPD pattern with little vacancy site can be fabricated using a small η value with a constant voltage operating condition. However, the vacancy sites of the EPD pattern became large by the on-off voltage operating scheme. The quality of the EPD films cannot be improved using this strategy.

Keywords: colloidal particles; EPD; KMC; photonic crystal; zeta potential.

INTRODUCTION

Electromagnetic wave (EM) propagation is impacted by periodic dielectric or metallo-dielectric nanostructures seen in photonic crystals. Since photonic crystals are intended to modify photon propagation properties, the word "photonic" has been added. It is said to be a “crystal” because the structure of the photonic crystal has highly ordered assemblies of particles. Some applications of photonic crystals are as optical fibre, nanoscopic lasers, radio frequency antennas, reflectors, light-emitting diodes, and photonic integrated circuits. The majority of colloidal particle-based photonic crystals feature highly ordered particle arrays that can be generated over a large area of about a centimeter. There are several approaches to producing photonic colloidal crystals. Self-assembly strategy is known as the best method to



fabricate photonic colloidal crystals. Colloidal crystals can self-assemble using a variety of techniques, including gravity sedimentation, vertical deposition, electrophoresis, spin coating, and crystallization in physically restrained cells [1–3]. Compared to other methods, electrophoretic deposition (EPD) is one of the effective and efficient methods to fabricate large-scale colloidal crystals with controllable thickness. The quality of the produced EPD films is affected by the process variables and parameters, i.e. the applied voltage to the EPD system [4–7]. If the quality of the films needs to be improved, the EPD process has to be understood well. However, the phenomena of particle deposition in solution are complex and difficult to clearly interpret.

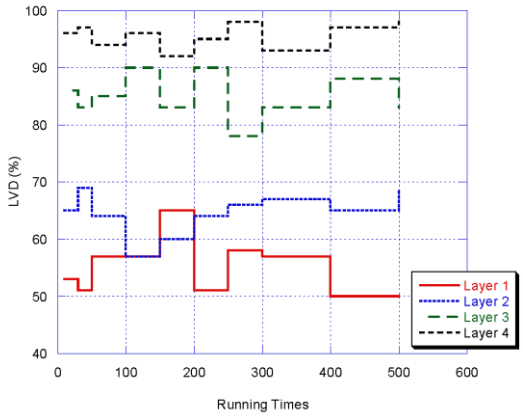
The deposition defects of the EPD films, such as vacancy, are still formed due to the complicated deposition process [8-9]. The formation of the vacancies is difficult to be observed using experimental approaches during the particle deposition process. In this work, the objective is to understand the defect formation of the films by the electrophoretic deposition process using the kinetic Monte Carlo (KMC) simulation method. The purpose of the KMC approach, one of the techniques used to analyze uncertainty propagation, is to discover how random variation and ignorance, performance, or reliability of the system that is being modeled [10–12]. The study involves the evaluation of the effects of the process operating variables, such as the zeta potential energy value of the solution and the electric force, on the particle distribution and deposition. The KMC simulation results are used to determine the EPD patterns under various operating situations. Effects of the operating variables on the electrophoretic particle deposition are analyzed and evaluated using the calculation data analysis. The simulation results can be further applied to the defect control of the fabrication of the EPD films.

METHOD

In order to simplify, we can simplify the structure of the simulation system by writing code with a simple and easy-to-understand approach and finally perform the analysis according to the computational result. The following table includes all of the necessary information for the constants utilized in this investigation.

Table 1. The constant data for this study

The constants	Value
Hamaker constant (SiO_2)	$0.46x10^{-20}$ Joule
Radius Particle	200 nm
Distance between particles	1 nm
Electric constant in water	$8.85\ x10^{-12}$ Farad/m
Relative dielectric constant in water	80 Farad/m



Boltzmann constant	1.38065 x10^{-23} J/K
Temperature	298^{0}K
Voltage	10 mV

Determine Moving Particles

The distribution of the colloidal particles in the EPD process using KMC simulation was demonstrated three-dimensional (3D). There are three main forces, the Van der Waals force, the electrostatic force, and the electric force, that are considered in the solution system. It is expected that the particles in 3D systems will most likely migrate in one of two ways: between layers for two directions (up and down), or the particles can move to four nearest neighbors' lattice side, or particles will steady on place. Forbidden for particles to move if neighbouring location are occupied by another particle, and the movement of particles will not be out boundary. Moreover, the moving particle can also remain in the same position during the migration process [13].

Simulation State

Derjaguin and Landau introduced a theory for the stability of colloidal dispersions, which can be used to explain how stable colloidal particles are in solutions. The idea explains the interaction of charged surfaces in a liquid medium. It combines the Van der Waals attractions and the electrostatic repulsion effects. The SiO_2 colloidal particle was selected as material which diluted in solution during the deposition process. The Hamaker constant for SiO_2 has been shown in **Table 1.** The effect of Van der Waals attraction energy in solution, the Hamaker statement can be used to estimate, which is described below

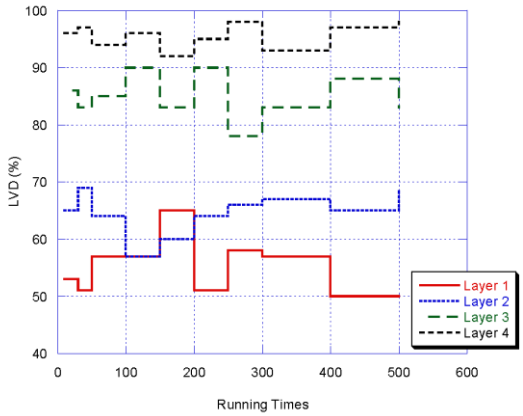
$$V_{vdW} = -\frac{A}{6}\left[\frac{2}{s^2-4}+\frac{2}{s^2}+ln\frac{s^2-4}{s^2}\right] \quad (1)$$

Where the distance s between the centers of the spheres is equal to ;

$$s = \frac{2a+h}{a} \quad (2)$$

Then, for the influence of electrostatic repulsive energy in the solution can be described using Coulomb's law as below;

$$V_{Elc} = \sum_{i=1}^{N} \quad \sum_{j=1\ j\neq i}^{N} \quad \frac{q_i \times q_j}{4\pi\varepsilon_0\varepsilon_{water}\, r_{ij}} \quad (3)$$



Therefore, the total energy of these particles in the solution system is the summation of the Van der Waals attraction energy and electrostatic repulsive energy, as shown [13];

$$V_T = Vv_{dW} + V_{Elc} \quad (4)$$

Electrostatic force will cause the charged particles in solution. The value of surface charge (q) depends on several variables, as described below;

$$q = \eta \times c \quad (5)$$

Where η is the zeta potential, and c is the capacitance of the sphere, defined by ;

$$c = 4\pi \times \varepsilon_0 \times \varepsilon_{water} \times rd \quad (6)$$

Another effect will influence potential charge in the solution is the electric field. When a colloidal suspension is exposed to an electric field, the particle motion is proportional to the strength of the applied field. The motion is called electrophoresis. Electrophoresis means motion due to an electric field, but the deposition method with the influence of electric fields is called electrophoretic deposition (EPD). A uniform electric field is constant at every point in this work. By aligning two conducting plates parallel to one another and keeping a voltage (potential difference) between them, it can be approximated; nevertheless, it is merely an approximation due to edge effects. Then the electric field energy can be described using the equation below;

$$V_{elf} = q\ x\ E_{elf}\ x\ d \quad (7)$$

Where V_{elf} is the electric field energy, then V is the voltage (Volt), and d is the distance separating the plates. A positive charge will feel a force away from the positively charged plate, going against the direction that the voltage raises, because the negative sign results from positive charges repelling one another [14-16]. In the case of the influence of the electric field, the position of the positive and negative electrode as Figure 1.

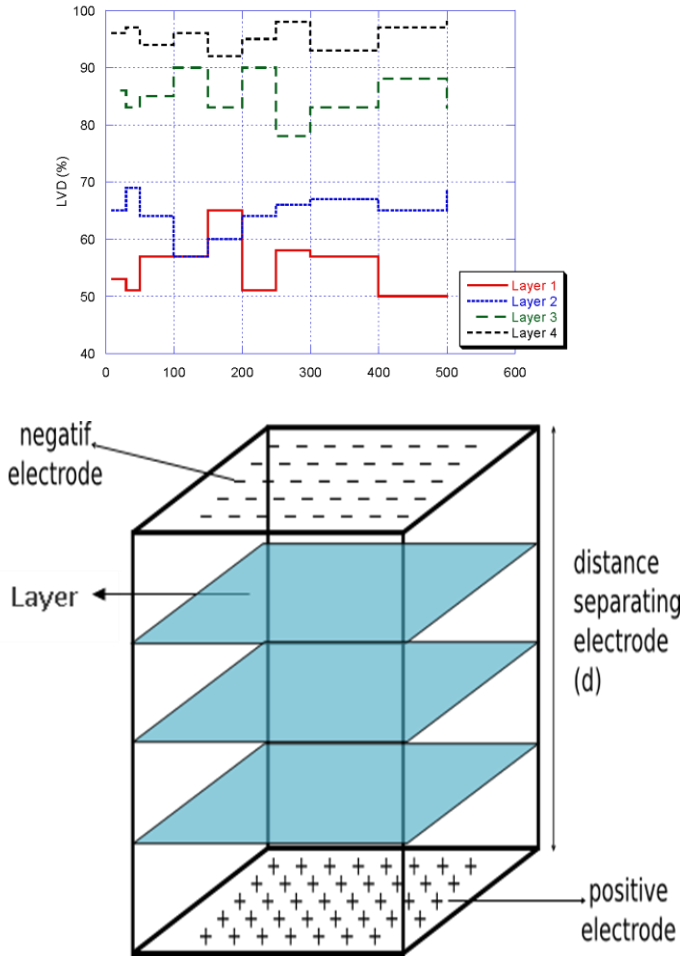


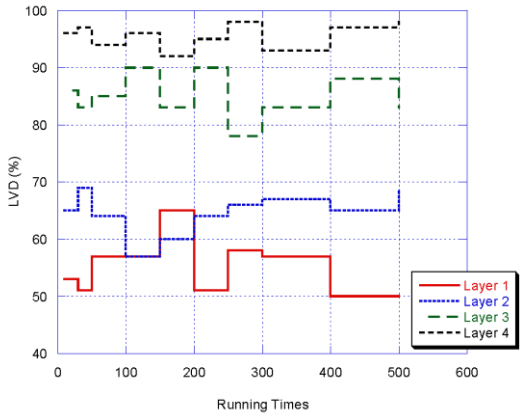
Figure 1. The position of electrode in deposition influence electric field

The simulation results distribution of particles during precipitation samples was determined and analyzed using a Kinetic Monte Carlo (KMC) simulation. The probable moving rate of a particle in a system state can be described by the Arrhenius equation as shown in the equation

$$W_T = v_0 \times exp\left(\frac{-\Delta E}{k_b T}\right) \tag{8}$$

The method for producing a weighted average in statistical KMC previous mechanics to the development of the Metropolis algorithm involved creating a large number of random configurations of the system, computing the properties of interest (such as energy or density) for each configuration, and then averaging the results using the formula exp(E/kbT), where E is the energy, T is the temperature, and kb is Boltzmann's constant.

This leads to improved convergence since the sampling concentrates on the low-energy configurations, which are the ones that contribute the most to the Boltzmann average. The authors developed the following approach to select configurations with a probability exp(E/kT) that can be weighed equally: The new energy is calculated for each configuration, after which the move is either always accepted if the new energy is lower than the prior configurations or rejected with a probability of exp(E/kT) if the new energy is higher. The most recent acceptable configurations is counted once again for statistical averages when a move is denied and is utilized as the foundation for the following move. To simplify it, define Metropolis algorithms can describe as below:



$$W = \{v_0 \qquad if\ \Delta E \le 0\ v_0\ \times exp\left(\frac{-\Delta E}{k_b \times T}\right) \qquad if\ \Delta E > 0 \tag{9}$$

Where W is the rate of probability of a particle moving. Another important thing in KMC is define event of a particle will move was described on equation 10 below

$$\sum_{x=1}^{m-1} W_x \le \xi W \sum_{y=1}^{N} W_y < \sum_{z=1}^{m} W_z \tag{10}$$

Selection of the moving path by linear as shown in Figure 2 search is represented by a cumulative and conditional probability process [13],[17].

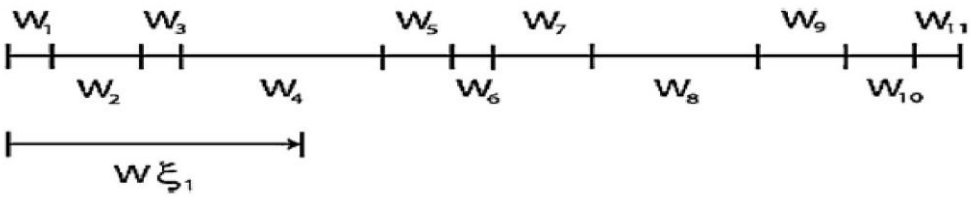


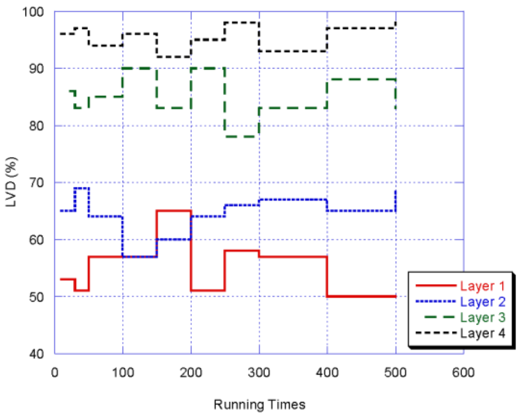
Figure 2. A particle moving step's decision-making event selection path is shown schematically.

RESULTS AND DISCUSSION

The particle migration process in two cases was studied for the 3D systems in this section. For the first case, the particles are considered by the effect of both the Van der Waals attractive force and the electrostatic repulsive force during the particle migration. The other case is to apply an external voltage on the system, which presents an electric field in the solution during the particle movement. The deposition patterns were evaluated by the findings of the KMC calculation. Additionally, the influence of the η values on the particle distribution was studied in this section.

Effect Of Zeta Potential (η) Values Without Applying External Voltage

The simulation experiments were also considered in the situation of different η values. The original condition is assumed that there are 100 particles located on the top layer in a rectangular system as shown in Figure 3**.** The simulation space system was set up in four layers, and there are 100 vacancies in each layer, which can be occupied by particles. When the particle migration began under the influence of η, randomly, the particles traveled from the top layer to the bottom layer in the system domain.




```
sumpq(:,:,1) =

     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
```

Frist Layer

```
sumpq(:,:,2) =

     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
```

Second Layer

```
sumpq(:,:,3) =

     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
     0     0     0     0     0     0     0     0     0     0
```

Third

```
sumpq(:,:,4) =

     1     1     1     1     1     1     1     1     1     1
     1     1     1     1     1     1     1     1     1     1
     1     1     1     1     1     1     1     1     1     1
     1     1     1     1     1     1     1     1     1     1
     1     1     1     1     1     1     1     1     1     1
     1     1     1     1     1     1     1     1     1     1
     1     1     1     1     1     1     1     1     1     1
     1     1     1     1     1     1     1     1     1     1
     1     1     1     1     1     1     1     1     1     1
     1     1     1     1     1     1     1     1     1     1
```

Fourth Layer

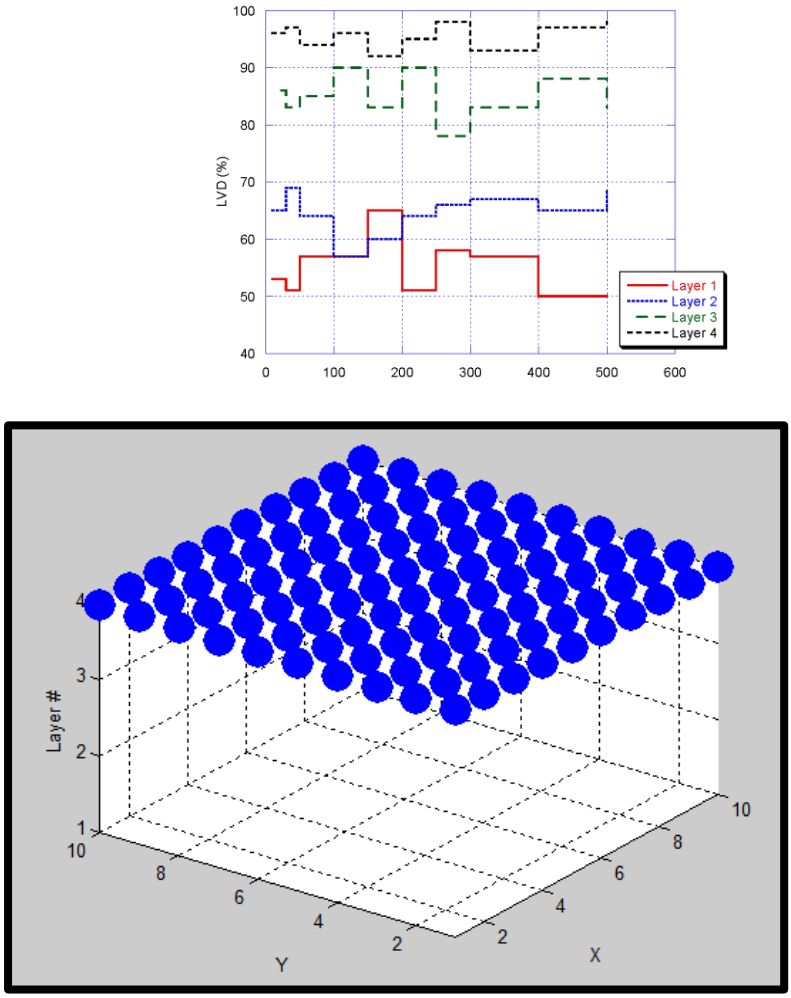


Figure 3. Pattern of the initial condition particles before moving when the difference in the value of the zeta potential in the 3D case.

The calculation results of the particle distribution under different η values are shown in Figure 4. Figure 4(a) shows the particle distribution when η = 0 mV. The particle distribution pattern demonstrates the particle aggregation in each layer. These particles in the solution have no surface charges under this condition, which means the electrostatic force is absent among these particles. Therefore, the Van der Waals attractive force is the dominant force acting among the particles. The particles can be attractive to each other and form the pattern.

If the η value increases to 1 mV, the particle distribution is shown in Figure 4(b). The particles distributed randomly in the system space. Some are aggregated in the corner, and some are spread on the edge. Even though the Van der Waals force still dominates the particle-particle interaction, the repulsive (electrostatic) force starts to affect the migration process in this small η case. This interprets that the Van der Waals force is strong enough to attract some particles to stay together, and some other particles moved to the edge area by the weak repulsive force to separate them, as shown in this Figure

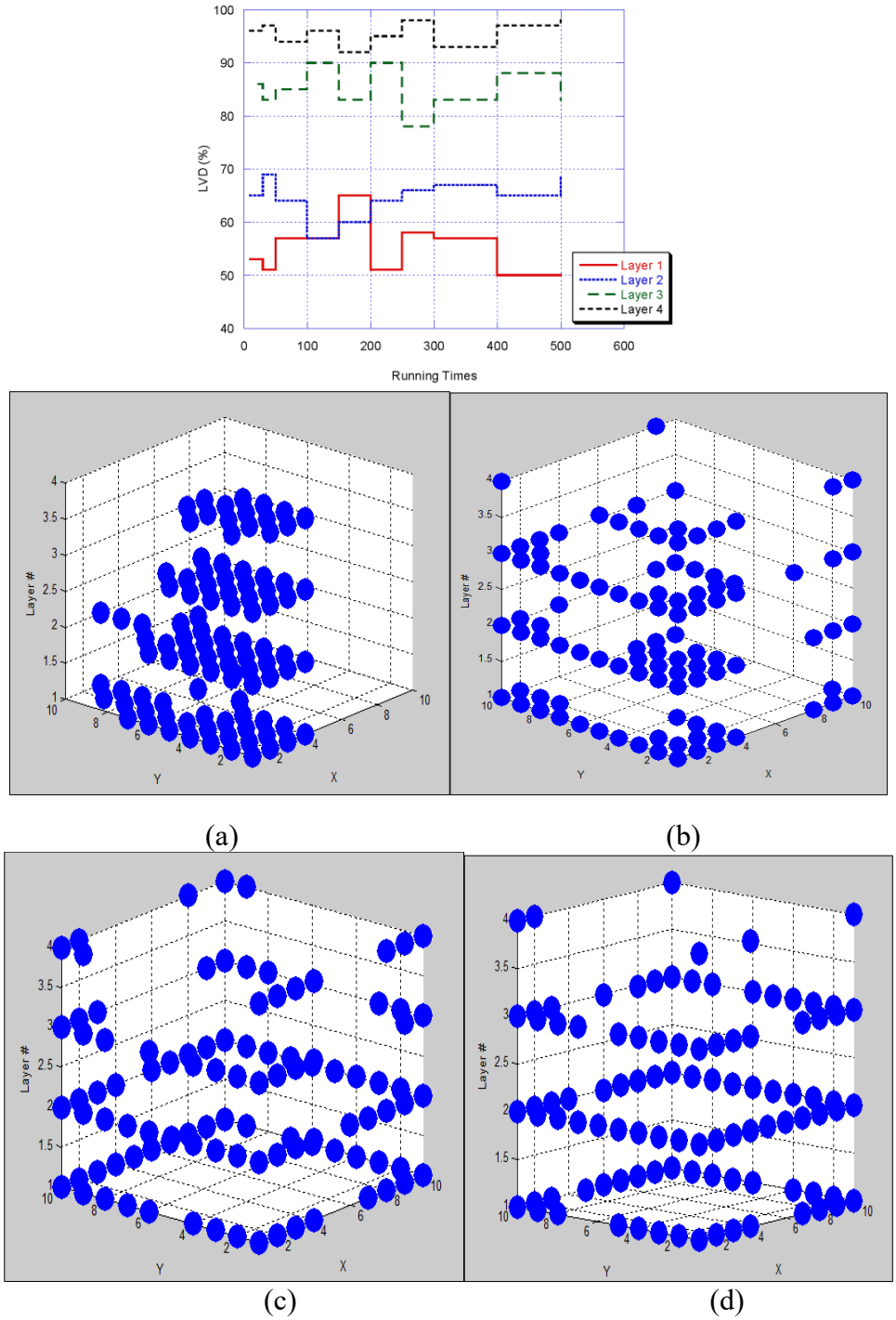
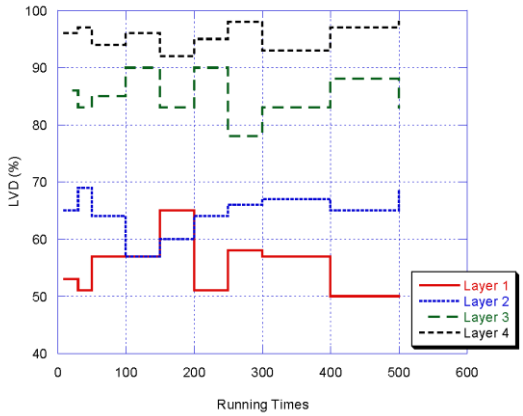


Figure 4. Pattern of the most possible position was occupied by particles in 3D cases when value of zeta potential (a) η =0.0 mV, (b) η =1 mV, (c) η =3 mV, (d) η =10 mV

Figure 4(c) illustrates the distribution pattern when the η value increases to 3 mV. The pattern shows that most of the particles are occupied around the boundary, but some particles stick together, located in the corner area. This is because the Van der Waals forces attract particles to stick the particles together, but the electrostatic forces push the particles into the system edges. This also implies that the Van der Waals force and the electrostatic force influence the



particle-particle interaction equally. The distribution pattern demonstrated the equilibrium condition of the two forces and reached the steady state.

If η changes to 10 mV, the particle distribution is shown in Figure 4(d). The particles scattered separately around the edges in each layer of the system boundary, and large vacancies appeared in the central area of the system. The strong repulsive force dominates the particle-particle interactions system due to high particle charges for the solution at a high zeta potential value. Therefore, the particles avoided particle agglomeration and were driven as separately as possible to satisfy the thermodynamic theory at this η value.

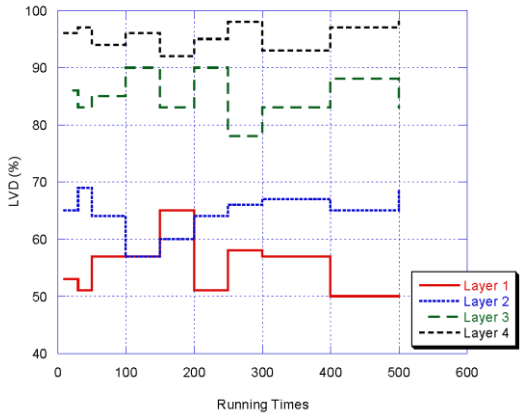
In a practical situation, the particles were diluted and dispersed in solution under a η condition. The above cases illuminated the effects of η values on the patterns of particle distribution in solution using KMC simulation. The simulation results represented the final state of the particle-particle interaction after the particle migration process. The patterns of the particle distribution showed the operation without any external voltage. The calculation results of these cases are to be used for the operation with the applied voltage applied to the electrophoretic deposition system in the following section.

Effect Of Zeta Potential (η) Value on Electrophoretic Deposition (EPD) Process

In the second section of 3D cases, the EPD is studied to investigate the particle deposition process under the influence of the externally applied voltage. The external voltage was applied at 10 mV between the top and bottom layers for all cases. Three main forces, the Van der Waals force, the electrostatic force, and electric force (induced by the applied voltage), are considered in the solution system. The probable events of the particles' migration in 3D systems are assumed to move both in the same layer and in four different orientations (right, left, front, and back). However, the particle can only jump down in the inter-layers for one direction in the system space due to the presence of the electric field caused by the voltage, as shown in Figure 2**.** Additionally, the moving particle can also remain in the same position during the migration process. Therefore, the total number of probable events is six in this case. The probability of the particle migration can be determined using the KMC method. The accumulative patterns of the particle migration stage can then be evaluated according to the calculation results.

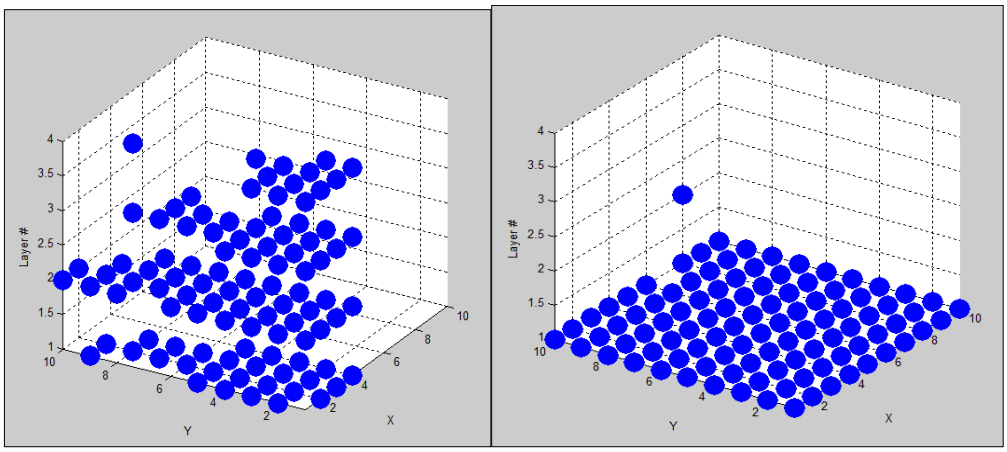
The particle deposition pattern can be estimated using the vacancy distribution of particles in each layer. Each layer's vacancy density (LVD) is described as [13];

$$LVD = \frac{N_s}{N_v}\, x\, 100\% \qquad (11)$$



Where Nv is the number of vacancy sites, and Ns is the number of total sites in a layer. The vacancies range in each layer is 100% as a maximum value, which means no particle is occupied in the layer, and 0% as a minimum value to represent no vacancy in the layer.

In the previous section, the particle distribution can be simulated in different η value conditions when the particles are dispersed in the solution. The final patterns of the particle distribution in Figure 3 are taken as the initial conditions for the EPD simulation, as shown in Figure 4. After particles dissolve in solution and approach a steady state, the electrophoretic deposition process starts due to the influence of the electric field in the solution system. The experiment simulation using KMC was programmed for a hundred times of particle jumps and repeated 500 times. Then, particles can move to the electrode located in the bottom layer because the electric force acts on the particles that contain surface charge. The particles can finally deposit on the electrode (bottom layer) as an EPD pattern.



(a) (b)

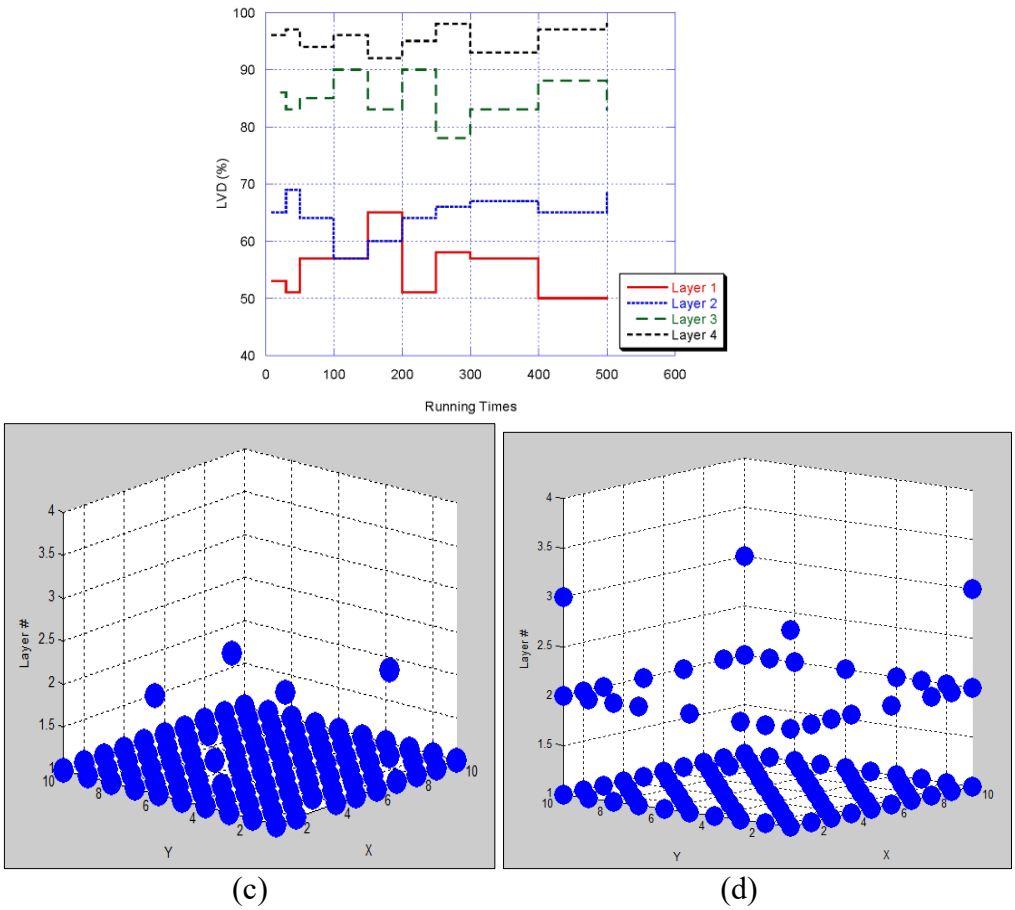
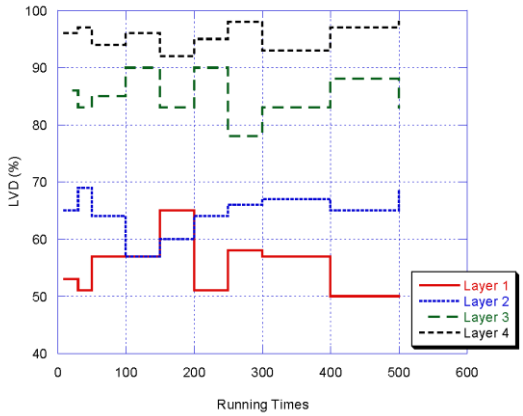


Figure 5. Pattern of the most possible position was occupied by particles in 3D cases when EPD in different zeta potential value (a) η = 0.0 mV, (b) η = 1 mV, (c) η = 3 mV and (d) η = 10 mV.

Figure 5(a) shows the EPD pattern for the η = 0 mV case. The initial condition of this case is shown in Figure 4(a). The final result of the deposition pattern is similar to the initial condition, as seen in Figure 5(a) and Figure 4(a). The deposition pattern shows the particle aggregation in each layer. This is because these particles in the solution have no surface charges under this condition. Thus, the particles were not influenced by the electric field. The Van der Waals attractive force is still the dominant force acting among the particles. Therefore, the particles can be attractive to each other and form the pattern. The deposition pattern shows that LVD values are high for each layer, i.e., 75% in the bottom layer, as listed in **Table 2**. This suggests that the deposition operation is not well-suited to form great quality of film for this case.

The EPD pattern can be significantly changed when the η value increases to 1 mV, as shown in Figure 5(b). The deposition pattern showed only one vacancy in the bottom layer. The deposition process begins with the initial condition as shown in Figure 4(b). All the particles can move down to the bottom layer due to the presence of the electric field. Additionally, although the Van der Waals force dominates the particle-particle interaction, the weak electrostatic force, together with the electric force, can compete with the Van der Waals force. The effect of the combination of forces can lead the particles into the vacancies in the bottom layer. Thus, the LVD can be the lowest value as listed in **Table 2** in this small η case.

When the η value increases to 3 mV, the EPD pattern is demonstrated in Figure 5(c), which transformed from the initial pattern as shown in Figure 4(c). Four particles were located in the second layer, which means 4 vacancies appeared in the bottom layer. The LVD value of this case is higher than that of the previous case, as shown in Table 2. This indicates that the EPD



pattern is slightly affected by the change of the η value. If η changed to 10 mV, the EPD pattern is shown in Figure 5(d). Several particles are located at the edges of the higher layers for this case. Most of the vacancies appear in the center of the bottom layer. This is because the surface charges of the particles formed a strong electrostatic force in the solution at this η value. Therefore, the repulsive force dominates the particle-particle interaction to repel the particle away from the central area.

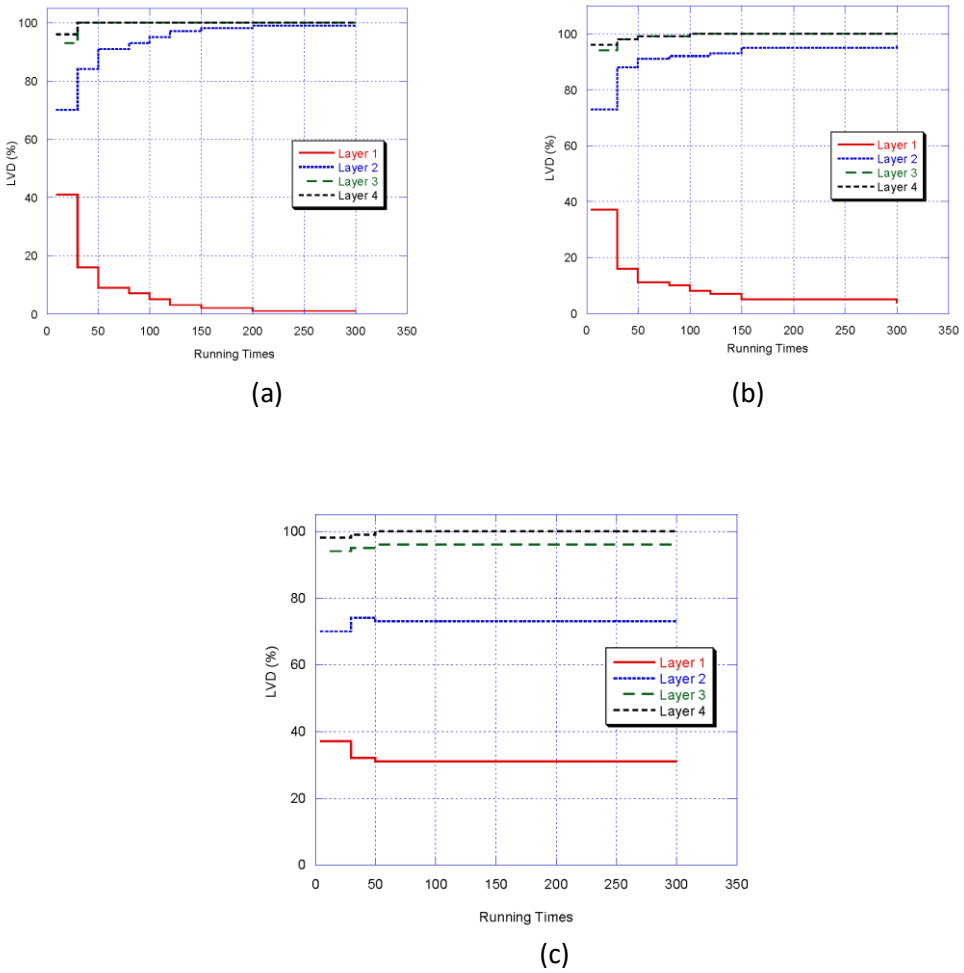
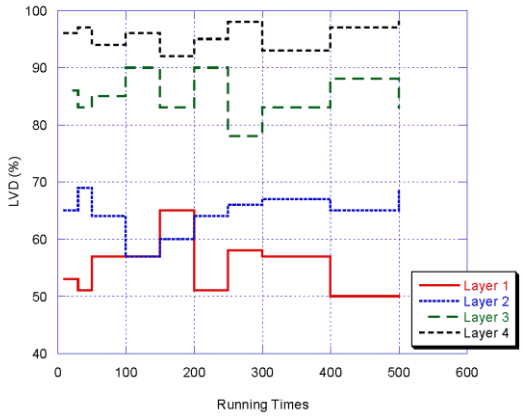


Figure 6. LVD data distribution of each layer during the EPD process for (a) η = 1 mV, (b) η = 3 mV, and (c) η = 10 mV cases.



The presence of vacancies is one kind of defect in this study. The quality of the films is influenced by the particle deposition pattern in the bottom layer. For high-quality particle films to be produced, voids in the bottom layer must be eliminated. From the KMC simulation data analysis, the vacancy distribution in each layer can be predicted during the EPD process. Figure 6 shows the moving track of the particles in each layer during the particle migration for different zeta potential cases. The LVD data in each layer for the EPD process are listed in **Table 2.**

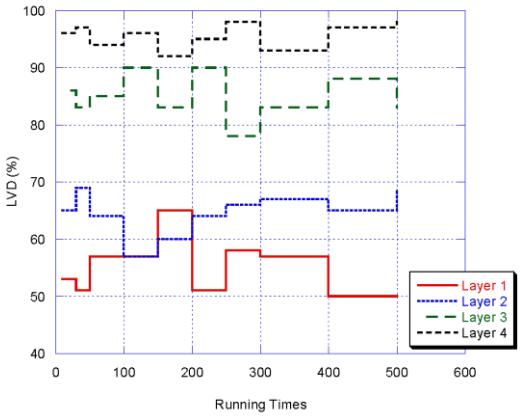
Table 2. LVD data for cases influence of electric field in different η value.

Sample	Zeta Potential (mV)	LVD (%)			
		Layer 1	Layer 2	Layer 3	Layer 4
1	0	75	74.8	74.6	75.5
2	1	2.3	97.3	100	100
3	3	5.9	94.4	99.8	99.8
4	5	18	81.7	99.3	100
5	10	31.4	72.8	95.8	99.8

The particles gradually began to move down from their places to the lowest layers. In all of the cases, the top layer's vacancy density (LVD) dramatically increased. The particle occupancy significantly went down to the bottom layer (layer 1). In each layer, the vacancy distribution reached a stable state after 200 repetitions of particle leaps without changing further for all of these cases. For η = 0 case, the vacancies remain in every layer due to no effect of the electric field on these particles. The distribution of the LVD values is similar for the two cases, η=1 mV and η=3 mV, as shown in Figure 6(a) and Figure 6(b). For η = 10 mV case, the LVD data in the first layer is the highest of these cases, which means the EPD pattern shows the worst defects for this operation.

On-off Voltage Operation Of Particles Deposition

The external voltage applied to the EPD system can significantly affect the deposition pattern. The number of vacancies, especially formed in the bottom layer, is taken as one type of the defects for the deposited film. The quality of the EPD films can be improved by applying the voltage to eliminate the vacancy generation during the EPD process. In the previous section, the vacancy of the EPD pattern was greatly reduced if a constant voltage was applied to the EPD process. An on-off voltage of a periodic operation scheme applied to the EPD process is proposed to evaluate the EPD pattern influenced by this scheme using the KMC simulation in this section.



The on-off voltage of the periodic operating scheme is shown in Figure 5 In this scheme, two types of operating conditions are involved in the EPD process according to the voltage states. If the voltage is in the "on" state, the particles in the EPD system are influenced by the electric field, which is induced by the voltage. On the contrary, when the voltage is in the "off" state, the EPD system is operated without the influence of the electric field. The EPD patterns were further analyzed to evaluate the effect of the on-off voltage operation on the particle deposition.

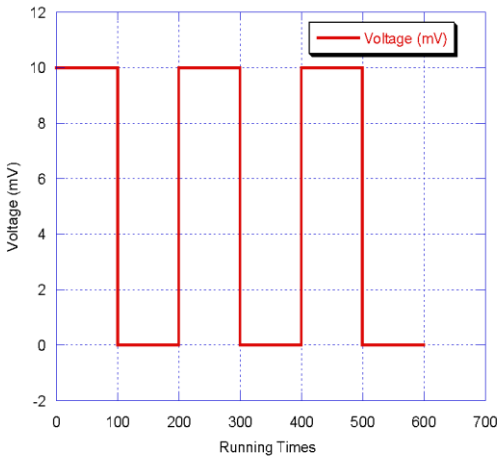
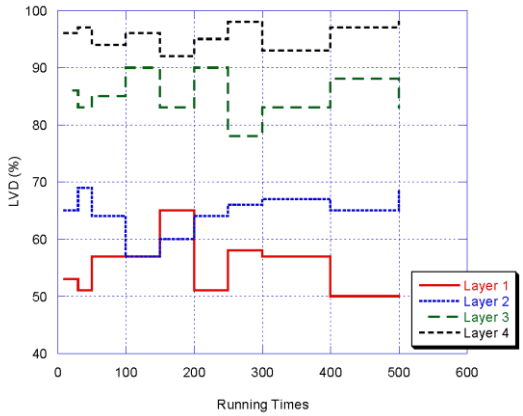


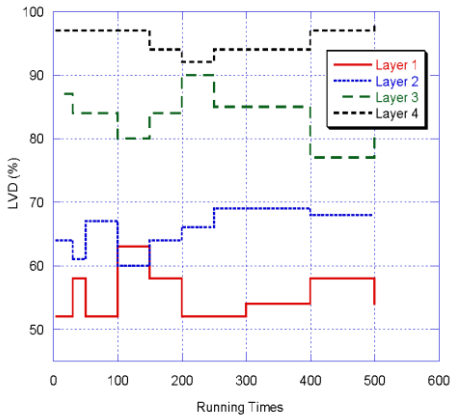
Figure 7. The scheme of on-off voltage operation, the first state is particle move 100 times in the voltage "on" and the second state is that particles move 100 times when the voltage is in the "off" state.

A design of experiment method is set up to carry out the study, as shown in Figure 7. The EPD system is designed as 4 layers with 100 particles, the same as the previous cases. Three factors, such as voltage on and off times, and the η value, were considered for the EPD experiments. Take sample 3 as an example. The initial conditions of all these cases in different η values were set up based on the results shown in Figure 4. The result shown under the condition of η = 1 mV, voltage on = 200 times, and voltage off = 100 times. The experimental outputs were the LVD data in each layer.

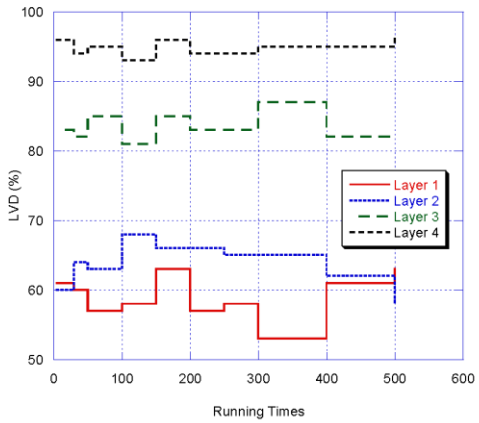
The LVD data in each layer for these cases are listed in **Table 3**. From the simulation data analysis, the LVD data of the on-off voltage cases showed significantly higher values than those of the constant voltage cases, as listed in **Table 2**. The lowest LVD values in the first layer (bottom layer) for each η value are samples 3, 6, and 9. All these samples are operated for a longer state of the voltage "on" (200 times) condition. Whereas, the highest LVD values in the first layer appeared for the samples under a longer state of the voltage "off" (200 times)



conditions, i.e. samples 2,5, and 8. These data also indicate that the on-off voltage operations produce the EPD films with high vacancy (defect) samples.



(b)



(c)

Figure 8. LVD data distribution of each layer in the periodic system during the deposition process, when operated for a longer state of the voltage "on" (200 times) condition in (a) η = 1 mV, (b) η = 3 mV, and (c) η = 10 mV cases.

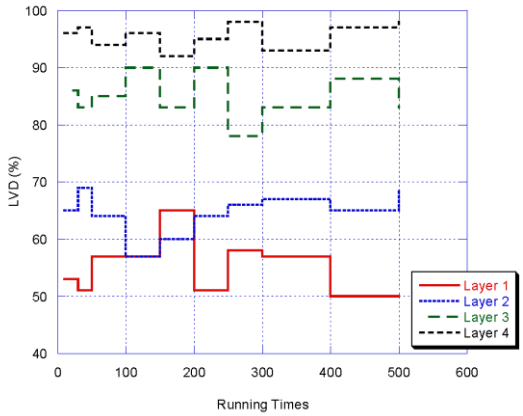
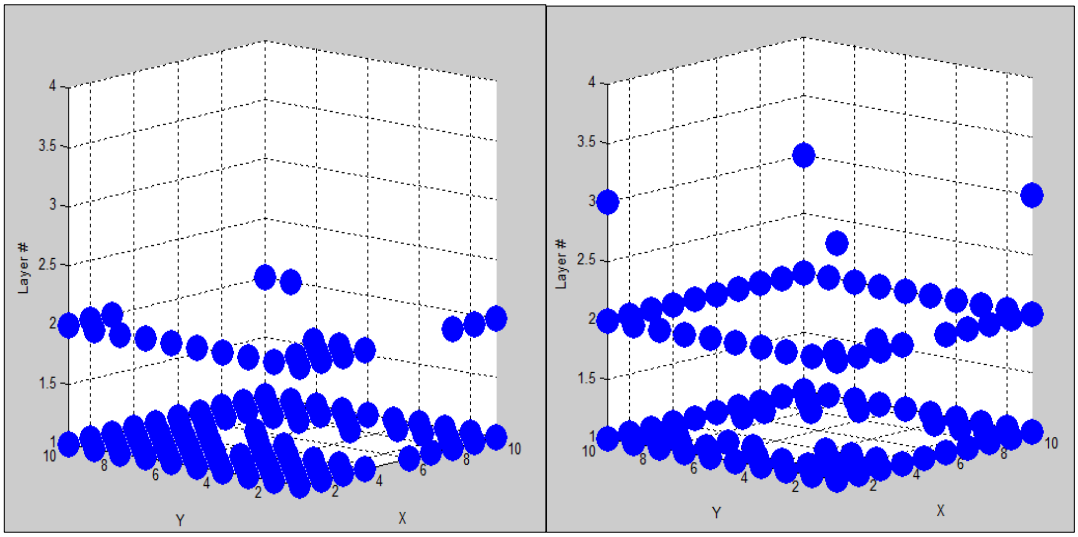
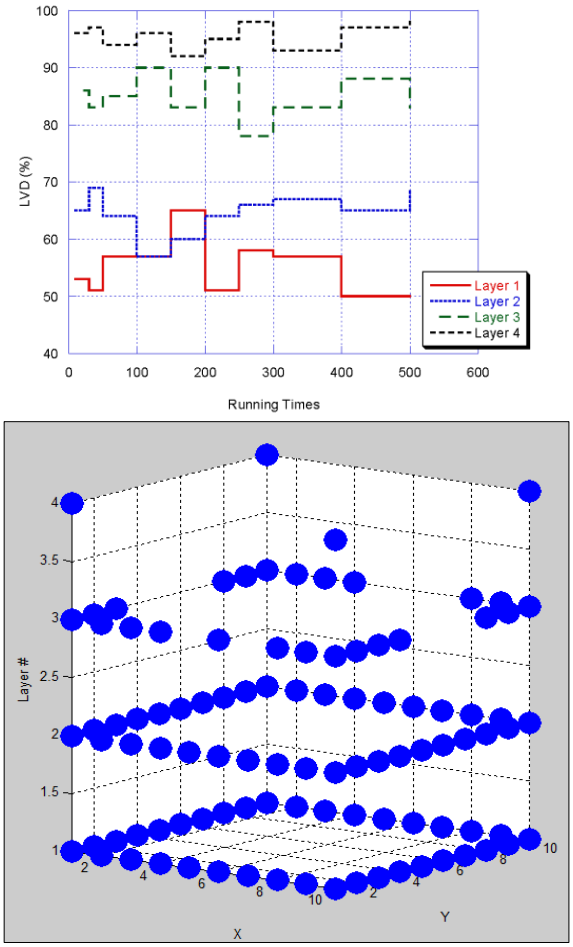


Figure 8 shows the dynamic change of LVD data in each layer for samples 3, 6, and 9. The particle distribution illustrates random migration in each layer due to the periodic voltage operation. The particle distribution did not reach the steady state, although the process ran for long running times. The EPD patterns of these cases are shown in Figure 9**.** The deposition patterns were still influenced by the η values during the deposition process. If η=1 mV, the aggregation happens on sample 3 as shown in Figure 9(a). Furthermore, in high η value (η=10 mV), all particles migrated to the boundary due to the strong repulsive forces among the particles. The vacancies of the EPD patterns appear greatly high using the on-off voltage operation according to the data listed in Table 3. Therefore, the quality of the EPD films cannot be improved by the on-off voltage operating scheme.



(a) (b)



(c)

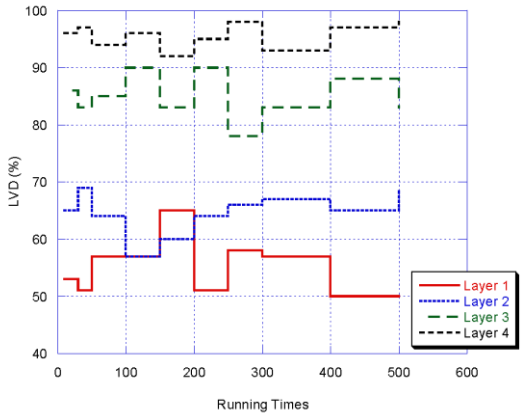
Figure 9. The distribution pattern of particles in each layer of the periodic system during the deposition process, when operated for a longer state of the voltage "on" (200 times) condition, in (a) η = 1 mV, (b) η = 3 mV, and (c) η = 10 mV cases.

The KMC result proved that the on-off voltage operations create a high vacancy in the EPD film. The periodic system cannot remove vacancies in the bottom layer of the deposition structure. How on-off voltage operations utilized to eliminate vacancy from the deposition process. Optimization by applying a constant voltage after on-off voltage operations was offered to decrease the vacancy in the bottom layer.

The deposition process was taken an important part in produce a good properties of film. The best way to produce a good quality of film is to eliminate the defect in the bottom layer. The defect, such as vacancies, is needed to take away from the deposition pattern.

Table 3. LVD data of each deposition layer for on-off operation

Sample	Zeta Potential (mV)	On-off voltage running cases	LVD (%)			
			Layer 1	Layer 2	Layer 3	Layer 4
1	1	(100,100)	58.7	70.3	82.6	89.4
2	1	(100,200)	66.1	71.5	77.4	85
3	1	(200,100)	54.9	64.6	84.9	95.6
4	3	(100,100)	59.8	61.5	82.7	89.9
5	3	(100,200)	66	69.7	80.4	83.8
6	3	(200,100)	55	65.6	83.6	95.7



7	10	(100,100)	65	66.4	79.7	88.7
8	10	(100,200)	69.4	69.8	75.5	85.2
9	10	(200,100)	59.1	63.7	83.3	94.9

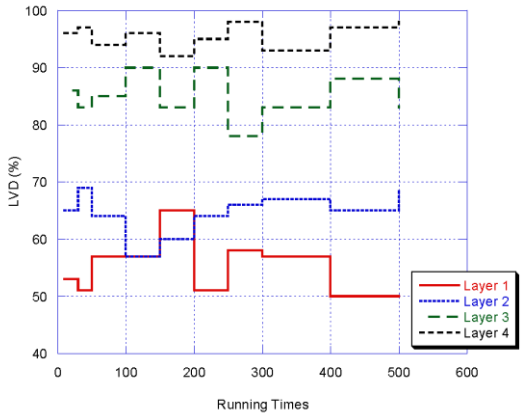
Although three kinds of on-off voltage operations were selected to operate on deposition process, the analysis result shown the LVD data showed larger vacancy in each layer. As seen in Figure 4(a), (b), and (c), the deposition pattern evolved from its beginning state to its final state, and the KMC result shown (listen in **Table 3**), proved vacancy site cannot be decrease when on-off voltage operation is compared with the single influence of electric field (listen in **Table 2**). This suggests that the periodic system cannot remove vacancies in the bottom layer of the deposition structure.

CONCLUSION

The EPD films with little vacancy were estimated under the small η value with the constant voltage operation, as revealed by the KMC simulation results. The particle distribution was greatly affected by the presence of the η value in the solution and the applied voltage in the EPD system. When a small η value was selected in the solution, the particles aggregated together because of the strong der Waals attractive forces among the particles; whereas, the particles scattered separately around the edge of the system boundary due to the strong electrostatic (repulsive) force. The particle distribution in different η values and the voltage applied to the EPD system can further influence the formation of deposition patterns. The great quality of the EPD film can be produced using a small η value with a constant voltage operating approach. However, the large number of vacancies in EPD patterns was generated using the on-off voltage operating scheme. The quality of the EPD films cannot be improved by this scheme for the EPD process due to the formation of a large number of vacancies. The simulation results can be applied to the optimal design and fabrication of photonic crystal films by the EPD approach.

ACKNOWLEDGMENTS

The authors acknowledge the understanding and help in terms of e-resources given by the Universitas Samudra (UNSAM) for the Penelitian Lektor (PDU) grant with contract number 641/UN54.6/PG/2023.



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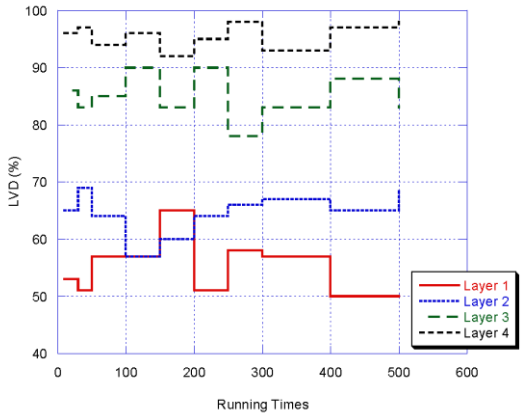

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