Journal of Nano- and Electronic Physics
Vol. **14** No 6, 06016(6pp) (2022)

Журнал нано- та електронної фізики
Том **14** № 6, 06016(6cc) (2022)

Numerical Modeling of Thin-Film Growth by Random Deposition with Particle Evaporation

A. Saoudi[1,2,*], S. Boulahrouz[1,3], S. Fares[4], M. Chitour[1], K. Mansouri[1,2], L. Aissani[5], A. Abboudi[1,2]

[1] *Mechanical Engineering Department, Abbes Laghrour-Khenchela University, PO Box 1252, 40004 Khenchela, Algeria*
[2] *Laboratory of Engineering and Sciences of Advanced Materials (ISMA), Abbes Laghrour University, 40004 Khenchela, Algeria*
[3] *Electromechanical Engineering Laboratory, 23000 Annaba, Algeria*
[4] *Laboratoire Microstructures et Défauts Dans les Matériaux, Univeristé Frères Mentouri Constantine 1 Route Ain El Bey, 25017 Constantine, Algeria*
[5] *Matter Science Department, Abbes Laghrour-Khenchela University, PO Box 1252, 40004 Khenchela, Algeria*

(Received 12 August 2022; revised manuscript received 22 December 2022; published online 27 December 2022)

In the present work, we provide a generalization of particle deposition to enhance the physics of the simulation process and make it more similar to real deposition processes, such as particle evaporation, which is tuned to attract vapor and gaseous particles by reducing air pressure and crowding of other air molecules. This not only reduces the energy required for evaporation but also allows for a more direct path to the area of deposition, as the vapor particles are not as frequently redirected by other particles within the chamber. Although we do not deal with bombardment in our approach, we provide a method to generate clusters of random shape and size, ranging from a single particle to a collection of particles, in order to make the simulation more representative of experimental reality. According to the results obtained from our study, interface growth in random vapor deposition follows two distinct regimes (the first clusters are grown randomly by building an interface which has grown as a result of deposition or evaporation of particles due to the difference between the average chemical vapor potential U_v and interface U_i). Growth (β) and roughness (α) exponents were stable with increasing substrate size (L) and a number of bombarded particles (N). These exponents are sensitive to the variation of U_i, where α decreases as U_i changes from 0 to − 6 inversely to the exponent β. All the surfaces obtained by this model have fractal properties. In addition, the technique of Greenwood and Williamson which consists in replacing the rough-rough contact by a rough-smooth contact is geometrically valid at the level of the interstices and less valid with respect to a thermal problem according to the roughness of interfaces of the surfaces in contact.

Keywords: Growth surface, Evaporation, Roughness, Scaling behavior, Fractal dimension, Interstices.

DOI: 10.21272/jnep.14(6).06016 PACS numbers: 62.20.Qp, 68.60.Dv

1. INTRODUCTION

The investigation of the morphology, structure, and other physical and chemical aspects of developing interfaces requires a perfect knowledge of non-equilibrium statistical physics [1]. Problems arose when the Hamiltonian formulation was impractical, or the detailed-balance requirement needed to be met. Surface growth and interface research are one of these issues. Understanding the fundamental principles of growth is essential for the fabrication of thin-film devices with crucial technological applications in academics and industry [2]. Growing interfaces can be found in a wide range of significant physical, chemical systems and processes.

When surface processes are controlled, new devices with real-world applications can be created. Thin-film and interface growth can be studied effectively with the use of theoretical and computational models, thanks to the fact that conventional statistical physics techniques can be used to explain these out-of-equilibrium processes. Developments in recent years have allowed a deeper comprehension of the fundamental phenomena that govern the deposition of particles forming a thin film at the nanoscale. Different types of particles have been used extensively in atomistic models in this area of research [3, 4]. Despite their apparent simplicity, numerous models give a solid foundation from which to investigate more complex systems inherent to experimental growth processes and methods. Lately, a great deal of attention has been focused on the dynamic roughening of surface growth in discrete models and stochastic Langevin equations [5]. Numerous statistical models, including random deposition, ballistic deposition, the Eden model, Cluster statistics models, Terrace statistics, and the solid-on-solid model, have been developed to explain the surface growth phenomenon [6]. Random deposition (RD) is the most straightforward process for surface growth, and its parameters can be easily estimated [7]. In this model, each particle is dropped randomly onto a flat surface and added to the top of the chosen column. Both the expansion of columns and surface fluctuations are independent of one another. Also, a particle is launched from a random location above the surface, further away than the maximum height of the interface. There is a correlation along the surface as the heights of the columns at each point influence one another. This means that the height of a new column will be comparable to or greater than the heights of the surrounding columns. The correlation length quantifies the average range over which two heights can know about one an-other. So, surface fluctuations rise over time, eventually reaching a maximum as the correlation

* saoudi_abdenour@univ-khenchela.dz

 © 2022 Sumy State University

length approaches the size of the system. Ghosh et al. [8] investigated the growth of thin films and their residual sur-face morphology in terms of the fractal parameters for random and ballistic deposition models.

Thin films are typically fabricated using vacuum based processes, including molecular beam epitaxy, sputtering, and evaporation. The process of evaporation involves the employment of several technologies to evaporate larger bits of the material in a vacuum chamber, ultimately leaving behind a uniformly thin coating. The most common method of producing a thin film involves heating the target material to the point of evaporation and then allowing it to condense on the substrate. Gedda et al. [9] investigated the crystalline structure and surface morphology, and kinetic roughening of Ag films using X-ray diffraction (XRD) and atomic force microscopy (AFM). It is revealed that surface roughness increases as film thickness increases. The Monte Carlo (MC) approach is one of the many methods that have been proposed to model the formation of thin films by random deposition. This technique is one of the most commonly used methods since it can be applied to systems that contain a significant number of atoms [10]. Tung et al. [11] developed a framework using kinetic Monte Carlo simulations to connect morphological properties and microscopic dynamics during island growth, coalescence, and formation of continuous heteroepitaxial films.

In the present work, the evaporation effect during the growth of a thin film obtained by random deposition has been studied, where the growth of the interface is dependent on the difference between the chemical vapor potentials U_v and interface U_i. Furthermore, we are interested in studying the morphology of surfaces generated by adding such randomly shaped clusters to an initially flat linear substrate. Finally, we study the validity of the technique of Greenwood and Williamson which consists in replacing a rough-rough contact by a rough-smooth contact.

2. GROWTH MODELING WITH EVAPORATION AND DYNAMIC SCALING

Fig. 1 represents the surface growth based on the random deposition model with evaporation, as reported by Kim [12]. The interface dynamics is initially characterized by a randomly selected site (i). The height of each site (i) is defined by the equation for the local chemical potential at the interface $U_i = 2h_i - h_i + 1 - h_i - 1$, which is then compared with that predetermined mean chemical vapor potential U_v. The growth of the surface interface is defined by the curvature between the three neighboring sites with the following probabilities.

- First, all particles were deposited randomly on a substrate of length (L) which contains a number of sites $i \in [1, \ldots, L]$, where the periodicity between the ends is preserved ($L + 1 \equiv 0$ and $0 \equiv L$).
- If $U_i < U_v$, the height of the site (i) increases by receiving a particle (deposit) and becomes $H_i = h_i + 1$.
- If $U_i > U_v$, the height of the site (i) decreases, i.e., a particle is removed as $H_i = h_i - 1$ (evaporation).
- For $U_i = U_v$, the height of site (i) increases or decreases with equal probability (deposition or evaporation).

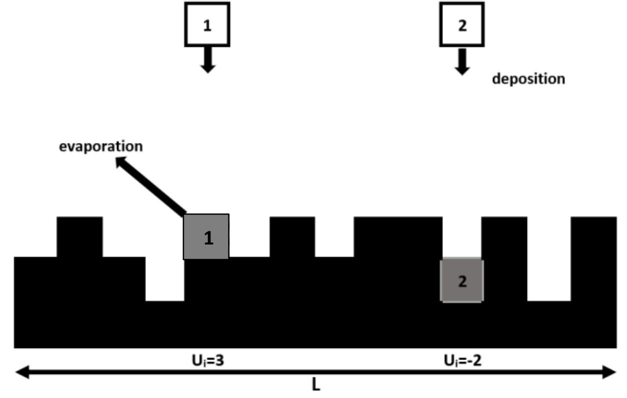

Fig. 1 – Illustration of interface growth conditions in a random vapor deposition model with $U_v = 1$ [12]

Ladder theory has been used to analyze the stochastic dynamics of fractal surfaces as a function of height standard deviation time [13]:

$$W(L,t) = \sqrt{\frac{1}{L}\sum_{i=1}^{L}\left[h(i,t) - \bar{h}(t)\right]^2}\,, \quad (1)$$

where W is the interface roughness, L is the size of the substrate, $h(i, t)$ is the height of the site surface (i) at time (t), $\bar{h}(t)$ is its average height which is calculated by the following equation:

$$\bar{h}(t) = \frac{1}{L}\sum_{i=1}^{L} h(i,t)\,. \quad (2)$$

For a general surface, the roughness increases as a power of time up to a time t_x, sometimes called saturation time:

$$W(L,t) \sim t^{\beta}, \left[t \ll t_x\right]. \quad (3)$$

The exponent β is called the growth exponent which describes the time-dependent roughness dynamics. Once the transit time is reached, the roughness is saturated giving the saturation value W_{sat}. The saturation value increases with increasing the substrate size (L), and the dependence also follows a power law:

$$W_{sat}(L) \sim t^{\alpha}, \left[t \gg t_x\right], \quad (4)$$

where α is the roughness exponent that describes the roughness after system saturation. The crossing time also depends on a power law:

$$t_x \sim L^z\,, \quad (5)$$

where z is the dynamic exponent [14]. The exponents are also related to each other, and the dynamic exponent is obtained as following:

$$z = \frac{\alpha}{\beta}\,. \quad (6)$$

This relationship between exhibitors is based on any growth process. The exponents α, β and z characterize the growth of the models and their self-affine structures and morphologies.

Height-height-surface correlation, it is important to include the spatiotemporal correlation function (C) by defining the height difference between two interface locations separated by a distance (x) at a deposition time (t) [15]:

$$C(x,t) = \left\langle \left(h(x_0 + x, t) - h(x_0, t) \right)^2 \right\rangle_{x_0} . \qquad (7)$$

3. RESULTS AND DISCUSSION

3.1 Morphology of Surfaces Produced

The growth in the model of random vapor deposition is completely different with those of deposition with diffusion or germination [16, 17], because every instant (t) particle is deposited or evaporated on the substrate due to the difference between the chemical vapor potentials U_v and interface U_i. The number of deposited particles (N) in the substrate remains constant for different lengths (L) of the substrate, just the substrate size that has been grown (Fig. 2a). However, the variation of the chemical vapor potential U_v is influenced by the number of particles deposited, for $U_v = 0$, the evaporated particles are larger than with those of the deposited particles and the latter take a maximum for $U_v = -6$ (Fig. 2b).

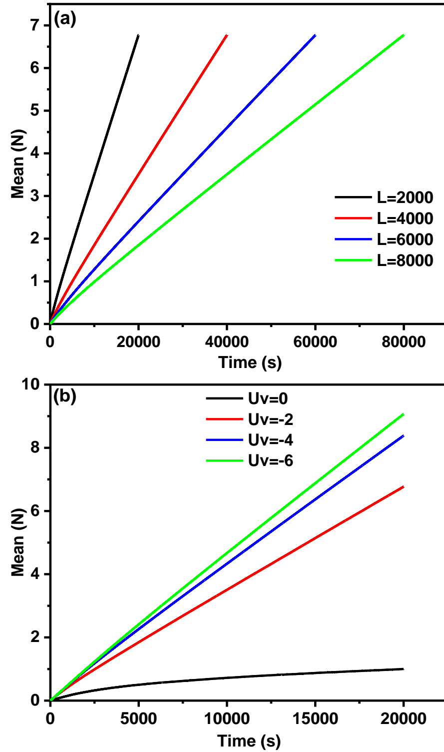

Fig. 2 – Average of the particles (N) deposited on the substrate for a) different sizes of the substrate (L), b) different chemical vapor potential U_v

Fig. 3 shows the surfaces produced for different chemical potentials U_v by fixing the length of the substrate $L = 2000$ and the number of bombarded particles $N = 20000$. From the obtained surfaces, we noticed that deposition and evaporation processes are dependent on the chemical potential U_v, where the evaporation process is dominant with $U_v = 0$ producing an incomplete surface in the islands shape. Furthermore, for $U_v = -6$, the deposition process is majority by building a rough surface of higher height.

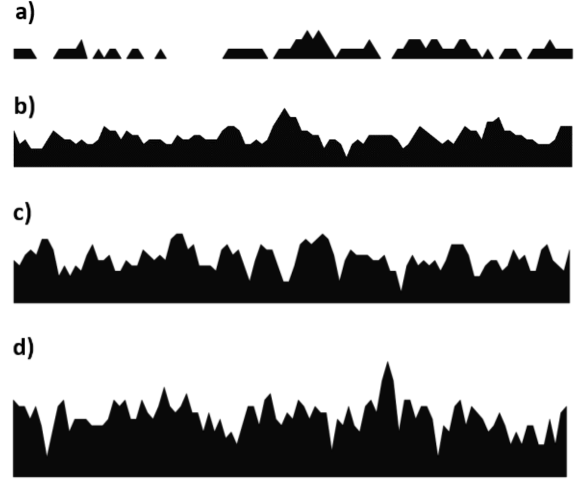

Fig. 3 – Morphology of the surfaces produced for a substrate length $L = 100$ and a number of particles $N = 1000$ for a) $U_v = 0$, b) $U_v = -2$, c) $U_v = -4$, d) $U_v = -6$

3.2 Scaling Behavior

The initial surface growth mode in random vapor deposition is close to random growth. Indeed, the particles do not change randomly occupied sites (transient autonomous regime). Subsequently, we witness a saturation regime due to the effect of the difference between the average chemical vapor potential U_v and the inter-

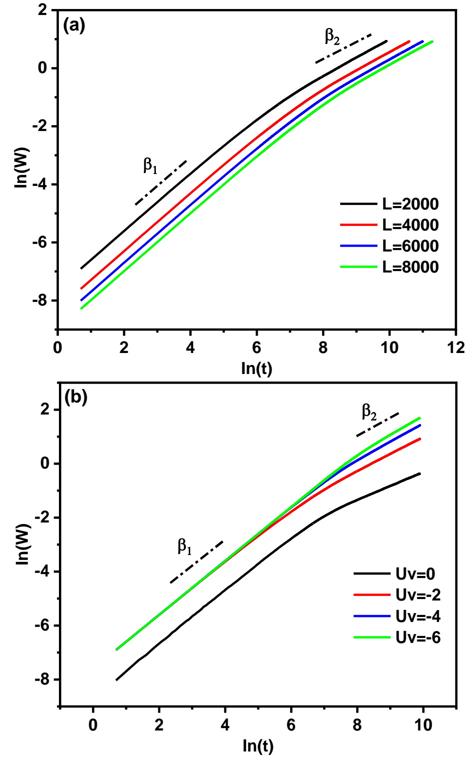

Fig. 4 – ln(W) as a function of ln(t) showing the growth regime for: a) different sizes of substrate (L), b) different chemical potentials of the vapor (U_v)

face U_i, where the particle follows two probabilities, either deposition or evaporation. We find that variation in the substrate size (L) has no impact on the surface growth regime; it is clearly observable that the growth system is increased for larger (L) (Fig. 4a).

However, the interface growth clearly influenced by the chemical potential value U_v, where particle deposition is dominant for $U_v = -6$. When $U_v = 0$, the majority of particles have been evaporated (Fig. 4b). The surface growth in this model is made through two distinct regimes (random regime followed by a saturation regime).

At the beginning of the deposition, the growth is random, which is characterized by the growth exponent β_1. Over time, growth is influenced by chemical vapor potential U_v by saturating the growth regime, which is determined by the growth exponent β_2. Fig. 4 shows the growth regimes of thin layers obtained by random vapor deposition, the variation of the size of the substrate (L) does not modify the growth regime.

Initially, a regime of random tendency with $\beta_1 \approx 0.50$, followed by a saturation regime slightly influenced by the value of U_v, where $\beta_2 = 0.3146$ for $U_v = 0$ and $\beta_2 = 0.3220$ at $U_v = -6$. In addition, the modification of the value of the chemical vapor potential U_v acted on the evolution of growth after saturation, at the beginning of a random regime $\beta_1 \approx 0.50$, followed by a saturation regime with the deposition effect and evaporation of the particles, the growth exponent is small $\beta_2 = 0.2577$ for $U_v = 0$ because the evaporation mechanism is predominant and reaches a value of $\beta_2 = 0.3579$ at $U_v = -6$.

Table 1 – Growth exponent values β for different substrate size (L) with $Uv = -2$

L	β_1	B_2
2000	0.4993	0.3146
4000	0.4990	0.3166
6000	0.4992	0.3207
8000	0.4991	0.3120

Table 2 – Growth exponent values β for different chemical vapor potentials (U_v) with $L = 2000$

U_v	β_1	B_2
0	0.4994	0.2577
– 2	0.4990	0.3166
– 4	0.4995	0.3413
– 6	0.4992	0.3579

The correlation was calculated by equation (7), where $x = L/2$, h is the surface height in the sites different values of (x) $\in [1, \ldots, L]$ for a given time (t). The results show that surface growth in the simple vapor deposition model is correlated. Furthermore, the correlation of heights between neighboring sites is independent of the substrate size (L) (Fig. 5a). Moreover, an interesting dependence of the correlation (C) on the value of the average chemical vapor potential U_v were observed (Fig. 5b).

The roughness exponent α characterizes the roughness of the interface after growth saturation, and the structures obtained are similar at all scales exhibiting fractal properties whose fractal dimension D_f is calculated by $D_f = d - \alpha$, where $d = 2$ [18]. From the results presented in Table 3 and Table 4, we find that the exponent $\alpha \approx 0.32$ and $z \approx 0.65$ remain stable by increasing the size of the substrate (L). The modification of the value of the average chemical vapor potential U_v has a direct effect on the roughness, which expresses a decrease in α, and z for U_v varies from 0 to -6. Furthermore, the fractal dimension D_f has been increased from 1.6299 to 1.7517.

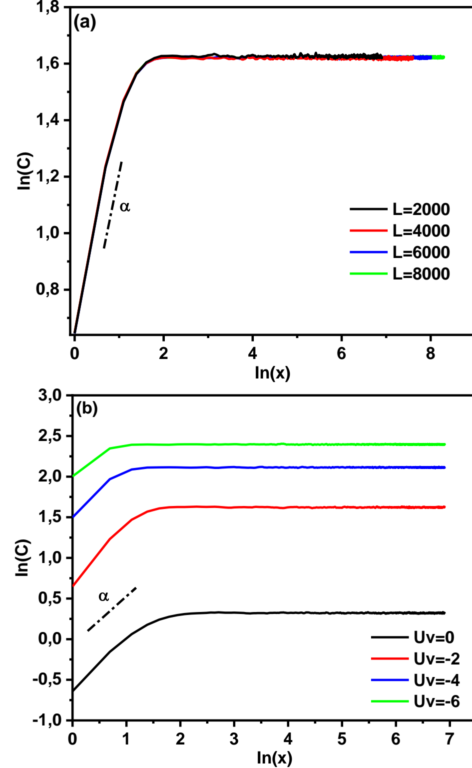

Fig. 5 – $\ln(C)$ vs $\ln(x)$ showing the correlation between two sites for: a) different sizes of substrate (L), b) different chemical potentials of vapor (U_v)

Table 3 – Values of roughness α, dynamic exponent z and fractal dimension D_f for different substrate size (L) with $U_v = -2$

L	α	z	D_f
2000	0.3256	0.6521	1.6744
4000	0.3232	0.6476	1.6768
6000	0.3249	0.6508	1.6751
8000	0.3240	0.6491	1.6760

Table 4 – Values of roughness α, dynamic exponent z and fractal dimension D_f for different chemical vapor potential (U_v) with $L = 2000$

U_v	α	z	D_f
0	0.3701	0.7410	1.6299
−2	0.3256	0.6525	1.6744
−4	0.2828	0.5661	1.7172
−6	0.2483	0.4973	1.7517

3.3 Rough-Rough to Rough-Smooth Contact

Among the research studies that have studied a contact between rough surfaces, we cite the technique of Greenwood and Williamson [19], which consists in comparing the sum of two rough surfaces in contact with a perfectly smooth surface, rather than considering the total problem between two rough surfaces in contact. To

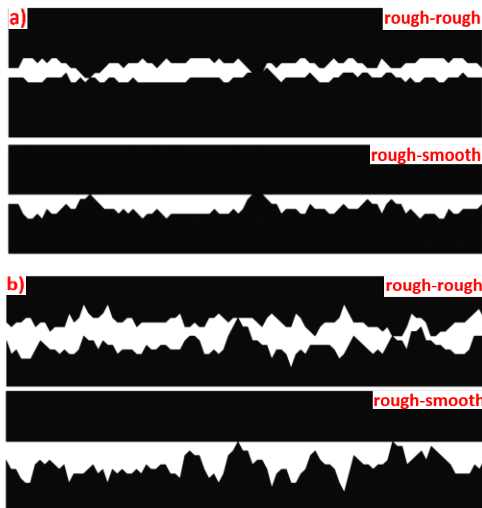

Fig. 6 – Rough-rough contact changed to rough-smooth for: a) $U_v = 0$, b) $U_v = -6$ with $L = 100$ and $N = 1000$

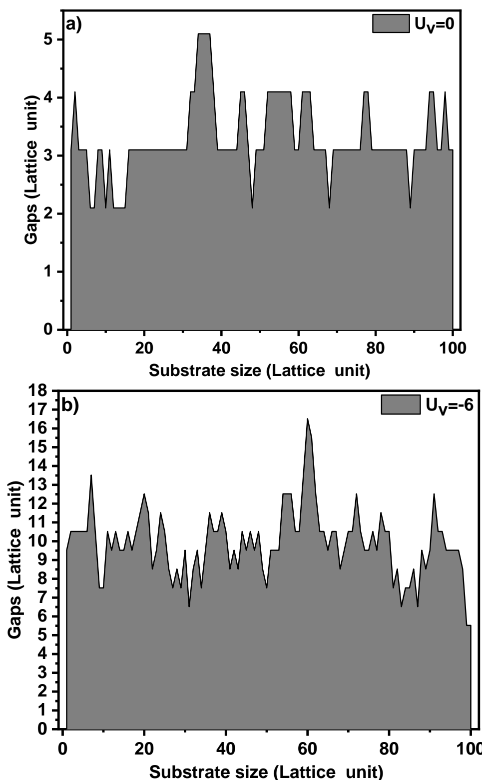

Fig. 7 – The gaps between rough-rough and rough-smooth contact for a) $U_v = 0$, b) $U_v = -6$, with $L = 100$ and $N = 1000$

prove the validity of this technique, we used the surfaces produced by the random vapor deposition model. We grow two assemblies, the first has a rough-rough contact between the surfaces *A* and *B*. The second assembly presents a rough-smooth contact between the sum of two rough surfaces ($G = A + B$) in contact with a smooth surface (*S*). The gaps between the bonded rough-rough and rough-smooth surfaces are calculated using I_1 = mean $(A - B)$ and I_2 = mean $(G - S)$. From the results shown in Fig. 7a, b, we find that the gaps are equal between rough-rough and rough-smooth assemblies for different chemical vapor potentials (U_v), which proves the geometric validity of the Greenwood and Williamson technique.

However, according to the work of Saoudi et al. [20], the validity of this technique does not resist a thermal problem, because for the self-affine geometry, the roughness makes this assumption less and less valid. In the opposite limit, using this technique on flat surfaces may be acceptable. Therefore, in general, the modeler must take into account the full complexity of the geometry.

4. CONCLUSIONS

In the present work, we studied the evolution of surface growth in random vapor deposition by varying the substrate size (*L*) and the chemical vapor potential (U_v). The surfaces produced by this model were used to construct two assemblies, rough-rough and rough-smooth, in order to analyze the validity of the technique of Greenwood and Williamson, which consists in replacing a contact between two rough surfaces by the sum of these surfaces in contact with a completely smooth surface. Furthermore, we have concluded the following.

1. The number of particles deposited or evaporated depends on the difference between the chemical vapor potential U_v and interface U_i.
2. For $U_v = 0$, the evaporated particles are greater than those deposited on the substrate, conversely for $U_v = -6$.
3. Surface growth follows two different regimes, initially an autonomous and transient random regime followed by a saturation regime.
4. The variation of the chemical vapor potential (U_v) has a strong effect on the exponents α, β and *z*.
5. The surfaces obtained are similar at all scales presenting fractal properties.
6. Greenwood and Williamson's technique is geometrically valid at interstices by replacing a rough-rough contact by rough-smooth, but it is less and less valid in a thermal problem for rough interfaces.

REFERENCES

1. J.W. Evans, P.A. Thiel, M.C. Bartelt, *Surf. Sci. Rep.* **61** No 1-2, 1 (2006).
2. F.L. Forgerini, W. Figueiredo, *phys. status solidi c* **8**, 3119 (2011).
3. Z. Ebrahiminejad, S.F. Masoudi, R.S. Dariani, S.S. Jahromi, *J. Chem. Phys.* **137** No 15, 154703 (2012).
4. F.L. Forgerini, W. Figueiredo, *Phys. Rev. E* **81** No 5, 051603 (2010).
5. M. Nasehnejad, M.G. Shahraki, G. Nabiyouni, *Appl. Surf. Sci.* **389**, 735 (2016).
6. S. Hosseinabadi, A.A. Masoudi, M.S. Movahed, *Physica B* **405**, 2072 (2010).
7. P. Meakin, Fractals, *Scaling and Growth Far from Equilibrium* (Cambridge University Press: New York: 1998).
8. K. Ghosh, R.K. Pandey, *AIP Conf. Proc.* **2265** No 1, 030508 (2020).
9. M. Gedda, N.V.V. Subbarao, D.K. Goswami, *Langmuir* **30**, 8735 (2014).
10. Y. Li, N. Vu, A.S. Kim, *Desalination* **249**, 416 (2009).
11. B.T.T. Tung, R. Almeida, S.O. Ferreira, F.D.A.A. Reis, *Appl. Surf. Sci.* **560**, 149946 (2021).
12. Y. Kim, *Prog. Theor. Phys.* **104** No 3, 495 (2000).

13. W. Kwak, J.M. Kim, *Physica A* **520**, 87 (2019).
14. A. Kolakowska, M.A. Novotny, P.S. Verma, *Phys. Rev. E* **73**, 011603 (2006).
15. D.A. Mirabella, C.M. Aldao, *Surf. Sci.* **646**, 282 (2016).
16. A. Saoudi, L. Aissani, S. Boulahrouz, L. Radjehi, O. Chahaoui, M. Mebarki, H. Djebaili, *J. Nano- Electron. Phys.* **14** No 5, 05021 (2022).
17. A. Saoudi, L. Aissani, G. Sorba, F. Chinesta, *Int. J. Comp. Mat. Sci. Eng.* **09**, 2050022 (2020).
18. K. Ghosh, R.K. Pandey, *AIP Conf. Proc.* **2115** No 1, 030280 (2019).
19. J. Greenwood, J. Williamson, *Proc. R. Soc. Appl.* **295** No 1442, 300 (1966).
20. A. Saoudi, A. Leon, G. Gregoire, A. Barasinski, H. Djebaili, F. Chinesta, *Surf. Topogr.: Metrol. Prop.* **5** No 4, 045010 (2017).

Чисельне моделювання росту тонких плівок шляхом випадкового осадження з випаровуванням частинок

A. Saoudi[1,2], S. Boulahrouz[1,3], S. Fares[4], M. Chitour[1], K. Mansouri[1,2], L. Aissani[5], A. Abboudi[1,2]

[1] *Mechanical Engineering Department, Abbes Laghrour-Khenchela University, PO Box 1252, 40004 Khenchela, Algeria*

[2] *Laboratory of Engineering and Sciences of Advanced Materials (ISMA), Abbes Laghrour University, 40004 Khenchela, Algeria*

[3] *Electromechanical Engineering Laboratory, 23000 Annaba, Algeria*

[4] *Laboratoire Microstructures et Défauts Dans les Matériaux, Univeristé Frères Mentouri Constantine 1 Route Ain El Bey, 25017 Constantine, Algeria*

[5] *Matter Science Department, Abbes Laghrour-Khenchela University, PO Box 1252, 40004 Khenchela, Algeria*

Робота містить узагальнення осадження частинок для покращення фізики процесу моделювання та для того, щоб зробити його більш схожим на реальні процеси осадження, такі як випаровування частинок, яке полягає у притяганні пароподібних і газоподібних частинок шляхом зменшення тиску повітря та скупчення інших молекул повітря. Це не тільки зменшує енергію, необхідну для випаровування, але також забезпечує більш прямий шлях до області осадження, оскільки частинки пари не так часто перенаправляються іншими частинками всередині камери. Хоча ми не маємо справу з бомбардуванням у нашому підході, ми пропонуємо метод генерації кластерів випадкової форми та розміру, починаючи від окремої частинки до набору частинок, щоб зробити моделювання більш репрезентативним для експериментальної реальності. Згідно з результатами, отриманими в нашому дослідженні, зростання інтерфейсу при випадковому осадженні з парової фази відбувається за двома різними режимами (перші кластери вирощуються випадковим чином шляхом створення інтерфейсу, який виріс в результаті осадження або випаровування частинок через різницю між середнім хімічним потенціалом пари U_v і потенціалом інтерфейсу U_i). Експоненти росту (β) і шорсткості (α) були стабільними зі збільшенням розміру підкладки (L) та кількості частинок (N), що бомбардуються. Ці експоненти чутливі до зміни U_i, де α зменшується, коли U_i змінюється від 0 до − 6, обернено до експоненти β. Усі поверхні, отримані в рамках цієї моделі, мають фрактальні властивості. Крім того, техніка Грінвуда та Вільямсона, яка полягає в заміні шорстко-шорсткого контакту шорстко-гладким, геометрично коректна на рівні міжвузлів і менш коректна по відношенню до теплової задачі відповідно по шорсткості інтерфейсів контактних поверхонь.

Ключові слова: Поверхня росту, Випаровування, Шорсткість, Поведінка масштабування, Фрактальна розмірність, Міжвузля.